Please note. Authors' responses are bold-faced. Authors' responses follow immediately below the editor's and reviewers' comments.

Editor Decision: Reconsider after major revisions (14 Nov 2015) by Prof. Mauro Giudici

Comments to the Author:

The reviewers expressed three positive assessments of the paper and two of them provided also a quite detailed list of modifications to be introduced in the paper before it can be accepted for publication.

I recommend the authors to properly account for the reviewers' comments in the revised text, above all with reference to the following general remarks:

1) improve the readability and the description of the mathematical development, possibly including some parts in an appendix;

2) provide more details about the numerical aspects for the pratical application of the proposed solutions;

3) fully describe the physical hypotheses on which the equations are based and discuss their relevance for practical applications and thier limitations.

Response:

Thanks for the remarks from the editor. We have improved the readability and elaborated on the description of the mathematical development. Moreover, we provide more details about the numerical aspects for the practical application of the proposed solutions. We also clearly and fully explain the physical hypotheses embedded in the governing equation used in the manuscript and discuss the relevance of the developed model for practical applications and their limitations. We have carefully addressed the comments from reviewers #1, #2 and #3 on a point-by-point basis in the revised manuscript.

Anonymous Referee #1

The authors derived an analytical solution of 2D transport coupled with first-order chain reactions and species-specific retardation factors. The derived solution is an advance over Sun et al. (1999) for considering species-specific retardation and over Srinivasan and Clement (2008) for higher-dimensional transport. Three examples are solid and convincing. Except for the 10-species chain used Srinivasan and Clement (2008), 4n+2 series and PCE-TCE-DCE-VC decay networks may not be consecutive. Authors may acknowledge analytical solution development in the review for branching and converging decay networks and state the limitation of this solution in the conclusion. I had hard time getting (A34). If there is no page limit, it will helpful to add the substitution.

Response:

Our manuscript develops a novel analytical model that considers different species-specific retardation factors for describing two-dimensional multispecies reactive transport coupled by a series of first-order chain reactions. Three example applications are considered to demonstrate the wide applicability of the derived parsimonious analytical model. We understand and fully agree the comments that some of the decay networks may not be consecutive. We appreciate the constructive suggestion that add acknowledge on analytical solution development for branching and converging decay networks and state the limitation of our solution. Thus, we have included the review of the analytical solution development for branching and converging decay network in the introduction and clearly indicate the limitation of our solution in the conclusion in the revised manuscript. Besides, we elaborate on the detailed mathematical manipulations and procedures to obtain the Eq. (34) in the revised manuscript for better readership.

Anonymous Referee #2

This is a nice piece of work advancing the multispecies plume (2D) migration from an analytical standpoint. The literature review is almost complete and through, the mathematical model is based on a technique developed by the same author (Dr. Chen) in 2012, but with substantially new materials and a physically based boundary condition (third-type or Rubin type) and extension to 2D. The solutions have been compared with carefully designed and proved numerical solutions. The examples used in the paper are relevant to actual applications and the details of all the derivation and programming are nicely documented. The figures are also well presented. The paper is well written and easy to follow.

The following revisions are necessary to improve the quality of the paper.

1. I think the title has to be changed. First of all, the word "parsimonious" should be deleted (as it is not parsimonious to me at all). Also, the author may want to add "two-dimensional" in the title as the problem investigated is 2D in nature.

Response:

Thanks for the constructive comment. This study present a novel analytical model with a parsimonious mathematical expression for describing multispecies plume migration. The concentration of arbitrary species can be directly evaluated from the unique mathematical expression. The parsimonious mathematical structures of the analytical are easy to code into a computer program for implementing the solution computations for arbitrary target species. This is quite different from previous analytical models in literature that generally used a distinct mathematical expression for distinct species of a decay chain. Thus, we think the word "parsimonious" can reflect the mathematical expression of our compact solution. The title is thus changed to as suggested "A parsimonious analytical model for simulating two-dimensional

multispecies plume migration".

2. I also think the use of "verified" or "verification" is inappropriate. A numerical solution cannot be used to verify an analytical solution per se, as it itself may involve the potential (and sometimes hidden) numerical errors. I think a better word is "compared" or "comparison" instead.

<u>Response</u>:

Thanks for the constructive comment. We fully agree that the analytical models are used to verify the numerical model. Thus, we have replaced "verified" or "verification" with "compared" or comparison.

3. Despite the fact that the authors have done a careful review of the previous studies.
Some important references are still missing. For instance, the paper of "Mieles, J., and
Zhan, H., Analytical solutions of one-dimensional multispecies reactive transport in a
permeable reactive barrier-aquifer system, Journal of Contaminant Hydrology, 134-135, 54-68, 2012. doi: 10.1016/j.jconhyd.2012.04.002" is closely related to this study and is a reference that should be included. The study of Mieles and Zhan (2002) dealt with the multispecies transport in a permeable reactive barrier (PRB)-aquifer system, with similar use of the third-type or mixed type boundary condition and other boundary conditions and the technique of Laplace transform.

<u>Response</u>:

Thanks for the valuable comment. Indeed, these are very important references. We have included these references in the revised manuscript.

4. In equations (13)-(15), there are a number of parameters introduced without explanation. Although the authors explained them in the Appendix, I still think it is necessary to explain a few key parameters

in the main text. For instance, the and terms, et al. Otherwise, it is difficult to follow the mathematics.

Response:

Thanks for the helpful suggestion. We have these parameters explained in the main text for better readership.

5. In section 3.3, the author mentioned three verifications at the first sentence, but then only discussed two cases in the first and second paragraphs. The third case is only mentioned from the third paragraph. It should be revised. I suggest moving the first sentence of the third paragraph “The third verification example is : : :.” to the first paragraph of section 3.3.

Response:

Thanks for the constructive comment. The first sentence of the third paragraph of section 3.3 is move to the first paragraph of section 3.3.

6. For the FORTRAN program, what type of FORTRAN program? (FORTRAN 95?). Also, since the summations terms involved (M and N) are so large for some cases, how long is it going to take for the program to generate the result? (CPU time? PC or Workstation?) This type of information should be mentioned for the application of the method.

Response:

The computer code is written in FORTRAN 90 language with double precision. The computation is not time-consuming. The computational time for evaluation of the solutions at 50 different observations only takes 3.782s, 11.325s, 23.95s and 67.23s computer clock time on an Intel Core i7-2600 3.40 MHz PC for species 1, 2, 3, and 4 in the comparison of example 1. We have added the discussions on the computational time in the revised manuscript.

In summary, I recommend a moderate revision.

(note: some special symbols are missing in this plain text version of the review)

Anonymous Referee #3

This manuscript summarizes a new analytical model that simulates the reactive transport of multiple interacting species in a 2D groundwater flow system. The authors describe the model (with derivations in the appendices), and then provide several examples showing model output, comparison with a numerical model, and a short sensitivity analysis to identify influential transport parameters. Overall, the manuscript is organized well and covers an important topic. However, before recommending publication the following points must be addressed:

- One of the main concerns is the lack of connection with real-world systems. The authors compare their model with other models, but the actual behavior of the chemical species (particularly the sequential first-order decay reactions) in actual aquifer systems is not discussed, nor is it discussed in the Methods, Results, or Discussion sections. Without this connection, it is difficult for the reader to have confidence that modeling results (and the model itself) can be useful if applied to real-world systems.

Response:

Analytical multispecies models are widely used to evaluate natural attenuation of plumes at chlorinated solvent sites. A study of 45 chlorinated solvent sites by McGuire et al. (2014) found that mathematical models were used at 60% of these sites and that the public domain model BIOCHLOR (Aziz et al., 2000) provided by the Center for Subsurface Modeling Support (CSMoS) of USEPA was the most commonly used model. The utility of the BIOCHLOR model to the real-world problems has been demonstrated by an example application that it can

reproduce plume movement from 1965 to 1998 at the contaminated site of Cape Canaveral Air Station, Florida. The illustrative example of the developed analytical solution in our study considered the example reported in the BIOCHLOR. BIOCHLOR model uses analytical solutions to a set of advection-dispersion equations coupled by sequential first-order decay reactions. The BIOCHLOR analytical solution is valid for the case of having identical retardation coefficients for all species. The same equations were considered in our study to develop new analytical solutions. Our new solutions can consider the case that each species has its own retardation coefficients. Thus, we assure that our analytical solutions have more practical applications than the BIOCHLOR model to the real-world system.

References:

McGuire, T. M., Newell, C. J., Looney, B. B., Vangeas, K. M., Sink, C. H., 2004: Historical analysis of monitored natural attenuation: A survey of 191 chlorinated solvent site and 45 solvent plumes. Remiat. J. 15: 99-122.

Aziz, C. E., Newell, C. J., Gonzales, J. R., Haas, P., Clement, T. P., Sun, Y., 2000: BIOCHLOR–Natural attenuation decision support system v1.0, User's Manual, US EPA Report, EPA 600/R-00/008, EPA Center for Subsurface Modeling Support (CSMOS), Ada, Oklahoma.

- In relation to the previous comment, the authors need to discuss limitations of their model. For example, I assume that the flow field used in the analytical model is steady state, and that sources and sinks within the groundwater system are ignored. When do these conditions actually occur? Under what field conditions can the model actually be applied? Again, without relating the model to reality, much of this is ignored by the authors.

Response:

All models have their limitations because they used physically-based mathematical equations to describe the transport processes in the subsurface system. The appropriateness of model depends on if transport behavior follows the basic assumptions of the physically-based mathematical equation. The analytical model in our study considers a steady uniform flow field and a boundary source. Thus, our model can be applied to a field site that has a steady uniform flow field and the contaminant source can be treated as a boundary source. Analytical model considers the same flow and source condition such as the BIOCHLOR model are widely used to assess many real-world problems. Detailed contaminated site applications were illustrated in the BIOCHLOR User's manual. We have elaborated a discussion on limitation of our analytical model in the revised manuscript for better readership.

- The use of the model requires a number of complicated numerical methods (correct?). So, at what point does the analytical solution actually become a numerical solution? Also, the authors never report the run-time of the model simulations in comparison with those of the numerical model (LTFD). Due to the complicated nature of the analytical model, I would assume that the run-times are substantial. Without this reported, it is hard to assess whether the newly developed analytical model is an improvement over numerical models. This must be reported and discussed.

Response:

The developed analytical model is straightforwardly evaluated by two series summations and does not require any complicated numerical method. The only numerical method involved in the code development is the determination of the eigenvalues which need to be obtained from the eigenvalues equation in Eq. (A19). The numerical method to solve the eigen-value equation

is quite easy and can be found in van Genuchten (1982). The computation is not time-consuming. The computational time for evaluation of the solutions at 50 different observations only takes 0.140 second computer clock time on an Intel Core i7-2600 3.40 MHz PC for species 1. We have added the discussions on the computational time in the revised manuscript.

Reference:

van Genuchten, M. Th., Alves, W. J., 1982: Analytical Solutions of the One-Dimensional Convective-Dispersive Solute Transport Equation, US Department of Agriculture, Washington, DC, Technical Bulletin No. 1661, 151 pp.

- The derivations are very hard to sort through as a reader, particularly if the reader is not well versed in the intricacies of the numerous transformations, etc... that are being performed. Please narrate the derivations in clear, concise language, with clear definitions and explanations. As written, most readers will skip over the derivations. - The first few sub-sections of the "Results and Discussion" section in fact seem like Methods. For example, 3.1 and 3.2 should be in the Methods section, since derivations are presented.

Response:

Thanks for the constructive comment. We elaborate on the detailed mathematical manipulations and procedures to obtain the final solutions in the revised manuscript for better readership. Moreover, Sections 3.1 and 3.2 are moved to Section 2 "Governing equations and analytical solutions".

- Overall, there are too many tables and figures. The large amount of model output shown in the tables probably is not needed, and instead can be replaced by metrics in several tables. The large amount of results is very tedious for a reader to sort through, and in the end discourages the reader from analyzing the model data and results critically.

Response:

Thanks for the helpful comments. Actually we have moved most of tables to the appendix. These figures and tables in the main text are important to illustrate the investigation of the convergence the derived solution, the accuracy of the computer code as well the transport processes affecting the transport behaviors.

A parsimonious analytical model for simulating two-dimensional multispecies plume migration *[response to comment of referee # 2]*

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Abstract

A parsimonious analytical model for rapidly predicting the two-dimensional plume behavior of decaying contaminant such as radionuclide and dissolved chlorinated solvent is presented in this study. Generalized analytical solutions in compact format are derived for the two-dimensional advection-dispersion equations coupled with sequential first-order decay reactions involving an arbitrary number of species in groundwater system. The solution techniques involve the sequential applications of the Laplace, finite Fourier cosine, and generalized integral transforms to reduce the coupled partial differential equation system to a set of linear algebraic equations. The system of algebraic equations is next solved for each species in the transformed domain, and the solutions in the original domain are then obtained through consecutive integral transform inversions. Explicit form solutions for a special case are derived using the generalized analytical solutions and are compared ***[response to comment of referee # 2]*** with the numerical solutions. The analytical results indicate that the parsimonious analytical solutions are robust and accurate. The solutions are useful for serving as simulation or screening tools for assessing plume behaviors of decaying contaminants including the radionuclides and dissolved chlorinated solvents in groundwater systems.

Keywords: Parsimonious analytical model; reactive transport; first-order decay reaction; Bateman-type source; radionuclide; dissolved chlorinated solvent.

1. Introduction

Experimental and theoretical studies have been undertaken to understand the fate and transport of dissolved hazardous substances in subsurface environments because that human health is threatened by a wide spectrum of contaminants in groundwater and soil. Analytical models are essential and efficient tools for understanding pollutants behavior in subsurface environments. Several analytical solutions for single-species transport problems have been reported for simulating the transport of various contaminants (Batu, 1989; 1993; 1996; Chen et al., 2008a; 2008b; 2011; Gao et al., 2010; 2012; 2013; Leij et al., 1991; 1993; Park and Zhan, 2001; Pérez Guerrero and Skaggs, 2010 ; Pérez Guerrero et al., 2013 ; van Genuchten and Alves, 1982; Yeh, 1981; Zhan et al., 2009; Ziskind et al., 2011). Transport processes of some contaminants such as radionuclides, dissolved chlorinated solvents and nitrogen generally involve a series of first-order or pseudo first-order sequential decay chain reactions. During migrations of decaying contaminants, mobile and toxic successor products may sequentially form and move downstream with elevated concentrations. Single-species analytical models do not permit transport behaviors of successor species of these decaying contaminants to be evaluated. Analytical models for multispecies transport equations coupled with first-order sequential decay reactions are useful tools for synchronous determination of the fate and transport of the predecessor and successor species of decaying contaminants. However, there are few analytical solutions for coupled multispecies transport equations compared to a large body of analytical solutions in the literature pertaining to the single-species advective-dispersive transport subject to a wide spectrum of initial and boundary conditions.

Mathematical approaches have been proposed in the literature to derive a limited number of one-dimensional analytical solutions or semi-analytical solutions for multispecies advective–dispersive transport equations sequentially coupled with first-order decay reactions. These include direct integral transforms with sequential substitutions (Cho, 1971; Lunn et al., 1996; van Genuchten, 1985, Mieles and Zhan, 2012) ***[response to comment of referee #2]***, decomposition by change-of-variables with the help of existing single-species analytical solutions (Sun and Clement, 1999; Sun et al., 1999a; 1999b), Laplace transform combined with decomposition of matrix diagonization (Quezada et al., 2004; Srinivasan and Clememt, 2008a; 2008b), decomposition by change-of-variables coupled with generalized integral transform (Pérez Guerrero et al., 2009; 2010), sequential integral transforms in association with algebraic decomposition (Chen et al., 2012a; 2012b).

Multi-dimensional solutions are needed for real world applications, making them more attractive than one-dimensional solutions. Bauer et al. (2001) presented the first set of semi-analytical solutions for one-, two-, and three-dimensional coupled multispecies transport problem with distinct retardation coefficients. Explicit analytical solutions were derived by Montas (2003) for multi-dimensional advective-dispersive transport coupled with first-order reactions for a three-species transport system with distinct retardation coefficients of species. Quezada et al. (2004) extended the Clement (2001) strategy to obtain Laplace-domain solutions for an arbitrary decay chain length. Most recently, Sudicky et al. (2013) presented a set of semi-analytical solutions to simulate the three-dimensional multi-species transport subject to first-order chain-decay reactions involving up to seven species and four decay levels. Basically, their solutions were obtained species by species using recursion relations between target species and its predecessor species. For a straight decay chain, they derived solutions for up to four species and no generalized expressions with compact formats for any target species were obtained. Note that their solutions were derived for the first-type (Dirichlet) inlet conditions which generally bring about physically improper mass conservation and significant errors in predicting the concentration distributions especially for a transport system with a large longitudinal dispersion coefficient (Barry and Sposito, 1988; Parlange et al., 1992). Moreover, in addition to some special cases, the numerical Laplace transforms are required to obtain the original time domain solution. Besides the straight decay chain, the analytical model by Clement (2001) and Sudicky (2013) can account more complicated decay chain problems such as diverging, converging and branched decay chains ***[response to comment of referee #2]***.

Based on the aforementioned reviews, this study presents a parsimonious explicit analytical model for two-dimensional multispecies transport coupled by a series of first-order decay reactions involving an arbitrary number of species in groundwater system. The derived analytical solutions have four salient features. First, the third-type (Robin) inlet boundary conditions which satisfy mass conservation are considered. Second, the solution is explicit, thus solution can be easily evaluated without invoking the numerical Laplace inversion. Third, the generalized solutions with parsimonious mathematical structures are obtained and valid for any species of a decay chain. The parsimonious mathematical structures of the generalized solutions are easy to code into a computer program for implementing the solution computations for arbitrary target species. Fourth, the derived solutions can account for any decay chain length. The explicit analytical solutions have applications for evaluation of concentration distribution of arbitrary target species of the real-world decaying contaminants. The developed parsimonious model is robustly verified with three example problems and applied to simulate the multispecies plume migration of dissolved radionuclides and chlorinated solvent.

2. Governing equations and analytical solutions

2.1 Derivation of analytical solutions

This study consider the problem of decaying contaminant plume migration. The source zone is located in the upstream of groundwater flow. The source zone can represent leaching of radionuclide from the deposit facility or release of chlorinated solvent from the residual NAPL phase into the aqueous phase. After these decaying contaminants enter the aqueous phase, they migrate by one-dimensional advection with flowing groundwater and by simultaneously longitudinal and transverse dispersion processes. While migration in the groundwater system, the contaminants undergo linear isothermal equilibrium sorption and a series of sequential first-order decaying reactions. Sudicky et al. (2013) provided the detailed modeling scenario. The scenario considered in this study can be ideally described as shown in Fig. 1. A steady and uniform velocity in the x direction is considered in Fig. 1. The governing equations describing two-dimensional reactive transport of the decaying contaminants and their successor species undergoing linear isothermal equilibrium sorption and a series of sequential first-order decaying reactions can be mathematically written as ***[response to comments of editor and referee #2]***

$$D_L \frac{\partial^2 C_1(x,y,t)}{\partial x^2} - v\frac{\partial C_1(x,y,t)}{\partial x} + D_T \frac{\partial^2 C_1(x,y,t)}{\partial y^2} - k_1 R_1 C_1(x,y,t) = R_1 \frac{\partial C_1(x,y,t)}{\partial t} \tag{1a}$$

$$D_L \frac{\partial^2 C_i(x,y,t)}{\partial x^2} - v\frac{\partial C_i(x,y,t)}{\partial x} + D_T \frac{\partial^2 C_i(x,y,t)}{\partial y^2} - k_i R_i C_i(x,y,t) + k_{i-1} R_{i-1} C_{i-1}(x,y,t) = R_i \frac{\partial C_i(x,y,t)}{\partial t} \quad i = 2 \ldots N. \tag{1b}$$

where $C_i(x,y,t)$ is the aqueous concentration of species i [$\mathbf{ML^{-3}}$]; x and y are the spatial coordinates in the groundwater flow and perpendicular directions [**L**], respectively; t is time [**T**]; D_L and D_T represent the longitudinal and transverse dispersion coefficients [$\mathbf{L^2T^{-1}}$], respectively; v is the average steady and uniform pore-water velocity [$\mathbf{LT^{-1}}$]; k_i is the first-order decay rate constant of species i [$\mathbf{T^{-1}}$]; R_i is the retardation coefficient of species i [**-**]. Note that these equations consider that the decay reactions occur simultaneously in both the aqueous and sorbed phases. If the decay reactions occur only in the aqueous phase, the retardation coefficients in the decay terms in the right-hand sides of Eqs. (1a) and (1b) become unity. For such case, k_i and k_{i-1} in the left-hand sides could be modified as $\frac{k_i}{R_i}$ and $\frac{k_{i-1}}{R_{i-1}}$ to facilitate the application of the derived analytical solutions obtained by Eqs. (1a) and (1b).

The initial and boundary conditions for solving Eqs. (1a) and (1b) are:

$$C_i(x, y, t=0) = 0 \quad 0 \le x \le L, 0 \le y \le W \qquad i = 1 \ldots N. \tag{2}$$

$$-D_L \frac{\partial C_i(x=0, y, t)}{\partial x} + vC_i(x=0, y, t) = vf_i(t)\left[H(y-y_1) - H(y-y_2)\right] \qquad t \ge 0 \qquad i = 1 \ldots N. \tag{3}$$

$$\frac{\partial C_i(x=L, y, t)}{\partial x} = 0 \qquad t \ge 0, 0 \le y \le W \qquad i = 1 \ldots N. \tag{4}$$

$$\frac{\partial C_i(x, y=0, t)}{\partial y} = 0 \qquad t \ge 0, 0 \le x \le L \qquad i = 1 \ldots N. \tag{5}$$

$$\frac{\partial C_i(x, y=W, t)}{\partial y} = 0 \qquad t \ge 0, 0 \le x \le L \qquad i = 1 \ldots N. \tag{6}$$

where $H(\bullet)$ is the Heaviside function, L and W are the length and width of the transport system under consideration. Eq. (2) implies that the transport system is free of solute mass at the initial time. Eq. (3) means that a third-type boundary condition satisfying mass conservation at the inlet boundary is considered. Eq. (4) considers the concentration gradient to be zero at the exit boundary based on the mass conservation principle. Such a boundary condition has been widely used for simulating solute transport in a finite-length system. Eqs. (5) and (6) assumes no solute flux across the lower and upper boundaries. It is noted that in Eq. (3), we assume arbitrary time-dependent sources of species i uniformly distributed at the segment ($y_1 \le y \le y_2$) of the inlet boundary ($x = 0$), the so-called Heaviside function source concentration profile. Relative to the first type boundary conditions used by Sudicky et al. (2013), the third-type boundary conditions which satisfy mass conservation at the inlet boundary (Barry and Sposito, 1988; Parlange et al., 1992) are used herein. Sudicky et al. (2013) considered the source concentration profiles as Gaussian or Heaviside step functions. If Gaussain distributions are desired, we can easily replace the Heaviside function in the right-hand side of Eq. (3) with a Gaussian distribution.

Eqs. (1)-(6) can be expressed in dimensionless form as

$$\frac{1}{Pe_L}\frac{\partial^2 C_1(X,Y,Z)}{\partial X^2}-\frac{\partial C_1(X,Y,Z)}{\partial X}+\frac{\rho^2}{Pe_T}\frac{\partial^2 C_1(X,Y,Z)}{\partial Y^2}-\kappa C_1(X,Y,Z)=R_1\frac{\partial C_1(X,Y,T)}{\partial T} \tag{7a}$$

$$\begin{aligned}&\frac{1}{Pe_L}\frac{\partial^2 C_i(X,Y,T)}{\partial X^2}-\frac{\partial C_i(X,Y,T)}{\partial X}+\frac{\rho^2}{Pe_T}\frac{\partial^2 C_i(X,Y,T)}{\partial Y^2}\\&-\kappa_i C_i(X,Y,T)+\kappa_{i-1}C_{i-1}(X,Y,T)=R_i\frac{\partial C_i(X,Y,T)}{\partial T}\end{aligned}\qquad i=2\ldots N. \tag{7b}$$

$$C_i(X,Y,T=0)=0 \quad 0\le X\le 1, 0\le Y\le 1 \quad i=1\ldots N. \tag{8}$$

$$-\frac{1}{Pe_L}\frac{\partial C_i(X=0,Y,T)}{\partial X}+C_i(X=0,Y,Z)=f_i(T)\left[H(Y-Y_1)-H(Y-Y_2)\right]\; T\ge 0\,, i=1\ldots N. \tag{9}$$

$$\frac{\partial C_i(X=1,Y,T)}{\partial X}=0 \quad T\ge 0, 0\le Y\le 1 \quad i=1\ldots N. \tag{10}$$

$$\frac{\partial C_i(X,Y=0,T)}{\partial Y}=0 \quad T\ge 0, 0\le X\le 1 \quad i=1\ldots N. \tag{11}$$

$$\frac{\partial C_i(X,Y=1,T)}{\partial Y}=0 \quad T\ge 0, 0\le X\le 1 \quad i=1\ldots N. \tag{12}$$

where $X=\frac{x}{L}$, $Y=\frac{y}{W}$, $Y_1=\frac{y_1}{W}$, $Y_2=\frac{y_2}{W}$, $T=\frac{vt}{L}$, $Pe_L=\frac{vL}{D_L}$, $Pe_T=\frac{vL}{D_T}$, $\rho=\frac{L}{W}$.

Our solution strategy used is extended from the approach proposed by Chen at al. (2012a; 2012b). The core of this approach is that the coupled partial differential equations are converted into an algebraic equation system via a series of integral transforms and the solutions in the transformed domain for each species are directly and algebraically obtained by sequential substitutions.

Following Chen et al. (2012a; 2012b), the generalized analytical solutions in compact formats can be obtained as follows (with detailed derivation provided in Appendix A)

$$
\begin{aligned}
&C_i(X,Y,T) \\
&= f_i(T)\Phi(n=0) + e^{\frac{Pe_L}{2}X}\sum_{l=1}^{\infty}\frac{K(\xi_l,X)}{N(\xi_l)}\left[p_i(\xi_l,n,T)+q_i(\xi_l,n,T)\right]\Phi(n=0)\Theta(\xi_l) \qquad (13)\\
&+2\sum_{n=1}^{n=\infty}\left\{f_i(T)\Phi(n)+e^{\frac{Pe_L}{2}X}\sum_{l=1}^{\infty}\frac{K(\xi_l,X)}{N(\xi_l)}\left[p_i(\xi_l,n,T)+q_i(\xi_l,n,T)\right]\Phi(n)\Theta(\xi_l)\right\}\cos(n\pi Y)
\end{aligned}
$$

where $\Phi(n)=\begin{cases} Y_2-Y_1 & n=0 \\ \dfrac{\sin(n\pi Y_2)-\sin(n\pi Y_1)}{n\pi} & n=1,2,3\ldots \end{cases}$, ξ_l is the eigenvalue, determined from the equation $\xi_l\cot\xi_l-\dfrac{\xi_l^2}{Pe_L}+\dfrac{Pe_L}{4}=0$, $\Theta(\xi_l)=\dfrac{Pe_L\xi_l}{\dfrac{Pe_L^2}{4}+\xi_l^2}$, $K(\xi_l,X)=\dfrac{Pe_L}{2}\sin(\xi_l X)+\xi_l\cos(\xi_l X)$,

$N(\xi_l)=\dfrac{2}{\dfrac{Pe_L^2}{4}+Pe_L+\xi_l^2}$,

$$p_i(\xi_l,n,T)=f_i(T)-\beta_i e^{-\alpha_i T}\int_0^T f_i(\tau)e^{\alpha_i\tau}d\tau \qquad (14)$$

and

$$q_i(\xi_l,n,T)=\sum_{k=0}^{k=i-2}\left(\beta_{i-k-1}\prod_{j_1=0}^{j_1=k}\sigma_{i-j_1}\right)\sum_{j_2=0}^{j_2=k+1}\frac{e^{-\alpha_{i-j_2}T}\int_0^T e^{\alpha_{i-j_2}\tau}f_{i-k-1}(\tau)d\tau}{\prod_{j_3=i-k-1,\,j_3\neq i-j_2}^{j_3=i}\left(\alpha_{j_3}-\alpha_{i-j_2}\right)} \qquad (15)$$

where $\alpha_i(\xi_l)=\dfrac{\kappa_i}{R_i}+\dfrac{\rho^2n^2\pi^2}{Pe_T R_i}+\dfrac{Pe_L}{4R_i}+\dfrac{\xi_l^2}{Pe_L R_i}$, $\beta_i(\xi_l)=\dfrac{Pe_L}{4R_i}+\dfrac{\xi_l^2}{Pe_L R_i}$, $\sigma_i=\dfrac{\kappa_{i-1}}{R_i}$ ***[response to referee #2]***.

Concise expressions for arbitrary target species such as described in Eqs. (13) to (15) facilitate the development of a computer code for implementing the computations of the analytical solutions.

The generalized solutions of Eq. (13) accompanied by two corresponding auxiliary functions

$p_i(\xi_l,n,T)$ and $q_i(\xi_l,n,T)$ in Eqs. (14)-(15) can be applied to derive analytical solutions for some special-case inlet boundary sources. Here the time-dependent decaying source which represents the specific release mechanism defined by the Bateman equations (van Genuchten, 1985) is considered. A Bateman-type source is described by

$$f_i(t) = \sum_{m=1}^{i} b_{im} e^{-\delta_m t} \tag{16a}$$

or in dimensionless form,

$$f_i(T) = \sum_{m=1}^{m=i} b_{in} e^{-\lambda_m T} \tag{16b}$$

The coefficients b_{im} and $\delta_m = \mu_m + \gamma_m$ account for the first-order decay reaction rate (μ_m) of each species in the waste source and the release rate (γ_m) of each species from the waste source, $\lambda_m = \dfrac{\delta_m L}{v}$.

By substituting Eq. (16b) into Eqs. (13)-(15), we obtain

$$\begin{aligned} &C_i(X,Y,T) \\ &= \sum_{m=1}^{m=i} b_{im} e^{-\lambda_m T}\Phi(n=0) + e^{\frac{Pe_L}{2}X} \sum_{l=1}^{\infty} \frac{K(\xi_l,X)}{N(\xi_l)} \left[p_i(\xi_l,n,T) + q_i(\xi_l,n,T)\right]\Phi(n=0)\Theta(\xi_l) \\ &+ 2\sum_{n=1}^{n=\infty} \left\{ \sum_{m=1}^{m=i} b_{im} e^{-\lambda_m T}\Phi(n) + e^{\frac{Pe_L}{2}X} \sum_{l=1}^{\infty} \frac{K(\xi_l,X)}{N(\xi_l)} \left[p_i(\xi_l,n,T) + q_i(\xi_l,n,T)\right]\Phi(n)\Theta(\xi_l) \right\} \cos(n\pi Y) \end{aligned} \tag{17}$$

where

$$p_i(\xi_l,n,T) = \sum_{m=1}^{m=i} b_{i,m} \cdot e^{-\lambda_m T} - \beta_i \sum_{m=1}^{m=i} b_{i,m} \frac{e^{-\lambda_m T} - e^{-\alpha_i T}}{\alpha_i - \lambda_m} \tag{18}$$

and

$$p_i(\xi_l,n,T)=\sum_{k=0}^{k=i-2}\left(\beta_{i-k-1}\prod_{j_1=0}^{j_1=k}\sigma_{i-j_1}\right)\sum_{j_2=0}^{j_2=k+1}\frac{\sum_{m=1}^{m=i-k-1}\frac{b_{i-k-1,m}\left(e^{-\lambda_m T}-e^{-\alpha_{i-j_2}T}\right)}{\alpha_{i-j_2}-\lambda_m}}{\prod_{j_3=i-k-1,\, j_3\neq i-j_1}^{j_3=i}\left(\alpha_{j_3}-\alpha_{i-j_2}\right)} \quad (19)$$

2.2 Convergence behavior of the Bateman-type source solution

Based on the special-case analytical solutions in Eq. (17) supported by two auxiliary functions, defined in Eqs. (18) and (19), a computer code was developed in FORTRAN 90 ***[response to referee # 2]*** language with double precision. The details of the FORTRAN computer code is described in Supplement. The derived analytical solutions in Eqs. (17)-(19) consist of summations of double infinite series expansions for the finite Fourier cosine and generalized integral transform inversions, respectively. It is straightforward to sum up these two infinite series expansions term by term. To avoid time-consuming summations of these infinite series expansions, the convergence tests should be routinely executed to determine the optimal number of the required terms for evaluating analytical solutions to the desired accuracies. Two-dimensional four-member radionuclide decay chain $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ is considered herein as convergence test example 1 to demonstrate the convergence behavior of the series expansions. This convergence test example 1 is modified from a one-dimensional radionuclide decay chain problem originated by Higashi and Pigford (1980) and later applied by van Genuchten (1985) to illustrate the applicability of their derived solution. The important model parameters related to this test example are listed in Tables 1 and 2. The inlet source is chosen to be symmetrical with respect to the x -axis and conveniently arranged in the $40\,m \le y \le 60\,m$ segment at the inlet boundary.

In order to investigate the required term number of series expansions to achieve accurate numerical evaluation for the finite Fourier cosine transform inverse, a sufficiently large number of series expansions for the generalized transform inverse are used to exclude the influence of the number of terms in series expansions for the generalized integral transform inverse on convergence of finite Fourier cosine transform inverse. A similar concept is used when investigating the required number of terms in the series expansions for the generalized integral transform inverse. An alternative approach is conducted by simultaneously varying the term numbers of series expansions for the generalized integral transform inverse and the finite Fourier cosine transform inverse.

Tables 3, 4 and 5 give results of the convergence tests up to 3 decimal digits of the solution computations along the three transects (inlet boundary at x =0 m, x =25 m, and exit boundary at x =250m). In these tables M and N are defined as the numbers of terms summed for the generalized integral transform inverse and finite Fourier cosine transform inverse, respectively. It is observed that M and N are related closely to the true values of the solutions. For smaller true values, the solutions must be computed with greater M and N. However, convergences can be drastically speeded up if lower calculation precision (e.g. 2 decimal digits accuracy) is acceptable. For example, $(M,N)=(100,200)$ is sufficient for 2 decimal digits accuracy, while for 3 decimal digits accuracy we need $(M,N)=(1600,8000)$. Two decimal digits accuracy is acceptable for most practical problems. It is also found that M increases and N decreases with increasing x.

To further examine the series convergence behavior, example 2 considers a transport system of large aspect ratio ($\frac{L}{W}=\frac{2{,}500m}{100m}$) and a narrower source segment, $45\,m \le y \le 55\,\mathrm{m}$, on the inlet boundary. Tables 6 and 7 present results of the convergence tests of the solution computations along two transects (inlet boundary and x =250 m). Tables 6 and 7 also show similar results for the dependences of M and N on x. Note that larger M and N are required for each species in this test example, suggesting that the evaluation of the solution for a large aspect ratio requires more series expansion terms to achieve the same accuracy as compared to example 1. Detailed results of the convergence test examples 1 and 2 are provided in Supplement.

Using the required numbers determined from the convergence test, the computational time for evaluation of the solutions at 50 different observations only takes 3.782s, 11.325s, 23.95s and 67.23s computer clock time on an Intel Core i7-2600 3.40 MHz PC for species 1, 2, 3, and 4 in the comparison of example 1 ***[response to comments of referees # 2 and #3]***.

3. Results and discussion

3.1 Comparison of the analytical solutions with the numerical solutions ***[response to comment of referee # 2]***

Three comparison ***[response to comment of referee # 2]*** examples are considered to examine the correctness and robustness of the analytical solutions and the accuracy of the computer code. The first comparison ***[response to comment of referee # 2]*** example is the four-member radionuclide transport problem used in the convergence test example 1. The second comparison example considers the four-member radionuclide transport problem used in the convergence test example 2. The third comparison example is used to test the accuracy of the computer code for simulating the reactive contaminant transport of a long decay chain ***[response to comment of referee # 2]***. The three comparison examples are executed by comparing the simulated results of the derived analytical solutions with the numerical solutions obtained using the Laplace transformed finite difference (LTFD) technique first developed by Moridis and Reddell (1991). A computer code for the LTFD solution are written in FORTTRAN language with double precision. The details of the FORTRAN computer code is described in Supplement.

Figures 2, 3 and 4 depicts the spatial concentration distribution along one longitudinal direction ($y = 50$ m) and two transverse directions ($x = 0$ m and $x = 25$ m) for convergence test example 1 at $t = 1{,}000$ year obtained from analytical solutions and numerical solutions. Figures 5, 6 and 7 present the spatial concentration distribution along one longitudinal direction ($y = 50$ m) and two transverse directions ($x = 0$ m and $x = 25$ m) for the convergence test example 2 at t = 1,000 year obtained from analytical solutions and numerical solutions. Excellent agreements between the two solutions for both examples are observed for a wide spectrum of concentration, thus warranting the accuracy and robustness of the developed analytical model.

The third ***[response to comments of referee # 2]*** example involves a 10 species decay chain previously presented by Srinivasan and Clement (2008a) to evaluate the performance of their one-dimensional analytical solutions. The relevant model parameters are summarized in Tables 8 and 9. Our computer code is also compared ***[response to comment of referee # 2]*** against the LTFD solutions for this example. Figure 8 depicts the spatial concentration distribution at t = 20 days obtained analytically and numerically. Again there is excellent agreement between the analytical and numerical solutions, demonstrating the performance of our computer code for simulating transport problems with a long decay chain. The three comparison results clearly establish the correctness of the analytical model and the accuracy and capability of the computer code.

3.2 Assessing physical and chemical parameters on the radionuclide plume migration

Physical processes and chemical reactions affect the extent of contaminant plumes, as well as concentration levels. To illustrate how the physical processes and chemical reactions affect multispecies plume development, we consider the four-member radionuclide decay chain used in the previous convergence test and solution verification. The model parameters are the same, except that the longitudinal (D_L) and transverse (D_T) dispersion coefficients are varied. Three sets of longitudinal and transverse dispersion coefficients D_L =1,000, D_T =100; D_L =1,000, D_T =200; D_L =2000, D_T =200 (all in m^2/year) are tested, all for a simulation time of 1,000 years.

Figure 9 illustrates the spatial concentration of four species at t = 1,000 year for the three sets of dispersion coefficients. The mobility of plumes of ^{234}U and ^{230}Th is retarded because of their stronger sorption ability. Hence the least retarded ^{226}Ra plume extensively migrated to 200 m × 60 m area in the simulation domain, whereas the ^{234}U and ^{230}Th plumes are confined within 60 m × 50 m area in the simulation domain. The moderate mobility of ^{238}Pu reflects the fact that it is a medial sorbed member of this radionuclide decay chain. The high concentration level of ^{234}U accounts for the high first-order decay rate constant of its parent species ^{238}Pu and its own low first-order decay rate constant. The plume extents and concentration levels may be sensitive to longitudinal and transverse dispersion. Increase of the longitudinal and/or transverse dispersion coefficients enhances the spreading of the plume extensively along the longitudinal and/or transverse directions, thereby lowering the plume concentration level. Because the concentration levels of the four radionuclides are influenced by both source release rates and decay chain reactions, ^{230}Th has the least extended plume area, while ^{226}Ra has the greatest plume area for all three set of dispersion coefficients. These dispersion coefficients only affect the size of plumes of the four radionuclide, but the order of their relative plume size remains the same (i.e. ^{226}Ra > ^{238}Pu > ^{234}U > ^{230}Th for the simulated condition). Indeed, in the reactive contaminant transport, the chemical parameters of sorption and decay rate are more important than the physical parameters of dispersion coefficients that govern the order of the plume extents and the concentration levels.

3.3 Simulating the natural attenuation of chlorinated solvent plume migration

Natural attenuation is the reduction in concentration and mass of the contaminant due to naturally occurring processes in the subsurface environment. The process is monitored for regulatory purposes to demonstrate continuing attenuation of the contaminant reaching the site-specific regulatory goals within reasonable time, hence, the use of the term monitored natural attenuation

(MNA). MNA has been widely accepted as a suitable management option for chlorinated solvent contaminated groundwater. Mathematical model are widely used to evaluate the natural attenuation of plumes at chlorinated solvent sites. The multispecies transport analytical model developed in this study provides an effective tool for evaluating performance of the monitoring natural attenuation of plumes at a chlorinated solvent site because a series of daughter products produced during biodegradation of chlorinated solvent such as $PCE \rightarrow TCE \rightarrow DCE \rightarrow VC \rightarrow ETH$. Thus simulation of the natural attenuation of plumes a chlorinated solvent constitutes an attractive field application example of our multispecies transport model.

A study of 45 chlorinated solvent sites by McGuire et al. (2014) found that mathematical models were used at 60% of these sites and that the public domain model BIOCHLOR (Aziz et al., 2000) provided by the Center for Subsurface Modeling Support (CSMoS) of USEPA was the most commonly used model. The utility of the BIOCHLOR model to the real-world problems has been demonstrated by an example application that it can reproduce plume movement from 1965 to 1998 at the contaminated site of Cape Canaveral Air Station, Florida ***[response to comment of referee # 3]***.

An illustrated example from BIOCHLOR (Aziz et al., 2000) is considered to demonstrate the application of the developed analytical model. The simulation conditions and transport parameters for this example application are summarized in Table 10. Constant source concentrations rather than exponentially declining source concentration of five-species chlorinated solvents are specified in the $90.7\ m \le y \le 122.7\ m$ segment at the inlet boundary ($x = 0$). This means that the exponents (λ_{im}) of Bateman-type sources in Eqs. (16a) or (16b) need to be set to zero for the constant source concentrations and source intensity constants (b_{im}) are set to zero when subscript *i* does not equal to subscript *m*. Table 11 lists the coefficients of Bateman-type boundary source used for this example application involving the five-species dissolved chlorinated solvent problem. Spatial concentration contours of five-species at t = 1 year obtained from the derived analytical solutions for natural attenuation of chlorinated solvent plumes are depicted in Fig. 10. It is observed that the mobility of plumes is quite sensitive to the species retardation factors, whereas the decay rate constants determine the plume concentration level. The plumes can migrate over a larger region for species having a low retardation factor such as VC. The low decay rate constants such as ETH have higher concentration distribution than the VC. It should be noted that a larger extent of plume observed for ETH in Fig. 10 is mainly attributed the plume mass accumulation from the predecessor species VC that have a larger plume extent. The effect of high retardation of the ETH is hindered by the mass accumulation of the predecessor species VC.

4. Conclusions

We present an analytical model with a parsimonious mathematical format for two-dimensional multispecies advective-dispersive transport of decaying contaminants such as radionuclides, chlorinated solvents and nitrogen. The developed model is capable of accounting for the temporal and spatial development of an arbitrary number of sequential first-order decay reactions. The solution procedures involve applying a series of Laplace, finite Fourier cosine and generalized integral transforms to reduce a partial differential equation system to an algebraic system, solving for the algebraic system for each species, and then inversely transforming the concentration of each species in transformed domain into the original domain. Explicit special solutions for Bateman type source problems are derived via the generalized analytical solutions. The convergence of the series expansion of the generalized analytical solution is robust and accurate. These explicit solutions and the computer code are comparing with the results computed by the numerical solutions. The two solutions agree well for a wide spectrum of concentration variations for three test examples. The analytical model is applied to assess the plume development of radionuclide and dissolved chlorinated solvent decay chain. The results show that dispersion only moderately modifies the size of the plumes, without altering the relative order of the plume sizes of different contaminant. It is suggested that retardation coefficients, decay rate constants and the predecessor species plume distribution mainly govern the order of plume size in groundwater. Although there are a number of numerical reactive transport models that can account for multispecies advective-dispersive transport, our analytical model with a computer code that can directly evaluate the two-dimensional temporal-spatial concentration distribution of arbitrary target species without involving the computation of other species. The analytical model developed in this study effectively and accurately predicts the two-dimensional radionuclide and dissolved chlorinated plume migration. It is a useful tool for assessing the ecological and environmental impact of the accidental radionuclide releases such as the Fukushima nuclear disaster where multiple radionuclides leaked through the reactor, subsequently contaminating the local groundwater and ocean seawater in the vicinity of the nuclear plant. It is also a screening model that simulates remediation by natural attenuation of dissolved solvents at chlorinated solvent release sites.

It should be noted the derived analytical model still have its application limitations for that the groundwater flow in the study site is non-uniform or the study or the site have multiple distinct zones. Furthermore, the developed model cannot simulate the more complicated decay chain problems such as diverging, converging and branched decay chains. The analytical model for more complicated decay chain problems can be pursued in the near future ***[response to comment of referee # 1]***.

Appendix A

Derivation of analytical solutions

In this appendix, we elaborate on the mathematical procedures for deriving the analytical solutions.

The Laplace transforms of Eqs. (7a), (7b), (9)-(12) yield

$$\frac{1}{Pe_L}\frac{\partial^2 G_1(X,Y,s)}{\partial X^2}-\frac{\partial G_1(X,Y,s)}{\partial X}+\frac{\rho^2}{Pe_T}\frac{\partial^2 G_1(X,Y,s)}{\partial Y^2}-(R_1 s+\kappa_1)G_1(X,Y,s)=0 \qquad \text{(A1a)}$$

$$\frac{1}{Pe_L}\frac{\partial^2 G_i(X,Y,s)}{\partial X^2}-\frac{\partial G_i(X,Y,s)}{\partial X}+\frac{\rho^2}{Pe_T}\frac{\partial^2 G_i(X,Y,s)}{\partial Y^2} \qquad i=2,3,\ldots N \qquad \text{(A1b)}$$
$$-\kappa_i G_i(X,Y,s)+\kappa_{i-1}G_{i-1}(X,Y,s)=R_i s G_i(X,Y,s)$$

$$-\frac{1}{Pe_L}\frac{\partial G_i(X=0,Y,s)}{\partial X}+G_i(X=0,Y,s)=F_i(s)\left[H(Y-Y_1)-H(Y-Y_2)\right] \quad 0\le Y\le 1 \quad i=1\ldots N.$$

(A2)

$$\frac{\partial G_i(X=1,Y,s)}{\partial X}=0 \qquad 0\le Y\le 1 \qquad i=1\ldots N. \qquad \text{(A3)}$$

$$\frac{\partial G_i(X,Y=0,s)}{\partial Y}=0 \qquad 0\le X\le 1 \qquad i=1\ldots N. \qquad \text{(A4)}$$

$$\frac{\partial G_i(X,Y=1,s)}{\partial Y}=0 \qquad 0\le X\le 1 \qquad i=1\ldots N. \qquad \text{(A5)}$$

where s is the Laplace transform parameter, and $G_i(X,Y,s)$ and $F_i(s)$ are defined by the Laplace transformation relations as

$$G_i(X,Y,s)=\int_0^\infty e^{-sT}C_i(X,Y,T)dT \qquad \text{(A6)}$$

$$F_i(s)=\int_0^\infty e^{-sT}f_i(T)dT \qquad \text{(A7)}$$

The finite Fourier cosine transform is used here because it satisfies the transformed governing equations in Eqs. (A1a) and (A2b) and their corresponding boundary conditions in Eqs. (A4) and (A5). Application of the finite Fourier cosine transform on Eqs. (A1)-(A3) leads to

$$\frac{1}{Pe_L}\frac{d^2H_1(X,n,s)}{\partial X^2}-\frac{dH_1(X,n,s)}{dX}-\left(R_1 s+\kappa_1+\frac{\rho^2 n^2\pi^2}{Pe_T}\right)H_1(X,n,s)=0 \tag{A8a}$$

$$\frac{1}{Pe_L}\frac{d^2H_i(X,n,s)}{dX^2}-\frac{dH_i(X,n,s)}{dX}-\left(R_i s+\kappa_i+\frac{\rho^2 n^2\pi^2}{Pe_T}\right)H_i(X,n,s)+\kappa_{i-1}H_{i-1}(X,n,s)=0 \tag{A8b}$$

$$-\frac{1}{Pe_L}\frac{dH_i(X=0,n,s)}{dX}+H_i(X=0,n,s)=F_i(s)\Phi(n) \tag{A9}$$

$$\frac{dH_i(X=1,n,s)}{dX}=0 \tag{A10}$$

where $\Phi(n)=\begin{cases} Y_2-Y_1 & n=0 \\ \dfrac{\sin(n\pi Y_2)-\sin(n\pi Y_1)}{n\pi} & n=1,2,3... \end{cases}$, n is the finite Fourier cosine transform parameter, $H_i(X,n,s)$ is defined by the following conjugate equations (Sneddon, 1972)

$$H_i(X,n,s)=\int_0^1 G_i(X,Y,s)\cos(n\pi Y)dY \tag{A11}$$

$$G_i(X,Y,s)=H_i(X,n=0,s)+2\sum_{n=1}^{n=\infty}H_i(X,n,s)\cos(n\pi Y) \tag{A12}$$

Using changes-of-variables, similar to those applied by Chen and Liu (2011), the advective terms in Eqs. (A8a) and A(8b) as well as nonhomogeneous terms in Eq. (A9) can be easily removed. Thus, substitutions of the change-of-variable into Eqs. (A8a), (A8b), (A9) and (A10) result in diffusive-type equations associated with homogeneous boundary conditions

$$\begin{aligned}&\frac{1}{Pe_L}\frac{d^2U_1(X,n,s)}{dX^2}-\left(R_1 s+\kappa_1+\frac{\rho^2 n^2\pi^2}{Pe_T}+\frac{Pe_L}{4}\right)U_1(X,n,s)\\&=e^{-\frac{Pe_L}{2}X}\left(R_1 s+\kappa_1+\frac{\rho^2 n^2\pi^2}{Pe_T}\right)F_1(s)\Phi(n)\end{aligned} \tag{A13a}$$

$$\frac{1}{Pe_L}\frac{d^2U_i(X,n,s)}{d\mathrm{X}^2}-\left(\frac{Pe_L}{4}+R_is+\kappa_1+\frac{\rho^2n^2\pi^2}{Pe_T}\right)U_i(X,n,s)$$
$$=e^{-\frac{Pe_L}{2}X}\left(R_is+\kappa_i+\frac{\rho^2n^2\pi^2}{Pe_T}\right)F_i(s)\Phi(n)-e^{-\frac{Pe_L}{2}X}\kappa_{i-1}F_{i-1}(s)\Phi(n)-\kappa_{i-1}U_{i-1}(X,n,s) \quad \text{(A13b)}$$

$$-\frac{dU_i(X=0,n,s)}{dX}+\frac{Pe}{2}U_i(X=0,n,s)=0 \quad \text{(A14)}$$

$$\frac{dU_i(X=1,n,s)}{dX}+\frac{Pe_L}{2}U_i(X=1,n,s)=0 \quad \text{(A15)}$$

where $U_i(X,n,s)$ is defined as the following change-of-variable relation

$$H_i(X,n,s)=F_i(s)\Phi(n)+e^{\frac{Pe_L}{2}X}U_i(X,n,s) \quad \text{(A16)}$$

As detailed in Ozisik (1989), the generalized integral transform pairs for Eqs. (A13a) and (A13b) and its associated boundary conditions (A14) and (A15) are defined as

$$Z_i(\xi_l,n,s)=\int_0^1 K(\xi_l,X)U_i(X,n,s)dX \quad \text{(A17)}$$

$$U_i(X,n,s)=\sum_{l=1}^{\infty}\frac{K(\xi_l,X)}{N(\xi_l)}Z_i(\xi_l,n,s) \quad \text{(A18)}$$

where $K(\xi_l,X)=\frac{Pe_L}{2}\sin(\xi_lX)+\xi_l\cos(\xi_lX)$ is the kernel function, $N(\xi_l)=\dfrac{2}{\frac{{Pe_L}^2}{4}+Pe_L+{\xi_l}^2}$,

ξ_l is the eigenvalue, determined from the equation

$$\xi_l\cot\xi_l-\frac{{\xi_l}^2}{Pe_L}+\frac{Pe_L}{4}=0 \quad \text{(A19)}$$

The generalized integral transforms of Eqs. (13a) and (13b) give

$$-\left(R_1s+\kappa_1+\frac{\rho^2n^2\pi^2}{Pe_T}+\frac{Pe_L}{4}+\frac{{\xi_l}^2}{Pe_L}\right)Z_i(\xi_l,n,s)=\left(R_1s+\kappa_1+\frac{\rho^2n^2\pi^2}{Pe_T}\right)F_1(s)\Phi(n)\Theta(\xi_l) \quad \text{(A20)}$$

$$
\begin{aligned}
&-\left(R_i s+\kappa_i+\frac{\rho^2 n^2 \pi^2}{Pe_T}+\frac{Pe_L}{4}+\frac{\xi_l{}^2}{Pe_L}\right)Z_i(\xi_l,n,s) \\
&=\left(R_i s+\kappa_i+\frac{\rho^2 n^2 \pi^2}{Pe_T}\right)F_i(s)\Phi(n)\Theta(\xi_l)-\kappa_{i-1}F_{i-1}(s)\Phi(n)\Theta(\xi_l)-\kappa_{i-1}Z_{i-1}(\xi_l,n,s)
\end{aligned}
\tag{A21}
$$

where $\Theta(\xi_l)=\dfrac{Pe_L\xi_l}{\dfrac{Pe_L{}^2}{4}+\xi_l{}^2}$.

Solving for Eqs. (A20) and (A21) algebraically for each species, $Z_i(\xi_l,n,s)$, in sequence, leads to

$$
Z_1(\xi_l,n,s)=-\frac{s+\alpha_1-\beta_1}{s+\alpha_1}F_1(s)\Phi(n)\Theta(\xi_l)
\tag{A22}
$$

$$
Z_2(\xi_l,n,s)=\left[-\frac{s+\alpha_2-\beta_2}{s+\alpha_2}F_2(s)+\frac{\sigma_2\beta_1}{(s+\alpha_2)(s+\alpha_1)}F_1(s)\right]\Phi(n)\Theta(\xi_l)
\tag{A23}
$$

$$
\begin{aligned}
Z_3(\xi_l,n,s)=&\left[-\frac{s+\alpha_3-\beta_3}{s+\alpha_3}F_3(s)+\frac{\sigma_3\beta_2}{(s+\alpha_3)(s+\alpha_2)}F_2(s)\right. \\
&\left.\frac{\sigma_3\sigma_2\beta_1}{(s+\alpha_3)(s+\alpha_2)(s+\alpha_1)}F_1\right]\Phi(n)\Theta(\xi_l)
\end{aligned}
\tag{A24}
$$

$$
\begin{aligned}
Z_4(\xi_l,n,s)=&\left[-\frac{s+\alpha_4-\beta_4}{s+\alpha_4}F_4(s)+\frac{\sigma_4\beta_3}{(s+\alpha_4)(s+\alpha_3)}F_3(s)\right. \\
&\left.+\frac{\sigma_4\sigma_3\beta_2}{(s+\alpha_4)(s+\alpha_3)(s+\alpha_2)}F_2(s)+\frac{\sigma_4\sigma_3\sigma_2\beta_1}{(s+\alpha_4)(s+\alpha_3)(s+\alpha_2)(s+\alpha_1)}F_1(s)\right]\Phi(n)\Theta(\xi_l)
\end{aligned}
\tag{A25}
$$

where $\alpha_i(\xi_l)=\dfrac{\kappa_i}{R_i}+\dfrac{\rho^2 n^2\pi^2}{Pe_T R_i}+\dfrac{Pe_L}{4R_i}+\dfrac{\xi_l{}^2}{Pe_L R_i}$, $\beta_i(\xi_l)=\dfrac{Pe_L}{4R_i}+\dfrac{\xi_l{}^2}{Pe_L R_i}$, $\sigma_i=\dfrac{\kappa_{i-1}}{R_i}$.

Upon inspection of Eqs. (A22)-(A25), compact expressions valid for all species can be generalized as

$$
Z_i(\xi_l,n,s)=\left[P_i(\xi_l,n,s)+Q_i(\xi_l,n,s)\right]\Phi(n)\Theta(\xi_l)\quad i=1,2\ldots N
\tag{A26}
$$

where $P_i(\xi_l, n, s) = -\dfrac{s+\alpha_i-\beta_i}{s+\alpha_i} F_i(s)$ and $Q_i(\xi_l, n, s) = \sum_{k=0}^{k=i-2} \dfrac{\beta_{i-k-1} \prod_{j_1=0}^{j_1=k} \sigma_{i-j_1}}{\prod_{j_2=0}^{j_2=k+1} \left(s+\alpha_{i-j_2}\right)} F_{i-k-1}(s)$.

The solutions in the original domain are obtained by a series of integral transform inversions in combination with changes-of-variables.

The inverse generalized integral transform of Eq. (A26) gives

$$W_i(X, n, s) = \sum_{m=1}^{\infty} \frac{K(\xi_l, X)}{N(\xi_l)} \left[P_i(\xi_l, n, s) + Q_i(\xi_l, n, s)\right] \Phi(n) \Theta(\xi_l) \tag{A27}$$

Using change-of-variable relation of Eq. (A16), one obtains

$$H_i(\xi_l, n, s) = F_i(s)\Phi(n) + e^{\frac{Pe_L}{2} x_D} \sum_{m=1}^{\infty} \frac{K(\xi_l, x_D)}{N(\xi_l)} \left[P_i(\xi_l, n, s) + Q_i(\xi_l, n, s)\right] \Phi(n) \Theta(\xi_l) \tag{A28}$$

The finite Fourier cosine inverse transform of Eq. (A28) results in

$$\begin{aligned} &G_i(X, Y, s) \\ &= F_i(s)\Phi(n=0) + e^{\frac{Pe_L}{2} X} \cdot \sum_{l=1}^{\infty} \frac{K(\xi_l, X)}{N(\xi_l)} \left[P_i(\xi_l, n, s) + Q_i(\xi_l, n, s)\right] \Phi(n=0) \Theta(\xi_l) \\ &+ 2 \sum_{n=1}^{n=\infty} \left\{ F_i(s)\Phi(n) + e^{\frac{Pe_L}{2} X} \sum_{l=1}^{\infty} \frac{K(\xi_l, X)}{N(\xi_l)} \left[P_i(\xi_l, n, s) + Q_i(\xi_l, n, s)\right] \Phi(n) \Theta(\xi_l) \right\} \cos(n\pi Y) \end{aligned} \tag{A29}$$

The analytical solutions in the original domain will be completed by taking the Laplace inverse transform of Eq. (A29). $P_i(\xi_l, n, s)$ in Eq. (29) is in the form of the product of two functions . The Laplace transform of $\dfrac{s+\alpha_i-\beta_i}{s+\alpha_i}$ can be easily obtained as

$$L^{-1}\left[\frac{s+\alpha_i-\beta_i}{s+\alpha_i}\right] = \delta(T) - \beta_i e^{-\alpha_i T} \tag{A30}$$

Thus, the Laplace inverse of $P_i(\xi_l, n, s)$ can be achieved using the convolution theorem as

$$p_i(\xi_l,n,T)=L^{-1}[P_i(\xi_l,n,s)]=L^{-1}\left[-\frac{s+\alpha_i-\beta_i}{s+\alpha_i}F_i(s)\right]=-f_i(T)+\beta_i e^{-\alpha_i T}\int_0^T f_i(\tau)e^{\alpha_i\tau}d\tau \qquad \text{(A31)}$$

The Laplace inverse of $Q_i(\xi_l,n,s)$ can be also approached using the similar method. By taking Laplace inverse transform on $Q_i(\xi_l,n,s)$, we have

$$q_i(\xi_l,n,T)=L^{-1}[Q_i(\xi_l,n,s)]=L^{-1}\left[\sum_{k=0}^{k=i-2}\frac{\beta_{i-k-1}\prod_{j_1=0}^{j_1=k}\sigma_{i-j_1}}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}F_{i-k-1}(s)\right]$$

$$=\sum_{k=0}^{k=i-2}\beta_{i-k-1}\prod_{j_1=0}^{j_1=k}\sigma_{i-j_1}L^{-1}\left[\frac{1}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}F_{i-k-1}(s)\right] \qquad \text{(A32)}$$

Expressing $\frac{1}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}$ as the summation of partial fractions and applying the inverse Laplace transform formula, one gets

$$L^{-1}\left[\frac{1}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}\right]=L^{-1}\left[\sum_{j_2=0}^{j_2=k+1}\frac{1}{\prod_{j_3=i-k-1,\, j_3\neq i-j_2}^{j_3=i}(\alpha_{j_3}-\alpha_{i-j_2})\left(s+\alpha_{i-j_2}\right)}\right]$$

$$=\sum_{j_2=0}^{j_2=k+1}\frac{e^{-\alpha_{i-j_1}T}}{\prod_{j_3=i-k-1,\, j_3\neq i-j_1}^{j_3=i}(\alpha_{j_3}-\alpha_{i-j_1})} \qquad \text{(A33)}$$

Recall that the inverse Laplace transform of $F_{i-k-1}(s)$ is $f_{i-k-1}(T)$. Thus, the Laplace inverse transform of $\frac{1}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}F_{i-k-1}(s)$ in Eq. (1) can be achieved using the convolution integral equation as

$$L^{-1}\left[\frac{1}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}F_{i-k-1}(s)\right]=\sum_{j_2=0}^{j_2=k+1}\frac{e^{-\alpha_{i-j_1}T}\int_0^T e^{\alpha_{i-j_1}\tau}f_{i-k-1}(\tau)d\tau}{\prod_{j_3=i-k-1,\,j_3\neq i-j_2}^{j_3=i}\left(\alpha_{j_3}-\alpha_{i-j_2}\right)} \quad \text{(A34)}$$

Putting Eq. (A34) into Eq. (A2) we can obtain the following form:

$$q_i(\xi_l,n,T)=\sum_{k=0}^{k=i-2}\beta_{i-k-1}\prod_{j_1=0}^{j_1=k}\sigma_{i-j_1}\sum_{j_2=0}^{j_2=k+1}\frac{e^{-\alpha_{i-j_1}T}\int_0^T e^{\alpha_{i-j_1}\tau}f_{i-k-1}(\tau)d\tau}{\prod_{j_3=i-k-1,\,j_3\neq i-j_2}^{j_3=i}\left(\alpha_{j_3}-\alpha_{i-j_2}\right)} \quad \text{(A35)}$$

Thus, the final solution can be expressed as Eq.(13) with the corresponding functions defined in Eqs.(14) and (15).

Note that Eq. (A33) is invalid for some of α_{i-j_2} being identical. For such conditions, we can still reduce $\frac{1}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}$ to a sum of partial fraction expansion. However, it will lead to different Laplace inverse formulae. For example, the following formulae is used for all α_{i-j_2} being identical

$$L^{-1}\left[\frac{1}{\prod_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j}\right)}\right]=\frac{T^k e^{-\alpha_{i-j_2}T}}{k!} \quad \text{(A36)}$$

The generalized formulae for the cases with some of α_{i-j_2} being identical will not be provided herein because there are a large number of combinations of α_{i-j_2}. We suggest that the readers can pursue the solutions by following the similar steps for such specific conditions case by case . ***[response to comment of referee # 1]***

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Table 1

Transport parameters used for convergence test example 1 involving the four-species radionuclide decay chain problem used by van Genuchten (1985)

Parameter	Value
Domain length, L [m]	250
Domain width, W [m]	100
Seepage velocity, v [m year^{-1}]	100
Longitudinal Dispersion coefficient, D_L [m^2 year^{-1}]	1,000
Transverse Dispersion coefficient, D_T [m^2 year^{-1}]	100
Retardation coefficient, R_i	
^{238}Pu	10,000
^{234}U	14,000
^{230}Th	50,000
^{226}Ra	500
Decay constant, k_i [year^{-1}]	
^{238}Pu	0.0079
^{234}U	0.0000028
^{230}Th	0.0000087
^{226}Ra	0.00043
Source decay constant, λ_m [year^{-1}]	
^{238}Pu	0.0089
^{234}U	0.00100280
^{230}Th	0.00100870
^{226}Ra	0.00143

Table 2

Values for coefficients of Bateman-type boundary source for four-species transport problem used by van Genuchten (1985)

Species, *i*	b_{im}			
	m=1	*m=2*	*m=3*	*m=4*
^{238}Pu , *i=1*	1.25			
^{234}U , *i=2*	-1.25044	1.25044		
^{230}Th , *i=3*	0.443684×10^{-3}	0.593431	-0.593874	
^{226}Ra , *i=4*	-0.516740×10^{-6}	0.120853×10^{-1}	-0.122637×10^{-1}	0.178925×10^{-3}

Table 3

Solution convergence of each species concentration at transect of inlet boundary ($x = 0$) for four-species radionuclide transport problem considering simulated domain of $L = 250$ m, $W = 100$ m, subject to Bateman-type sources located at $40\,m \le y \le 60\,m$ for t = 1,000 year (*M* = number of terms summed for inverse generalized integral transform; *N* = number of terms summed for inverse finite Fourier cosine transform). When we investigate the required *M* for inverse generalized integral transform, *N*=16,000 for the finite Fourier cosine transform inverse are used. When we investigate the required *N* for inverse finite Fourier cosine transform, *M*=1,600 for the generalized transform inverse are used.

^{238}Pu

x [m]	y [m]	M =100	M =200	M =400	M =800	M =1,600
0	30	2.714E-07	2.712E-07	2.711E-07	2.710E-07	2.710E-07
0	34	3.412E-06	3.412E-06	3.411E-06	3.411E-06	3.411E-06
0	38	2.677E-05	2.677E-05	2.677E-05	2.677E-05	2.677E-05
0	46	1.608E-04	1.609E-04	1.609E-04	1.609E-04	1.609E-04
0	50	1.637E-04	1.637E-04	1.637E-04	1.637E-04	1.637E-04
x [m]	y [m]	N =1,000	N =2,000	N =4,000	N =8,000	N =16,000
0	30	2.723E-07	2.713E-07	2.711E-07	2.710E-07	2.710E-07
0	34	3.413E-06	3.412E-06	3.411E-06	3.411E-06	3.411E-06
0	38	2.677E-05	2.677E-05	2.677E-05	2.677E-05	2.677E-05
0	46	1.609E-04	1.609E-04	1.609E-04	1.609E-04	1.609E-04
0	50	1.637E-04	1.637E-04	1.637E-04	1.637E-04	1.637E-04

^{234}U

x [m]	y [m]	M =25	M =50	M =100	M =200	M =400
0	32	1.092E-03	1.091E-03	1.090E-03	1.090E-03	1.090E-03
0	34	4.829E-03	4.827E-03	4.826E-03	4.826E-03	4.825E-03
0	38	5.745E-02	5.753E-02	5.753E-02	5.753E-02	5.753E-02
0	46	3.999E-01	4.004E-01	4.005E-01	4.005E-01	4.005E-01
0	50	4.044E-01	4.049E-01	4.049E-01	4.049E-01	4.049E-01
x [m]	y [m]	N =500	N =1,000	N =2,000	N =4,000	N =8,000
0	32	1.107E-03	1.094E-03	1.091E-03	1.090E-03	1.090E-03
0	34	4.850E-03	4.831E-03	4.827E-03	4.826E-03	4.825E-03
0	38	5.761E-02	5.755E-02	5.753E-02	5.753E-02	5.752E-02

0	46	4.0005E-01	4.005E-01	4.005E-01	4.005E-01	4.005E-01
0	50	4.049E-01	4.049E-01	4.049E-01	4.049E-01	4.049E-01

^{230}Th

x [m]	y [m]	M =100	M =200	M =400	M =800	M =1,600
0	34	1.498E-06	1.495E-06	1.493E-06	1.492E-06	1.492E-06
0	38	4.269E-05	4.267E-05	4.267E-05	4.266E-05	4.266E-05
0	42	6.847E-04	6.848E-04	6.848E-04	6.848E-04	6.848E-04
0	46	7.259E-04	7.260E-04	7.260E-04	7.260E-04	7.260E-04
0	50	7.273E-04	7.274E-04	7.274E-04	7.274E-04	7.274E-04
x [m]	y [m]	N =1,000	N =2,000	N =4,000	N =8,000	N =16,000
0	34	1.514E-06	1.497E-06	1.493E-06	1.492E-06	1.492E-06
0	38	4.274E-05	4.268E-05	4.267E-05	4.266E-05	4.266E-05
0	42	6.847E-04	6.848E-04	6.848E-04	6.848E-04	6.848E-04
0	46	7.259E-04	7.260E-04	7.260E-04	7.260E-04	7.260E-04
0	50	7.274E-04	7.274E-04	7.274E-04	7.274E-04	7.274E-04

^{226}Ra

x [m]	y [m]	M =50	M =100	M =200	M =400	M =800
0	18	3.084E-08	3.082E-08	3.082E-08	3.081E-08	3.081E-08
0	24	1.294E-07	1.293E-07	1.293E-07	1.293E-07	1.293E-07
0	28	3.492E-07	3.492E-07	3.492E-07	3.492E-07	3.492E-07
0	44	2.217E-05	2.222E-05	2.223E-05	2.223E-05	2.223E-05
0	50	2.425E-05	2.430E-05	2.431E-05	2.431E-05	2.431E-05
x [m]	y [m]	N =1,000	N =2,000	N =4,000	N =8,000	N =16,000
0	18	3.086E-08	3.082E-08	3.082E-08	3.081E-08	3.081E-08
0	24	1.294E-07	1.293E-07	1.293E-07	1.293E-07	1.293E-07
0	28	3.493E-07	3.492E-07	3.492E-07	3.492E-07	3.492E-07
0	44	2.223E-05	2.223E-05	2.223E-05	2.223E-05	2.223E-05
0	50	2.431E-05	2.431E-05	2.431E-05	2.431E-05	2.431E-05

Table 4

Solution convergence of each species concentration at transect of $x = 25$ m for four-species radionuclide transport problem considering simulated domain of $L = 250$ m, $W = 100$ m, subject to Bateman-type sources located at $40\,m \le y \le 60\,m$ for t = 1,000 year (M = number of terms summed for inverse generalized integral transform; N = number of terms summed for inverse finite Fourier cosine transform). When we investigate the required M for inverse generalized integral transform, N=160 for the finite Fourier cosine transform inverse are used. When we investigate the required N for inverse finite Fourier cosine transform, M=1,600 for the generalized transform inverse are used.

^{238}Pu

x [m]	y [m]	M =100	M =200	M =400	M =800	M =1,600
25	28	5.531E-08	5.576E-08	5.580E-08	5.580E-08	5.580E-08
25	30	2.319E-07	2.312E-07	2.312E-07	2.311E-07	2.311E-07
25	38	1.106E-05	1.106E-05	1.106E-05	1.106E-05	1.106E-05
25	46	3.430E-05	3.430E-05	3.430E-05	3.430E-05	3.430E-05
25	50	3.616E-05	3.616E-05	3.616E-05	3.616E-05	3.616E-05
x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	28	-7.841E-07	9.961E-08	5.579E-08	5.580E-08	5.580E-08
25	30	-4.063E-07	2.616E-07	2.312E-07	2.311E-07	2.311E-07
25	38	1.195E-05	1.114E-05	1.106E-05	1.106E-05	1.106E-05
25	46	3.404E-05	3.441E-05	3.430E-05	3.430E-05	3.430E-05
25	50	3.817E-05	3.606E-05	3.616E-05	3.616E-05	3.616E-05

^{234}U

x [m]	y [m]	M =100	M =200	M =400	M =800	M =1,600
25	30	9.734E-05	9.612E-05	9.594E-05	9.592E-05	9.592E-05
25	34	1.727E-03	1.725E-03	1.724E-03	1.724E-03	1.724E-03
25	38	1.167E-02	1.167E-02	1.167E-02	1.167E-02	1.167E-02
25	46	4.023E-02	4.024E-02	4.024E-02	4.024E-02	4.024E-02
25	50	4.177E-02	4.178E-02	4.178E-02	4.178E-02	4.178E-02
x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	30	-9.427E-04	1.728E-04	9.610E-05	9.592E-05	9.592E-05
25	34	3.154E-03	1.588E-03	1.725E-03	1.724E-03	1.724E-03
25	38	1.324E-02	1.186E-02	1.167E-02	1.167E-02	1.167E-02

25	46	3.984E-02	4.049E-02	4.024E-02	4.024E-02	4.024E-02
25	50	4.487E-02	4.153E-02	4.178E-02	4.178E-02	4.178E-02

^{230}Th

x [m]	y [m]	M =100	M =200	M =400	M =800	M =1,600
25	30	1.822E-08	1.379E-08	1.312E-08	1.305E-08	1.305E-08
25	34	3.288E-07	3.207E-07	3.195E-07	3.193E-07	3.193E-07
25	38	2.766E-06	2.740E-06	2.735E-06	2.735E-06	2.735E-06
25	46	1.013E-05	1.015E-05	1.015E-05	1.015E-05	1.015E-05
25	50	1.043E-05	1.045E-05	1.045E-05	1.045E-05	1.045E-05

x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	30	-2.948E-07	4.484E-08	1.320E-08	1.305E-08	1.305E-08
25	34	7.000E-07	2.632E-07	3.196E-07	3.193E-07	3.193E-07
25	38	3.246E-06	2.816E-06	2.735E-06	2.735E-06	2.735E-06
25	46	1.005E-05	1.025E-05	1.015E-05	1.015E-05	1.015E-05
25	50	1.134E-05	1.035E-05	1.045E-05	1.045E-05	1.045E-05

^{226}Ra

x [m]	y [m]	M =25	M =50	M =100	M =200	M =400
25	10	2.681E-08	2.757E-08	2.767E-08	2.765E-08	2.765E-08
25	14	6.580E-08	6.665E-08	6.676E-08	6.674E-08	6.674E-08
25	18	1.606E-07	1.615E-07	1.617E-07	1.617E-07	1.617E-07
25	42	1.686E-05	1.658E-05	1.656E-05	1.656E-05	1.656E-05
25	50	2.315E-05	2.278E-05	2.277E-05	2.277E-05	2.277E-05

x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	10	-5.355E-08	3.027E-08	2.766E-08	2.765E-08	2.765E-08
25	14	7.068E-08	6.392E-08	6.675E-08	6.674E-08	6.674E-08
25	18	2.642E-07	1.640E-07	1.617E-07	1.617E-07	1.617E-07
25	42	1.624E-05	1.655E-05	1.656E-05	1.656E-05	1.656E-05
25	50	2.311E-05	2.275E-05	2.277E-05	2.277E-05	2.277E-05

Table 5

Solution convergence of each species concentration at transect of exit boundary ($x = 250$ m) for four-species radionuclide transport problem considering simulated domain of $L = 250$ m, $W = 100$ m subject to Bateman-type sources located at $40\,m \le y \le 60\,m$ for t = 1000 year (*M* = number of terms summed for inverse generalized integral transform and *N* = number of terms summed for inverse finite Fourier cosine transform). When we investigate the required *M* for inverse generalized integral transform, *N*=16 for the finite Fourier cosine transform inverse are used. When we investigate the required *N* for inverse finite Fourier cosine transform, *M*=6,400 for the generalized transform inverse are used.

^{226}Ra

x [m]	y [m]	M =400	M =800	M =1,600	M =3,200	M =6,400
250	2	2.289E-08	1.842E-08	1.814E-08	1.812E-08	1.812E-08
250	14	5.617E-08	5.060E-08	5.025E-08	5.022E-08	5.022E-08
250	26	1.528E-07	1.420E-07	1.413E-07	1.413E-07	1.413E-07
250	38	3.757E-07	2.743E-07	2.678E-07	2.674E-07	2.674E-07
250	50	1.645E-07	3.208E-07	3.306E-07	3.312E-07	3.312E-07

x [m]	y [m]	N =1	N =2	N =4	N =8	N =16
250	2	1.529E-07	-1.848E-09	1.892E-08	1.812E-08	1.812E-08
250	14	1.529E-07	5.348E-08	4.946E-08	5.022E-08	5.022E-08
250	26	1.529E-07	1.627E-07	1.414E-07	1.413E-07	1.413E-07
250	38	1.529E-07	2.666E-07	2.680E-07	2.674E-07	2.674E-07
250	50	1.529E-07	3.089E-07	3.303E-07	3.312E-07	3.312E-07

Table 6

Solution convergence of each species concentration at transect of inlet boundary ($x = 0$ m) for four-species radionuclide transport problem considering simulated domain of $L = 2{,}500$ m, $W = 100$ m subject to Bateman-type sources located at $45\,m \le y \le 55\,m$ for t = 1,000 year (M = number of terms summed for inverse generalized integral transform; N = number of terms summed for inverse finite Fourier cosine transform). When we investigate the required M for inverse generalized integral transform, N=12,800 for the finite Fourier cosine transform inverse are used. When we investigate the required N for inverse finite Fourier cosine transform, M=6,400 for the generalized transform inverse are used.

^{238}Pu

x [m]	y [m]	M =400	M =800	M =1,600	M =3,200	M =6,400
0	36	5.395E-07	5.391E-07	5.389E-07	5.387E-07	5.387E-07
0	38	1.908E-06	1.908E-06	1.908E-06	1.907E-06	1.907E-06
0	42	1.640E-05	1.642E-05	1.642E-05	1.642E-05	1.642E-05
0	46	1.203E-04	1.199E-04	1.198E-04	1.198E-04	1.198E-04
0	50	1.522E-04	1.524E-04	1.525E-04	1.525E-04	1.525E-04
x [m]	y [m]	N =2,000	N =4,000	N =8,000	N =16,000	N =32,000
0	36	5.392E-07	5.389E-07	5.388E-07	5.387E-07	5.387E-07
0	38	1.908E-06	1.908E-06	1.907E-06	1.907E-06	1.907E-06
0	42	1.642E-05	1.642E-05	1.642E-05	1.642E-05	1.642E-05
0	46	1.198E-04	1.198E-04	1.198E-04	1.198E-04	1.199E-04
0	50	1.525E-04	1.525E-04	1.525E-04	1.525E-04	1.525E-04

^{234}U

x [m]	y [m]	M =800	M =1,600	M =3,200	M =6,400	M =12,800
0	36	4.817E-04	4.815E-04	4.815E-04	4.814E-04	4.814E-04
0	38	2.348E-03	2.348E-03	2.348E-03	2.348E-03	2.348E-03
0	44	1.011E-01	1.012E-01	1.012E-01	1.012E-01	1.012E-01
0	48	3.704E-01	3.705E-01	3.705E-01	3.705E-01	3.705E-01
0	50	3.862E-01	3.864E-01	3.864E-01	3.864E-01	3.864E-01
x [m]	y [m]	N =4,000	N =8,000	N =16,000	N =32,000	N =64,000
0	36	4.818E-04	4.816E-04	4.815E-04	4.814E-04	4.814E-04
0	38	2.348E-03	2.348E-03	2.348E-03	2.348E-03	2.348E-03

0	44	1.013E-01	1.013E-01	1.012E-01	1.012E-01	1.012E-01
0	48	3.705E-01	3.705E-01	3.705E-01	3.705E-01	3.705E-01
0	50	3.864E-01	3.864E-01	3.864E-01	3.864E-01	3.864E-01

^{230}Th

x [m]	y [m]	M =400	M =800	M =1,600	M =3,200	M =6,400
0	40	3.429E-06	3.427E-06	3.424E-06	3.423E-06	3.423E-06
0	42	1.773E-05	1.783E-05	1.782E-05	1.782E-05	1.782E-05
0	44	1.028E-04	1.089E-04	1.093E-04	1.093E-04	1.093E-04
0	48	7.095E-04	7.089E-04	7.090E-04	7.090E-04	7.090E-04
0	50	7.210E-04	7.205E-04	7.206E-04	7.206E-04	7.206E-04
x [m]	y [m]	N =2,000	N =4,000	N =8,000	N =16,000	N =32,000
0	40	3.430E-06	3.425E-06	3.424E-06	3.423E-06	3.423E-06
0	42	1.783E-05	1.782E-05	1.782E-05	1.782E-05	1.782E-05
0	44	1.093E-04	1.093E-04	1.093E-04	1.093E-04	1.093E-04
0	48	7.090E-04	7.090E-04	7.090E-04	7.090E-04	7.090E-04
0	50	7.206E-04	7.206E-04	7.206E-04	7.206E-04	7.206E-04

^{226}Ra

x [m]	y [m]	M =400	M =800	M =1,600	M =3,200	M =6,400
0	24	3.557E-08	3.556E-08	3.556E-08	3.555E-08	3.555E-08
0	28	9.276E-08	9.274E-08	9.273E-08	9.273E-08	9.273E-08
0	40	2.159E-06	2.159E-06	2.159E-06	2.159E-06	2.159E-06
0	44	7.739E-06	7.809E-06	7.813E-06	7.813E-06	7.813E-06
0	50	2.072E-05	2.082E-05	2.083E-05	2.084E-05	2.084E-05
x [m]	y [m]	N =1,000	N =2,000	N =4,000	N =8,000	N =16,000
0	24	3.559E-08	3.557E-08	3.556E-08	3.555E-08	3.555E-08
0	28	9.278E-08	9.275E-08	9.274E-08	9.273E-08	9.273E-08
0	40	2.159E-06	2.159E-06	2.159E-06	2.159E-06	2.159E-06
0	44	7.815E-06	7.814E-06	7.813E-06	7.813E-06	7.813E-06
0	50	2.084E-05	2.084E-05	2.084E-05	2.084E-05	2.084E-05

Table 7

Solution convergence of each species concentration at transect of $x = 250$ m for four-species radionuclide transport problem considering simulated domain of $L = 2{,}500$ m, $W = 100$ m subject to Bateman-type sources located at $45\,m \le y \le 55\,m$ for t = 1,000 year (M = number of terms summed for inverse generalized integral transform; N = number of terms summed for inverse finite Fourier cosine transform). When we investigate the required M for inverse generalized integral transform, N=160 for the finite Fourier cosine transform inverse are used. When we investigate the required N for inverse finite Fourier cosine transform, M=12,800 for the generalized transform inverse are used.

^{238}Pu

x [m]	y [m]	M =200	M =400	M =800	M =1,600	M =3,200
25	32	2.578E-08	2.569E-08	2.564E-08	2.563E-08	2.563E-08
25	34	1.153E-07	1.162E-07	1.161E-07	1.161E-07	1.161E-07
25	40	3.485E-06	3.661E-06	3.661E-06	3.661E-06	3.661E-06
25	46	2.262E-05	2.176E-05	2.163E-05	2.163E-05	2.163E-05
25	50	2.752E-05	2.920E-05	2.929E-05	2.929E-05	2.929E-05
x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	32	-7.217E-07	4.318E-08	2.558E-08	2.563E-08	2.563E-08
25	34	-1.422E-06	1.470E-07	1.162E-07	1.161E-07	1.161E-07
25	40	4.741E-06	3.665E-06	3.661E-06	3.661E-06	3.661E-06
25	46	2.175E-05	2.155E-05	2.163E-05	2.163E-05	2.163E-05
25	50	2.713E-05	2.938E-05	2.929E-05	2.929E-05	2.929E-05

^{234}U

x [m]	y [m]	M =200	M =400	M =800	M =1,600	M =3,200
25	34	3.937E-05	4.038E-05	4.022E-05	4.019E-05	4.019E-05
25	36	2.029E-04	2.162E-04	2.160E-04	2.159E-04	2.159E-04
25	42	5.649E-03	7.897E-03	7.936E-03	7.936E-03	7.936E-03
25	46	2.695E-02	2.593E-02	2.565E-02	2.564E-02	2.564E-02
25	50	2.913E-02	3.552E-02	3.585E-02	3.586E-02	3.586E-02
x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	34	-2.184E-03	1.134E-04	4.038E-05	4.019E-05	4.019E-05
25	36	-2.113E-03	1.975E-04	2.158E-04	2.159E-04	2.159E-04

25	42	1.118E-02	8.092E-03	7.936E-03	7.936E-03	7.936E-03
25	46	2.580E-02	2.544E-02	2.564E-02	2.564E-02	2.564E-02
25	50	3.262E-02	3.608E-02	3.586E-02	3.586E-02	3.586E-02

^{230}Th

x [m]	y [m]	M =800	M =1,600	M =3,200	M =6,400	M =12,800
25	36	3.192E-08	3.181E-08	3.180E-08	3.179E-08	3.179E-08
25	38	1.578E-07	1.576E-07	1.576E-07	1.576E-07	1.576E-07
25	44	3.838E-06	3.914E-06	3.914E-06	3.914E-06	3.914E-06
25	48	8.531E-06	8.539E-06	8.539E-06	8.539E-06	8.539E-06
25	50	9.253E-06	9.261E-06	9.261E-06	9.262E-06	9.262E-06
x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	36	-6.448E-07	2.862E-08	3.167E-08	3.179E-08	3.179E-08
25	38	-1.271E-07	1.141E-07	1.577E-07	1.576E-07	1.576E-07
25	44	4.705E-06	3.925E-06	3.914E-06	3.914E-06	3.914E-06
25	48	7.869E-06	8.534E-06	8.540E-06	8.539E-06	8.539E-06
25	50	8.345E-06	9.353E-06	9.261E-06	9.262E-06	9.262E-06

^{226}Ra

x [m]	y [m]	M =100	M =200	M =400	M =800	M =1600
25	12	1.268E-08	1.273E-08	1.272E-08	1.272E-08	1.272E-08
25	18	4.817E-08	4.822E-08	4.821E-08	4.821E-08	4.821E-08
25	26	2.830E-07	2.824E-07	2.824E-07	2.824E-07	2.824E-07
25	42	8.794E-06	7.484E-06	7.578E-06	7.579E-06	7.579E-06
25	50	1.761E-05	1.449E-05	1.494E-05	1.497E-05	1.497E-05
x [m]	y [m]	N =10	N =20	N =40	N =80	N =160
25	12	8.791E-08	1.264E-08	1.272E-08	1.272E-08	1.272E-08
25	18	-1.512E-07	4.713E-08	4.821E-08	4.821E-08	4.821E-08
25	26	5.221E-07	2.830E-07	2.824E-07	2.824E-07	2.824E-07
25	42	7.960E-06	7.587E-06	7.578E-06	7.579E-06	7.579E-06
25	50	1.458E-05	1.498E-05	1.494E-05	1.497E-05	1.497E-05

Table 8

Transport parameters used for verification example 2 involving the ten-species transport problem used by Srinivasan and Clement (2008b)

Parameter	Value
Domain length, L [m]	250
Domain width, W [m]	100
Seepage velocity, v [m year^{-1}]	5
Longitudinal Dispersion coefficient, D_L [m^2 year^{-1}]	50
Transverse Dispersion coefficient, D_T [m^2 year^{-1}]	50
Retardation coefficient, R_i i=1, 2,…,10	1.9, 1, 1.4, 1, 5, 8, 1.4, 3.1, 1, 1
Decay constant, k_i [year^{-1}] i=1, 2,…,10	3, 2, 1.5, 1.25, 2.75, 1, 0.75, 0.5, 0.25, 0.1
Source decay constant, λ_m [year^{-1}] m=1, 2,…,10	0.1, 0.75, 0.5, 0.25, 0, 0, 0.3, 1, 0, 0.65

Table 9

Coefficients of Bateman-type boundary source for ten-species transport problem used by Srinivasan and Clement (2008b)

Species, *i*	b_{im}									
	m=1	*m*=2	*m*=3	*m*=4	*m*=5	*m*=6	*m*=7	*m*=8	*m*=9	*m*=10
Species 1	10									
Species 2	0	5								
Species 3	0	0	2.5							
Species 4	0	0	0	0						
Species *5*	0	0	0	0	10					
Species *6*	0	0	0	0	0	5				
Species *7*	0	0	0	0	0	0	2.5			
Species *8*	0	0	0	0	0	0	0	0		
Species *9*	0	0	0	0	0	0	0	0	0	
Species *10*	0	0	0	0	0	0	0	0	0	0

Table 10

Transport parameters used for example application involving the five-species dissolved chlorinated solvent problem used by BIOCHLOR.

Parameter	Value
Domain length, L [m]	330.7
Domain width, W [m]	213.4
Seepage velocity, v [m year^{-1}]	34.0
Longitudinal dispersion coefficient, D_L [m^2 year^{-1}]	449
Transverse dispersion coefficient, D_T [m^2 year^{-1}]	44.9
Retardation coefficient, R_i [-]	
PCE	7.13
TCE	2.87
DCE	2.8
VC	1.43
ETH	5.35
Decay constant, k_i [year^{-1}]	
PCE	2
TCE	1
DCE	0.7
VC	0.4
ETH	0
Source decay rate constant, λ_m [year^{-1}]	
PCE	0
TCE	0
DCE	0
VC	0
ETH	0

Table 11

Coefficients of Bateman-type boundary source used for example application involving the five-species dissolved chlorinated solvent problem used by BIOCHLOR.

Species, *i*	b_{im}				
	m=1	*m=2*	*m=3*	*m=4*	*m=5*
PCE , *i=1*	0.056				
TCE , *i=2*		15.8			
DCE , *i=3*			98.5		
VC , *i=4*				3.08	
ETH , *i=5*					0.03

Figures Captions

Fig. 1. Schematic representation of two-dimensional transport of decaying contaminants in a uniform flow field with flux boundary source located at of the inlet boundary.

Fig. 2. Comparison of spatial concentration profiles of four species along the longitudinal direction (=50 m) at t = 1,000 years obtained from derived analytical solutions and numerical solutions for convergence test example 1 of four-member radionuclide decay chain $^{238}Pu \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$.

Fig. 3. Comparison of spatial concentration profiles of four species along the transverse direction (=0 m) at t = 1,000 years obtained from derived analytical solutions and numerical solutions for convergence test example 1 of four-member radionuclide decay chain $^{238}Pu \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$.

Fig. 4. Comparison of spatial concentration profiles of four species along the transverse direction (=25 m) at t = 1,000 years obtained from derived analytical solutions and numerical solutions for convergence test example 1 of four-member radionuclide decay chain $^{238}Pu \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$.

Fig. 5. Comparison of spatial concentration profiles of four species along the longitudinal direction (=50 m) at t = 1,000 years obtained from derived analytical solutions and numerical solutions for convergence test example 2 of four-member radionuclide decay chain $^{238}Pu \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$.

Fig. 6. Comparison of spatial concentration profiles of four species along the transverse direction (=0

m) at t = 1,000 years obtained from derived analytical solutions and numerical solutions for convergence test example 2 of four-member radionuclide decay chain $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$.

Fig. 7. Comparison of spatial concentration profiles of four species along the transverse direction (=25 m) at t = 1,000 years obtained from derived analytical solutions and numerical solutions for convergence test example 2 of four-member radionuclide decay chain $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$.

Fig. 8. Comparison of spatial concentration profiles of ten-species along x-direction at t = 20 days obtained from derived analytical solutions and numerical solutions for the test example 3 of ten species decay chain used by Srinivasan and Clement (2008b).

Fig. 9. Effects of physical processes and chemical reactions on the concentration contours of four-species at t = 1,000 years obtained from derived analytical solutions for four-member decay chain $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$.

Fig. 10. Spatial concentration contours of five-species at t = 1 year obtained from derived analytical solutions for natural attenuation of chlorinated solvent plumes $PCE \rightarrow TCE \rightarrow DCE \rightarrow VC \rightarrow ETH$.

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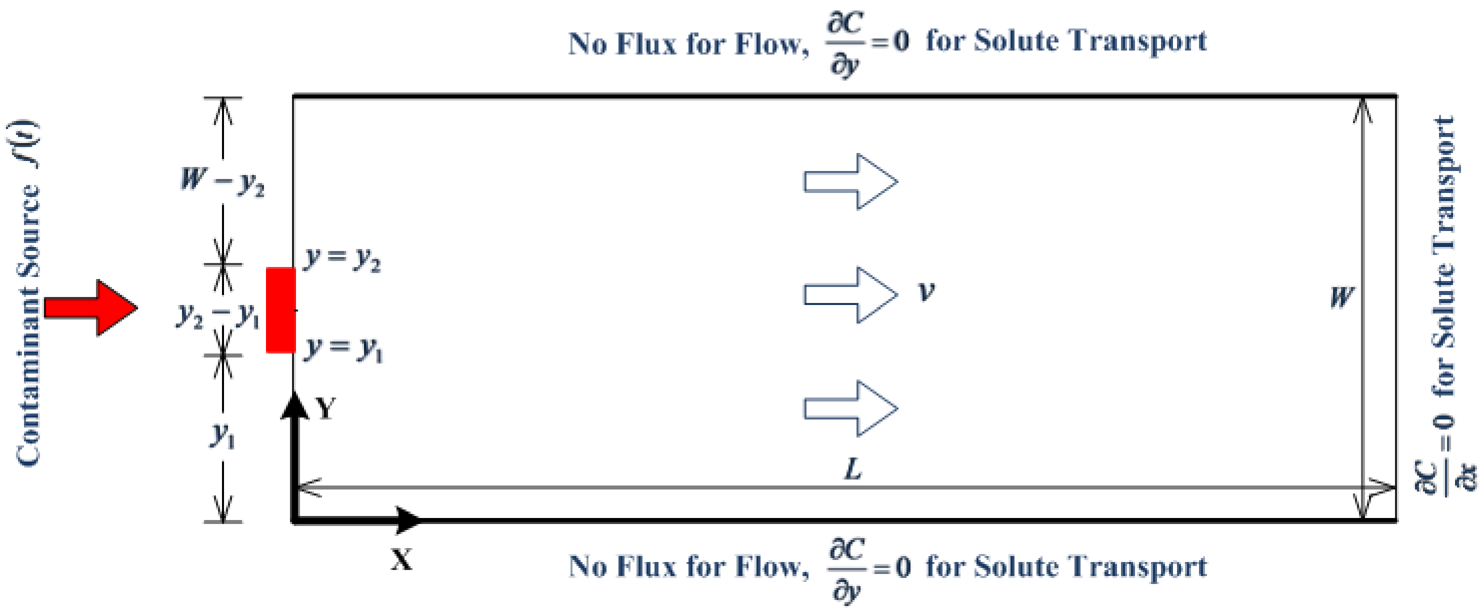
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Contaminant Source $f(t)$
$W-y_2$
$y=y_2$
y_2-y_1
$y=y_1$
y_1
Y
X
v
W
L
$\frac{\partial C}{\partial x}=0$ for Solute Transport
No Flux for Flow, $\frac{\partial C}{\partial y}=0$ for Solute Transport

Fig. 1.

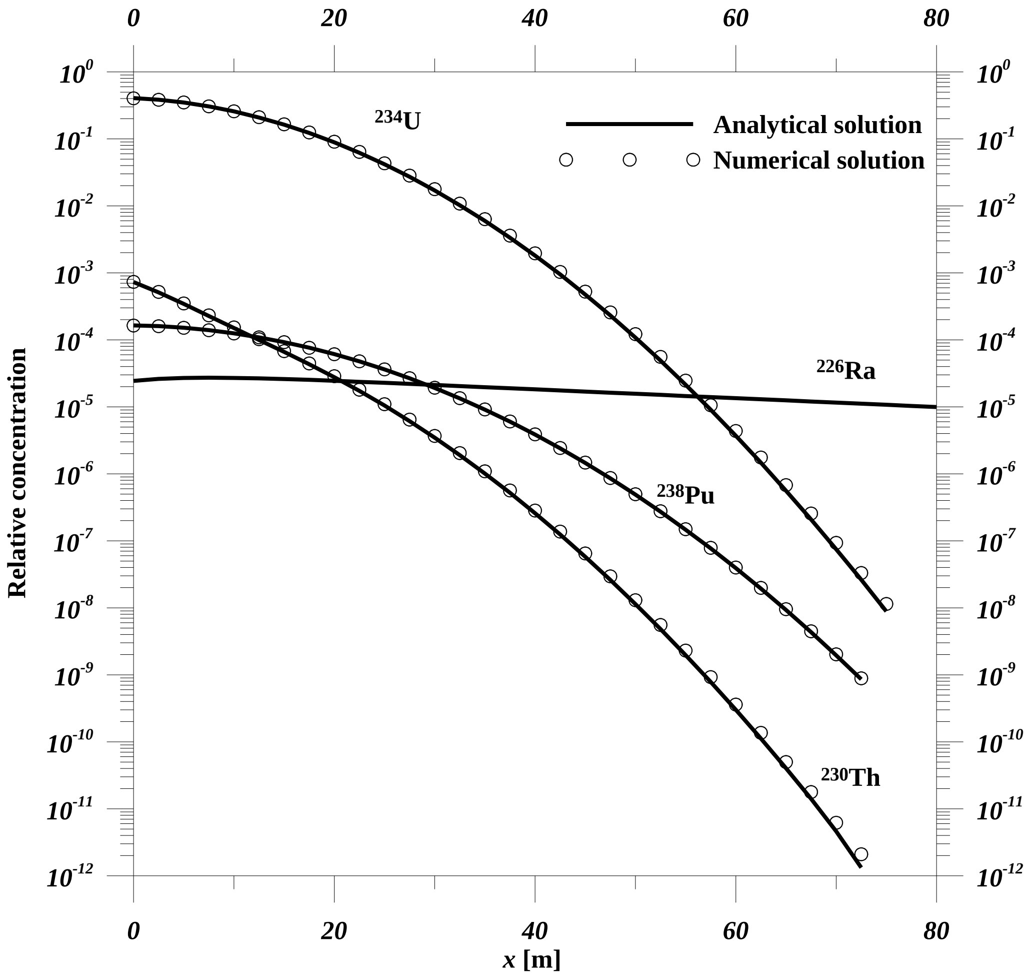
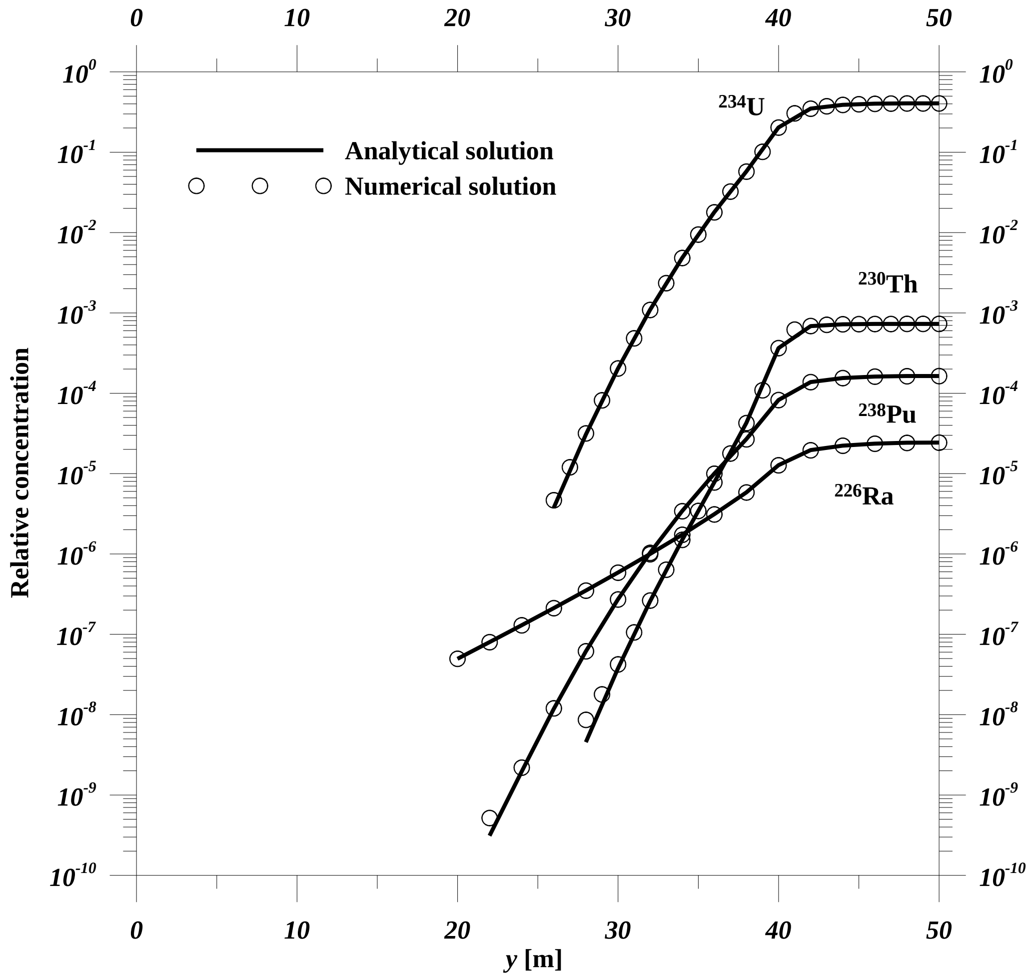

0
20
40
60
80
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-12}
Relative concentration
^{234}U
Analytical solution
Numerical solution
^{226}Ra
^{238}Pu
^{230}Th
x [m]

Fig. 2.



Analytical solution
Numerical solution
234U
230Th
238Pu
226Ra
Relative concentration
y [m]

Fig. 3

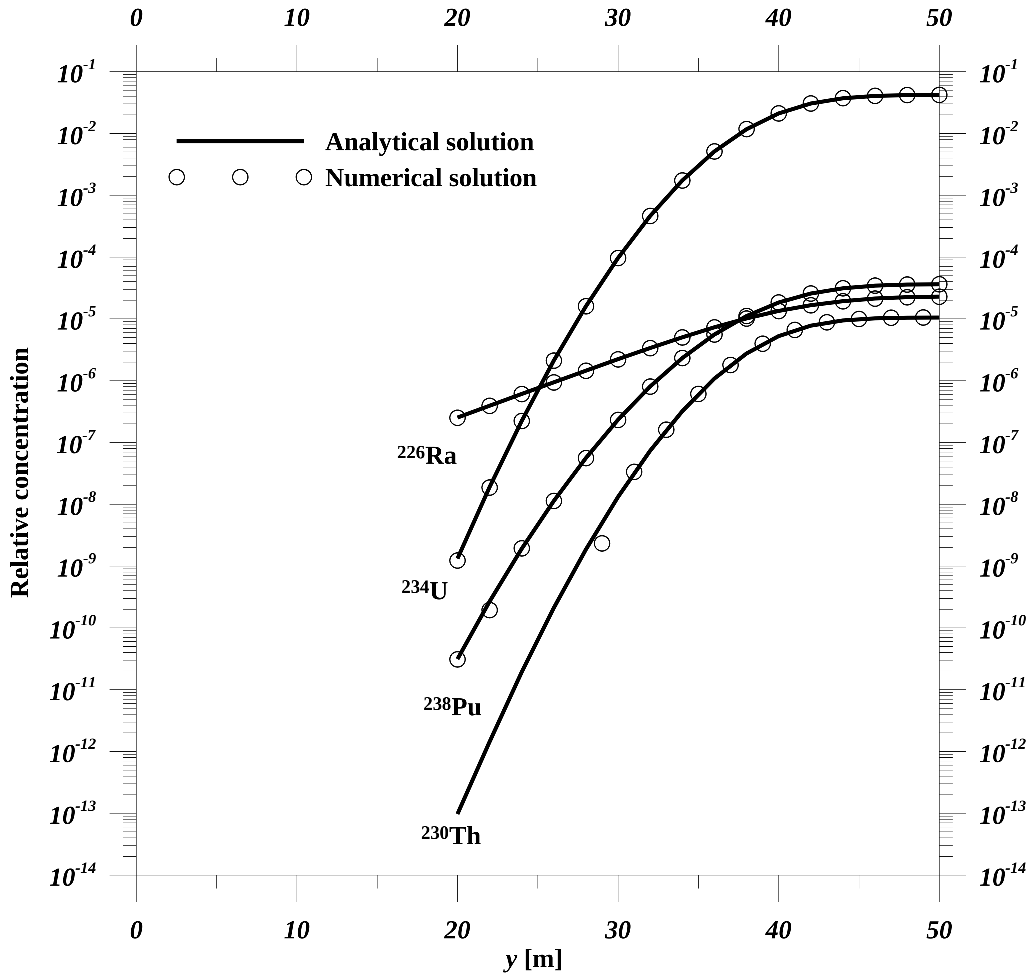
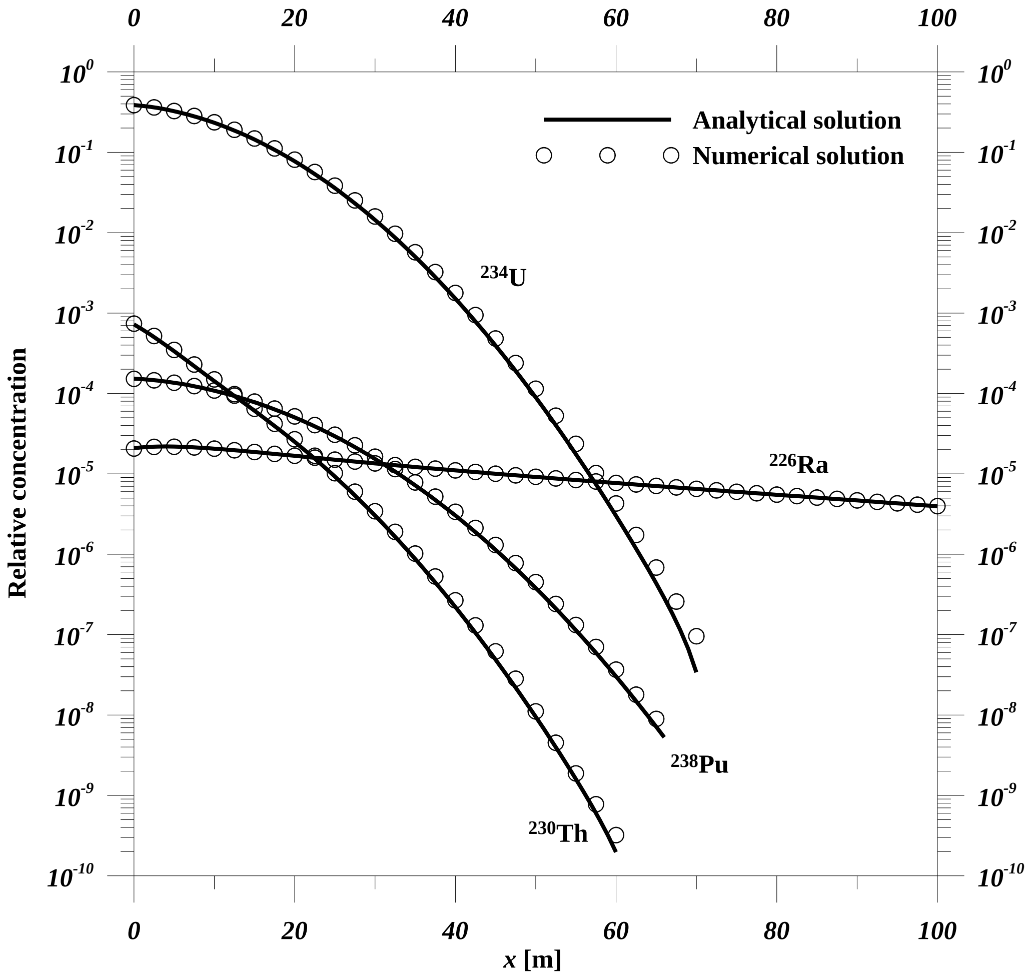

Analytical solution
Numerical solution
Relative concentration
y [m]
226Ra
234U
238Pu
230Th

Fig. 4



Analytical solution
Numerical solution
234U
226Ra
238Pu
230Th
Relative concentration
x [m]
0
20
40
60
80
100
10^0
10^-1
10^-2
10^-3
10^-4
10^-5
10^-6
10^-7
10^-8
10^-9
10^-10

Fig. 5

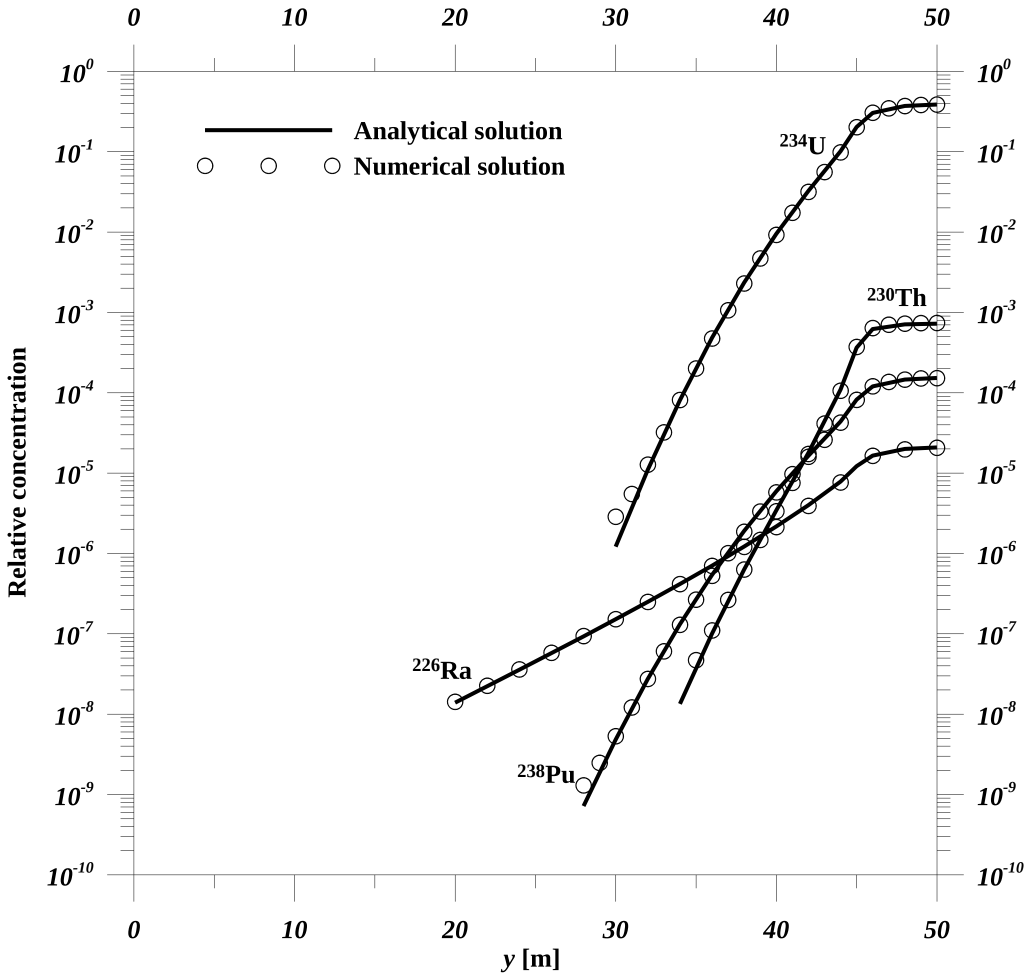
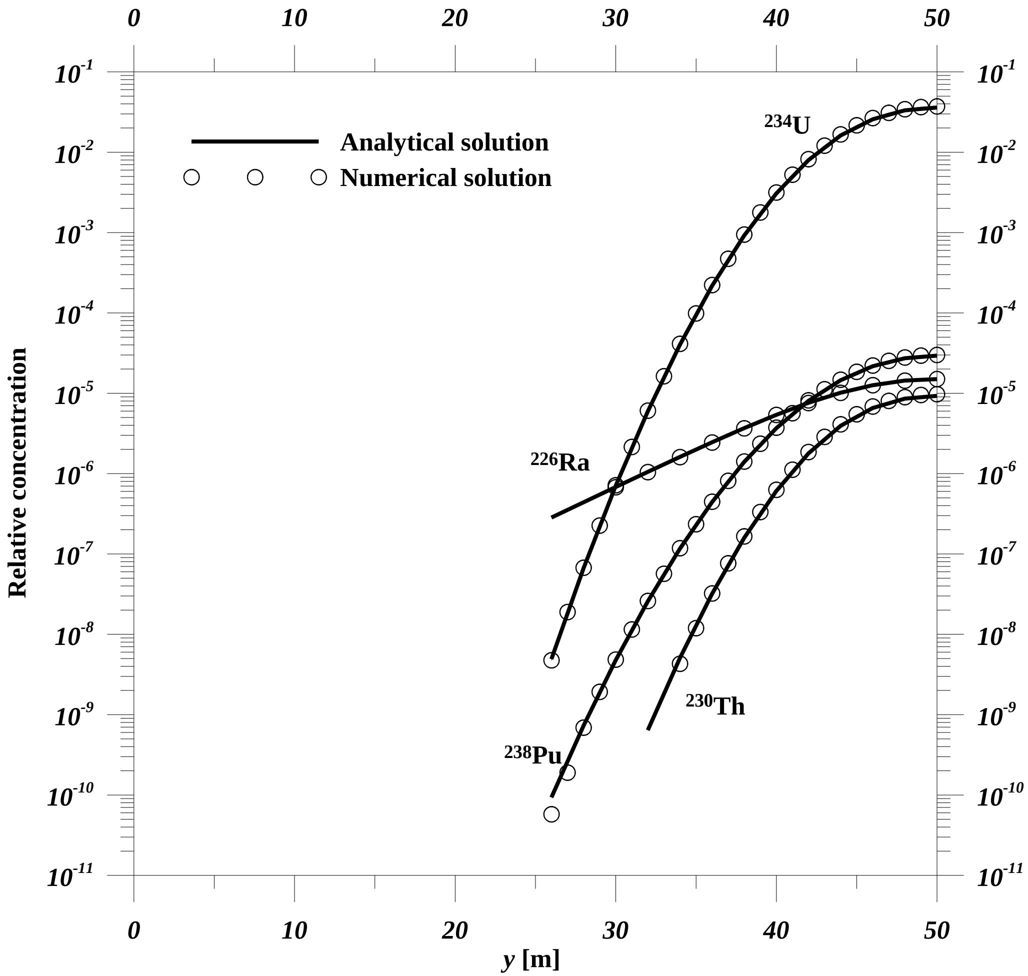
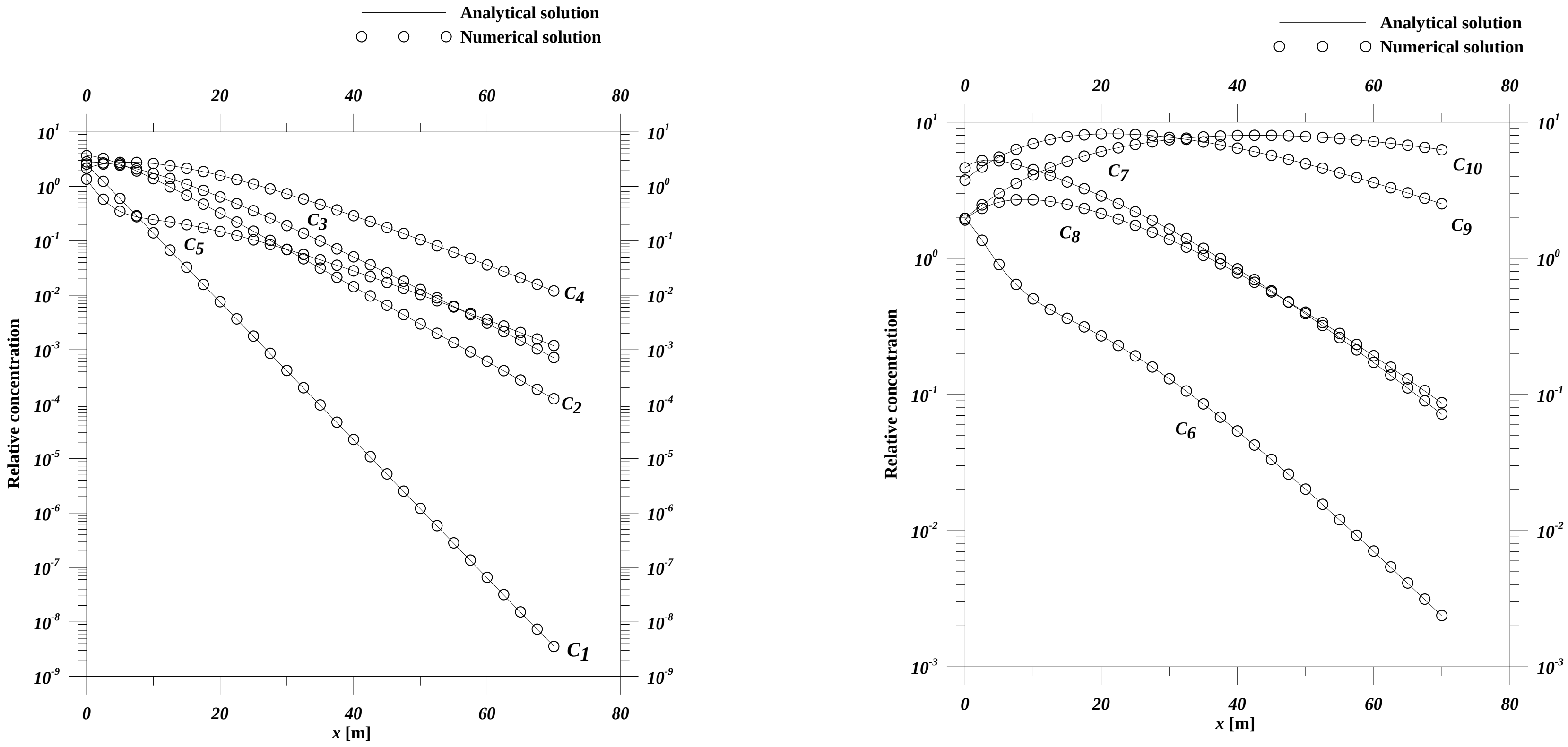

0
10
20
30
40
50
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
Analytical solution
Numerical solution
^{234}U
^{230}Th
^{226}Ra
^{238}Pu
Relative concentration
y [m]

Fig. 6



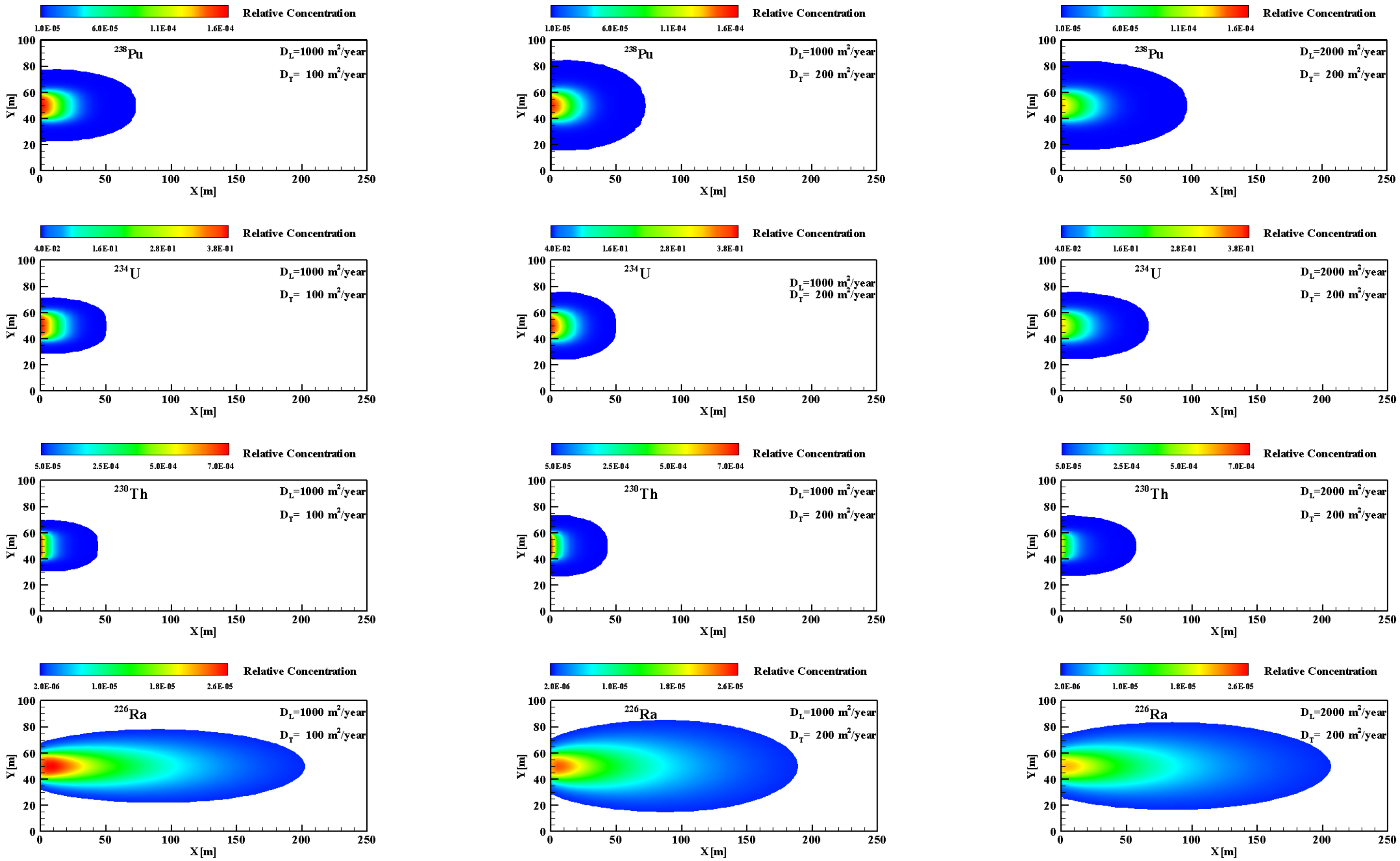
Analytical solution
Numerical solution
234U
226Ra
238Pu
230Th
Relative concentration
y [m]

Fig. 7



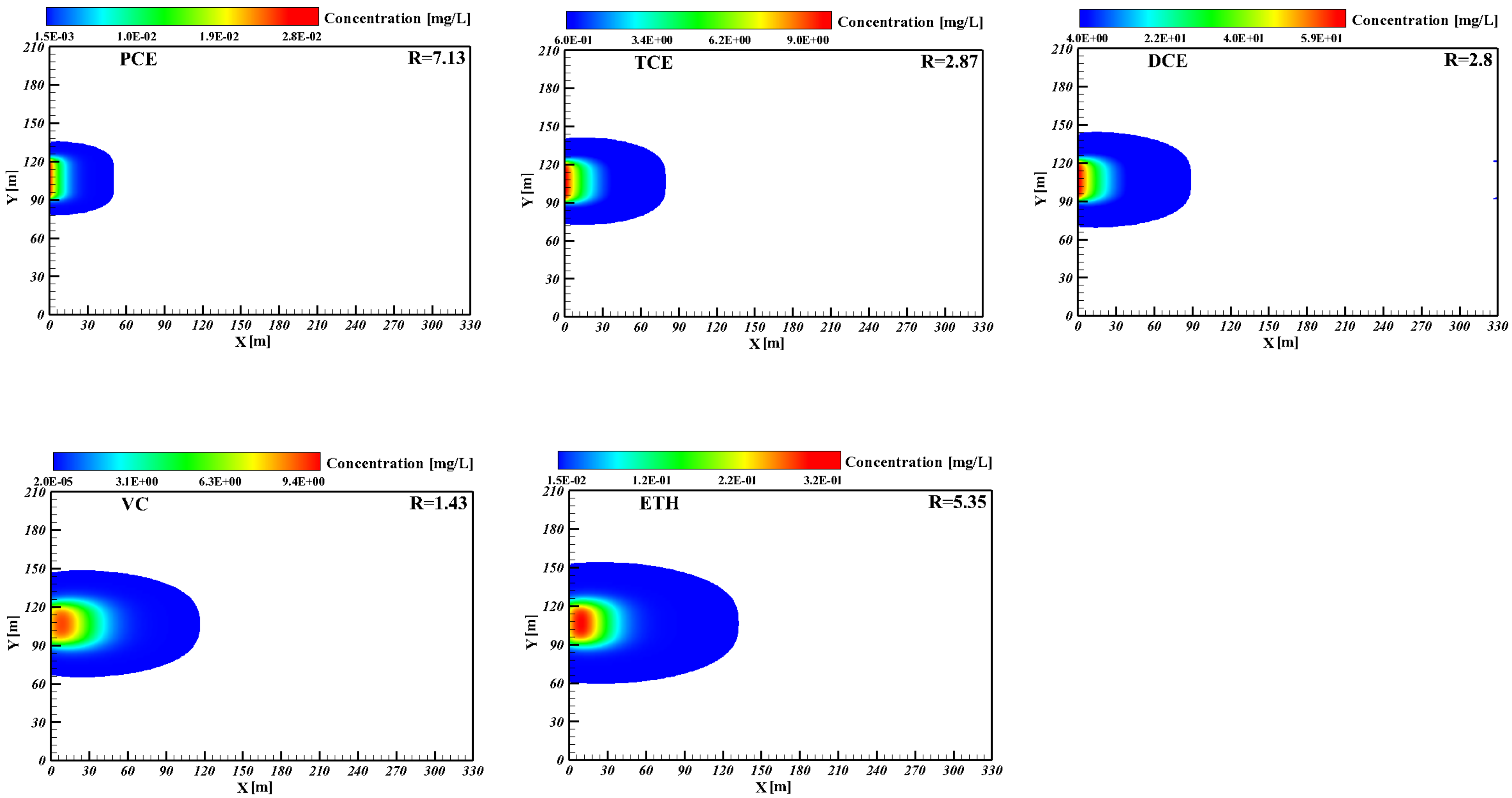
Analytical solution
Numerical solution
Relative concentration
x [m]
C_1
C_2
C_3
C_4
C_5
C_6
C_7
C_8
C_9
C_{10}

Fig. 8



Relative Concentration
238Pu
234U
230Th
226Ra
D_L=1000 m²/year
D_T= 100 m²/year
D_L=1000 m²/year
D_T= 200 m²/year
D_L=2000 m²/year
D_T= 200 m²/year
X [m]
Y [m]

Fig. 9



Concentration [mg/L]
1.5E-03
1.0E-02
1.9E-02
2.8E-02
PCE
R=7.13
Concentration [mg/L]
6.0E-01
3.4E+00
6.2E+00
9.0E+00
TCE
R=2.87
Concentration [mg/L]
4.0E+00
2.2E+01
4.0E+01
5.9E+01
DCE
R=2.8
Concentration [mg/L]
2.0E-05
3.1E+00
6.3E+00
9.4E+00
VC
R=1.43
Concentration [mg/L]
1.5E-02
1.2E-01
2.2E-01
3.2E-01
ETH
R=5.35
Y [m]
X [m]

Fig. 10