

Transport of Reactive Tracer in Compacting Multi-Fraction Bottom Sediments



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Abstract The one-dimensional transport of reactive tracer (radionuclide) in the multi-fractional aquatic sediments is considered. In the model a transfer of tracer in sediment is governed by the molecular diffusion of dissolved phase in pore water, biodiffusion, phase exchange between dissolved and solid phases of tracers, exchange between fast and slow reversible phases of tracer, and compaction under gravity force resulting in a decrease of porosity with sediment layer depth. The transport equations for 2-step reactions were completed by an equation for fractions of sediments assuming that mixing in the bottom sediment is intraphase. The compaction dynamics was parameterized using an approximation of porosity by travelling wave profile. The results of an idealized numerical experiment on the constant deposition of contaminated sediments show that in the case of 2-step reactions the exchange processes in multifraction sediments were far from equilibrium.

Keywords Reactive tracer · Multifraction sediments · Compaction

1 Introduction

An exchange between the dissolved and solid phases is a significant process in the transport of reactive tracers in the marine environment. Settling of contaminated suspended sediments and resuspension by waves and currents results in tracer exchanges between the bottom and suspended sediment. The transfer of activity between the water column and the pore water in the bottom sediment is governed by the bottom boundary layer turbulence, which regulated diffusional processes [1]. The migration of tracer in the sediments is due to molecular diffusion, transport driven by bioturbation, bioirrigation, and also due to advection driven by surface waves and by subsurface groundwater flow [2]. Bottom sediments are important for the burial of particle-reactive radionuclides. After the Fukushima Dai-ichi accident, they are a

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long-term source of radionuclide contamination through resuspension, bioturbation, and losses of soluble radionuclides from the pore water.

An important factor in the transport of tracer in the sediments is compaction processes caused by gravity forces resulting in a decrease of porosity with the depth of the sediment layer and corresponding fluxes of pore water and sediment particles. In general, the dynamics of this process is described by the nonlinear Gibson equation [3]. However, for a slow varied compaction process, the profile can be parameterized using an approximation of porosity by travelling wave profile [4]. Most of the studies of migration of reactive tracer were carried out for a single fraction of sediments. At the same time, the kinetics of fast reversible exchange between water and solids depends on the particle size. Therefore, it is necessary to extend transport models of reactive transport on compacting multifraction bottom sediments.

In the paper, the one-dimensional transport of reactive tracer (radionuclide) in the multi-fractional compacting aquatic sediments is considered. The model is described in Sect. 2. The results of an idealized numerical experiment are given in Sect. 3. Conclusions are drawn in Sect. 4.

2 Model Formulation

Consider one-dimensional transport of reactive tracer (radionuclide) in the multi-fractional aquatic sediments. It is assumed that reversible phase exchange is described by reactions of the first order. The bottom sediment layer is characterized by porosity $\varepsilon^w(z, t)$, solid volume fraction $\varepsilon^s(z, t)$, ($\varepsilon^w + \varepsilon^s = 1$), a fraction of sediment particles of i -th class $\phi_i(z, t)$ ($\sum_{i=1}^n \phi_i = 1$), and density of sediment fraction $\rho_{s,i}$. In the model, a transfer of tracer in sediment is governed by (i) molecular diffusion of dissolved phase in porewater, (ii) bioturbation (mixing of solid sediment particles by bottom organisms), (iii) phase exchange between dissolved and solid phases of tracers, (iv) exchange between fast and slow reversible phases of tracer, and (v) compaction under gravity force resulting in a decrease of porosity with sediment layer depth. The transport equations for dissolved tracer concentration in pore water C_d^b (Bq m⁻³), fast particulate concentration in i -th fraction of the bottom sediments $C_{s,i}^b$ (Bq kg⁻¹), and slow particulate concentration in i -th fraction of the bottom sediments $\tilde{C}_{s,i}^b$ are written following [2] as

$$\begin{aligned} \frac{\partial \varepsilon^w C_d^b}{\partial t} = & -\frac{\partial \varepsilon^w w_f C_d^b}{\partial z} + \frac{\partial}{\partial z} \left(v_B + \frac{v_D}{\psi^2} \right) \varepsilon^w \frac{\partial C_d^b}{\partial z} \\ & - a_{ds} \theta \varepsilon^s \left(C_d^b \hat{K}_d^b - \hat{C}_s^b \right) - \lambda \varepsilon^w C_d^b, \end{aligned} \quad (1)$$

$$\begin{aligned} \frac{\partial \varepsilon^s \phi_i C_{s,i}^b}{\partial t} = & -\frac{\partial \varepsilon^s w_s \phi_i C_{s,i}^b}{\partial z} + \frac{\partial}{\partial z} v_B \varepsilon^s \frac{\partial \phi_i C_{s,i}^b}{\partial z} + a_{ds} \theta \varepsilon^s \phi_i \left(C_d^b K_{d,i}^b - C_{s,i}^b \right) \\ & - a_{fs} \varepsilon^s \phi_i C_{s,i}^b + a_{sf} \varepsilon^s \phi_i \tilde{C}_{s,i}^b - \lambda \varepsilon^s \phi_i C_{s,i}^b, \end{aligned} \quad (2)$$

$$\frac{\partial \varepsilon^s \phi_i \tilde{C}_{s,i}^b}{\partial t} = -\frac{\partial \varepsilon^s w_s \phi_i \tilde{C}_{s,i}^b}{\partial z} + \frac{\partial}{\partial z} \nu_B \varepsilon^s \frac{\partial \phi_i \tilde{C}_{s,i}^b}{\partial z} + a_{sf} \varepsilon^s \phi_i C_{s,i}^b - a_{fs} \varepsilon^s \phi_i \tilde{C}_{s,i}^b - \lambda \varepsilon^s \phi_i C_{s,i}^b \quad (3)$$

where t is time (s), z is vertical coordinate directed upward (m), w_f is a vertical pore water velocity (m s⁻¹), w_s is a particle vertical velocity (m s⁻¹), λ (s⁻¹) is the tracer decay constant. The phase exchange between dissolved and reversible tracer phase is written in terms of desorption rate a_{ds} (s⁻¹) and distribution coefficient $K_{d,i}$ (m³kg⁻¹) [5], whereas a_{fs} and a_{sf} (s⁻¹) are the direct and reverse exchange rates between fast and slow reversible phases of tracer, θ is a correction factor for desorption rate. The free solution diffusion coefficient ν_D was corrected for tortuosity ψ following [1] as $\psi^2 = 1 - 2 \ln \varepsilon^w$. The bioturbation is parameterized by the bioturbation coefficient $\nu_B(z)$. The total concentration of tracer \hat{C}_s^b (Bq m⁻³) is defined as

$$\hat{C}_s^b = \sum_{i=0}^n \rho_{s,i} \phi_i C_{s,i}^b, \quad (4)$$

The total distribution coefficient \hat{K}_d^b is defined as

$$\hat{K}_d^b = \sum_{i=1}^n \rho_{s,i} \phi_i K_{d,i}, \quad (5)$$

The dependence of $K_{d,i}$ on sediment particle diameter d_i is written following [5] as.

$$K_{d,i} = \frac{\chi}{a_{ds} \rho_{s,i}} \frac{6}{d_i}, \quad (6)$$

where χ is an exchange velocity (m s⁻¹), d_i (m) is the sediment particle diameter.

The transport equation for fractions of bottom sediments at $\rho_{s,i} = \rho_s$ is described as

$$\frac{\partial \varepsilon^s \phi_i}{\partial t} = -\frac{\partial \varepsilon^s w_s \phi_i}{\partial z} + \frac{\partial}{\partial z} \nu_B \varepsilon^s \frac{\partial \phi_i}{\partial z} \quad (7)$$

Summing this equation on all fractions we obtain the transport equation for ε^s .

$$\frac{\partial \varepsilon^s}{\partial t} = -\frac{\partial \varepsilon^s w_s}{\partial z} \quad (8)$$

It was assumed in the derivation of (1)–(3), (7) that mixing in the bottom sediment is intraphase (i.e. porosity is not affected by biodiffusivity [5]). Therefore, an equation

for porosity can be written as

$$\frac{\partial \varepsilon^w}{\partial t} = - \frac{\partial \varepsilon^w w_f}{\partial z} \quad (9)$$

Combining (8) and (9) and assuming that $w_f \rightarrow 0$, $w_s \rightarrow 0$ at $z \rightarrow -\infty$ yields

$$\varepsilon^w w_f + \varepsilon^s w_s = 0. \quad (10)$$

Introducing moving coordinate system

$$\zeta = H(t) - z, \quad (11)$$

where $H(t)$ is the coordinate of the water–sediment interface. Assume that in moving system coordinate profile of porosity was a profile of travelling wave, which does not depend on time. Following [5] this profile was approximated by exponent

$$\varepsilon^w(\zeta) = (\varepsilon_0^w - \varepsilon_\infty^w) \exp(-k_\varepsilon \zeta) + \varepsilon_\infty^w, \quad (12)$$

where k_ε is an attenuation coefficient, $\varepsilon_0^s = \varepsilon^s(0)$, ε_∞^s is an asymptotic value at large depth.

$$\frac{\partial}{\partial \zeta} [\varepsilon^s (\omega + w_s)] = 0, \quad (13)$$

where $\omega = \partial H / \partial t$ is displacement velocity of the water-bottom interface described as

$$\omega = \frac{1}{\varepsilon_0^s \rho_s} \sum_{i=0}^n W_{p,i} S_{p,i} - w_s(0), \quad (14)$$

where W_p (m s^{-1}) is the settling velocity of suspended sediment, $S_{p,i}$ (kg m^{-3}) is suspended sediment concentration, and $w_s(0)$ is consolidation velocity at sediment surface.

Integration of (13) and determination of integration constant from asymptotic conditions: $w_s \rightarrow 0$, $\varepsilon^s \rightarrow 0$ at $\zeta \rightarrow \infty$ leads to the relations between vertical velocities of particles and pore water and displacement velocity of the water sediment interface

$$w_f = - \frac{\varepsilon^w - \varepsilon_\infty^w}{\varepsilon^s} \omega, \quad w_s = \frac{\varepsilon_\infty^s - \varepsilon^s}{\varepsilon^s} \omega. \quad (15)$$

As seen in Eq. (15) the consolidation velocity $w_s(0)$ can be comparable with sedimentation velocity. Taking into account (15) the displacement velocity was rewritten as

$$\omega = \frac{1}{\varepsilon_\infty^s \rho_s} \sum_{i=0}^n W_{p,i} S_{p,i} \quad (16)$$

Finally, governing system of equations describing transport of reactive transport of tracer in compacting sediments

$$\begin{aligned} \varepsilon^w \frac{\partial C_d^b}{\partial t} &= -\varepsilon^w \omega \frac{\partial C_d^b}{\partial \zeta} + \frac{\partial}{\partial \zeta} \left(v_B + \frac{v_D}{\psi^2} \right) \varepsilon^w \frac{\partial C_d^b}{\partial \zeta} \\ &\quad - a_{ds} \theta \varepsilon^s \left(C_d^b \hat{K}_d^b - \hat{C}_s^b \right) - \lambda \varepsilon^w C_d^b, \end{aligned} \quad (17)$$

$$\begin{aligned} \varepsilon^s \frac{\partial \phi_i C_{s,i}^b}{\partial t} &= -\varepsilon^s \omega \frac{\partial \phi_i C_{s,i}^b}{\partial \zeta} + \frac{\partial}{\partial \zeta} v_B \varepsilon^s \frac{\partial \phi_i C_{s,i}^b}{\partial \zeta} \\ &\quad + a_{ds} \theta \varepsilon^s \phi_i \left(C_d^b K_{d,i}^b - C_{s,i}^b \right) - \lambda \varepsilon^s \phi_i C_{s,i}^b, \end{aligned} \quad (18)$$

$$\begin{aligned} \varepsilon^s \frac{\partial \phi_i \tilde{C}_{s,i}^b}{\partial t} &= -\varepsilon^s \omega \frac{\partial \phi_i \tilde{C}_{s,i}^b}{\partial \zeta} + \frac{\partial}{\partial \zeta} v_B \varepsilon^s \frac{\partial \phi_i \tilde{C}_{s,i}^b}{\partial \zeta} + a_{fs} \varepsilon^s \phi_i C_{s,i}^b \\ &\quad - a_{sf} \varepsilon^s \phi_i \tilde{C}_{s,i}^b - \lambda \varepsilon^s \phi_i \tilde{C}_{s,i}^b, \end{aligned} \quad (19)$$

$$\varepsilon^s \frac{\partial \phi_i}{\partial t} = -\varepsilon^s \omega \frac{\partial \phi_i}{\partial \zeta} + \frac{\partial}{\partial z} \varepsilon^s v_B \frac{\partial \phi_i}{\partial z}. \quad (20)$$

The boundary conditions at interface water–sediment $\zeta = 0$ are

$$\left(v_B + \frac{v_D}{\psi^2} \right) \frac{\partial C_d^b}{\partial \zeta} = -W_{pw} (C_d^w - C_d^b), \quad (21)$$

$$v_B \frac{\partial C_{s,i}^b}{\partial \zeta} = -\frac{\omega}{\varepsilon_\infty^s} (C_{p,i}^w - C_{s,i}^b), \quad (22)$$

$$v_B \frac{\partial \tilde{C}_{s,i}^b}{\partial \zeta} = -\frac{\omega}{\varepsilon_\infty^s} (\tilde{C}_{p,i}^w - \tilde{C}_{s,i}^b), \quad (23)$$

$$v_B \frac{\partial \phi_i}{\partial \zeta} = -\frac{\omega}{\varepsilon_\infty^s} (\phi_i(0) - \phi_i), \quad (24)$$

where C_d^w (Bq m^{-3}) is tracer concentration in the water, C_d^b (Bq m^{-3}) is tracer concentration in the water, $C_{p,i}^w$ is the particulate concentration in i -th fraction of the deposited from water column sediments. The exchange rate W_{pw} is estimated from

corrected for surface roughness relation [6] as

$$W_{PW} = 0.1778u_*\text{Re}^{-0.2} \text{Sc}^{-0.604}. \quad (25)$$

Here u_* is friction velocity, $\text{Sc} = \nu_M/\nu_D$ is the Schmidt number, ν_M is kinematic viscosity, ν_D is free solution diffusion coefficient, $\text{Re} = u_*\delta_*\nu_M^{-1}$ is the Reynolds number, δ_* is an average height of roughness elements, ν_M is kinematic viscosity. Alternatively, the values of $C_{s,i}^b$, $\tilde{C}_{s,i}^b$ and ϕ_i can be prescribed [7]. At the lower boundary of the sediment layer, the zero diffusion fluxes of whole variables are prescribed. The system of Eqns. (17)–(20) is solved using the finite-difference implicit scheme of second order.

3 Results of Modelling

Consider results of an idealized numerical experiment where the constant flux of contaminated by ^{137}Cs sediments of two fractions caused the formation of contaminated layer and redistribution of activity in different forms. The sediment layer thickness is 1 m, density of sediment particles $\rho_s = 2600 \text{ kg m}^{-3}$, surface porosity $\varepsilon_0^w = 0.8$, bottom porosity $\varepsilon_\infty^w = 0.4$, $\omega = 6.4 \cdot 10^{-10} \text{ m s}^{-1}$, $\nu_D = 1.45 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$, $\nu_M = 1.6 \cdot 10^{-6} \text{ m}^2 \text{ s}^{-1}$, $\phi_1 = 0.2$, $d_1 = 10^{-5} \text{ m}$, $d_2 = 4 \cdot 10^{-5} \text{ m}$, $u_* = 10^{-2} \text{ m s}^{-1}$, $\delta_* = 10^{-3} \text{ m}$. The biodiffusivity is described by the relation

$$\nu_B = \nu_{B0} \exp(-\zeta^2/\zeta_{eff}^2), \quad (26)$$

where $\nu_{B0} = 10^{-12} \text{ m}^2 \text{ s}^{-1}$, $\zeta_{eff} = 0.02 \text{ m}$. Kinetic parameters were chosen for ^{137}Cs [2, 5] as: $a_{ds} = 1.16 \cdot 10^{-5} \text{ s}^{-1}$, $\chi = 3.8 \cdot 10^{-6} \text{ m s}^{-1}$, $a_{fs} = 0.25 \cdot 10^{-7} \text{ s}^{-1}$, $a_{sf} = 0.25 \cdot 10^{-8} \text{ s}^{-1}$, $\theta = 0.1$, $\lambda = 1.06 \cdot 10^{-8} \text{ s}^{-1}$. Initially, the sediment layer was not contaminated.

Two scenarios were considered. In the first scenario, the 1-step reactions were taken into account. It was assumed that the concentration of ^{137}Cs in the settled to the bottom sediment particles was in equilibrium with a concentration in water. The corresponding interface values of variables were $C_d^b(0) = 23.4 \text{ Bq m}^{-3}$, $C_{s,1}^b(0) = 372.9 \text{ Bq kg}^{-1}$, $C_{s,2}^b(0) = 31.7 \text{ Bq kg}^{-1}$, $\phi_1 = 0.2$. In the second scenario, the 2-step reactions were taken into account where corresponding interface values of variables were $C_d^b(0) = 23.3 \text{ Bq m}^{-3}$, $C_{s,1}^b(0) = 371.2 \text{ Bq kg}^{-1}$, $C_{s,2}^b(0) = 31.5 \text{ Bq kg}^{-1}$, $\tilde{C}_{s,1}^b(0) = \tilde{C}_{s,2}^b(0) = 0$, $\phi_1 = 0.2$.

The simulation results for the concentration of ^{137}Cs in solid and dissolved phases calculated using 1-step and 2-step reaction models after 10 years of deposition are shown in Figs. 1 and 2. In the case of a 1-step reaction (Fig. 1) the exchange processes between dissolved and particulate phases of radionuclide were close to equilibrium excluding a thin layer near the interface. However, accounting of 2-step reactions (Fig. 2) results in the slow evolution of both slow and fast phases and

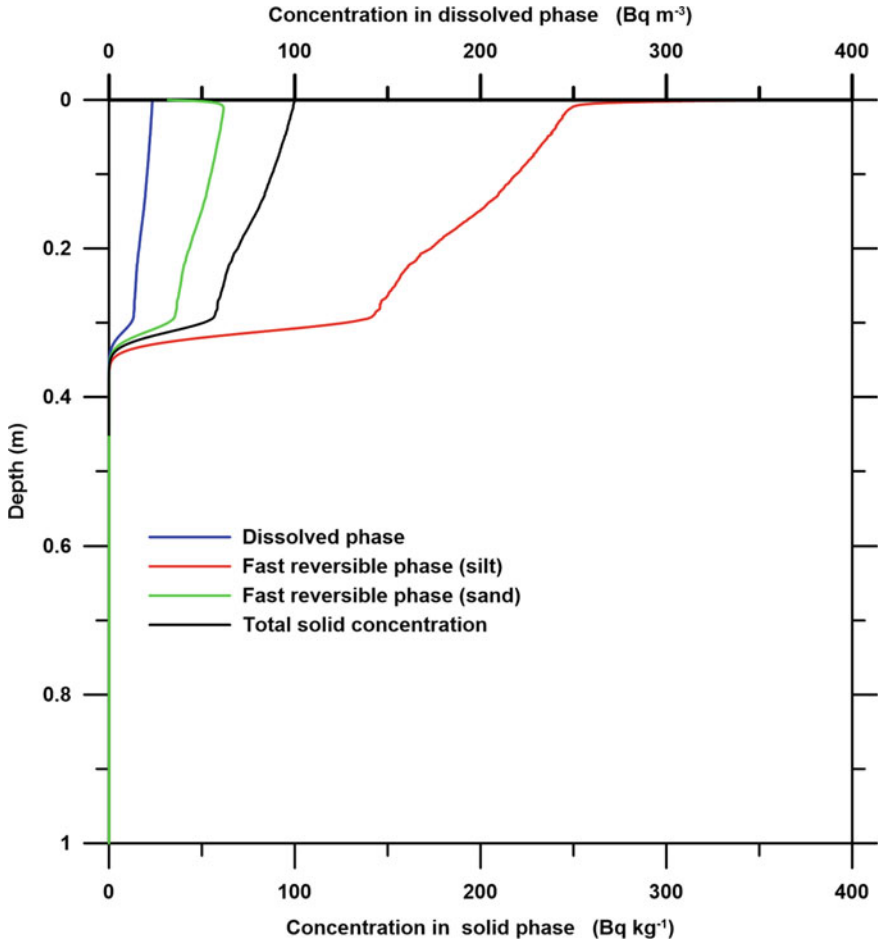


Fig. 1 The concentration of ¹³⁷Cs in solid and dissolved phases calculated using 1-step reaction models after 10 years of deposition

exchange between fractions of sediments through the pore water. In that case, the exchange processes in multifraction sediments were far from equilibrium. A decrease of porosity with depth caused by the compaction process changes the profile of the concentration of ¹³⁷Cs in solid and dissolved phases.

The effect of seasonal variations of deposition of sediment fraction was simulated using the periodic representation of fractions. The corresponding relation for silt fraction was approximated as

$$\phi_1 = \phi_{01} + \phi_{01} \sin\left(\frac{2\pi t}{T}\right), \tag{27}$$

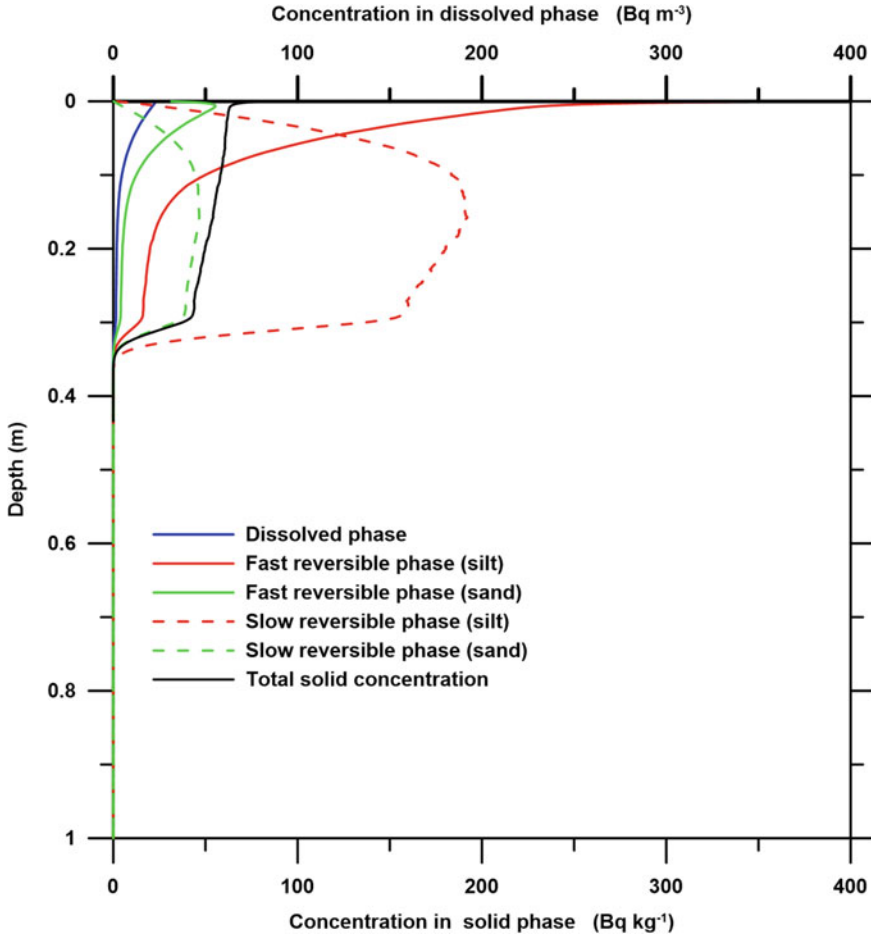


Fig. 2 The concentration of ^{137}Cs in solid and dissolved phases calculated using 2-step (b) reaction models after 10 years of deposition

where $\phi_{01} = 0.5$, $\phi_{02} = 0.3$, $T = 3.1536 \cdot 10^7$ s. As seen in Fig. 3 accounting variation in depositing sediments results in more complicated profiles of the concentration of ^{137}Cs for different fractions, although the profile of total solid concentration is more smooth due to mutual compensation by different fractions. An accounting of 2-step reactions (Fig. 4) results in similar variations of profiles of the concentration of ^{137}Cs for different fractions with decaying seasonal effects with depth due to diffusivity and slow kinetics.

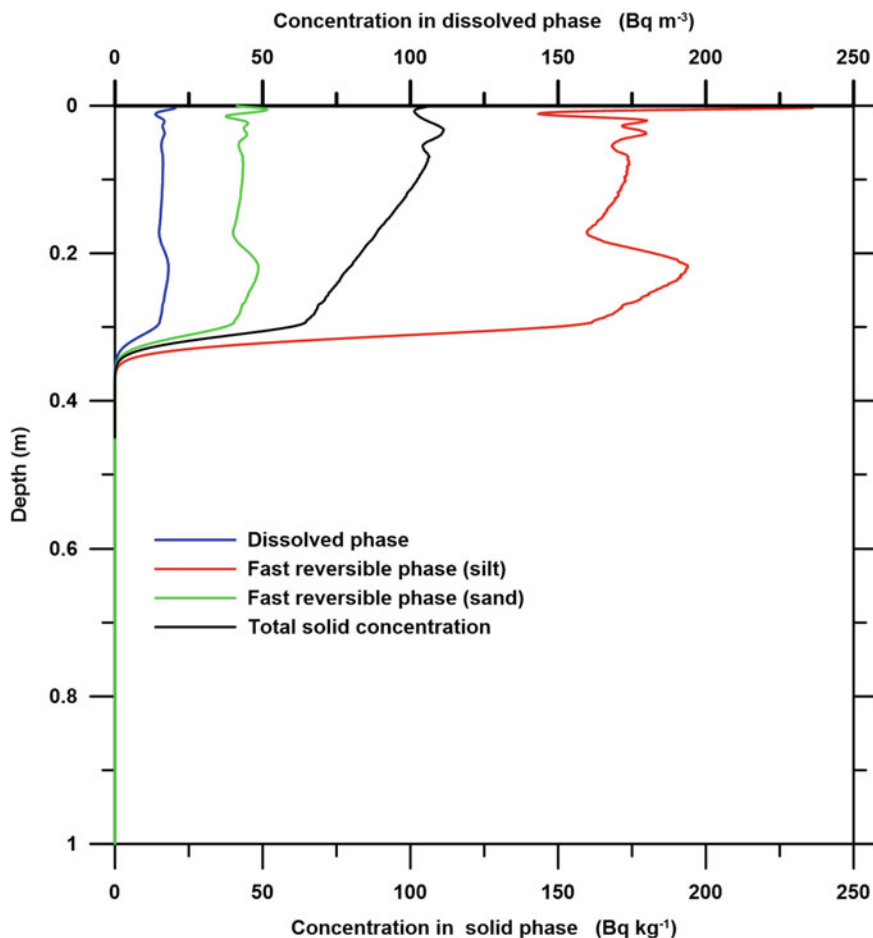


Fig. 3 The concentration of ^{137}Cs in solid and dissolved phases calculated using 2-step reaction models after 10 years of seasonally varying deposition of fractions

4 Conclusions

In the paper, the one-dimensional transport of reactive tracer (radionuclide) in the multi-fractional aquatic sediments is considered. In the model, a transfer of tracer in sediment is governed by the molecular diffusion of dissolved phase in pore water and biodiffusion. The phase exchange of tracer is described in the frame of 2-step reactions of first-order between dissolved phase and fast reversible phase of tracer on particle and exchange between fast and slow reversible phases of tracer on the particles. The transport equations for 2-step reactions were completed by the equation for sediment fraction transport assuming that mixing in the bottom sediment is

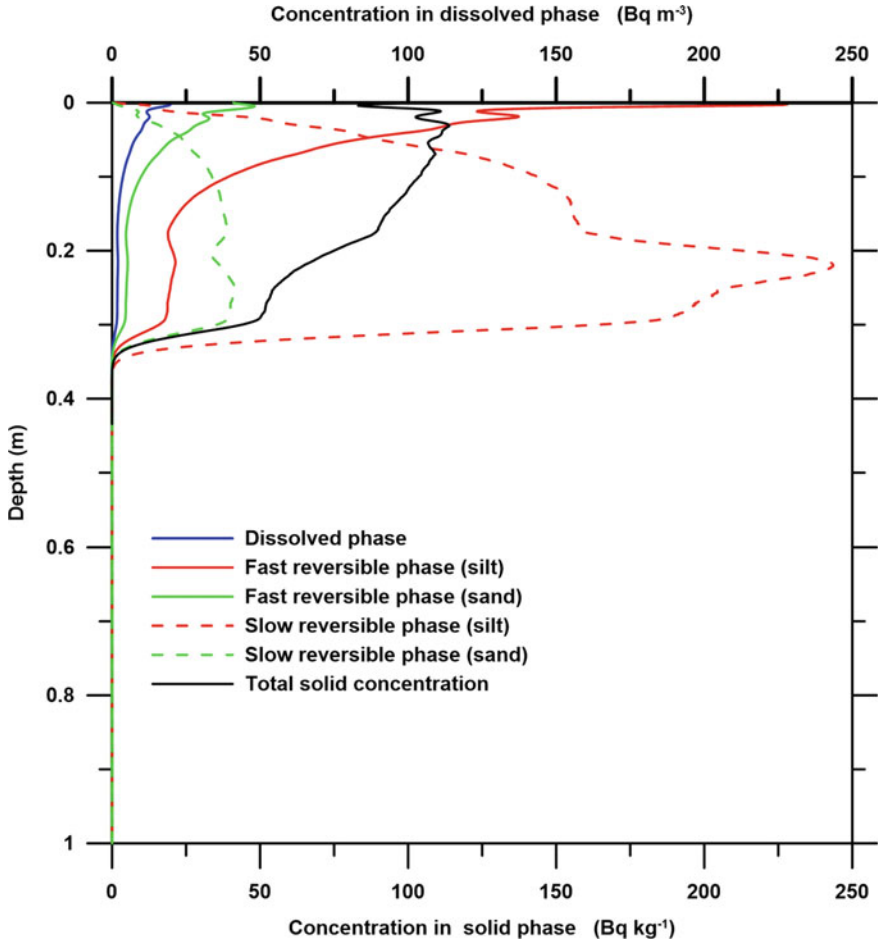


Fig. 4 The concentration of ^{137}Cs in solid and dissolved phases calculated using 2-step reaction models after 10 years of seasonally varying deposition of fractions

intraphase. It was found that the consolidation velocity due to gravity can be comparable with sedimentation velocity. An accounting of 2-step reactions results in the slow evolution of both slow and fast phases and exchange between fractions of sediments through the pore water. Therefore, the exchange processes in the multifraction sediments were far from equilibrium. Time variations in depositing fractions make profiles of reactive tracer result in more complicated profiles.

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