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Reconstruction of long-term dynamics of Chernobyl-derived ^{137}Cs in the Upa River using bottom sediments in the Scheckino reservoir and semi-empirical modelling

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Abstract. Two cores of bottom sediments were collected in 2018 to a depth of ∼200 cm in the deepest part of the Scheckino reservoir on the Upa River (9500 km^2), Tula region, Russia. This area was severely contaminated by radiocesium (r-Cs) after the Chernobyl accident in 1986. The fact that ^{137}Cs activity concentrations in a specific horizon of the bottom sediments correspond to ^{137}Cs concentrations associated with suspended matter delivered to the reservoir, provides a basis for constructing the dynamics of particulate ^{137}Cs activity concentrations in the Upa River catchment from 1986 to 2017. Over the time since the Chernobyl accident, the particulate ^{137}Cs concentrations have decreased by more than an order of magnitude, with only minor changes occurring during the last 15 years. Using a typical value for the distribution coefficient K_d for the rivers of the Chernobyl contamination zone, dissolved ^{137}Cs activity concentrations in the Upa River have been estimated and their changes over the past 30 years since the accident have been studied. The resulting estimates of dissolved ^{137}Cs concentrations in the Upa River have been found to be in good agreement with measured data over the period 1987–1991. The proposed and tested method provides a basis for reconstructing the long-term dependence of radionuclide concentrations in rivers and reservoirs based on their vertical distribution in bottom sediments. Reconstructed time dependencies of particulate and dissolved ^{137}Cs activity concentrations in the Upa River were found to be described well by the proposed semi-empirical "diffusion" model based on an assumption that the time dependency of particulate r-Cs in the river corresponds to the time dependency of its concentration in top soil layers across the catchment which can be approximated by a dispersion-convection equation with physically meaningful parameters.

1 Introduction

Large areas of land were contaminated with radionuclides from the Chernobyl fallout (Izrael, 1998). These areas became long-term sources of radionuclides to natural waters and aquatic ecosystems in the Central European Russian landscape (Ivanov et al., 2017). Wash-off by surface runoff is the primary pathway for contamination of water bodies after initial fallout deposition from the atmosphere (Konoplev et al., 1992). Initial radioactive contamination of water bodies after the Chernobyl accident was relatively high as a result of direct fallout onto river and reservoir surfaces.

Over longer time periods after fallout, radionuclides bound in catchment surface soils are slowly transferred to rivers by the erosion of soil particles (particulate phase) and by desorption from soil to solution (dissolved phase). The rates of radionuclide transfer are influenced by the extent of soil erosion, the strength of radionuclide binding to catchment soils

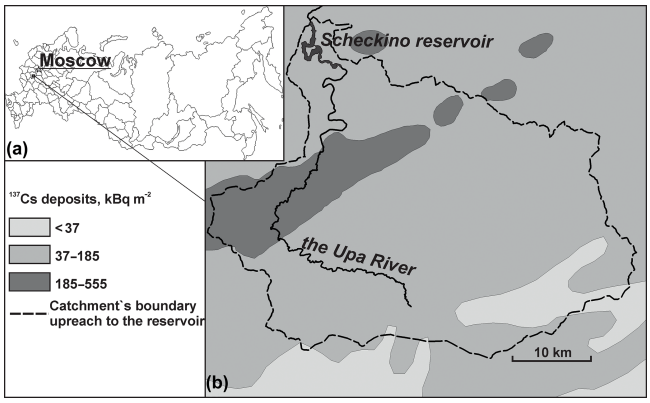

Figure 1. Location of the Upa river basin **(a)** and map of Chernobyl-derived ^{137}Cs deposition in the Upa River catchment upstream the Scheckino reservoir (Izrael, 1998) decay corrected for 1 May 1986 **(b)**.

and the potential for migration down the soil profile (Golosov et al., 2018). Detailed analysis of Chernobyl data over an extended period provides a basis for long-term prediction of changes in environmental radioactive contamination as a result of the Fukushima accident, or any other emergency involving radioactive emissions to the surrounding environment.

Unfortunately, long-term data are not always available regarding changes in radioactive contamination, and this is the case for ^{137}Cs in the Chernobyl zone rivers. Against this background, this study attempted to reconstruct, from 1986 to the present day, changes in ^{137}Cs concentrations in the Upa River as a result of the Tula-Oryol contamination zone, based on the current vertical distribution of ^{137}Cs (as of 2018) in bottom sediments of the Scheckino reservoir. These changes are described by a semi-empirical diffusion model.

2 Materials and methods

2.1 Sampling and processing of bottom sediments

The Upa River basin lies in the northern part of the Central Russian Upland within the Tula region (Fig. 1). Mean annual precipitation in the basin is ~ 540 mm yr^{-1}. The southern part of the basin, with a high proportion of arable land, was heavily contaminated by the Chernobyl fallout in April–May 1986. The northern forested part appeared to be less contaminated. Tula city is located in the north-east part of the Upa basin. The upper part of the Upa river (1362 km^2) is restricted by the Scheckino reservoir.

The reservoir has an area of 6 km^2 and is elongated upwards of the dam following the drowned valley of the Upa River. For the investigation of radionuclide concentrations in bottom sediments, the upper part of the reservoir was selected since local sources of sediment such as bank erosion and inputs resulting from the transport of sediment by small streams were judged to be minor contributors.

The bottom sediments were collected in February 2018 using a pistol sampler in the deepest parts of the Scheckino reservoir in which higher rates of sedimentation were expected. The cores were wrapped in cellophane film and transported to laboratory. In laboratory, the cores were sliced into 2 cm sections, weighed, dried at 105 °C for 8 h and weighed again for determining moisture content and calculating dry sediment mass. The samples were then ground, sieved through 2 mm mesh and placed in a container of defined geometry for measurement of ^{137}Cs activity using a semi-conductor γ-spectrometer with high purity Ge detector.

2.2 Semi-empirical "diffusional" model of ^{137}Cs dynamics in river water

2.2.1 Particulate ^{137}Cs in surface runoff and rivers

Over the long term, it can be assumed that a major source of suspended matter in surface runoff and rivers is the upper layer of catchment soils. Radionuclide concentrations in top soil decrease over time due to vertical migration to deeper layers. Hence vertical migration in catchment soils is a critical process responsible for the decline in particulate-associated radionuclide concentrations in surface waters. According to the convection-diffusion model (Crank, 1975; Prokhorov, 1981; Bulgakov et al., 2002) a change in radionuclide concentration in the top soil layer C_s with time is given by the following equation:

$$C_s(t) = \frac{\sigma}{\sqrt{\pi D_{eff} t}} e^{-\left(\frac{v^2}{4D_{eff}} + \lambda\right)t} \quad (1)$$

where D_{eff} is the effective dispersion coefficient; v is the effective velocity of convective transport; λ is the radioactive decay constant; σ is the radionuclide deposition density, and t is time.

The time dependence of particulate radionuclide concentrations C_p in surface runoff and river water can be described by this equation using averaged values of D_{eff} and v over the catchment area.

Studies of r-Cs vertical migration in soils of river catchments show that, as a rule, its transport due to dispersion prevails over its convective transport (Konshin, 1992; Ivanov et al., 1997; Konoplev et al., 2016). Therefore,

$$\frac{v^2}{4D_{eff}} \ll \lambda \quad (2)$$

Eq. (1) can be simplified as follows:

$$C_p(t) = \frac{\sigma}{\sqrt{\pi D_{eff} t}} e^{-\lambda t} \sim \frac{e^{-\lambda t}}{\sqrt{t}} \quad (3)$$

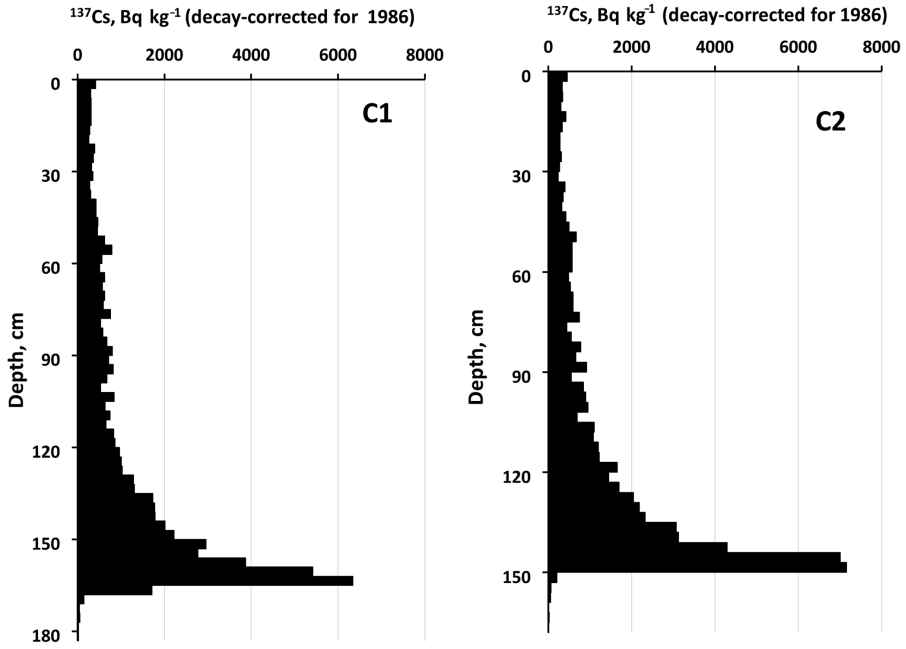

Figure 2. Vertical distribution of ^{137}Cs, decay-corrected for 1986, in the reservoir cores. The total inventory of ^{137}Cs in core C1 is 1030 kBq m^{-2} compared with 1160 kBq m^{-2} in core C2.

2.2.2 Dissolved ^{137}Cs in surface runoff and rivers

Dissolved radionuclides in surface runoff and rivers are transferred from soil to water due to cation exchange. As this takes place, only the exchangeable fraction of radionuclides is involved in exchanges with the dissolved phase. Therefore, dissolved radionuclide concentrations in water C_d relate to particulate concentrations as follows (Konoplev and Bulgakov, 2000):

$$C_d(t) = \frac{C_p(t) \cdot \alpha_{ex}}{K_d^{ex}} = \frac{C_p(t)}{K_d^{tot}} \quad (4)$$

where α_{ex} is the exchangeable fraction of the radionuclide, and K_d^{tot} and K_d^{ex} are, respectively, the total and exchangeable distribution coefficients of the radionuclide.

Over longer time periods, when equilibrium between exchangeable and nonexchangeable forms of r-Cs is reached and when condition (2) is valid, the equation for radionuclide dissolved concentrations can be presented as follows:

$$C_d(t) = \frac{\alpha_{ex}(\infty)\sigma}{K_d^{ex}\sqrt{\pi D_{eff} t}} e^{-\lambda t} = \frac{\sigma}{K_d^{tot}\sqrt{\pi D_{eff} t}} e^{-\lambda t} \sim \frac{e^{-\lambda t}}{\sqrt{t}} \quad (5)$$

The advantage of this semi-empirical "diffusion-based approach" is that all phases after the accident can be described by the same equation using the same physically-based parameters which can be estimated or determined in the field or using laboratory studies. Generally speaking, the decay-corrected particulate and dissolved ^{137}Cs activity concentrations in both surface runoff and rivers should follow the inverse square root of the time function.

3 Results and discussion

Figure 2 presents the vertical distribution of ^{137}Cs activity concentrations in the two bottom sediment cores collected from the study reservoir (C1 and C2). Accumulation of sediments entering the reservoir is associated with an increase in the total inventory of ^{137}Cs. In both cores, the ^{137}Cs inventory is more than twice the corresponding maximum inventory for the Upa River basin (Fig. 2)

Regretfully, no systematic long-term monitoring of radioactive contamination has been conducted on the Upa River. Data are only available for the first few years after the accident (1987–1991) for total activity concentrations of ^{137}Cs in water (dissolved + particulate) on the Upa River (Vakulovsky et al., 1994). The data obtained by the present study regarding the vertical distribution of ^{137}Cs in bottom sediments in deep-water zones, where sediments tend to accumulate, can provide a basis for reconstructing temporal changes in both particulate and dissolved ^{137}Cs concentrations in the Upa River water following the Chernobyl accident. Here, we make an assumption that annual sediment accumulation on the bottom of the reservoir was not changing over the years since the accident. The shape of the ^{137}Cs vertical distribution in the cored sediment accumulation zones

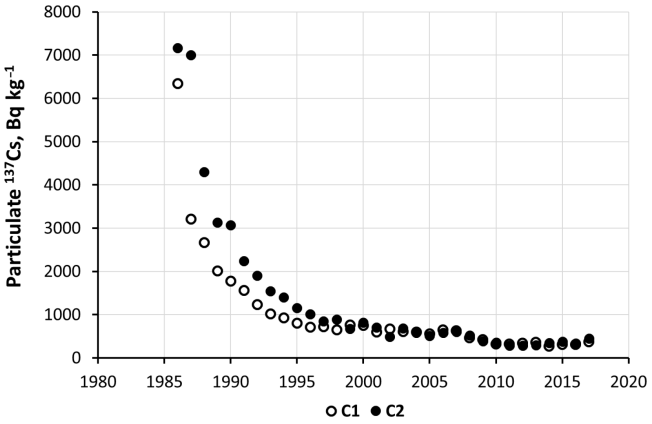

Figure 3. Reconstructed time dependencies of particulate ^{137}Cs activity concentrations (decay-corrected for 1986) in the Upa River based on ^{137}Cs vertical distributions in cores C1 and C2 collected in February 2018.

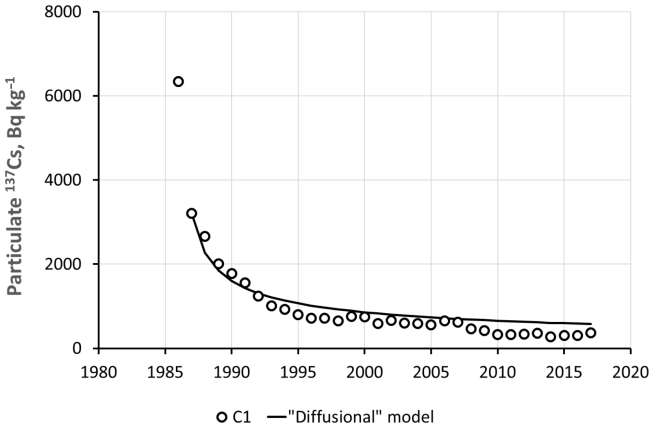

Figure 4. Diffusion based semi-empirical modelling of reconstructed particulate ^{137}Cs in the Upa River (line) based on the depth profile for the bottom sediments in core C1 (points).

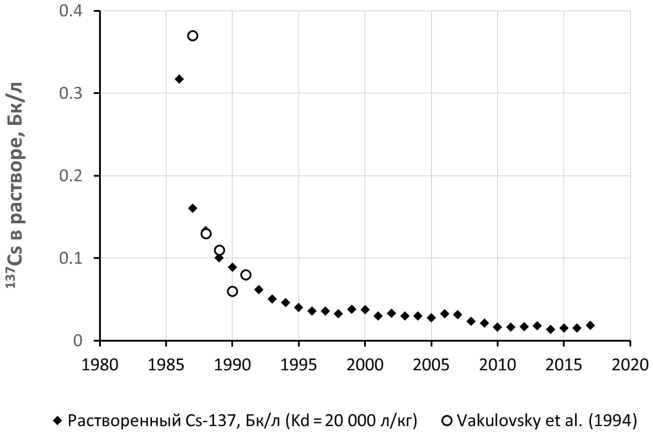

Figure 5. Comparison of the predicted time dependence of dissolved ^{137}Cs activity concentrations in the Upa River based on reconstructed particulate ^{137}Cs from sediments in core C1 with published measurements for 1987–1991 (Vakulovsky et al., 1994). To calculate dissolved ^{137}Cs activity concentrations, the total distribution coefficient $K_d^{tot} = 20\,000\,\mathrm{L\,kg^{-1}}$ (Konoplev, 2015) was used.

(Fig. 2) indicates that depositing sediments do not mix vertically and that the ^{137}Cs incorporated in them does not migrate, since the ^{137}Cs peak is well defined. Considering the mean rate of sediment accumulation is about 5 cm yr^{-1}, based on the position of the peak radionuclide concentration (Fig. 2), the bottom sediment layers can be attributed to a certain period of sediment accumulation. If ^{137}Cs activity concentration in a respective layer matches its concentration on suspended matter in this time period, changes in ^{137}Cs concentrations on suspended matter in the Upa River from 1986 to 2017 can be easily derived.

Figure 3 shows a reconstructed time dependence of particulate ^{137}Cs concentration in the Upa river, based on the data for the two bottom sediment cores collected in February 2018. As can be seen, over the time since the accident, ^{137}Cs concentrations have decreased by more than an order of magnitude, with only minor changes during the last 15 years.

Figure 4 shows the results of calculating temporal changes in ^{137}Cs concentrations in suspended material in the Upa River by the described model (line) with concentrations reconstructed based on the sediment core C1. As can be seen, the "diffusion" model provides a good representation of the time dependency of particulate ^{137}Cs concentrations in the Upa River reconstructed using the depth profile in the bottom sediments.

Further, using the typical value of the distribution coefficient K_d for the rivers of the Chernobyl contaminated zone, it is possible to estimate the concentration of dissolved ^{137}Cs in the Upa River and its variation over the 30 years following the accident. A K_d of 20 000 L kg^{-1} was taken as being a representative value on the basis of analysis of data for a number of rivers in Chernobyl contaminated areas (Konoplev, 2015). Using this value, and reconstructed data for particulate ^{137}Cs activity concentrations, it is possible to estimate the concentrations of dissolved ^{137}Cs in the Upa River for the time window from 1986 to 2017. The results of these estimations are presented in Fig. 5. The published measurement data for 1987–1991 (Vakulovsky et al., 1994) is also given for comparison. It can be seen that the predicted activity concentrations of dissolved ^{137}Cs in the Upa River and their temporal trend are consistent with the measured data.

Data availability. The data can be accessed by request to authors.

Author contributions. AVK suggested the idea and semi-empirical model, MMI, VNG and EAK designed the sampling and provided radiocesium analysis, AVK analyzed the data and prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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