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Model of Non-Equilibrium Multiphase Radionuclide Transport in Lake Water-Sediment System

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Introduction

The advanced model of radioactive contaminant transport between the abiotic components of the lake water-sediment system is presented.

The model includes *a concept of dynamics* of the contaminant sorption in lake water and sediment compartments, considering the specific porous structure of sediments, *the contaminant material exchange* between the liquid and solid phases of sediments.

The key processes included in the model are: sedimentation, resuspension, diffusive exchange of solute at the lake water-sediment interface and advection-diffusion in sediment solute.

The aim of this work is to develop the contaminant flow model in the lake water-sediment system including:

- the concept of *non-equilibrium* dynamics of contaminant sorption process in lake water and sediment compartments;
- to consider the porous structure of the solid phases, contaminant material exchange between the liquid and solid phases in the water and sediment spheres;
- to pay special attention to the contamination balance between the two spheres in the interface area.

The concept of the model

The model has a representation of the lake water system as continuously mixing water in the reservoir.

In this model, the direction of transport of pollution is not important. The amount of water in the influent and effluent flows of the system is determined by *variation* of the *water volume*.

The compartment model consists of two spheres (the lake water and bottom sediments underneath the water) and of four compartments (two compartments in each of the spheres).

The lake water and suspended matter compartments are in the lake water sphere. The liquid phase compartment and solid porous sediment compartment are in the sediment sphere (Fig. 1).

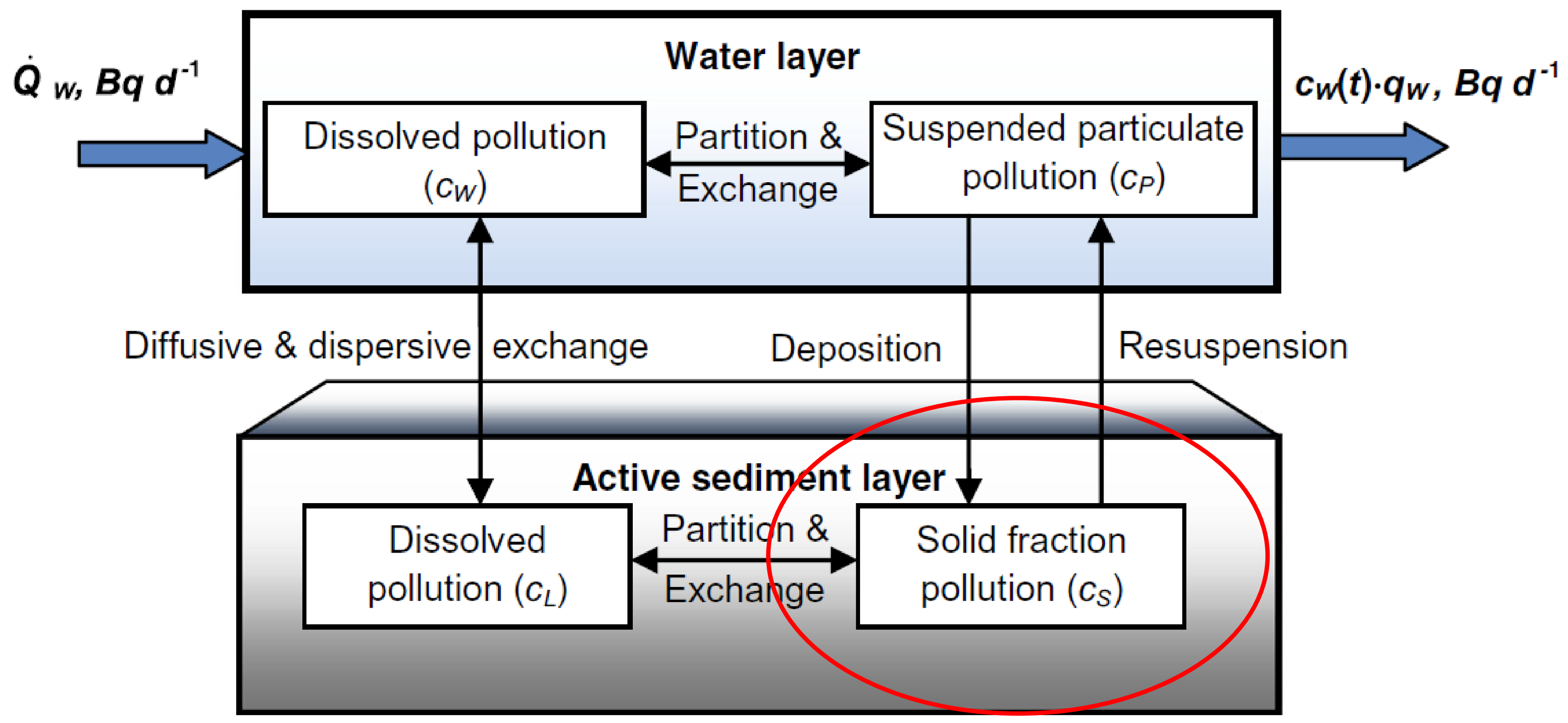

Fig. 1. The flowchart of the lake water and sediment interaction model.

Constantly is no equilibrium !!!!!!

Vertical distribution profile in sediment layer is considered as a process having two stages (I and II):

- the first stage (I) is related with the migration of the contaminant from water to the sediments,
- whereas, the second stage (II) – with the further transformation of the deposited contaminant.

First stage (I)

Is a fast sorption process. It is modelled by making the assumption of the instantaneous equilibrium, and the concentration sorbed on particles is in the equilibrium with the dissolved contamination of the lake water c_W [Bq m^{-3}].

Second stage (II)

The interaction or a slow contaminant sorption-desorption-degradation mechanism of the porous structure, the activity conservation equation is used.

Mathematical description

$$\frac{\partial c_L}{\partial t} = D^*\frac{\partial^2 c_L}{\partial x^2} - v^*\frac{\partial c_L}{\partial x} + \kappa_{31}c_W + \kappa_{21}c_{SK} - \mu_1 c_L;$$

$$\frac{\partial c_{SK}}{\partial t} = D_S\frac{\partial^2 c_{SK}}{\partial x^2} - v_S\frac{\partial c_{SK}}{\partial x} + \kappa_{42}c_{PK} + \kappa_{12}c_L - \mu_2 c_{SK};$$

$$\frac{\partial c_W}{\partial t} = \frac{G_W(t)}{R_W V_L} + \kappa_{43}c_{PK} + \kappa_{13}c_L - \mu_3 c_W;$$

$$\frac{\partial c_{PK}}{\partial t} = \frac{G_{PK}(t)}{V_L} + \kappa_{24}c_{SK} + \kappa_{34}c_W - \mu_4 c_{PK};$$

As the system of *equations* has the second-order derivative with respect to the depth *x* for the liquid phase of sediments, two boundary conditions are required:

$$\left[-D\frac{\partial c_L(t,x)}{\partial x} + v c_L(t,x)\right]\Big|_{x=0} = v c_W(t);$$

$$\frac{\partial c_L(t,x)}{\partial x}\Big|_{x\to\infty} = 0.$$

The first one of the two boundary conditions describes the contaminants in the sediment liquid phase and the lake water liquid flow balance at the water sediment interface (x=0), and the other one – removes the possibility of existence of the infinite volumetric activities away from the interface area limits.

Experimental validation

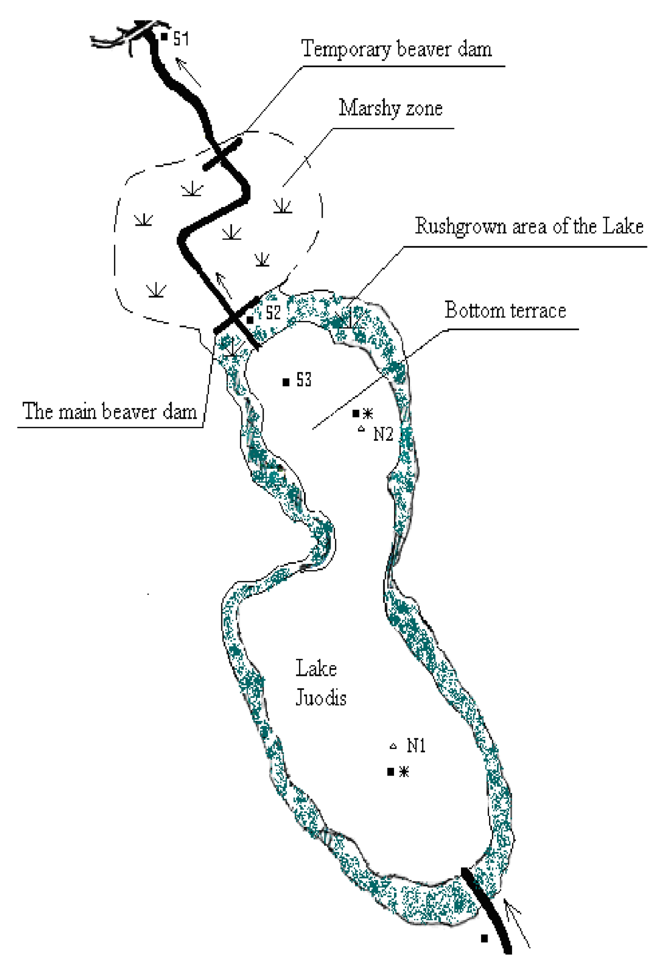

Fig. 2. Lake Juodis in Lithuania

Parameter	Value
Parameters for stage I	
L – Length of the sediment layer	100 cm
v – Darcy velocity	0.008 cm day^{-1}
D – dispersion (diffusion) coefficient	0.2 cm^2 day^{-1}
ρ_s –sediment dry matter density	1.0 g cm^{-3}
ρ_b –sediment balk density	0.043 g cm^{-3}
$\theta = 1 - \rho_b / \rho_s$ – porosity	0.96
k_D – exchangeable part KD	500 cm^3 g^{-1} or L kg^{-1}
$\alpha_{12} \equiv \kappa_f$ – exchang. part velocity	$\kappa_f = \ln(2)/20 = 0.0347$ day^{-1}
$\alpha_{21} \equiv \kappa_s$ – exchang. part velocity	$\kappa_s = \kappa_f / (k_D * \rho_b) = 0.0016$ day^{-1}
λ_f – degradation (decay) constant	ln(2) / (30*365.25) = 0.000063 day^{-1}
λ_s – degradation (decay) constant	0.000063 day^{-1}
g_e – initial water activity in Lake	1.0 Bq cm^{-3}
α_e – activity decrease in Lake const.	ln(2) / 30 = 0.023 day^{-1}
v_0 – activity deposition rate in Lake water	0.1 cm day^{-1}
Additional parameters for stage II	
f_{e1} – initial activity at x=0 (water ph.)	0.697 Bq cm^{-3}
α_{e1} – expon. decrease rate by depth	0.0807 cm^{-1}
f_{e2} – initial activity at x=0 (solid ph.)	0.591 Bq cm^{-3}
α_{e2} – expon. decrease rate by depth	0.0875 cm^{-1}
ω_{21} – from solid phase to water phase	0.0385 day^{-1}
μ_1 – degradation+decay+exch. const.	0.0362 day^{-1}
ω_{12} – from water phase to solid phase	0.0346 day^{-1}
μ_2 – degradation+decay+exch. const.	0.0369 day^{-1}

Results

Samples – sediment cores, 40 cm deep. The measured vertical distribution of ^{137}Cs in lake sediments $c_{S(t,x)}$ shows a peculiar deposition after the Chernobyl accident and nuclear weapons testing peaks (Fig. 3):

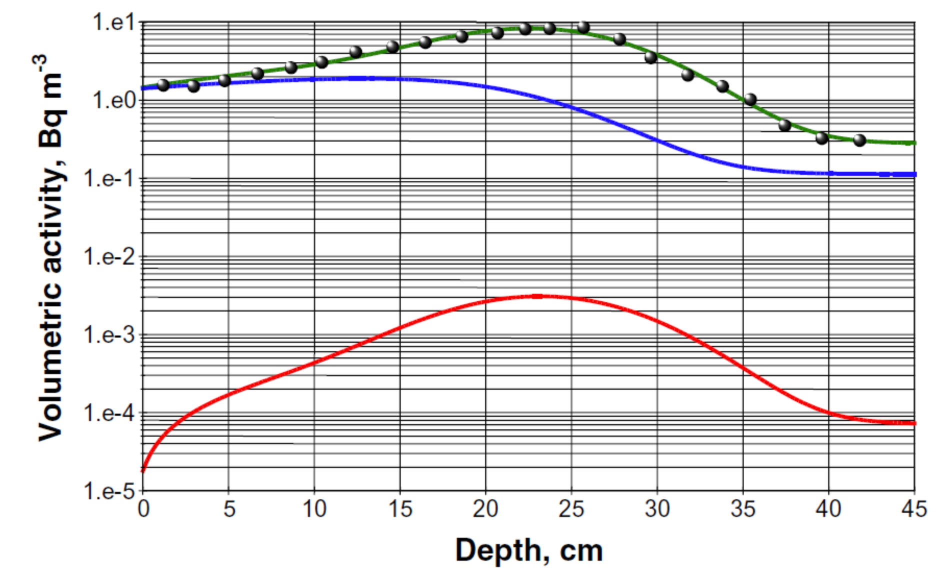

Fig. 3. Volumetric activities [Bq m^{-3}] depending on the depth of sediments *x* [cm] for the liquid phase of the sediments c_L (red), for the solid-phase kinetic parts c_{SK} (blue) of the total aggregated volumetric activities in solid phase (kinetic and fast) c_S (green) and measured volumetric activities (total aggregated) c_S (points).

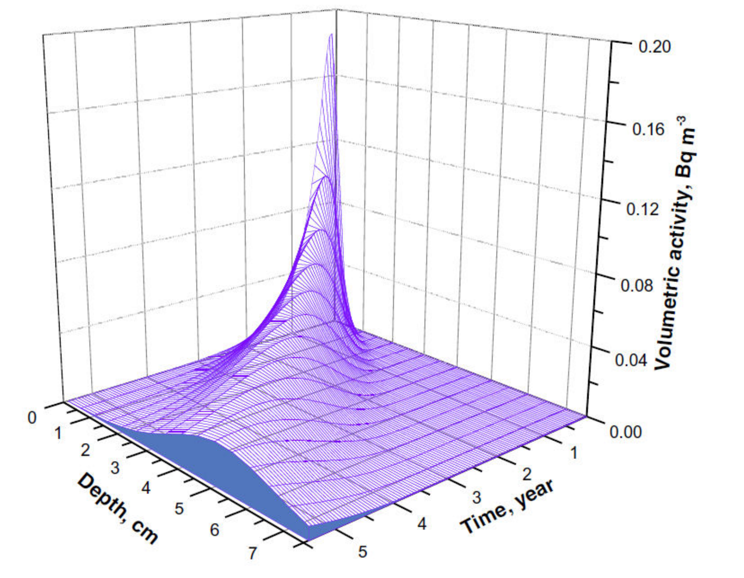

Fig. 4. Formation of ^{137}Cs volume activity c_L [Bq m^{-3}] in sediment *liquid* phase, depending on the sediment depth and on time after the acute release to the lake, obtained using the model.

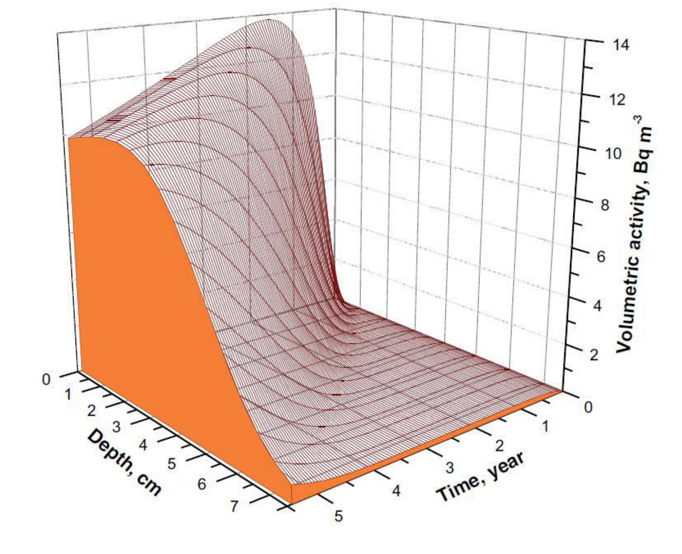

Fig. 5. Formation of ^{137}Cs volume activity c_L [Bq m^{-3}] in sediment *solid* phase, depending on the sediment depth and on time after the acute release to the lake, obtained using the model.

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The advantages of the model can also be attributed not only to the ability to apply modification of a two-site sorption/desorption method for both water and sediment spheres, but also to provide its own determination of the lake water contamination for various forms of contamination sources (intake to the lake) in the model for the whole investigation period.

Conclusion

The multiphase model characterizing the radionuclide migration between the abiotic components of the lake was developed.
The model can be used in experimental measurements interpreting the contaminant profile in lake sediments as well as a part associated with the comprehensive determination of the volumetric activity, in the estimation of irradiation doses due to radionuclides released into the lake water.
The presented model can be extended by including the biota typical processes (e.g., bioturbation) into the model!!!!!!!

References:
1. B. Lukšienė et al. J Radioanal Nucl Chem (2014) 300:277–286
2. V. Filistovič et al. Water Air Soil Pollut (2015) 226:202

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