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

**Editor Decision: Publish subject to technical corrections** (30 Jan 2016) by Prof. Mauro Giudici

Comments to the Author:

I remain convinced that the term parsimonious is not used in the proper way; in fact parsimonious models are usually models which require few input parameters or simple approximations of the physical processes. Any analytical model is usually considered to be "parsimonious". However, this is a problem of minor importance from the scientific point of view.

<u>Response:</u>

**Thanks the constructive comment from editor. We have corrected the title as "**

Lines 364-365: please, check the sentence.

<u>Response</u>

This sentence has been corrected as "It should be noted the derived analytical model still has its application limitations. The model cannot handle the site with non-uniform groundwater flow or with multiple distinct zones."

# An analytical model for simulating two-dimensional multispecies plume migration

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**Abstract**

The two-dimensional advection-dispersion equations coupled with sequential first-order decay reactions involving arbitrary number of species in groundwater system is considered to predict the two-dimensional plume behavior of decaying contaminant such as radionuclide and dissolved chlorinated solvent. Generalized analytical solutions in compact format are derived through the sequential application of the Laplace, finite Fourier cosine, and generalized integral transform 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 with the numerical solutions. The analytical results indicate that the analytical solutions are robust, accurate and useful for simulation or screening tools to assess plume behaviors of decaying contaminants.

## 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

64    and Zhan, 2012), decomposition by change-of-variables with the help of existing single-species

65    analytical solutions (Sun and Clement, 1999; Sun et al., 1999a; 1999b), Laplace transform combined

66    with decomposition of matrix diagonization (Quezada et al., 2004; Srinivasan and Clememt, 2008a;

67    2008b), decomposition by change-of-variables coupled with generalized integral transform (Pérez

68    Guerrero et al., 2009; 2010), sequential integral transforms in association with algebraic decomposition

69    (Chen et al., 2012a; 2012b).

70    Multi-dimensional solutions are needed for real world applications, making them more attractive

71    than one-dimensional solutions. Bauer et al. (2001) presented the first set of semi-analytical solutions

72    for one-, two-, and three-dimensional coupled multispecies transport problem with distinct retardation

73    coefficients. Explicit analytical solutions were derived by Montas (2003) for multi-dimensional

74    advective-dispersive transport coupled with first-order reactions for a three-species transport system

75    with distinct retardation coefficients of species. Quezada et al. (2004) extended the Clement (2001)

76    strategy to obtain Laplace-domain solutions for an arbitrary decay chain length. Most recently, Sudicky

77    et al. (2013) presented a set of semi-analytical solutions to simulate the three-dimensional multi-

78    species transport subject to first-order chain-decay reactions involving up to seven species and four

79    decay levels. Basically, their solutions were obtained species by species using recursion relations

80    between target species and its predecessor species. For a straight decay chain, they derived solutions

81    for up to four species and no generalized expressions with compact formats for any target species were

82    obtained. Note that their solutions were derived for the first-type (Dirichlet) inlet conditions which

83    generally bring about physically improper mass conservation and significant errors in predicting the

84    concentration distributions especially for a transport system with a large longitudinal dispersion

85    coefficient (Barry and Sposito, 1988; Parlange et al., 1992). Moreover, in addition to some special

86    cases, the numerical Laplace transforms are required to obtain the original time domain solution.

87    Besides the straight decay chain, the analytical model by Clement (2001) and Sudicky (2013) can

4

88 account for more complicated decay chain problems such as diverging, converging and branched decay

89 chains.

90  Based on the aforementioned reviews, this study presents a parsimonious explicit analytical model

91 for two-dimensional multispecies transport coupled by a series of first-order decay reactions involving

92 an arbitrary number of species in groundwater system. The derived analytical solutions have four

93 salient features. First, the third-type (Robin) inlet boundary conditions which satisfy mass conservation

94 are considered. Second, the solution is explicit, thus solution can be easily evaluated without invoking

95 the numerical Laplace inversion. Third, the generalized solutions with parsimonious mathematical

96 structures are obtained and valid for any species of a decay chain. The parsimonious mathematical

97 structures of the generalized solutions are easy to code into a computer program for implementing the

98 solution computations for arbitrary target species. Fourth, the derived solutions can account for any

99 decay chain length. The explicit analytical solutions have applications for evaluation of concentration

100 distribution of arbitrary target species of the real-world decaying contaminants. The developed

101 parsimonious model is robustly verified with three example problems and applied to simulate the

102 multispecies plume migration of dissolved radionuclides and chlorinated solvent.

103

## 2. Governing equations and analytical solutions

*2.1 Derivation of analytical solutions*

106  This study consider the problem of decaying contaminant plume migration. The source zone is

107 located in the upstream of groundwater flow. The source zone can represent leaching of radionuclide

108 from a radioactive waste disposal facility or release of chlorinated solvent from the residual NAPL

109 phase into the aqueous phase. After these decaying contaminants enter the aqueous phase, they migrate

110 by one-dimensional advection with flowing groundwater and by simultaneously longitudinal and

111 transverse dispersion processes. While migrating in the groundwater system, the contaminants undergo

112 linear isothermal equilibrium sorption and a series of sequential first-order decaying reactions. Sudicky

113 et al. (2013) provided the detailed modeling scenario. The scenario considered in this study can be

114 ideally described as shown in Fig. 1. A steady and uniform velocity in the $x$ direction is considered

115 in Fig. 1. The governing equations describing two-dimensional reactive transport of the decaying

116 contaminants and their successor species undergoing linear isothermal equilibrium sorption and a series

117 of sequential first-order decaying reactions can be mathematically written as

118
$$D_L \frac{\partial^2 C_1(x,y,t)}{\partial x^2} - v \frac{\partial C_1(x,y,t)}{\partial x} + D_T \frac{\partial^2 C_1(x,y,t)}{\partial y^2} - k_1 R_1 C_1(x,y,t)$$
$$= R_1 \frac{\partial C_1(x,y,t)}{\partial t}$$
(1a)

119
$$D_L \frac{\partial^2 C_i(x,y,t)}{\partial x^2} - v \frac{\partial C_i(x,y,t)}{\partial x} + D_T \frac{\partial^2 C_i(x,y,t)}{\partial y^2} - k_i R_i C_i(x,y,t)$$
$$+ k_{i-1} R_{i-1} C_{i-1}(x,y,t) = R_i \frac{\partial C_i(x,y,t)}{\partial t} \qquad i = 2...N.$$
(1b)

120 where $C_i(x,y,t)$ is the aqueous concentration of species $i$ [**ML$^{-3}$**]; $x$ and $y$ are the spatial

121 coordinates in the groundwater flow and perpendicular directions [**L**], respectively; $t$ is time [**T**];

122 $D_L$ and $D_T$ represent the longitudinal and transverse dispersion coefficients [**L$^2$T$^{-1}$**], respectively;

123 $v$ is the average steady and uniform pore-water velocity [**LT$^{-1}$**]; $k_i$ is the first-order decay rate

124 constant of species $i$ [**T$^{-1}$**]; $R_i$ is the retardation coefficient of species $i$ [**-**]. Note that these equations

125 consider that the decay reactions occur simultaneously in both the aqueous and sorbed phases. If the

126 decay reactions occur only in the aqueous phase, the retardation coefficients in the decay terms in the

127 right-hand sides of Eqs. (1a) and (1b) become unity. For such case, $k_i$ and $k_{i-1}$ in the left-hand sides

128 could be modified as $\dfrac{k_i}{R_i}$ and $\dfrac{k_{i-1}}{R_{i-1}}$ to facilitate the application of the derived analytical solutions

129 obtained by Eqs. (1a) and (1b).

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

131    $C_i(x, y, t = 0) = 0 \quad 0 \leq x \leq L, 0 \leq y \leq W \qquad i = 1...N.$ (2)

132    $-D_L \dfrac{\partial C_i(x = 0, y, t)}{\partial x} + v C_i(x = 0, y, t) = v f_i(t)[H(y - y_1) - H(y - y_2)] \qquad t \geq 0 \qquad i = 1...N.$ (3)

133    $\dfrac{\partial C_i(x = L, y, t)}{\partial x} = 0 \qquad t \geq 0, 0 \leq y \leq W \qquad i = 1...N.$ (4)

134    $\dfrac{\partial C_i(x, y = 0, t)}{\partial y} = 0 \qquad t \geq 0, 0 \leq x \leq L \qquad i = 1...N.$ (5)

135    $\dfrac{\partial C_i(x, y = W, t)}{\partial y} = 0 \qquad t \geq 0, 0 \leq x \leq L \qquad i = 1...N.$ (6)

136    where $f_i(t)$ is the arbitrary time-dependent source concentration of species $i$ applied at the source

137    segment ($H(y - y_1) - H(y - y_2)$) at boundary ($x = 0$) which will be specified later [L], $H(\bullet)$ is the

138    Heaviside function, $L$ and $W$ are the length and width of the transport system under consideration

139    [L]. Eq. (2) implies that the transport system is free of solute mass at the initial time.

140    Eq. (3) means that a third-type boundary condition satisfying mass conservation at the inlet boundary

141    is considered. Eq. (4) considers the concentration gradient to be zero at the exit boundary based on

142    the mass conservation principle. Such a boundary condition has been widely used for simulating

143    solute transport in a finite-length system. Eqs. (5) and (6) assume no solute flux across the lower and

144    upper boundaries. It is noted that in Eq. (3), we assume arbitrary time-dependent sources of species $i$

145    uniformly distributed at the segment ($y_1 \leq y \leq y_2$) of the inlet boundary ($x = 0$), the so-called

146    Heaviside function source concentration profile. Relative to the first type boundary conditions used

147    by Sudicky et al. (2013), the third-type boundary conditions which satisfy mass conservation at the

148    inlet boundary (Barry and Sposito, 1988; Parlange et al., 1992) are used herein. Sudicky et al. (2013)

149    considered the source concentration profiles as Gaussian or Heaviside step functions. If Gaussain

150    distributions are desired, we can easily replace the Heaviside function in the right-hand side of Eq.

151  (3) with a Gaussian distribution.

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

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

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

155  $$C_i(X,Y,T=0) = 0 \quad 0 \le X \le 1, 0 \le Y \le 1 \qquad i = 1...N. \qquad (8)$$

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

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

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

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

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

161  Our solution strategy used is extended from the approach proposed by Chen at al. (2012a; 2012b).

162  The core of this approach is that the coupled partial differential equations are converted into an

163  algebraic equation system via a series of integral transforms and the solutions in the transformed

164  domain for each species are directly and algebraically obtained by sequential substitutions.

165  Following Chen et al. (2012a; 2012b), the generalized analytical solutions in compact formats can

166  be obtained as follows (with detailed derivation provided in Appendix A)

$$C_i(X,Y,T)$$

$$= f_i(T)\Phi(n=0) + e^{\frac{Pe_L}{2}X} \sum_{l=1}^{\infty} \frac{K(\xi_l,X)}{N(\xi_l)} [p_i(\xi_l,n,T) + q_i(\xi_l,n,T)]\Phi(n=0)\Theta(\xi_l) \tag{13}$$

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

where $\Phi(n) = \begin{cases} Y_2 - Y_1 & n = 0 \\ \dfrac{\sin(n\pi Y_2) - \sin(n\pi Y_1)}{n\pi} & n = 1,2,3... \end{cases}$, $\xi_l$ is the eigenvalue, determined from the

equation $\xi_l \cot \xi_l - \dfrac{\xi_l^2}{Pe_L} + \dfrac{Pe_L}{4} = 0$, $\Theta(\xi_l) = \dfrac{Pe_L \xi_l}{\dfrac{Pe_L^2}{4} + \xi_l^2}$, $K(\xi_l,X) = \dfrac{Pe_L}{2}\sin(\xi_l X) + \xi_l \cos(\xi_l X)$,

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

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

and

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

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

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

179    special-case inlet boundary sources.   Here the time-dependent decaying source which represents the

180    specific release mechanism defined by the Bateman equations (van Genuchten, 1985) is considered.

181    A Bateman-type source is described by

182
$$f_i(t) = \sum_{m=1}^{i} b_{im} e^{-\delta_m t} \tag{16a}$$

183    or in dimensionless form,

184
$$f_i(T) = \sum_{m=1}^{m=i} b_{in} e^{-\lambda_m T} \tag{16b}$$

185    The coefficients $b_{im}$ and $\delta_m = \mu_m + \gamma_m$ account for the first-order decay reaction rate ($\mu_m$) of each

186    species in the waste source and the release rate ($\gamma_m$) of each species from the waste source,

187    $\lambda_m = \dfrac{\delta_m L}{v}$ .

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

$C_i(X,Y,T)$

189
$$= \sum_{m=1}^{m=i} b_{im} e^{-\lambda_m T} \Phi(n=0) + e^{\frac{Pe_L}{2} X} \sum_{l=1}^{\infty} \frac{K(\xi_l,X)}{N(\xi_l)} \left[ p_i(\xi_l,n,T) + q_i(\xi_l,n,T) \right] \Phi(n=0)\Theta(\xi_l)$$
$$+ 2\sum_{n=1}^{n=\infty} \left\{ \sum_{m=1}^{m=i} b_{im} e^{-\lambda_m T} \Phi(n) + e^{\frac{Pe_L}{2} X} \sum_{l=1}^{\infty} \frac{K(\xi_l,X)}{N(\xi_l)} \left[ p_i(\xi_l,n,T) + q_i(\xi_l,n,T) \right] \Phi(n)\Theta(\xi_l) \right\} \cos(n\pi Y)$$

190                                                                                                         (17)

191    where

192
$$p_i(\xi_l,n,T) = \sum_{m=1}^{m=i} b_{i,m} \cdot e^{-\lambda_m T} - \beta_i \sum_{m=1}^{m=i} b_{i,m} \frac{e^{-\lambda_m T} - e^{-\alpha_i T}}{\alpha_i - \lambda_m} \tag{18}$$

193    and

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

195

### *2.2 Convergence behavior of the Bateman-type source solution*

197    Based on the special-case analytical solutions in Eq. (17) supported by two auxiliary functions,

198    defined in Eqs. (18) and (19), a computer code was developed in FORTRAN 90 language with double

199    precision. The details of the FORTRAN computer code are described in Supplement. The derived

200    analytical solutions in Eqs. (17)-(19) consist of summations of double infinite series expansions for

201    the finite Fourier cosine and generalized integral transform inversions, respectively. It is

202    straightforward to sum up these two infinite series expansions term by term. To avoid time-consuming

203    summations of these infinite series expansions, the convergence tests should be routinely executed to

204    determine the optimal number of the required terms for evaluating analytical solutions to the desired

205    accuracies.        Two-dimensional        four-member        radionuclide        decay        chain

206    $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$  is considered herein as convergence test example 1 to demonstrate

207    the convergence behavior of the series expansions. This convergence test example 1 is modified from

208    a one-dimensional radionuclide decay chain problem originated by Higashi and Pigford (1980) and

209    later applied by van Genuchten (1985) to illustrate the applicability of their derived solution.    The

210    important model parameters related to this test example are listed in Tables 1 and 2. The inlet source

211    is chosen to be symmetrical with respect to the  $x$ -axis and conveniently arranged in the

212    $40\,m \leq y \leq 60\,m$  segment at the inlet boundary.

213    In order to determine the optimal term number of series expansions for the finite Fourier cosine

214    transform inverse to achieve accurate numerical evaluation, we specify a sufficiently large number of

215 series expansions for the generalized transform inverse so that the influence of the number of series

216 expansions for the generalized integral transform inverse on convergence of series expansion for finite

217 Fourier cosine transform inverse can be excluded. A similar concept is used when investigating the

218 required number of terms in the series expansions for the generalized integral transform inverse. An

219 alternative approach is conducted by simultaneously varying the term numbers of series expansions

220 for the generalized integral transform inverse and the finite Fourier cosine transform inverse.

221     Tables 3, 4 and 5 give results of the convergence tests up to 3 decimal digits of the solution

222 computations along the three transects (inlet boundary at $x=0$ m, $x=25$ m, and exit boundary at $x$

223 $=250$m). In these tables $M$ and $N$ are defined as the numbers of terms summed for the generalized

224 integral transform inverse and finite Fourier cosine transform inverse, respectively. It is observed that

225 $M$ and $N$ are related closely to the true values of the solutions. For smaller true values, the solutions

226 must be computed with greater $M$ and $N$. However, convergences can be drastically speeded up if

227 lower calculation precision (e.g. 2 decimal digits accuracy) is acceptable. For example,

228 $(M,N) = (100,200)$ is sufficient for 2 decimal digits accuracy, while for 3 decimal digits accuracy we

229 need $(M,N) = (1600,8000)$. Two decimal digits accuracy is acceptable for most practical problems.

230 It is also found that $M$ increases and $N$ decreases with increasing $x$.

231     To further examine the series convergence behavior, example 2 considers a transport system of

232 large aspect ratio ($\dfrac{L}{W} = \dfrac{2,500m}{100m}$) and a narrower source segment, $45\,m \le y \le 55\,\mathrm{m}$, on the inlet

233 boundary. Tables 6 and 7 present results of the convergence tests of the solution computations along

234 two transects (inlet boundary and $x=250$ m). Tables 6 and 7 also show similar results for the

235 dependences of $M$ and $N$ on $x$. Note that larger $M$ and $N$ are required for each species in this

236 test example, suggesting that the evaluation of the solution for a large aspect ratio requires more series

237 expansion terms to achieve the same accuracy as compared to example 1. Detailed results of the

12

238     convergence test examples 1 and 2 are provided in Supplement.

239     Using the required numbers determined from the convergence test, the computational time for

240     evaluation of the solutions at 50 different observations only takes 3.782s, 11.325s, 23.95s and 67.23s

241     computer clock time on an Intel Core i7-2600 3.40 MHz PC for species 1, 2, 3, and 4 in the comparison

242     of example 1.

243

## 244     3. Results and discussion

245     *3.1 Comparison of the analytical solutions with the numerical solutions*

246     Three comparison examples are considered to examine the correctness and robustness of the

247     analytical solutions and the accuracy of the computer code. The first comparison example is the four-

248     member radionuclide transport problem used in the convergence test example 1. The second

249     comparison example considers the four-member radionuclide transport problem used in the

250     convergence test example 2. The third comparison example is used to test the accuracy of the computer

251     code for simulating the reactive contaminant transport of a long decay chain. The three comparison

252     examples are executed by comparing the simulated results of the derived analytical solutions with the

253     numerical solutions obtained using the Laplace transformed finite difference (LTFD) technique first

254     developed by Moridis and Reddell (1991). A computer code for the LTFD solution are written in

255     FORTTRAN language with double precision. The details of the FORTRAN computer code is

256     described in Supplement.

257     Figures 2, 3 and 4 depicts the spatial concentration distribution along one longitudinal direction

258     ( $y = 50$ m) and two transverse directions ( $x = 0$ m and $x = 25$ m) for convergence test example 1

259     at $t = 1,000$ year obtained from analytical solutions and numerical solutions. Figures 5, 6 and 7 present

260     the spatial concentration distribution along one longitudinal direction ( $y = 50$ m) and two transverse

261     directions ( $x = 0$ m and $x = 25$ m) for the convergence test example 2 at $t = 1,000$ year obtained

13

262    from analytical solutions and numerical solutions. Excellent agreements between the two solutions for

263    both examples are observed for a wide spectrum of concentration, thus warranting the accuracy and

264    robustness of the developed analytical model.

265        The third example involves a 10 species decay chain previously presented by Srinivasan and

266    Clement (2008a) to evaluate the performance of their one-dimensional analytical solutions. The

267    relevant model parameters are summarized in Tables 8 and 9. Our computer code is also compared

268    against the LTFD solutions for this example. Figure 8 depicts the spatial concentration distribution at

269    $t = 20$ days obtained analytically and numerically. Again there is excellent agreement between the

270    analytical and numerical solutions, demonstrating the performance of our computer code for

271    simulating transport problems with a long decay chain. The three comparison results clearly establish

272    the correctness of the analytical model and the accuracy and capability of the computer code.

273

274    *3.2 Assessing physical and chemical parameters on the radionuclide plume migration*

275        Physical processes and chemical reactions affect the extent of contaminant plumes, as well as

276    concentration levels. To illustrate how the physical processes and chemical reactions affect

277    multispecies plume development, we consider the four-member radionuclide decay chain used in the

278    previous convergence test and solution verification.   The model parameters are the same, except that

279    the longitudinal ( $D_L$ ) and transverse ( $D_T$ ) dispersion coefficients are varied. Three sets of

280    longitudinal and transverse dispersion coefficients  $D_L$ =1,000,  $D_T$ =100;  $D_L$ =1,000,  $D_T$  =200;

281    $D_L$ =2000,  $D_T$ =200 (all in m$^2$/year) are tested, all for a simulation time of 1,000 years.

282        Figure 9 illustrates the spatial concentration of four species at  $t = 1,000$ year for the three sets of

283    dispersion coefficients.   The mobility of plumes of $^{234}U$ and $^{230}Th$ is retarded because of their stronger

284    sorption ability. Hence the least retarded   $^{226}Ra$ plume extensively migrated to 200 m  ×  60 m area

14

285 in the simulation domain, whereas the $^{234}U$ and $^{230}Th$ plumes are confined within 60 m × 50 m area

286 in the simulation domain. The moderate mobility of $^{238}Pu$ reflects the fact that it is a medial sorbed

287 member of this radionuclide decay chain. The high concentration level of $^{234}U$ accounts for the high

288 first-order decay rate constant of its parent species $^{238}Pu$ and its own low first-order decay rate constant.

289 The plume extents and concentration levels may be sensitive to longitudinal and transverse dispersion.

290 Increase of the longitudinal and/or transverse dispersion coefficients enhances the spreading of the

291 plume extensively along the longitudinal and/or transverse directions, thereby lowering the plume

292 concentration level. Because the concentration levels of the four radionuclides are influenced by both

293 source release rates and decay chain reactions, $^{230}Th$ has the least extended plume area, while $^{226}Ra$

294 has the greatest plume area for all three set of dispersion coefficients. These dispersion coefficients

295 only affect the size of plumes of the four radionuclide, but the order of their relative plume size remains

296 the same (i.e. $^{226}Ra > {}^{238}Pu > {}^{234}U > {}^{230}Th$ for the simulated condition). Indeed, in the reactive

297 contaminant transport, the chemical parameters of sorption and decay rate are more important than the

298 physical parameters of dispersion coefficients that govern the order of the plume extents and the

299 concentration levels.

300

301 *3.3 Simulating the natural attenuation of chlorinated solvent plume migration*

302      Natural attenuation is the reduction in concentration and mass of the contaminant due to

303 naturally occurring processes in the subsurface environment. The process is monitored for regulatory

304 purposes to demonstrate continuing attenuation of the contaminant reaching the site-specific

305 regulatory goals within reasonable time, hence, the use of the term monitored natural attenuation

306 (MNA). MNA has been widely accepted as a suitable management option for chlorinated solvent

307 contaminated groundwater. Mathematical model are widely used to evaluate the natural attenuation

308 of plumes at chlorinated solvent sites. The multispecies transport analytical model developed in this

309    study provides an effective tool for evaluating performance of the monitoring natural attenuation of

310    plumes at a chlorinated solvent site because a series of daughter products produced during

311    biodegradation of chlorinated solvent such as $PCE{\rightarrow}TCE{\rightarrow}DCE{\rightarrow}VC{\rightarrow}ETH$. Thus simulation of

312    the natural attenuation of plumes a chlorinated solvent constitutes an attractive field application

313    example of our multispecies transport model.

314    A study of 45 chlorinated solvent sites by McGuire et al. (2014) found that mathematical

315    models were used at 60% of these sites and that the public domain model BIOCHLOR (Aziz et al.,

316    2000) provided by the Center for Subsurface Modeling Support (CSMoS) of USEPA was the most

317    commonly used model. An illustrated example from BIOCHLOR manual (Aziz et al., 2000) is

318    considered to demonstrate the application of the developed analytical model. This example

319    application demonstrated that BIOCHLOR can reproduce plume movement from 1965 to 1998 at the

320    contaminated site of Cape Canaveral Air Station, Florida. The simulation conditions and transport

321    parameters for this example application are summarized in Table 10. Constant source concentrations

322    rather than exponentially declining source concentration of five-species chlorinated solvents are

323    specified in the $90.7\,m \le y \le 122.7\,m$ segment at the inlet boundary ( $x = 0$ ). This means that the

324    exponents ( $\lambda_{im}$ ) of Bateman-type sources in Eqs. (16a) or (16b) need to be set to zero for the constant

325    source concentrations and source intensity constants ( $b_{im}$ ) are set to zero when subscript $i$ does not

326    equal to subscript $m$. Table 11 lists the coefficients of Bateman-type boundary source used for this

327    example application involving the five-species dissolved chlorinated solvent problem. Spatial

328    concentration contours of five-species at $t = 1$ year obtained from the derived analytical solutions for

329    natural attenuation of chlorinated solvent plumes are depicted in Fig. 10. It is observed that the

330    mobility of plumes is quite sensitive to the species retardation factors, whereas the decay rate

331    constants determine the plume concentration level. The plumes can migrate over a larger region for

332    species having a low retardation factor such as VC. The low decay rate constants such as ETH have

16

333 higher concentration distribution than the VC. It should be noted that a larger extent of plume

334 observed for ETH in Fig. 10 is mainly attributed the plume mass accumulation from the predecessor

335 species VC that have a larger plume extent. The effect of high retardation of the ETH is hindered by

336 the mass accumulation of the predecessor species VC.

337

338 **4. Conclusions**

339 We present an analytical model with a parsimonious mathematical format for two-dimensional

340 multispecies advective-dispersive transport of decaying contaminants such as radionuclides,

341 chlorinated solvents and nitrogen. The developed model is capable of accounting for the temporal and

342 spatial development of an arbitrary number of sequential first-order decay reactions. The solution

343 procedures involve applying a series of Laplace, finite Fourier cosine and generalized integral

344 transforms to reduce a partial differential equation system to an algebraic system, solving for the

345 algebraic system for each species, and then inversely transforming the concentration of each species

346 in transformed domain into the original domain. Explicit special solutions for Bateman type source

347 problems are derived via the generalized analytical solutions. The convergence of the series expansion

348 of the generalized analytical solution is robust and accurate. These explicit solutions and the computer

349 code are comparing with the results computed by the numerical solutions. The two solutions agree well

350 for a wide spectrum of concentration variations for three test examples. The analytical model is applied

351 to assess the plume development of radionuclide and dissolved chlorinated solvent decay chain. The

352 results show that dispersion only moderately modifies the size of the plumes, without altering the

353 relative order of the plume sizes of different contaminant. It is suggested that retardation coefficients,

354 decay rate constants and the predecessor species plume distribution mainly govern the order of plume

355 size in groundwater. Although there are a number of numerical reactive transport models that can

356 account for multispecies advective-dispersive transport, our analytical model with a computer code

17

357    that can directly evaluate the two-dimensional temporal-spatial concentration distribution of arbitrary

358    target species without involving the computation of other species. The analytical model developed in

359    this study effectively and accurately predicts the two-dimensional radionuclide and dissolved

360    chlorinated plume migration. It is a useful tool for assessing the ecological and environmental impact

361    of the accidental radionuclide releases such as the Fukushima nuclear disaster where multiple

362    radionuclides leaked through the reactor, subsequently contaminating the local groundwater and ocean

363    seawater in the vicinity of the nuclear plant. It is also a screening model that simulates remediation by

364    natural attenuation of dissolved solvents at chlorinated solvent release sites. It should be noted the

365    derived analytical model still has its application limitations. The model cannot handle the site with

366    non-uniform groundwater flow or with multiple distinct zones. Furthermore, the developed model

367    cannot simulate the more complicated decay chain problems such as diverging, converging and

368    branched decay chains. The analytical model for more complicated decay chain problems can be

369    pursued in the near future.

370

371

372

373

374 **Appendix A**

375 **Derivation of analytical solutions**

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

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

378 $$\frac{1}{Pe_L}\frac{\partial^2 G_1(X,Y,s)}{\partial X^2} - \frac{\partial G_1(X,Y,s)}{\partial X} + \frac{\rho^2}{Pe_T}\frac{\partial^2 G_1(X,Y,s)}{\partial Y^2} - (R_1 s + \kappa_1)G_1(X,Y,s) = 0 \tag{A1a}$$

379 $$\frac{1}{Pe_L}\frac{\partial^2 G_i(X,Y,s)}{\partial X^2} - \frac{\partial G_i(X,Y,s)}{\partial X} + \frac{\rho^2}{Pe_T}\frac{\partial^2 G_i(X,Y,s)}{\partial Y^2} \qquad i = 2,3,...N \tag{A1b}$$
$$- \kappa_i G_i(X,Y,s) + \kappa_{i-1}G_{i-1}(X,Y,s) = R_i s G_i(X,Y,s)$$

380 $$-\frac{1}{Pe_L}\frac{\partial G_i(X=0,Y,s)}{\partial X} + G_i(X=0,Y,s) = F_i(s)\big[H(Y-Y_1)-H(Y-Y_2)\big] \ \ 0 \le Y \le 1 \ i=1...N.$$

381 $$\tag{A2}$$

382 $$\frac{\partial G_i(X=1,Y,s)}{\partial X} = 0 \qquad 0 \le Y \le 1 \quad i = 1...N. \tag{A3}$$

383 $$\frac{\partial G_i(X,Y=0,s)}{\partial Y} = 0 \qquad 0 \le X \le 1 \quad i = 1...N. \tag{A4}$$

384 $$\frac{\partial G_i(X,Y=1,s)}{\partial Y} = 0 \qquad 0 \le X \le 1 \quad i = 1...N. \tag{A5}$$

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

386 transformation relations as

387 $$G_i(X,Y,s) = \int_0^\infty e^{-sT} C_i(X,Y,T)dT \tag{A6}$$

388 $$F_i(s) = \int_0^\infty e^{-sT} f_i(T)dT \tag{A7}$$

389

390    The finite Fourier cosine transform is used here because it satisfies the transformed governing

19

391  equations in Eqs. (A1a) and (A2b) and their corresponding boundary conditions in Eqs. (A4) and (A5).

392  Application of the finite Fourier cosine transform on Eqs. (A1)-(A3) leads to

393
$$\frac{1}{Pe_L}\frac{d^2H_1(X,n,s)}{\partial X^2} - \frac{dH_1(X,n,s)}{dX} - \left(R_1 s + \kappa_1 + \frac{\rho^2 n^2 \pi^2}{Pe_T}\right)H_1(X,n,s) = 0 \tag{A8a}$$

394
$$\frac{1}{Pe_L}\frac{d^2H_i(X,n,s)}{dX^2} - \frac{dH_i(X,n,s)}{dX} - \left(R_i s + \kappa_i + \frac{\rho^2 n^2 \pi^2}{Pe_T}\right)H_i(X,n,s) + \kappa_{i-1}H_{i-1}(X,n,s) = 0 \tag{A8b}$$

395
$$-\frac{1}{Pe_L}\frac{dH_i(X=0,n,s)}{dX} + H_i(X=0,n,s) = F_i(s)\Phi(n) \tag{A9}$$

396
$$\frac{dH_i(X=1,n,s)}{dX} = 0 \tag{A10}$$

397  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

398  parameter, $H_i(X,n,s)$ is defined by the following conjugate equations (Sneddon, 1972)

399
$$H_i(X,n,s) = \int_0^1 G_i(X,Y,s)\cos(n\pi Y)dY \tag{A11}$$

400
$$G_i(X,Y,s) = H_i(X,n=0,s) + 2\sum_{n=1}^{n=\infty} H_i(X,n,s)\cos(n\pi Y) \tag{A12}$$

401  Using changes-of-variables, similar to those applied by Chen and Liu (2011), the advective terms

402  in Eqs. (A8a) and A(8b) as well as nonhomogeneous terms in Eq. (A9) can be easily removed. Thus,

403  substitutions of the change-of-variable into Eqs. (A8a), (A8b), (A9) and (A10) result in diffusive-type

404  equations associated with homogeneous boundary conditions

405
$$\frac{1}{Pe_L}\frac{d^2U_1(X,n,s)}{dX^2} - \left(R_1 s + \kappa_1 + \frac{\rho^2 n^2 \pi^2}{Pe_T} + \frac{Pe_L}{4}\right)U_1(X,n,s)$$
$$= e^{-\frac{Pe_L}{2}X}\left(R_1 s + \kappa_1 + \frac{\rho^2 n^2 \pi^2}{Pe_T}\right)F_1(s)\Phi(n) \tag{A13a}$$

406

$$\frac{1}{Pe_L}\frac{d^2 U_i(X,n,s)}{dX^2} - \left(\frac{Pe_L}{4} + R_i s + \kappa_1 + \frac{\rho^2 n^2 \pi^2}{Pe_T}\right)U_i(X,n,s)$$

$$= e^{-\frac{Pe_L}{2}X}\left(R_i s + \kappa_i + \frac{\rho^2 n^2 \pi^2}{Pe_T}\right)F_i(s)\Phi(n) - e^{-\frac{Pe_L}{2}X}\kappa_{i-1}F_{i-1}(s)\Phi(n) - \kappa_{i-1}U_{i-1}(X,n,s)$$

(A13b)

407

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

(A14)

408

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

(A15)

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

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

(A16)

411         As detailed in Ozisik (1989), the generalized integral transform pairs for Eqs. (A13a) and (A13b)

412    and its associated boundary conditions (A14) and (A15) are defined as

413    $$Z_i(\xi_l,n,s) = \int_0^1 K(\xi_l,X)U_i(X,n,s)dX$$

(A17)

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

(A18)

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

416    $\xi_l$   is the eigenvalue, determined from the equation

417    $$\xi_l\cot\xi_l - \frac{\xi_l^2}{Pe_L} + \frac{Pe_L}{4} = 0$$

(A19)

418

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

419    $$-\left(R_1 s + \kappa_1 + \frac{\rho^2 n^2 \pi^2}{Pe_T} + \frac{Pe_L}{4} + \frac{\xi_l^2}{Pe_L}\right)Z_i(\xi_l,n,s) = \left(R_1 s + \kappa_1 + \frac{\rho^2 n^2 \pi^2}{Pe_T}\right)F_1(s)\Phi(n)\Theta(\xi_l)$$

(A20)

21

$$-\left(R_i s + \kappa_i + \frac{\rho^2 n^2 \pi^2}{Pe_T} + \frac{Pe_L}{4} + \frac{\xi_l^2}{Pe_L}\right) Z_i(\xi_l, n, s)$$

420                                                                                              (A21)

$$= \left(R_i s + \kappa_i + \frac{\rho^2 n^2 \pi^2}{Pe_T}\right) F_i(s)\Phi(n)\Theta(\xi_l) - \kappa_{i-1} F_{i-1}(s)\Phi(n)\Theta(\xi_l) - \kappa_{i-1} Z_{i-1}(\xi_l, n, s)$$

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

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

423     to

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

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

        $Z_3(\xi_l, n, s) = \left[ -\dfrac{s + \alpha_3 - \beta_3}{s + \alpha_3} F_3(s) + \dfrac{\sigma_3 \beta_2}{(s + \alpha_3)(s + \alpha_2)} F_2(s) \right.$

426                                                                                              (A24)

        $\left. \dfrac{\sigma_3 \sigma_2 \beta_1}{(s + \alpha_3)(s + \alpha_2)(s + \alpha_1)} F_1 \right]\Phi(n)\Theta(\xi_l)$

        $Z_4(\xi_l, n, s) = \left[ -\dfrac{s + \alpha_4 - \beta_4}{s + \alpha_4} F_4(s) + \dfrac{\sigma_4 \beta_3}{(s + \alpha_4)(s + \alpha_3)} F_3(s) \right.$

427                                                                                              (A25)

        $\left. + \dfrac{\sigma_4 \sigma_3 \beta_2}{(s + \alpha_4)(s + \alpha_3)(s + \alpha_2)} F_2(s) + \dfrac{\sigma_4 \sigma_3 \sigma_2 \beta_1}{(s + \alpha_4)(s + \alpha_3)(s + \alpha_2)(s + \alpha_1)} F_1(s) \right]\Phi(n)\Theta(\xi_l)$

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

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

430     $Z_i(\xi_l, n, s) = \left[ P_i(\xi_l, n, s) + Q_i(\xi_l, n, s) \right]\Phi(n)\Theta(\xi_l)$     $i = 1,2...N$                         (A26)

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

432 The solutions in the original domain are obtained by a series of integral transform inversions in

433 combination with changes-of-variables.

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

435 $$W_i(X, n, s) = \sum_{m=1}^{\infty} \frac{K(\xi_l, X)}{N(\xi_l)} \left[P_i(\xi_l, n, s) + Q_i(\xi_l, n, s)\right] \Phi(n) \Theta(\xi_l) \tag{A27}$$

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

437 $$H_i(\xi_l, n, s) = F_i(s)\Phi(n) + e^{\frac{Pe_L}{2} x_D} \sum_{m=1}^{\infty} \frac{K(\xi_l, x_D)}{N(\xi_l)} \left[P_i(\xi_l, n, s) + Q_i(\xi_l, n, s)\right] \Phi(n) \Theta(\xi_l) \tag{A28}$$

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

$$G_i(X, Y, s)$$

439 $$= F_i(s)\Phi(n=0) + e^{\frac{Pe_L}{2} X} \cdot \sum_{l=1}^{\infty} \frac{K(\xi_l, X)}{N(\xi_l)} \left[P_i(\xi_l, n, s) + Q_i(\xi_l, n, s)\right] \Phi(n=0) \Theta(\xi_l) \tag{A29}$$

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

440 The analytical solutions in the original domain will be completed by taking the Laplace inverse

441 transform of Eq. (A29). $P_i(\xi_l, n, s)$ in Eq. (29) is in the form of the product of two functions . The

442 Laplace transform of $\dfrac{s + \alpha_i - \beta_i}{s + \alpha_i}$ can be easily obtained as

443 $$L^{-1}\left[\frac{s + \alpha_i - \beta_i}{s + \alpha_i}\right] = \delta(T) - \beta_i e^{-\alpha_i T} \tag{A30}$$

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

445 $\quad p_i(\xi_l, n, T) = L^{-1}\left[P_i(\xi_l, n, s)\right] = L^{-1}\left[-\dfrac{s + \alpha_i - \beta_i}{s + \alpha_i} F_i(s)\right] = -f_i(T) + \beta_i e^{-\alpha_i T} \int\limits_0^T f_i(\tau) e^{\alpha_i \tau} d\tau \quad$ (A31)

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

447 Laplace inverse transform on $Q_i(\xi_l, n, s)$, we have

448 $\quad q_i(\xi_l, n, T) = L^{-1}\left[Q_i(\xi_l, n, s)\right] = L^{-1}\left[\displaystyle\sum_{k=0}^{k=i-2} \dfrac{\beta_{i-k-1} \prod\limits_{j_1=0}^{j_1=k} \sigma_{i-j_1}}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s + \alpha_{i-j_2}\right)} F_{i-k-1}(s)\right]$

449 $\quad = \displaystyle\sum_{k=0}^{k=i-2} \beta_{i-k-1} \prod_{j_1=0}^{j_1=k} \sigma_{i-j_1} L^{-1}\left[\dfrac{1}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s + \alpha_{i-j_2}\right)} F_{i-k-1}(s)\right]$ (A32)

450

451 Expressing $\dfrac{1}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s + \alpha_{i-j_2}\right)}$ as the summation of partial fractions and applying the inverse

452 Laplace transform formula, one gets

453 $\quad L^{-1}\left[\dfrac{1}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s + \alpha_{i-j_2}\right)}\right] = L^{-1}\left[\displaystyle\sum_{j_2=0}^{j_2=k+1} \dfrac{1}{\prod\limits_{j_3=i-k-1, j_3 \neq i-j_2}^{j_3=i}\left(\alpha_{j_3} - \alpha_{i-j_2}\right)\left(s + \alpha_{i-j_2}\right)}\right]$

454 $\quad = \displaystyle\sum_{j_2=0}^{j_2=k+1} \dfrac{e^{-\alpha_{i-j_1} T}}{\prod\limits_{j_3=i-k-1, j_3 \neq i-j_1}^{j_3=i}\left(\alpha_{j_3} - \alpha_{i-j_1}\right)}$ (A33)

455

456 Recall that the inverse Laplace transform of $F_{i-k-1}(s)$ is $f_{i-k-1}(T)$. Thus, the Laplace inverse

457 transform of $\dfrac{1}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}F_{i-k-1}(s)$ in Eq. (1) can be achieved using the convolution integral

458 equation as

459 $$L^{-1}\left[\dfrac{1}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}F_{i-k-1}(s)\right]=\sum\limits_{j_2=0}^{j_2=k+1}\dfrac{e^{-\alpha_{i-j_1}T}\int\limits_{0}^{T}e^{\alpha_{i-j_1}\tau}f_{i-k-1}(\tau)d\tau}{\prod\limits_{j_3=i-k-1,\,j_3\neq i-j_2}^{j_3=i}\left(\alpha_{j_3}-\alpha_{i-j_2}\right)}\tag{A34}$$

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

461 $$q_i(\xi_l,n,T)=\sum\limits_{k=0}^{k=i-2}\beta_{i-k-1}\prod\limits_{j_1=0}^{j_1=k}\sigma_{i-j_1}\sum\limits_{j_2=0}^{j_2=k+1}\dfrac{e^{-\alpha_{i-j_1}T}\int\limits_{0}^{T}e^{\alpha_{i-j_1}\tau}f_{i-k-1}(\tau)d\tau}{\prod\limits_{j_3=i-k-1,\,j_3\neq i-j_2}^{j_3=i}\left(\alpha_{j_3}-\alpha_{i-j_2}\right)}\tag{A35}$$

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

463 and (15).

464      Note that Eq. (A33) is invalid for some of $\alpha_{i-j_2}$ being identical. For such conditions, we can

465 still reduce $\dfrac{1}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j_2}\right)}$ to a sum of partial fraction expansion. However, it will lead to

466 different Laplace inverse formulae. For example, the following formulae is used for all $\alpha_{i-j_2}$ being

467 identical

468 $$L^{-1}\left[\dfrac{1}{\prod\limits_{j_2=0}^{j_2=k+1}\left(s+\alpha_{i-j}\right)}\right]=\dfrac{T^k e^{-\alpha_{i-j_2}T}}{k!}\tag{A36}$$

469 The generalized formulae for the cases with some of $\alpha_{i-j_2}$ being identical will not be provided

470 herein because there are a large number of combinations of $\alpha_{i-j_2}$. We suggest that the readers can

471 pursue the solutions by following the similar steps for such specific conditions case by case.

472

473 **Acknowledgement**

477

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586

587

588 **Table 1**

589 Transport parameters used for convergence test example 1 involving the four-species radionuclide

590 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, $\lambda_m$ [year$^{-1}$] | |
| $^{238}Pu$ | 0.0089 |
| $^{234}U$ | 0.00100280 |
| $^{230}Th$ | 0.00100870 |
| $^{226}Ra$ | 0.00143 |

591

592

593 **Table 2**

594 Values for coefficients of Bateman-type boundary source for four-species transport problem used by

595 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 | | |
| $^{230}Th$ , $i=3$ | $0.443684 \times 10^{-3}$ | 0.593431 | $-0.593874$ | |
| $^{226}Ra$ , $i=4$ | $-0.516740 \times 10^{-6}$ | $0.120853 \times 10^{-1}$ | $-0.122637 \times 10^{-1}$ | $0.178925 \times 10^{-3}$ |

596

597

598 **Table 3**

599 Solution convergence of each species concentration at transect of inlet boundary ( $x = 0$ ) for four-

600 species radionuclide transport problem considering simulated domain of $L = 250$ m, $W = 100$ m,

601 subject to Bateman-type sources located at $40\,m \le y \le 60\,m$ for $t = 1,000$ year ($M$ = number of

602 terms summed for inverse generalized integral transform; $N$ = number of terms summed for inverse

603 finite Fourier cosine transform). When we investigate the required $M$ for inverse generalized integral

604 transform, $N$=16,000 for the finite Fourier cosine transform inverse are used. When we investigate the

605 required $N$ for inverse finite Fourier cosine transform, $M$=1,600 for the generalized transform inverse

606 are used.

607

$$^{238}Pu$$

| $x$ [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 |

608

$$^{234}U$$

| $x$ [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 |

| x [m] | y [m] | | | | |
|---|---|---|---|---|---|
| 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 |

609

$^{230}Th$

| 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 |

610

$^{226}Ra$

| 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 |

611

612

613

614 **Table 4**

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

623 $${}^{238}Pu$$

| $x$ [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 |

624 $${}^{234}U$$

| $x$ [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 |

625

626

$^{230}Th$

| $x$ [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 |

627

$^{226}Ra$

| $x$ [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 |

628

629

**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.

<div align="center"><sup>226</sup> <em>Ra</em></div>

| $x$  [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  $45\,m \leq y \leq 55\,m$  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 |

39

| x [m] | y [m] |  |  |  |  |  |
|-------|-------|--------|--------|--------|--------|--------|
| 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 |

655

$$^{230}Th$$

| 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 |

656

$$^{226}Ra$$

| 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 |

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**Table 7**

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

668
$$^{238}Pu$$

| $x$ [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 |

669
$$^{234}U$$

| $x$ [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 |

| x [m] | y [m] | | | | |
|-------|-------|---------|---------|---------|---------|---------|
| 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 |

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$$^{230}Th$$

| 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 |

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$$^{226}Ra$$

| 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 |

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675 **Table 8**

676 Transport parameters used for verification example 2 involving the ten-species transport problem

677 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, $\lambda_m$ [year$^{-1}$] $m$=1, 2,…,10 | 0.1, 0.75, 0.5, 0.25, 0, 0, 0.3, 1, 0, 0.65 |

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682 **Table 9**

683 Coefficients of Bateman-type boundary source for ten-species transport problem used by Srinivasan

684 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 |

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690 **Table 10**

691 Transport parameters used for example application involving the five-species dissolved chlorinated

692 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, $\lambda_m$ [year$^{-1}$] | |
| PCE | 0 |
| TCE | 0 |
| DCE | 0 |
| VC | 0 |
| ETH | 0 |

693 **Table 11**

694 Coefficients of Bateman-type boundary source used for example application involving the five-

695 species dissolved chlorinated solvent problem used by BIOCHLOR.

|  | | | $b_{im}$ | | |
|---|---|---|---|---|---|
| **Species, $i$** | | | | | |
| | $m=1$ | $m=2$ | $m=3$ | $m=4$ | $m=5$ |
| *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 |

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704 **Figures Captions**

705 Fig. 1. Schematic representation of two-dimensional transport of decaying contaminants in a uniform

706     flow field with flux boundary source located at of the inlet boundary.

707 Fig. 2. Comparison of spatial concentration profiles of four species along the longitudinal direction

708     (=50 m) at $t$ = 1,000 years obtained from derived analytical solutions and numerical

709     solutions for convergence test example 1 of four-member radionuclide decay chain

710     $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ .

711 Fig. 3. Comparison of spatial concentration profiles of four species along the transverse direction (=0

712     m) at $t$ = 1,000 years obtained from derived analytical solutions and numerical solutions

713     for convergence test example 1 of four-member radionuclide decay chain

714     $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ .

715 Fig. 4. Comparison of spatial concentration profiles of four species along the transverse direction

716     (=25 m) at $t$ = 1,000 years obtained from derived analytical solutions and numerical

717     solutions for convergence test example 1 of four-member radionuclide decay chain

718     $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ .

719 Fig. 5. Comparison of spatial concentration profiles of four species along the longitudinal direction

720     (=50 m) at $t$ = 1,000 years obtained from derived analytical solutions and numerical

721     solutions for convergence test example 2 of four-member radionuclide decay chain

722     $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ .

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

724 m) at $t = 1{,}000$ years obtained from derived analytical solutions and numerical solutions

725 for convergence test example 2 of four-member radionuclide decay chain

726 $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ .

727 Fig. 7. Comparison of spatial concentration profiles of four species along the transverse direction

728 (=25 m) at $t = 1{,}000$ years obtained from derived analytical solutions and numerical

729 solutions for convergence test example 2 of four-member radionuclide decay chain

730 $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ .

731 Fig. 8. Comparison of spatial concentration profiles of ten-species along $x$-direction at $t = 20$ days

732 obtained from derived analytical solutions and numerical solutions for the test example 3

733 of ten species decay chain used by Srinivasan and Clement (2008b).

734 Fig. 9. Effects of physical processes and chemical reactions on the concentration contours of four-

735 species at $t = 1{,}000$ years obtained from derived analytical solutions for four-member decay

736 chain $^{238}Pu \rightarrow ^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ .

737 Fig. 10. Spatial concentration contours of five-species at $t = 1$ year obtained from derived analytical

738 solutions for natural attenuation of chlorinated solvent plumes $PCE \rightarrow TCE \rightarrow DCE \rightarrow VC$

739 $\rightarrow ETH$.

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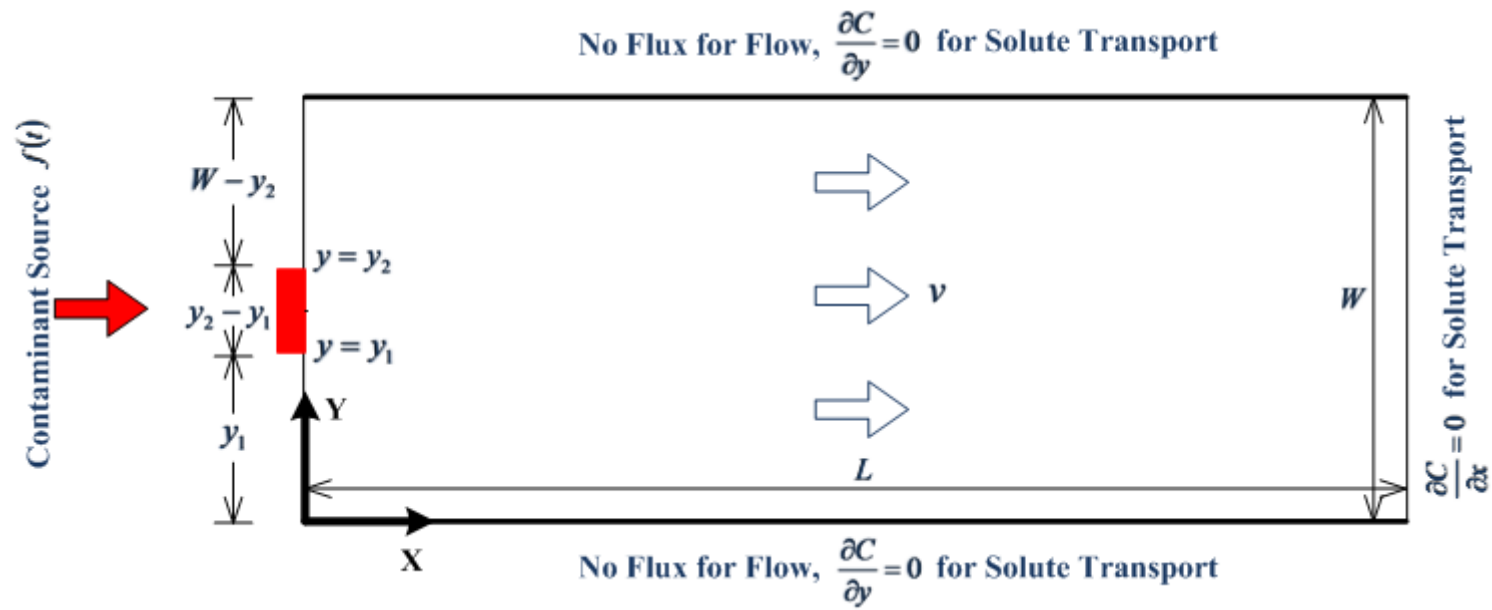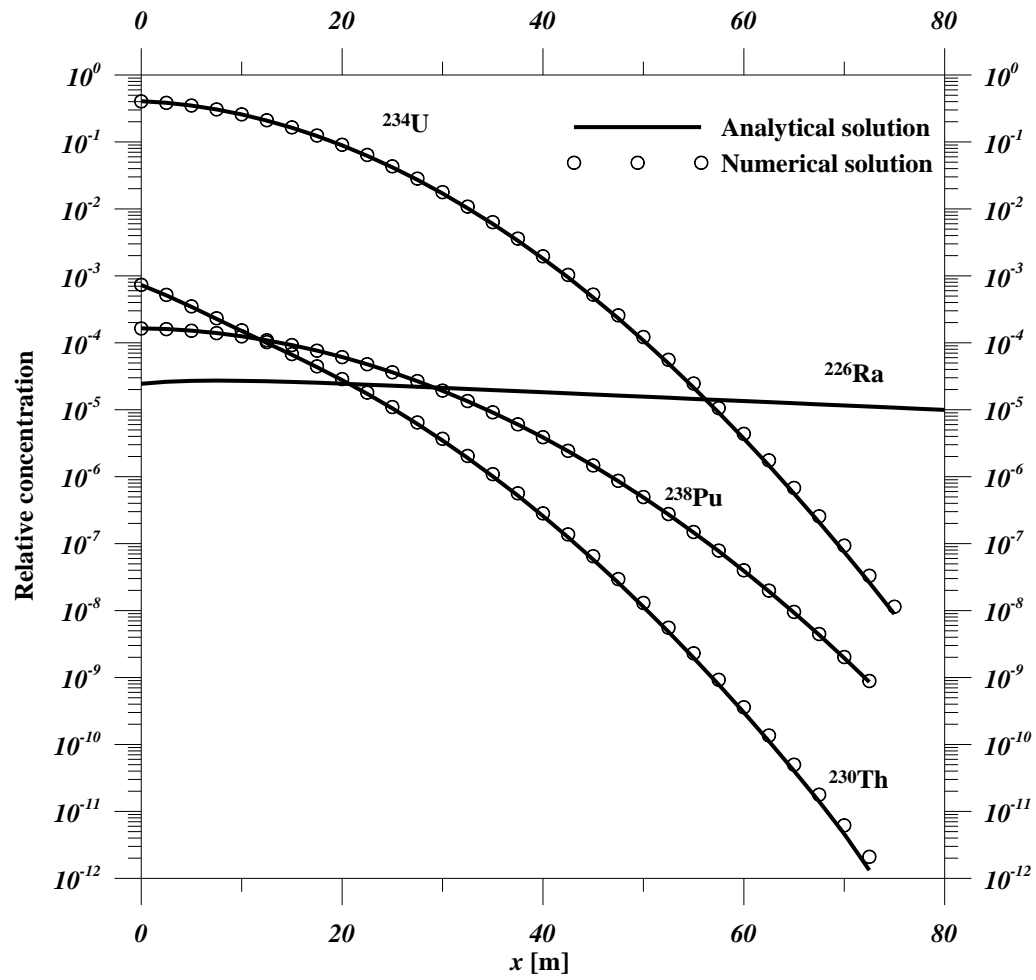
**Fig. 1.**

**Fig. 2.**
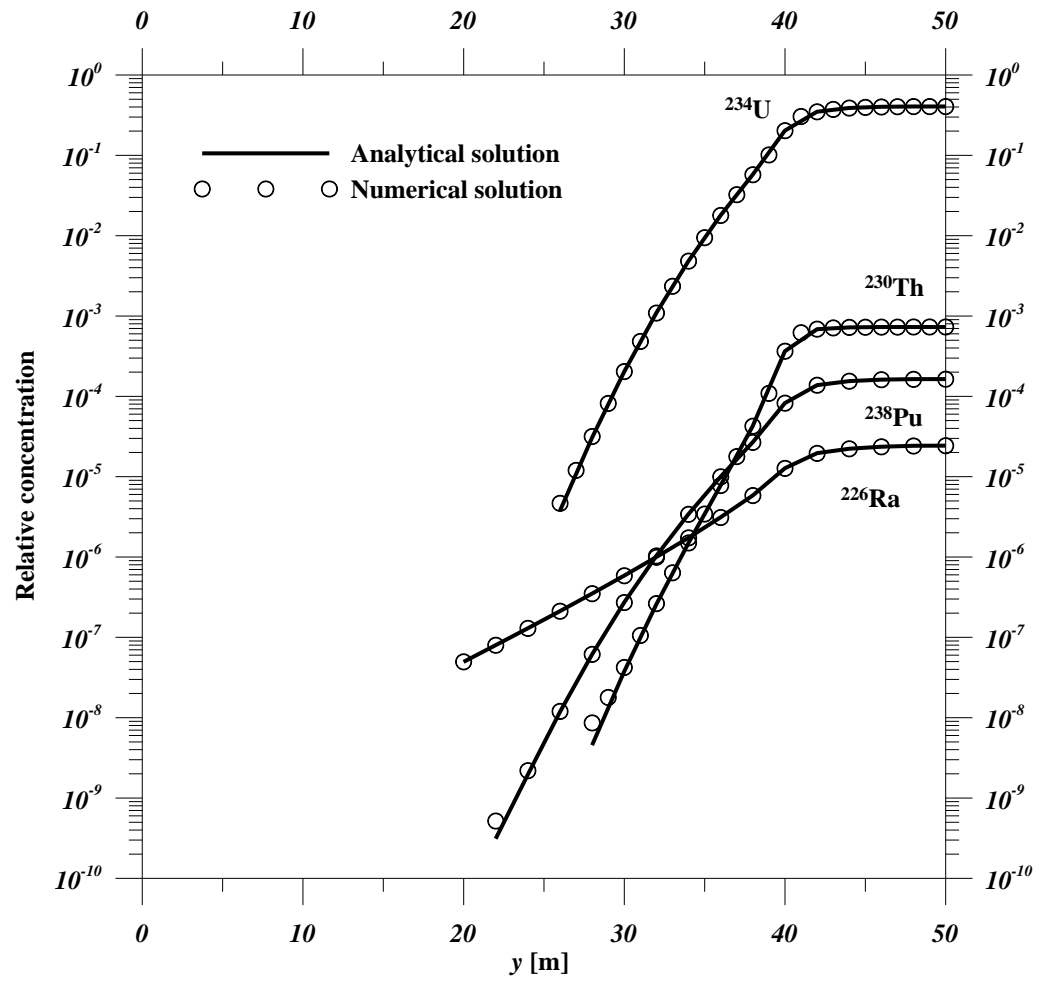
**Fig. 3**
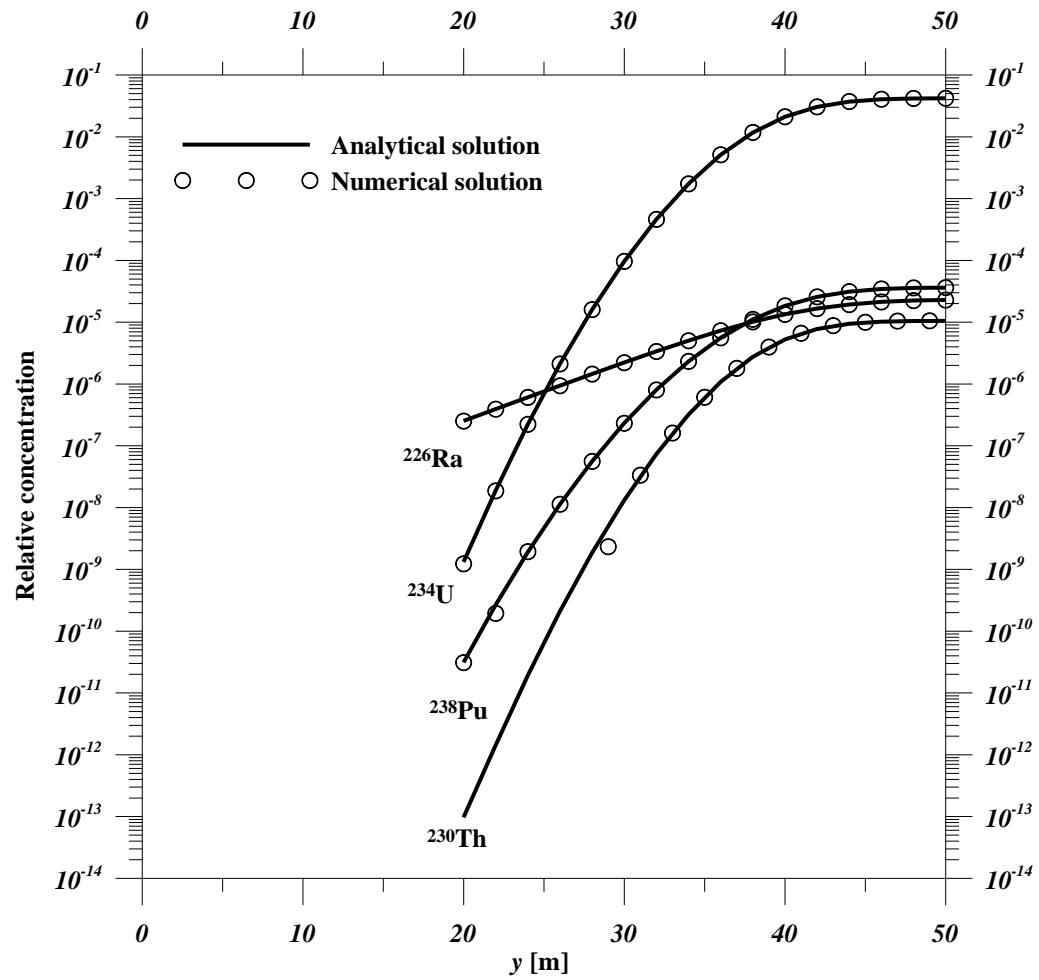
**Fig. 4**
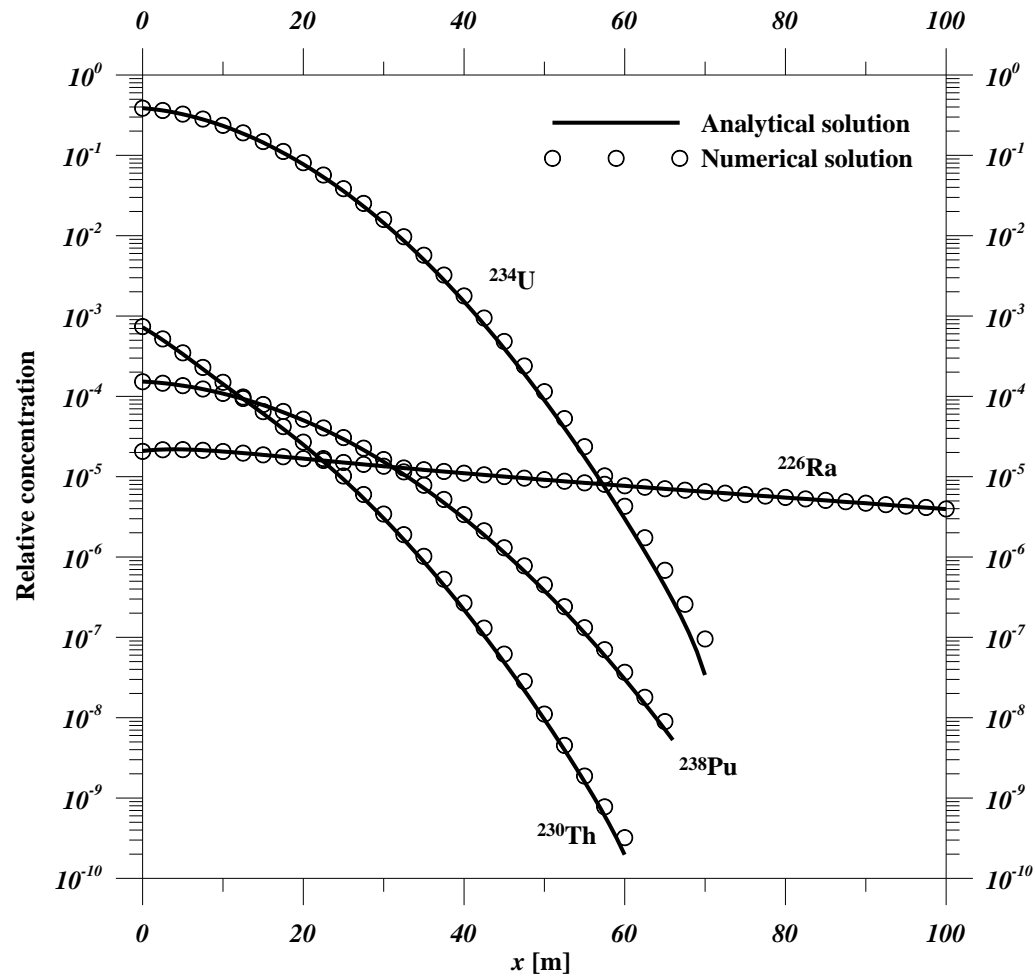
**Fig. 5**
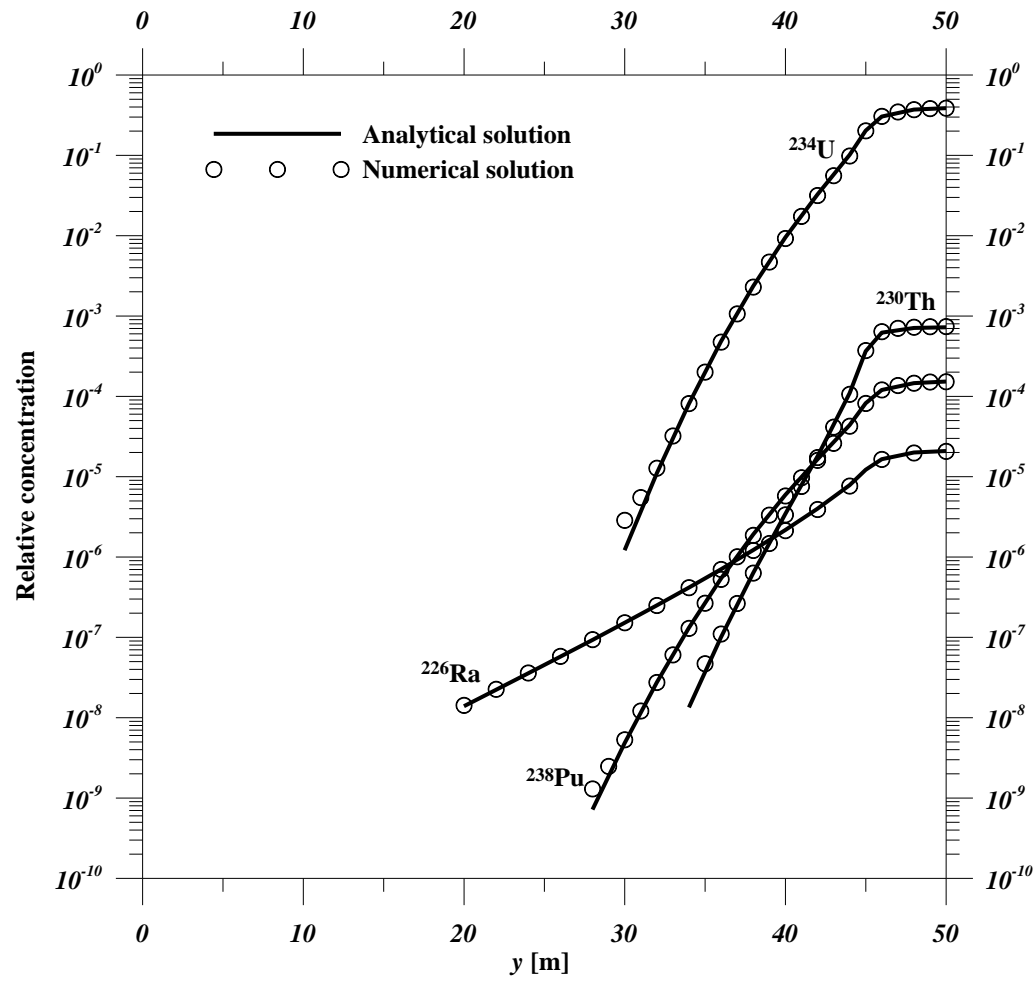
**Fig. 6**

**Fig. 7**

**Fig. 8**

**Fig. 9**

**Fig. 10**