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# Using reservoir sediment deposits to determine the longer-term fate of chernobyl-derived $^{137}$Cs fallout in the fluvial system☆

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ABSTRACT

Vast areas of Europe were contaminated by the fallout of $^{137}$Cs and other radionuclides, as a result of the Chernobyl accident in 1986. The post-fallout redistribution of Chernobyl-derived $^{137}$Cs was associated with erosion and sediment transport processes within the fluvial system. Bottom sediments from lakes and reservoirs can provide a valuable source of information regarding the post-fallout redistribution and fate of $^{137}$Cs released by the Chernobyl accident. A detailed investigation of sediment-associated $^{137}$Cs in the bottom sediments of a reservoir in a Chernobyl-affected area in Central Russia has been undertaken. A new approach, based on the vertical distribution of $^{137}$Cs activity concentrations in the reservoir bottom sediment makes it possible to separate the initially deposited bottom sediment, where the $^{137}$Cs activity reflects the direct fallout of Chernobyl-derived $^{137}$Cs to the reservoir surface and its subsequent incorporation into sediment deposited immediately after the accident, from the sediment mobilized from the catchment deposited subsequently. The deposits representing direct fallout from the atmosphere was termed the "Chernobyl peak". Its shape can be described by a diffusion equation and it can be distinguished from the remaining catchment-derived $^{137}$Cs associated with sediment accumulated with sediments during the post-Chernobyl period. The $^{137}$Cs depth distribution above the "Chernobyl peak" was used to provide a record of changes in the concentration of sediment-associated $^{137}$Cs transported from the upstream catchment during the post-Chernobyl period. It was found that the $^{137}$Cs activity concentration in the sediment deposited in the reservoir progressively decreased during the 30-year period after the accident due to a reduction in the contribution of sediment eroded from the arable land in the catchment. This reflects a reduction in both the area of cultivated land area and the reduced incidence of surface runoff from the slopes during spring snowmelt due to climate warming.



## 1. Introduction

The radioactive isotope $^{137}$Cs is one of the primary radioactive contaminants released into the environment as a result of nuclear weapons tests and accidents at nuclear facilities, such as that at Chernobyl (Ritchie, McHenry, 1990; Devell et al., 1995). Because of its close affinity with soil and sediment particles, the post-fallout redistribution and fate of $^{137}$Cs is closely associated with the processes of erosion and sediment transport. As a result, this radioisotope has been widely used as a tracer in soil erosion and sediment budget investigations (Walling and Quine, 1991; Owens et al., 1997; Golosov et al., 1999). Fluvial transport is the primary mechanism of $^{137}$Cs redistribution within and beyond contaminated areas (Monte, 1995; Sansone et al., 1996; Taniguchi et al., 2019). When considering the longer-term fate of this radioactive contaminant, it is important to recognize that the sediment yield

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from a catchment or river basin represents the output from a complex set of processes, varying both spatially and temporally (Walling, 1983). The mobility and redistribution of $^{137}$Cs and other sediment-associated pollutants generally increase in areas with a high proportion of cultivated land, due to the increased rates of soil loss (Evrard et al., 2015).

The land-use system and temporal changes in that system must be taken into account in studies and projections of longer-term $^{137}$Cs redistribution. The ploughing of cultivated soil after fallout receipt or in subsequent years results in the mixing of the upper 20–25 cm of soil, which leads to a decrease in the content of $^{137}$Cs in the surface horizons of the soil mobilized by erosion (He and Walling, 1997; Golosov and Ivanova, 2002). As a result, a progressive reduction in the $^{137}$Cs activity associated with mobilized sediment (Vetrov et al., 1990) and with sediment deposited in sediment sinks (Belyaev et al., 2013; Konoplev et al., 2018) can occur. Accumulation of substantial amounts of sediment in sediment sinks such as dry valleys and reservoir bottoms can result in high $^{137}$Cs inventories, which can represent a potential hazard if this sediment is subsequently remobilized into the fluvial system (Panin et al., 2001; Golosov et al., 2018a,b). Problems associated with the longer-term redistribution of $^{137}$Cs fallout have again attracted attention after the accident at the Fukushima nuclear power plant in March 2011, which led to radioactive contamination of the river basins of the north-east coast of Honshu Island (Mouri et al., 2014; Sakaguchi et al., 2015; Konoplev et al., 2016).

Bottom sediments from lakes and reservoirs can potentially provide a valuable source of information regarding the post-fallout redistribution and fate of $^{137}$Cs released by the Chernobyl and Fukushima Nuclear Power Plant accidents, covering periods of more than 30 years and ten years, respectively. Such information can permit reconstruction of changes in the $^{137}$Cs activity of mobilized or transported sediment, as well as changes in sediment loads and yields (Konoplev et al., 2019). Most studies that have investigated contaminated sediment deposited in lakes and reservoirs after the Chernobyl accident (e.g Rank et al., 1990; Broberg and Andersson, 1991; Smith et al., 1999; Putyrskaya et al., 2009; Saxen et al., 2010) and the Fukushima Daichi Power Plant accident (e.g. Ochiai et al., 2013; Yoshimura and Yokoduka, 2014; Wakiyama et al., 2017; Funakia et al., 2019) have focused on either the magnitude of $^{137}$Cs storage or the spatial variation of $^{137}$Cs accumulation, although attention has also been directed to post-fallout mobilization in the waterbody (Vray et al., 2003; Klaminder et al., 2012), and redistribution of $^{137}$Cs within the upstream catchment (Konoplev et al., 2019).

For locations affected by the Chernobyl accident and where post-depositional mixing of the sediment deposited in lakes and reservoirs has been negligible, the maximum activity concentration of $^{137}$Cs in the bottom sediment column can be expected to reflect the receipt of $^{137}$Cs fallout by the surface of the water body in 1986. The existence of this time marker makes it possible to establish sedimentation rates for the post-Chernobyl period (Walling, 1990; He et al., 1996; Belyaev et al., 2004; Golosov et al., 2012) and to document changes in the $^{137}$Cs concentration of sediment delivered to the reservoir and deposited during this period (Konoplev et al., 2019).

The main objective of the study reported here was to document variations in the $^{137}$Cs activity concentration of sediment deposited in a reservoir during the post-Chernobyl period and to use this information to investigate the rate of decline of the $^{137}$Cs activity of suspended sediment transported by the river feeding the reservoir during this period and thus the post-fallout sediment mobilization and transport of $^{137}$Cs from the contributing catchment.

## 2. Materials and methods

### 2.1. Meeting the requirements for a study reservoir

In order to use the vertical distribution of $^{137}$Cs in the bottom sediment of a reservoir to provide information on the export of Chernobyl-derived $^{137}$Cs from the catchment feeding that reservoir, a number of conditions and constraints need to be recognized (e.g. Walling and He, 1992; He et al., 1996). To meet these requirements, the water body must have been in existence at the time of the fallout from the Chernobyl accident and sedimentation in the reservoir should have continued with little change or disturbance through to the present. More particularly, the reservoir should not have been drained or its bottom sediments disturbed since 1986. Similarly, the sediment column should not have been subject to significant post-depositional disturbance or mixing due to both natural processes (e.g. bioturbation, creep, erosion, ice ploughing, major drawdown etc.) or human impact. As such, the sediment column should provide a representative record of sediment deposition. This is essential to ensure that a clear peak in $^{137}$Cs activity corresponding to the occurrence of Chernobyl-derived fallout in 1986 (i.e. the Chernobyl peak) exists and that the overlying sediment column is directly representative of the sediment input to the reservoir and its progressive deposition since 1986.

### 2.2. The study reservoir and its catchment

The Schekino reservoir selected for the study is located in the upper reaches of the Upa river basin in the central Tula region, which lies ca 200 km (south) of Moscow, Russia (Fig. 1A) The reservoir has a surface area of 5.9 km$^2$ and a design volume of 20.7x10$^6$ m$^3$. It was constructed in 1950 as an element of the Schekino State District Power Plant, located in the city of Sovetsk (see Fig. 1B). Water from the reservoir is used for cooling purposes and for the heating system serving the city of Sovetsk. The reservoir is impounded by a concrete gravity dam, 10 m high and 36 m long. The water level in the reservoir is maintained relatively constant within a range of 2–3 m by a spillway equipped with a radial gate. The reservoir is subdivided into two sections by an earth dam constructed across the central part of the reservoir. The two sections are connected by an artificial channel. This configuration aims to restrict siltation primarily to the upper section of the reservoir. The trap efficiency of the reservoir is estimated to be more than 90%, which is typical for reservoirs of this size in the forest-steppe zone of European Russia (Prytkova, 1984).

The catchment of the reservoir represents the upper reaches of the Upa River (Fig. 1C). The $^{137}$Cs fallout within the reservoir catchment area (1362 km$^2$) associated with the Chernobyl accident ranged from 37 to 555 kBq m$^{-2}$ (Fig. 1C). The map of the initial $^{137}$Cs contamination of the study area (Fig. 1C) was constructed based on information derived from both very detailed aerial surveys and dense field soil sampling programmes (de Cort et al., 1998). The mean annual precipitation for the catchment is about 540 mm yr$^{-1}$, with values showing a reduction from west to east. The catchment area is characterized by a relatively homogeneous soil cover, represented primarily by Phaeozems: grey forest soils and northern Chernozems developed on carbonate loess-like heavy loams with a <0.01 mm fraction greater than 50% (Mamikhin et al., 2016). Arable land occupies between 30 and 40% of the area in the northern part of the catchment, increasing to 80–90% in the southern part (Ivanov et al., 2019).

The reservoir shores are stable, with little or no evidence of bank erosion. The side slopes of the Upa River valley adjacent to the reservoir are mainly occupied by meadows, with some areas of forest. The residential area of Sovetsk town adjoins the left bank of

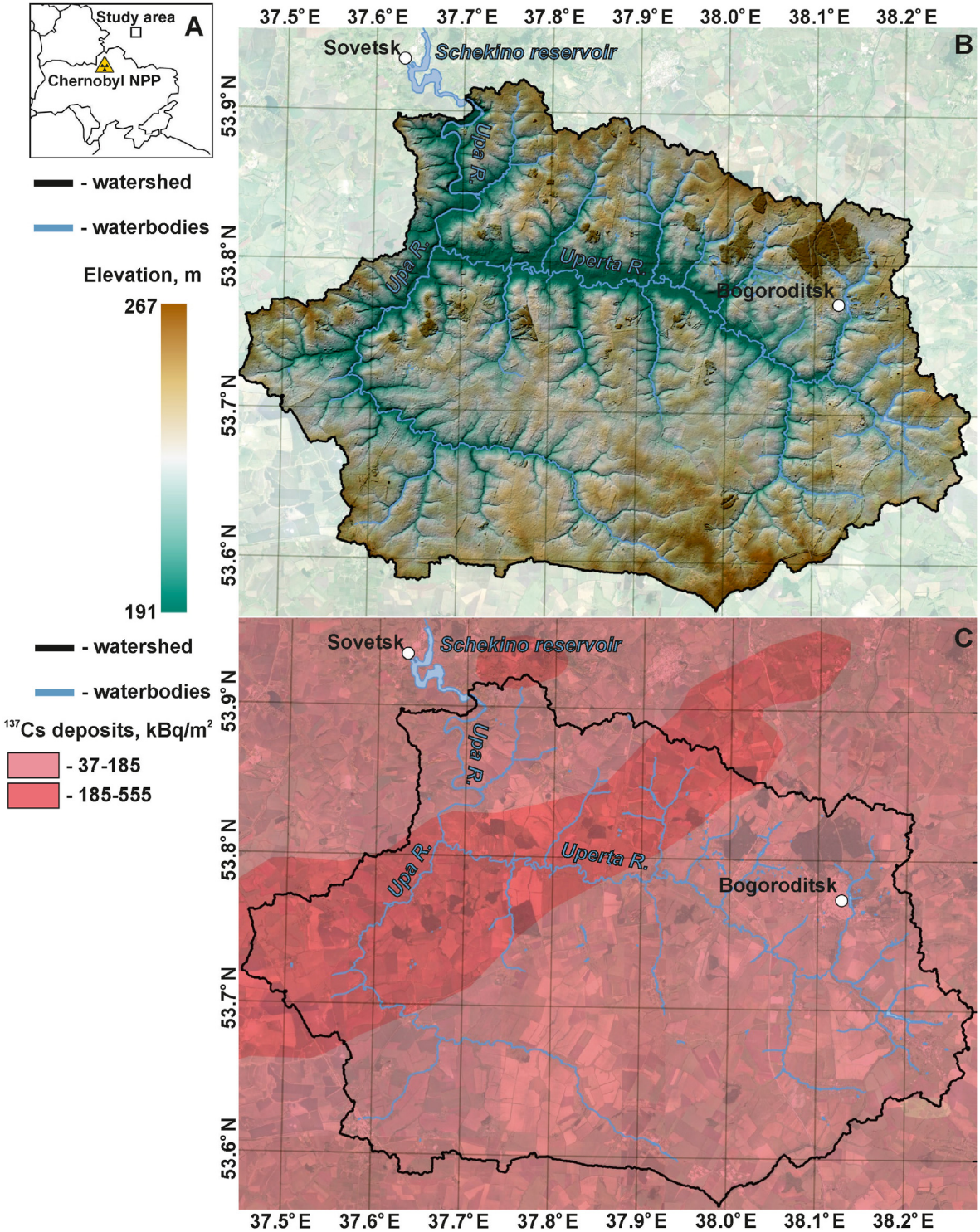


**Fig. 1.** A – Location of Chernobyl NPP and study area; B – A Digital Elevation Model of the Schekino reservoir catchment; C – The spatial distribution of $^{137}$Cs contamination within the catchment of the Schekino reservoir resulting from fallout from the Chernobyl accident (based on de Cort et al., 1998).

the reservoir in the section located close to the main concrete dam. The contribution of local sediment sources to the sediment deposited in the reservoir is therefore judged to be negligible. The dominant source of sediment accumulating in the reservoir bottom is the sediment input associated with the Upa River, which is derived from the upstream catchment. There are two primary sediment sources within the Upa River catchment. The first represents the arable land, where soil erosion is observed during periods of snowmelt in March–April and heavy rainstorms during the months of May–October. The second comprises the river channels and more particularly eroding banks and channel beds. The contribution of gully erosion to the sediment yield from the catchment of the reservoir is judged to have been negligible during the period after 1986, since Golosov (2006) indicates that most of

the gullies in the area ceased to grow during that period. The majority of the sediment is delivered to the reservoir during the spring floods (Ivanov et al., 2019). However, the relative contribution of the summer and autumn floods has increased in recent decades, primarily due to a considerable reduction of surface runoff from the slopes during spring snowmelt. According to the available long-term records of surface runoff from the cultivated slopes, the mean annual runoff coefficient reduced from 0.36 before 1980 (period of observation 1959–1980) to 0.09 after 1980 (period of observation 1981–2014) (Barabanov et al., 2018). The reduction of the frozen soil depth, due to increasing air temperature in winter, is the main cause of the reduction in surface runoff during spring snowmelt after 1980 (Golosov et al., 2018a,b). Locally, heavy storm rainfall can lead to high soil losses from cultivated land in the catchment of the Upa River (Belyaev et al., 2008). However, the connectivity between the cultivated slopes and the permanent watercourses is limited and most of the eroded sediment is redeposited along the intervening pathways (Panin et al., 2001; Golosov and Ivanova, 2002).

### 2.3. Sample collection and processing

Sediment cores were collected from the deepest parts of the upper part of the reservoir, which corresponded with the location of the former channel of the Upa River. At each sampling point, two sediment cores were collected, to provide duplicate sediment columns. The distance between the duplicate cores was 3 m. The collection of duplicate cores was aimed at avoiding sampling errors and non-representative conditions and other potential problems, including local post-depositional disturbance. Sediment cores were collected from four sampling points, selected to characterize sections of the reservoir with different conditions of sediment deposition (Fig.SM1). The water depth in the reservoir was slightly different at the individual sampling points (S-1, 2–3.5 m; S-3, 4–3.2 m; S-5, 6–3 m; S-7, 8–3.2 m).

Sediment coring was undertaken from the ice in February 2018 and February 2019 using a piston sampler (Nesje, 1992). The design of the sampler permits samples to be collected from a given depth, bypassing the overlying part of the sediment column. This made it possible to obtain a single column of bottom sediment, consisting of several sequential cores (each of 1 m). The collected cores were wrapped in cellophane film, frozen and delivered to the laboratory for further processing. In the laboratory, the sediment cores were sliced into 3 cm depth incremental sections. The 3 cm depth increment for slicing was trialed in 2018 to confirm its ability to provide individual samples of sufficient mass and to achieve good depth resolution, whilst limiting the number of samples requiring analysis. This trial indicated that, due to the relatively high rates of sedimentation in the reservoir, a 3 cm depth increment was appropriate for core sectioning. Core sections were weighed, dried at 105 °C for 8 h, and re-weighed to determine their moisture content and calculate the density of the dry sediment. Subsequently, the samples were disaggregated and sieved to <2 mm. The < 2 mm fraction was placed in container of fixed geometry for subsequent measurement of $^{137}$Cs activity using a semiconductor gamma-ray spectrometer equipped with an HPGe detector.

To confirm the absence of significant mixing in the sediment column, the depth distribution of unsupported or excess $^{210}$Pb ($^{210}Pb_{ex}$) was examined in the longest core (S-4). Because of the low mass of sample available, it proved necessary to combine individual sections to represent 9 cm depth incremental samples. The results of the $^{210}Pb_{ex}$ measurements presented in the Supplementary Material (see Fig. SM2).

Spectrometric measurements were performed using a semiconductor γ-spectrometer with a high-purity (HPGe) detector SKS-07(09) "Green Star Instruments" (Russia). The specific activity of the radionuclides was measured for samples presented in standard geometries. The time periods associated with the Energy spectra measurements depended on the activity of the sample but were not less than 60000 s activity was determined from the net full energy peak for $^{137}$Cs at 661.7 keV, $^{226}$Ra at 609.3 and 186.2 keV and $^{210}$ Pb at 46.5 keV using standard procedures. The uncertainty of the γ-spectrometry measurements did not exceed + or - 10% at the 95% level of confidence. The detector was calibrated using IAEA reference materials containing $^{137}$Cs (IAEA-447 & 448). The limit of detection (MDA) was 0.88 Bq for $^{137}$Cs, 1.3 Bq for $^{226}$Ra and 1.34 Bq for $^{210}$Pb. The activity concentration of $^{137}$Cs in all the sediment samples was decay corrected to the time of fallout in 1986.

The grain-size composition of the samples representing the individual depth-incremental sections of the sediment cores was determined using a Malvern Mastersizer 3000 particle size analyzer. Preparation of samples for analysis included treatment with a 4% sodium pyrophosphate solution to disperse the clay fraction. A representative aliquot of the prepared suspension was pipetted into the liquid cuvette of the analyzer dispersion unit. The sediment in the cuvette was processed for 100 s with a 40 W ultrasound and intensively mixed by a centrifugal pump using a pump speed of 2400 rpm. After turning off the ultrasound, ten repeated measurements were made, and the results were averaged by the Mastersizer v.3.62 application. The distribution of particle size fractions was calculated based on the Fraunhofer diffraction model.

### 2.4. Analysis of the $^{137}$Cs depth distribution in the sediment cores

With the exception of cores S-5 and S-6, all the $^{137}$Cs depth distributions obtained for the reservoir sediment cores are characterized by the presence of a clearly defined depth increment containing the maximum $^{137}$Cs activity concentration and representing the "Chernobyl peak". For cores S-5 and S-6 part of the depth increment containing the maximum $^{137}$Cs maximum activity concentration was lost, because the "Chernobyl peak" was located between two separate 1 m sections of the cores collected from those sites.

The vertical distribution of $^{137}$Cs in reservoir sediment cores can be used to verify existing models of the post-fallout redistribution of radionuclides mobilized from a catchment by erosion and transported to the catchment outlet in association with the sediment load of the river. Since the sediment load of the rivers in the Upa River basin is comprised primarily of sediment eroded from the arable land, the post-fallout behaviour of $^{137}$Cs in the cultivated soil profile is a critical factor in controlling the $^{137}$Cs content of the eroded soil and therefore the $^{137}$Cs content of the suspended sediment load of the river. Post-fallout changes in the activity concentration of $^{137}$Cs in the upper horizons of the cultivated soil are effectively controlled by four key factors. These represent the process of radioactive decay, the regular mixing of the cultivated or plough layer by tillage, soil erosion and associated soil loss and surface lowering, and downward migration of radiocaesium within the soil profile. Information on the depth distribution of $^{137}$Cs in reservoir bottom sediment can be used to evaluate changes in the $^{137}$Cs content of the sediment transported by the river. Since the main source of the suspended sediment load of a river is commonly soil erosion from areas of cultivated land (Dedkov and Mozzherin, 1996), information on changes in the $^{137}$Cs content of the suspended sediment load of a river can be used to evaluate changes in the $^{137}$Cs content of the surface soil in cultivated areas. Following (Konoplev et al., 2019, 2020), the dynamics of changes in the decay corrected activity concentration of sediment-associated $^{137}$Cs in surface runoff and in a river can be approximately described by the

inverse root function of time.

Land use maps for the catchment of the Upa River basin upstream from the Schekino reservoir were compiled for three timelines, namely, 1985 (the year preceding the deposition of Chernobyl-derived $^{137}$Cs), 2000 (the mid-point of the time interval between Chernobyl fallout and the field sampling component of this study) and 2015.

## 3. Results and discussion

### 3.1. Possible $^{137}$Cs losses during sampling

The sediment and its associated radionuclide inventory overlying the $^{137}$Cs "Chernobyl peak" directly reflect sediment accumulation during the post-Chernobyl period. However, determination of the precise position of the "Chernobyl peak" can involve some uncertainty. The core processing and sample preparation techniques necessarily involve dividing the 1 m core sections into depth increments of equal thickness, for which the mean $^{137}$Cs content is determined. The assumed surface, which represents the sediment surface at the time of fallout receipt in 1986, cannot be precisely defined in terms of its mass depth, but must be defined by the upper and lower bounds of the depth increment characterized by the maximum activity concentration. If sediment accumulation rates are high, the potential uncertainty associated with identifying the depth of the $^{137}$Cs maximum activity concentration is likely to be of limited importance.

Losses of sediment from the cores during their collection can introduce further uncertainty in defining the depth distribution of $^{137}$Cs within the bottom sediment of the reservoir. To provide evidence of possible losses of sediment from the cores, the depth distributions of $^{137}$Cs in the duplicate cores were compared. Assuming that the sedimentation is relatively uniform within a small area, the position of the "Chernobyl peak" in the duplicate cores, which were collected 3 m apart, should be at the same mass depth. The most reliable and comprehensive information will be that provided by cores collected from locations with the greatest sedimentation rates. Where comparison of the mass depths associated with the sediment section characterized by the maximum $^{137}$Cs activity concentration demonstrated differences between the two duplicate cores (Table 1), the $^{137}$Cs depth distribution associated with the shorter core was corrected by adding the difference in mass depth shown in Table 1 (see Fig. 2).

In the case of cores S-5 and S-6, which comprised two sections, the highest $^{137}$Cs activity was found in the bottom segment of the upper part (1 m) of the core. As a result of the sampling technology, there is a risk of losing depth-incremental core sections located at the boundary. Because of this, the most reliable determination of the position of the "Chernobyl peak" will be provided by cores where a sharp increase and decrease in the activity concentration of $^{137}$Cs occurs within a single section. Nevertheless, the vertical distribution of $^{137}$Cs activity concentration above the designated peaks was seen to be almost identical. It is noteworthy that both plots indicate a minor peak in $^{137}$Cs activity at around 269 kg/m$^2$ (1030 Bq/kg) on the S-5 graph and 262 kg/m$^2$ (881 Bq/kg) on the adjusted S-6 curve (Fig. 2). The above similarity adds confidence to the correction of the depth distribution for Core S-5 based on the position of the "Chernobyl peak".

**Table 1**
Mass depth of the "Chernobyl peak" in cores collected from the Schekino Reservoir.

| Core | Mass depth of "Chernobyl peak", kg/m$^2$ | Difference, kg/m$^2$ |
|---|---|---|
| S-1 | 312 | 94 |
| S-2 | 406 | |
| S-3 | 792 | 53 |
| S-4 | 845 | |
| S-5 | 520 | 55 |
| S-6 | 465 | |
| S-7 | 629 | 90 |
| S-8 | 538 | |

### 3.2. Interpreting the downcore variations of $^{137}$Cs in the sediment cores

When interpreting the information on the downcore variation in the $^{137}$Cs activity of the sediment deposits from the reservoir provided by the sediment cores, it is important to confirm that the sediment column has not been disturbed by post depositional mixing or bioturbation. In this study, the very clearly defined form of the "Chernobyl peaks" depicted in Fig. 2 was seen as providing confirmation that no significant post depositional mixing or disturbance such as bioturbation had occurred. This conclusion was further supported by the measurements of unsupported $^{210}$Pb$_{ex}$ measurements undertaken on core S-4. The depth distribution of $^{210}$Pb$_{ex}$ in this core, presented in Figure SM2, shows the expected exponential decrease in activity with depth, although this is complicated by minor variations in $^{210}$Pb$_{ex}$ activity associated with temporal variation in the relative contribution of direct fallout-derived and catchment-derived $^{210}$Pb$_{ex}$ and of sediment from different sources within the catchment characterized by different $^{210}$Pb$_{ex}$ activity. However, the overall shape confirms the absence of substantial post depositional disturbance. The sediment columns represented by the cores were therefore seen as providing suitability records of the changes in the $^{137}$Cs activity of sediment deposited in the reservoir during the post Chernobyl period.

### 3.3. Origin and interpretation of the "Chernobyl peak"

Existing understanding indicates that the "Chernobyl peak", as reported above, represents $^{137}$Cs fallout to the reservoir surface at the time of the Chernobyl accident. The fraction of $^{137}$Cs fallout in solution would be adsorbed and then fixed by suspended sediment in the water column. Insoluble dust particles in the atmospheric fallout which incorporated $^{137}$Cs would combine with the suspended sediment and this sediment and its associated $^{137}$Cs would be subsequently deposited on the surface of the existing bottom sediment within the reservoir to produce a thin layer characterized by high $^{137}$Cs activity. Because of the limited amount of rainfall, the contribution of $^{137}$Cs, both particulate and in solution, associated with storm runoff entering the reservoir during and immediately after the period of fallout are judged to have been negligible when compared with the direct atmospheric fallout to the water surface of the reservoir (Konoplev et al., 1992). The shape of the "Chernobyl peak" can be described by a Gauss function, reflecting subsequent dispersion of the deposited $^{137}$Cs, both upwards and downwards within the sediment column.

$$C_x = C_0 * e^{-\frac{(x-x_0)^2}{2\sigma^2}}, \quad (1)$$

Where: $C_x$ is the $^{137}$Cs activity concentration at depth x, Bq/kg; $C_0$ is the $^{137}$Cs activity concentration at the maximum ("Chernobyl peak"), Bq/kg; $x$ is the mass-depth of the given sediment layer, kg/m$^2$; and $x_0$ is the mass-depth of the "Chernobyl peak", kg/m$^2$.

Using this approach, the precise position and shape of "the Chernobyl peak" in the sediment columns represented by the individual cores can be specified by overlaying the Gauss curve onto the empirically derived $^{137}$Cs depth distribution. It was assumed that equation (1) should reliably describe the $^{137}$Cs depth

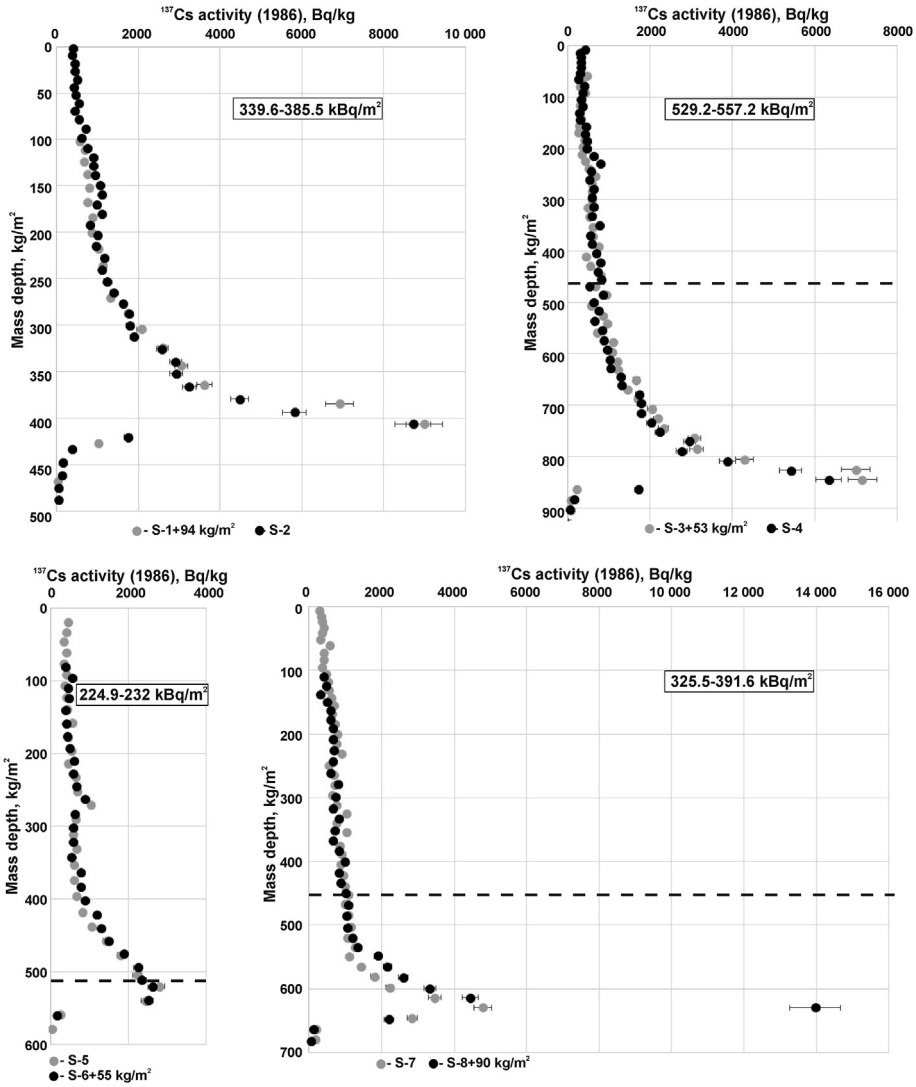


**Fig. 2.** $^{137}$Cs depth distribution curves for the sediment cores (the dashed line shows the boundary between the lower and upper parts of the core, if the core was collected in two 1 m sections.

distribution immediately below the "Chernobyl peak". However, the effects of cross-contamination between the core slices are likely to result in overestimation of the $2\sigma^2$ values. The results of these calculations are presented in Table 2. Cores S-5 and S-6 were excluded from these calculations, because the sediment slice characterized by the maximum activity concentration of $^{137}$Cs was partly lost.

Where: *$C_1$ is the $^{137}$Cs activity concentration in the slice of bottom sediment located below the sediment layer with the maximum $^{137}$Cs activity concentration (i.e. the "Chernobyl peak"); $C_0$ is the $^{137}$Cs activity concentration associated with the "Chernobyl peak" $x_1$ is the mas depth of the sediment layer located immediately below the sediment layer with the maximum $^{137}$Cs activity concentration; $x_0$ is the mass depth of the "Chernobyl peak".*

**Table 2**
The parameters of the Gauss functions (Equation (1)) fitted to the cores from the Schekino reservoir.

| Core | $x_0$, kg/m$^2$ | $C_0$, Bq/kg | $x_1$, kg/m$^2$ | $C_1$, Bq/kg | $2\sigma^2$ |
|---|---|---|---|---|---|
| S-1 | 406 | 9000 | 426 | 1010 | 189 |
| S-2 | | 8720 | 420 | 1740 | 125 |
| S-3 | 845 | 7160 | 864 | 206 | 102 |
| S-4 | | 6340 | 864 | 1720 | 183 |
| S-7 | 629 | 4790 | 646 | 2830 | 532 |
| S-8 | | 14000 | 648 | 2190 | 193 |

The fitted values of $2\sigma^2$ were used to represent the distribution of direct Chernobyl fallout $^{137}$Cs in the reservoir bottom sediment and to generate the corresponding depth distribution (see Fig. 3). However, it can be seen that in some cases the slicing of cores into 3 cm depth increments may not provide sufficient resolution to determine the precise location of the "Chernobyl peak." For example, Fig. 4 indicates that for core S-3 two adjacent core slices were characterized by very similar $^{137}$Cs activity concentrations. This suggests that the "Chernobyl peak" is located close to the boundary between these two slices of the core and that the value for the $2\sigma^2$ parameter may therefore be incorrect. This could result in underestimation of the initial direct input of Chernobyl-derived $^{137}$Cs fallout to the reservoir bottom sediment. This potential source of uncertainty should therefore be taken into account when sectioning a core. In studies where the capacity to determine the $^{137}$Cs content of a large number of samples exists, there can clearly be advantages in slicing the core into thinner sections and applying

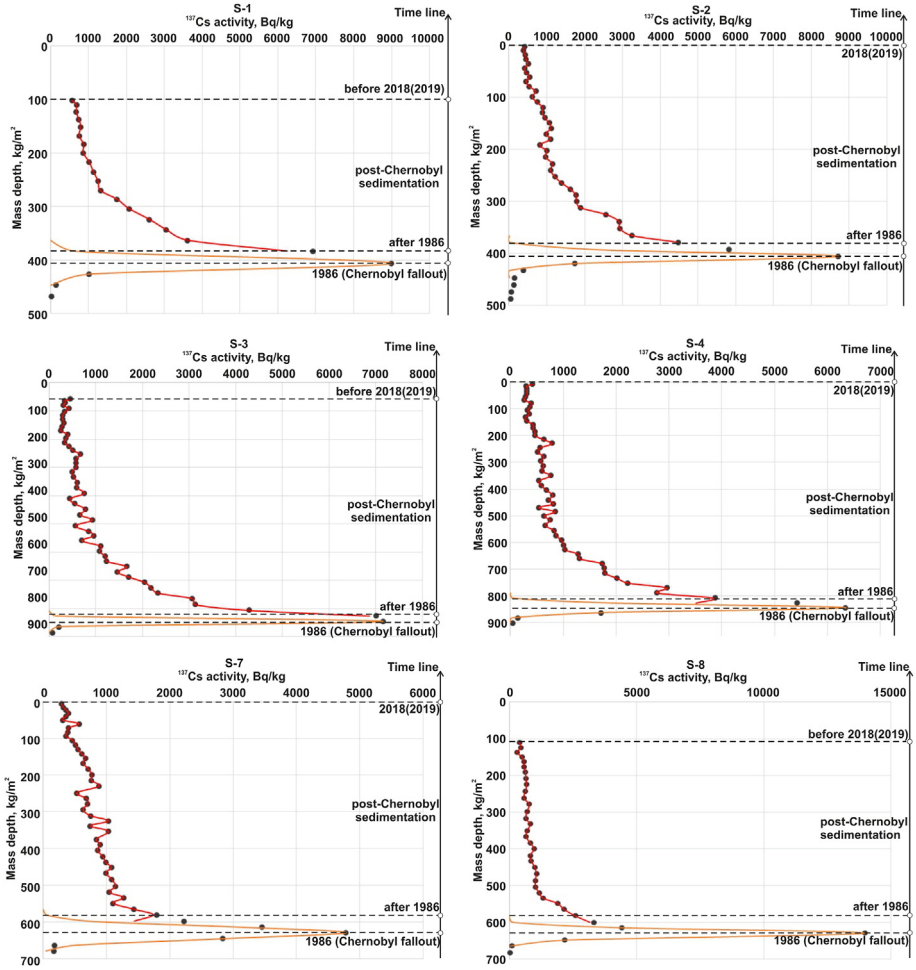


**Fig. 3.** Separation of the Chernobyl-derived $^{137}$Cs contained in the sediment cores recovered from the Schekino reservoir into two component representing the direct Chernobyl fallout to the reservoir surface (the "Chernobyl peak" based on Eq. 2) and the subsequent longer-term mobilization of Chernobyl fallout from the upstream catchment and its transport to the reservoir and accumulation in the sediment column.

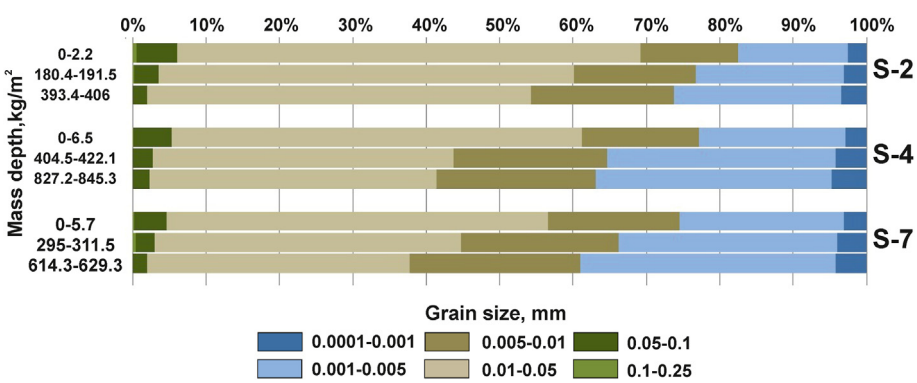


**Fig. 4.** The grain-size composition of sediment representative of different mass-depths in the sediment cores collected from the Schekino reservoir.

additional dating techniques such as examination of excess lead-210 ($^{210}Pb_{ex}$) activities (Huang et al., 2019).

Fig. 3 separates the Chernobyl-derived $^{137}$Cs present in the bottom sediments of the Schekino reservoir into two components, namely the short-lived direct fallout to the reservoir surface resulting from the accident (the orange line, based on Eq. 2) and the subsequent and longer-term post-fallout mobilization of $^{137}$Cs from the upstream catchment by erosion and its transport to the reservoir and accumulation in its bottom sediment (the red line). The latter shows the expected progressive reduction in $^{137}$Cs activity over time during the post-Chernobyl period and therefore towards the sediment surface for all cores including cores with larger losses (S-1; S-3; S-8), where the upper part of the core was deposited before 2018(2019).

Legend: S-1 = Core number (see Fig. SM1 for location); black points = measured values of $^{137}$Cs activity in the depth incremental

**Table 3**
The magnitude of the inventories of the sediment cores associated with the initial direct Chernobyl-derived $^{137}$Cs fallout to the reservoir surface and with the post-Chernobyl increase in inventory representing $^{137}$Cs mobilized from the catchment upstream of the reservoir during the post-Chernobyl period.

| Core | Initial Chernobyl-derived $^{137}$Cs deposition, kBq/m$^2$ | post-Chernobyl $^{137}$Cs deposition with sediment, kBq/m$^2$ | Total $^{137}$Cs deposition kBq/m$^2$ |
|---|---|---|---|
| S-1 | 104 | 235 | 340 |
| S-2 | 82 | 303 | 386 |
| S-3 | 61 | 496 | 557 |
| S-4 | 89 | 441 | 529 |
| S-7 | 93 | 233 | 326 |
| S-8 | 164 | 228 | 392 |

slices of the cores; orange line = "Chernobyl peak"; red line = depth distribution of Chernobyl-derived $^{137}$Cs mobilized from the upstream catchment in the sediment above the "Chernobyl peak"; notes on the right side of the figures link to the time scale.

### 3.4. Post-chernobyl increases in the $^{137}$Cs inventories of the bottom sediment of the schekino reservoir

The integral of Eq. (1) represents the $^{137}$Cs fallout deposited directly on the reservoir surface at the time of the Chernobyl accident. These values are indicated in Table 3 and are consistent with available information on the Chernobyl-derived fallout in the study area (Fig. 1C). The magnitude of the post-Chernobyl increase in the $^{137}$Cs inventory of the reservoir bottom sediment can be estimated by subtracting the inventory associated with direct fallout to the reservoir from the total inventory of the core (Table 3). This increase primarily reflects the post-fallout mobilization of Chernobyl-derived $^{137}$Cs from the upstream catchment by erosion and its delivery to the reservoir in the sediment load of the Upa River. Chernobyl-derived $^{137}$Cs fallout was characterized by high spatial variability, since it reflected the spatial distribution of rainfall during a short period immediately after the accident and the complex processes associated with mixing of the $^{137}$Cs release in the atmosphere (Silantiev and Silantiev 1997; Golosov and Markelov, 2000). Values of $^{137}$Cs inventory can therefore be significantly different, even for adjacent sampling locations separated by only a short distance. In the catchment of the Upa River above the Schekino Reservoir, Chernobyl-derived $^{137}$Cs inventories measured as part of this study varied from 61 to 164 kBq m$^{-2}$. This range is consistent with the range of Chernobyl $^{137}$Cs fallout indicated by available maps of radioactive contamination (de Cort et al., 1998) (see Fig. 1C). In contrast, the $^{137}$Cs inventories of the reservoir bottom sediment, which reflect deposition of sediment mobilized from the upstream catchment, can be expected to show much less variability, due to the mixing of contaminated sediment mobilized from different parts of the catchment during its transfer via the stream network to the reservoir. $^{137}$Cs inventories found in the reservoir bottom sediment can, however, be expected to reflect variations in sedimentation rates. The greatest $^{137}$Cs inventories associated with the bottom sediment of the Schekino Reservoir were found in the central part of the reservoir in cores S-3 and S-4, where the highest mean annual rates of sedimentation are found. These sedimentation rates were >25 kg m$^{-2}$yr$^{-1}$ and >26 kg m$^{-2}$yr$^{-1}$, respectively. In general, the magnitude of the post Chernobyl $^{137}$Cs inventories for the duplicate cores were very similar (see Table 3). The most significant difference was detected between cores S-3 and S-4. In this case, it is likely that the initial direct Chernobyl-derived $^{137}$Cs fallout was underestimated for core S-3, as discussed above.

### 3.5. Temporal changes in $^{137}$Cs activity concentration associated with the Upa River suspended sediment

Assuming that the contribution of the initial direct Chernobyl-derived $^{137}$Cs fallout to the reservoir surface to the $^{137}$Cs content of its bottom sediment can be distinguished, the up-column variation of the $^{137}$Cs activity concentration in the reservoir bottom sediment can be used to provide information on temporal changes in the $^{137}$Cs content of the suspended sediment transported by the Upa River and delivered to the reservoir since late 1986. Sediment in the cores located above the "Chernobyl peak" was deposited during the period following the input of $^{137}$Cs fallout in 1986 and before the time of sampling in 2018–2019. As discussed above, problems exist in attributing a particular slice of the sediment column to a specific date. However, the up-column changes in $^{137}$Cs activity can be seen as providing a valuable and detailed record of changes in the $^{137}$Cs content of suspended sediment transported by the Upa River over the ca. 32 years that have elapsed since the Chernobyl accident. This record provides important information regarding the redistribution and fate of Chernobyl fallout in the catchment of the reservoir. Because of potential interaction between the Chernobyl peak and the sediment immediately overlying the peak, only sediment located above the peak can be identified as sediment deposited after 1986. Also, sediment, located at the top of each core cannot necessarily be attributed to the exact time of core collection, due to the potential for mixing at the sediment-water interface and loss of some sediment at the time of sampling. The sediment column therefore provides a record of the sediment transported by the Upa River and its $^{137}$Cs content which extends from a date that is strictly "younger" than 1986 to a date that is possibly slightly "older" than 2018–2019 (see Fig. 3).

Three cores with the least losses in the upper part were selected for comparison, and the up-core changes in the $^{137}$Cs content of the sediment indicate that this decreased through time (Table 4). Application of the semi-empirical diffusion equation (Konoplev et al., 2019, 2020) to predict the evolution of the $^{137}$Cs content of surface soil in the catchment predicts a much higher $^{137}$Cs activity concentration for the surface soil – $C_{2018(2019)}/C_{1986} > 0.17$ which could be expected to be reflected by a smaller decrease in the $^{137}$Cs activity concentration of sediment transported by the river and deposited in the reservoir. The reduction through time of the $^{137}$Cs activity concentrations in the sediment transported by the Upa River indicated by the sediment cores is considerably greater than that predicted by the semi-empirical diffusion model.

Two factors are likely to account for the discrepancy between the reduction in the $^{137}$Cs activity concentration of the deposited sediment indicated by the reservoir core and that predicted by the semi-empirical diffusion model. The first is the mixing of the soil by regular ploughing of the arable land. This factor is seen to be the most critical, because soil erosion from arable land will represent the primary source of the contaminated sediment delivered to the reservoir (Litvin et al., 2003). The $^{137}$Cs activity concentration in the surface soil in the catchment of the reservoir will have reduced

**Table 4**
$^{137}$Cs activity concentrations in the bottom sediment of the Schekino reservoir attributed to different time windows.

| Core | $^{137}$Cs activity concentration in corresponding layer, Bq/kg | | $\frac{C<2018(2019)}{C>1986}$ |
|---|---|---|---|
| | $C_{>1986}$ | $C_{<2018(2019)}$ | |
| S-2 | 4450 | 405 | <0.09 |
| S-4 | 3830 | 422 | <0.11 |
| S-7 | 2220 | 285 | <0.13 |

$C_{>1986}$- layer above the Chernobyl peak; $C_{<2018(2019)}$- surface layer.

significantly as a result of the first ploughing after $^{137}$Cs fallout receipt and subsequent mixing by annual ploughing (Walling and He, 1999). Loss of soil from the surface of the plough layer by erosion will further increase the impact of annual ploughing in reducing the $^{137}$Cs content of eroded soil, since soil from below the plough layer containing little or no Chernobyl-derived $^{137}$Cs will be progressively incorporated into the plough layer, reducing its $^{137}$Cs activity concentration. In addition, deep ploughing was implemented within the most contaminated parts of the study area as a remediation measure after the Chernobyl accident, in order to bury the surface soil contaminated by Chernobyl fallout. It should be recognized that direct observations of $^{137}$Cs activity concentration in rivers reported for both Chernobyl-affected and Fukushima-affected zones represent a particular moment of time and duration after fallout during which radioactive decay will have taken place. The values reported here for the cores all relate to measurements made after core collection and therefore all incorporate the effects of radioactive decay to the present. This important difference must be taken into account when comparing the changes reported here for the cores with the much greater reductions in $^{137}$Cs activity concentrations activity reported for the sediment transported by rivers in both Chernobyl-affected (Vakulovsky et al., 1994) and Fukushima-affected areas (Taniguchi et al., 2019), which are based on direct observations involving measurements of $^{137}$Cs made at the time of sampling.

The second factor relates to potential changes in the contribution of different sediment sources to the sediment load of the Upa River during the period concerned. The sediment load of this river is comprised primarily of sediment mobilized and delivered from the cultivated slopes of the catchment and material eroded from the banks and beds of the river channels. Changes in the relative contributions of these two sources can be expected to result in changes in the $^{137}$Cs activity concentration of the sediment transported by the river. This will reduce if the relative contribution of sediment mobilized from the banks and bed of the river channels increases. Part of the arable land in the region was abandoned due to economic reasons after the collapse of the USSR in 1991. However, this change is unlikely to have greatly affected the catchment above the Schekino reservoir. The proportion of arable land area in the Schekino reservoir basin decreased from 68% of the total area in 1985 to 63% in 2000 and to 60% in 2015. The most important change in catchment behaviour was that surface runoff from the cultivated slopes during spring snowmelt has significantly decreased over the past three decades (Barabanov et al., 2018). This has also contributed to a reduction in soil erosion rates and sediment delivery from cultivated slopes to the rivers during spring floods (Golosov et al., 2018a,b).

A change in the relative contribution of sediment mobilized by erosion of river channel banks and beds should be reflected by a change in the grain-size composition of the sediment deposited in the Schekino reservoir. A clear trend of an increasing proportion of sand extending from the deeper to the surface layers is observed in the sediment cores collected from the reservoir (Fig. 4). This provides additional evidence that the contribution of the two main sediment sources changed during time window 1986–2018, with the contribution from the eroding river channels increasing during last decade. The alluvium in the valley bottoms, in which the river channels have developed is characterized by a higher proportion of coarse particles (>0.05 mm) than the surface soils of the arable fields. This contrast will be further emphasized by preferential mobilization of fines by the erosion occurring on the slopes and preferential deposition of coarser particles during the delivery of that eroded sediment to the river channels (Walling, 1983).

## 4. Conclusions

The activity concentration of $^{137}$Cs in the bottom sediments of the Schekino reservoir decreased through time during the 30-year period after the accident at the Chernobyl nuclear power plant accident. This partly reflects the effects of ploughing of the cultivated area in mixing the Chernobyl fallout into the plough layer and progressive reduction of the $^{137}$Cs content of the plough layer by soil loss. Changes in the grain-size of the reservoir bottom sediment, which became progressively coarser over the period extending from immediately after 1986 to the upper layers of the cores, provide a clear indicator of a reduction in the relative contribution of sediment from the cultivated areas in the catchment. Several factors involving land-use and climate change are likely to have caused this reduction. This reduction is consistent with the considerable decrease in surface runoff during snowmelt from arable land in the study area during the period since 1986. Scope exists to develop further and refine the approach employed in this study to exploit the potential to use reservoir bottom sediments to provide information on longer-term changes in the $^{137}$Cs activity concentration of the suspended sediment transported by rivers in the post-Chernobyl period. Such work could usefully focus on providing a detailed chronology for the sediment cores and the associated record of changes in $^{137}$Cs activity concentration. Because, sediment yields and therefore deposition rates are known to have changed during the post-Chernobyl period, there is not a simple relationship between age and sediment depth. Depth in the sediment profile cannot be used as a direct surrogate for time. However, the work of Huang et al. (2019) has, for example, demonstrated the potential to use excess lead-210 measurements to establish a detailed chronology for a sediment core collected from a reservoir. If a detailed chronology for the cores can be established, the $^{137}$Cs activity concentration data for a core could be adjusted for radioactive decay to provide a record equivalent to that obtained by contemporary sampling of the sediment transported by a river and reporting the $^{137}$Cs activity measured at that time.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.116588.

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