

GEOCHEMICAL INDICATION OF SEDIMENT FLUXES USING CHERNOBYL-DERIVED ^{137}Cs : THE CASE STUDY OF A SMALL AGRICULTURAL CATCHMENT IN THE TULA REGION, CENTRAL RUSSIA

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ABSTRACT. This paper explores the use of ^{137}Cs derived from Chernobyl as an indicator of sediment supply and transport within small agricultural catchments by analyzing the depth distribution of radionuclides, with a focus on post-Chernobyl changes in the activity concentration of radionuclides. To this end, depth-incremental sampling was carried out along routes of sediment transport within a small agricultural catchment subject to intense radioactive contamination in the Tula region. Some points were set to repeat the position of those made 27 years earlier and to understand the dynamics of deposition and the ^{137}Cs content in the sediment load. It has been suggested that a decrease in the activity concentration of ^{137}Cs can be used as an indicator of the relative age of deposits. Assuming this, the pattern of erosion product deposition on the sides and bottom of the dry valley was determined. This pattern was found to be stable and consistent with the observed geomorphic features and climate trends: the rate of accumulation in the valley bottom over the past 27 years has dropped almost twice, coinciding with a decrease in snowmelt runoff during springtime and no increase in intense rainfall. Grain-size analysis of the collected samples showed that selective transfer of clay particles may occur, but over a short delivery distance, it is unlikely that the sorting process will significantly alter the downward trend of ^{137}Cs concentrations. The proposed approach has the potential to significantly improve the accuracy of sediment budget estimations and environmental quality assessments.

KEYWORDS: erosion, sediment delivery, ^{137}Cs , Chernobyl contamination, sedimentation

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INTRODUCTION

Due to anthropogenic impact, which results in disturbances of the natural canopy, accelerated erosion on the interfluvial slopes plays a major role in the sediment budget (Vanwalleghe et al. 2017). Yet, the products of erosion are mostly re-deposited along pathways from cultivated slopes to the river channels (Sidorchuk 2018). Emerging sediment fluxes are discontinuous; they begin with single events of short erosion periods on the slopes, continue along the thalwegs of hollows and valleys toward permanent watercourses, and extend beyond the outlets of river catchments, where they partly mix with products of riverbed deformations and become trapped by floodplains and reservoirs. Exploration of accumulated deposits using high-resolution chronomarkers and tracers along the routes of sediment transport may help understand the

transformation of sediment budgets due to changes in the intensity of erosion and sediment delivery processes (Owens 2020).

Several artificial compounds that are brought into the environment are successfully used to investigate erosion and sedimentation, including heavy metals (Dai et al. 2013; Wang et al. 2019; Elbaz-Poulichet et al. 2020), fly ash (Olson et al. 2008; Davis & Fox 2009; Gennadiev et al. 2010), and radioactive isotopes (Zapata 2003; Alewell et al. 2017). The latter is closely linked to regular and occasional discharges from nuclear facilities (UNSCEAR report 2000) and nuclear weapons tests (Aoyama et al. 2006). Among other anthropogenic fallout radionuclides ^{137}Cs is most often used as a tracer (Zapata 2002).

The highest ^{137}Cs activity concentration usually occurs during the ^{137}Cs fallout from the atmosphere, unless the soil has been affected by perturbations and

erosion, intense migration of radionuclides (Jagercikova et al. 2015), and material from a more contaminated area has been transported and deposited at the sediment sinks. Considering the pointed limitations, precise dating of sediments becomes possible by using ^{137}Cs depth distribution (Foucher et al. 2020).

The distribution of radionuclides above the layer associated with massive fallout, such as that from the Chernobyl accident, can be seen as a record of changes in activity concentrations in sediments carried and deposited after the event. These variations are determined by activity concentration in the material from a specific sediment source, the proportion of sources contributing to the sediment flux, and the possible sorting of particles and aggregates during transportation. If the long-term trend in the behavior of ^{137}Cs in mobilized sediment for a selected location is predictable, then it is possible to link activity concentrations with the age of the accumulated sediment.

The aim of this research is to assess the potential use of the Chernobyl-derived ^{137}Cs depth distribution not only as an accurate chronomarker but also as a geochemical indicator of sediment fluxes in a small agricultural catchment affected by intense Chernobyl fallout in Central Russia. To this end, the following questions were raised:

1. How did the ^{137}Cs activity concentration change in sediments that reached the lower boundary of the cultivated fields and then entered a dry valley bottom over the post-Chernobyl period?
2. Is there a significant difference in the grain-size composition of material deposited in different geomorphic units, indicating the potential influence of sorting during transport on the radionuclide content?
3. What is the sedimentation rate in the dry valley along different sediment transfer routes, and can it be related to the activity concentration values of sediment believed to be older than 1986?
4. How long does it take for sediment to move from the arable slope to the catchment outlet, considering changes in erosion and sediment load from the slopes?

Materials and methods

The study area is located in the southern part of the Tula region near Plavsk town in the central part of a zone heavily contaminated with ^{137}Cs following the Chernobyl accident in 1986 (Fig. 1A). The chosen catchment has been studied over the past few decades using Chernobyl-derived ^{137}Cs as a soil erosion tracer (Golosoov et al. 1999a,b; Golosoov et al. 2000; Ivanova et al. 2000; Panin et al. 2001; Ivanov 2017; Ivanov, Ivanova 2023; Ivanov et al. 2023, 2024a). Despite this, the catchment still has high potential for in-depth exploration of erosion and sediment transport processes. Surveys from different years allow us to observe the long-term transformation of sediment and contaminant fluxes.

The area of the study site is 0.25 km², and the elevation difference is 52 m: from 187 m asl near its mouth (see Fig. 1B) to almost 236 m on the watershed surface. A major part of the drainage area is occupied with arable interfluvial slopes of 1°–7°. The rest of the catchment area is represented by

steep (up to 25°) sides and a gently sloping bottom of a dry valley (Fig. 1B).

The bedrock represented by Carboniferous limestone is covered by a loess-like loam (Ratnikov 1960) that serves as a soil-forming deposit for leached (Luvic Chernic Phaeozems) and podzolized (Luvic Greyzemic Chernic Phaeozems) according to the WRB-22 classification. According to the Plavsk weather station, the average annual precipitation is approximately 650 millimeters. Since the early 1990s, there has been a clear trend towards an increase in average winter air temperatures and a decrease in snowmelt runoff, up to its complete disappearance in some years (Barabanov et al. 2018), due to the lower depth of soil freezing and the higher infiltration capacity of the soil during the snowmelt season.

The lower boundary of arable slopes is usually outlined with lynchets: a ramparts emerged due to ploughing. Therefore, the transfer of mobilized sediments outside the slope occurs mainly through slope hollows, where slope runoff is concentrated (Panin et al. 2001). Before plowing ramparts (lynchets), erosion products accumulate at the foot of arable slopes. Accumulation in this zone plays a significant role in the sediment budget, comparable to the sediment load entering receiving watercourses (Ivanov et al. 2024b). The material that is carried outside the slope is deposited in the form of slopewash fans and covers on the sides of the valley. The rest of the sediments are transferred along the fluvial network and mainly deposited inside valleys.

As far as water flow is predominantly controlled by local topographic features, the specific routes of sediment transfer can be identified and studied separately. In our study, three routes were investigated with depth incremental sampling points (Fig. 1D, Table 1).

The first route is located in the western part of the catchment. The slope runoff from the neighboring slopes is concentrated along the lynchets, so its transfer to the valley bottom is observed in the corner of the cultivated field. Two sampling points were selected here: one at the foot of the slope before the fulfilled lynchets (LF-1) and one on the side of the dry valley (LS-1), where conveyance of mobilized material was expected. The second route passes through the central part of the study area and includes three sampling points: on the slope of the fulfilled lynchets (LF-2), on the side of the dry valley (LS-2), and in the upper reaches of the valley's bottom (LB-1). The third route starts at the lower reach of the bottom of a large slope hollow (LF-3), passes through a well-defined slopewash fan on the side of the valley (LS-3), and continues along the bottom dry valley bottom, where two sampling points were selected: in the upper reaches (LB-2) and near the mouth (LB-3). In the central part of the valley bottom, there is a local area with bottom gully incision. The position of the gully head has not changed significantly since it was first observed in 1997. The locations of LB-2 and LB-3 were selected to be close to the soil sections examined in 1997 by Golosoov et al. (1999a) for comparison purposes within the 1997–2024 time window (see Fig. 1C).

The depth incremental sampling was conducted in two ways. Soil cores were collected using a hand auger at points LF-1, LF-2, LS-2, LF-3, and LS-3. At points LS-1, LB-1,

Table 1. The routes of sediment delivery reaching the foot of arable slope

Route	Sampling points	Soil losses 1986–2022, t*
1	LF-1–LS-1	483.4
2	LF-2–LS-2–LB-1	358.3
3	LF-3–LS-3–LB-2–LB-3	7379.2

*after Ivanov et al. 2024b

LB-2, and LB-3, we dug pits to describe the soil profile and collect samples from the walls of the soil sections. Sampling was performed at 3–5 centimeter intervals: either from the wall of the soil section or by cutting a core directly with the hand auger. All samples were delivered to the laboratory and dried out. They were then weighed and ground before being placed in petri dishes for further examination of ^{137}Cs activity, using a gamma spectrometer with a high-purity germanium (HPG) detector manufactured by ORTEC (USA) with an error not exceeding 10%. All activity values were recalculated for 1986, taking into account radioactive decay. For the samples of Route 3 (LF-3, LS-3, LB-2 and LB-3) the grain-size composition was determined using a Malvern Mastersizer 3000 particle size analyzer to figure out any sorting during transport.

Results

After examining samples from set points along the first route, it was learned that some of the material was accumulated at the foot of the slope during the post-Chernobyl period, while the rest was deposited downstream in the valleys. The depth distribution of ^{137}Cs from LS-1 demonstrates heavily contaminated strata, whose thickness is several times greater than the depth of ploughing. Due to the intensive deposition, the sampling depth at this site was insufficient to collect all the soil material containing Chernobyl ^{137}Cs (Fig. 2A). On the valley's side, accumulation also occurred. The layer with highest activity concentration corresponding to the fallout was identified at depth of 25–30 cm. The upper 25 cm strata is argued to be deposited later (Fig. 2B).

Obviously, the concentration of activity in the accumulated material shows a downward trend. As can be seen from LF-1 (Fig. 2A), despite repeated cultivation resulting in mixing of the upper layer of soil, values

dropped from 2124 ± 34 to 1386 ± 27 Bq kg $^{-1}$. Given no disturbance after the accumulation in point LS-1, the activity concentration dropped by almost two times, from 2564 ± 101 to 1324 ± 30 Bq kg $^{-1}$. Assuming that the sediments redeposited at the studied points have the same origin, the activity concentration can be used as a parameter to correlate the ^{137}Cs depth distributions. Thereby the almost equal range of concentration indicates that accumulation in LF-1 and LS-2 took place simultaneously. Even if the lynchet had been morphologically pronounced sometime after the Chernobyl fallout, the concentration of runoff was enough to deliver sediment beyond the cultivated field.

For the second route, the situation is quite different. At the point LF-2, activity concentrations of ^{137}Cs exceeding 1500 Bq kg $^{-1}$ are only seen in the upper 30 centimeters and are distributed almost evenly. Downwards in the soil profile, ^{137}Cs content starts to drop (Fig. 3A). Therefore, it can be concluded that there has been no significant accumulation before the cultivated field boundary. On the adjacent side of the valley (point LS-2), the accumulation during the post-Chernobyl period has been no more than 9 cm (Fig. 3B). In addition, the concentration of ^{137}Cs in the upper 6 cm of sediment, which can be linked to post-Chernobyl accumulation, turned out to be higher than in the material deposited at the foot of the slope: 2392 ± 132 – 2752 ± 168 Bq kg $^{-1}$ versus 1791 ± 167 – 1940 ± 173 Bq kg $^{-1}$ (Fig. 3B). It may indicate that sediment deposition on the valley side occurred when the activity concentration of ^{137}Cs in the sediment runoff was higher. Currently, no accumulation is detected. Down by the route in the valley's bottom examination of sediments at the point LB-1 showed that a maximum of ^{137}Cs activity lies almost on the surface (Fig. 3C). The only upper 3 cm layer which can be argued to have accumulated after 1986 has a very high concentration of 3352 ± 72 Bq kg $^{-1}$. This is much higher than the concentrations in sediments that were accumulated nearby at the LS-2 point. Considering that

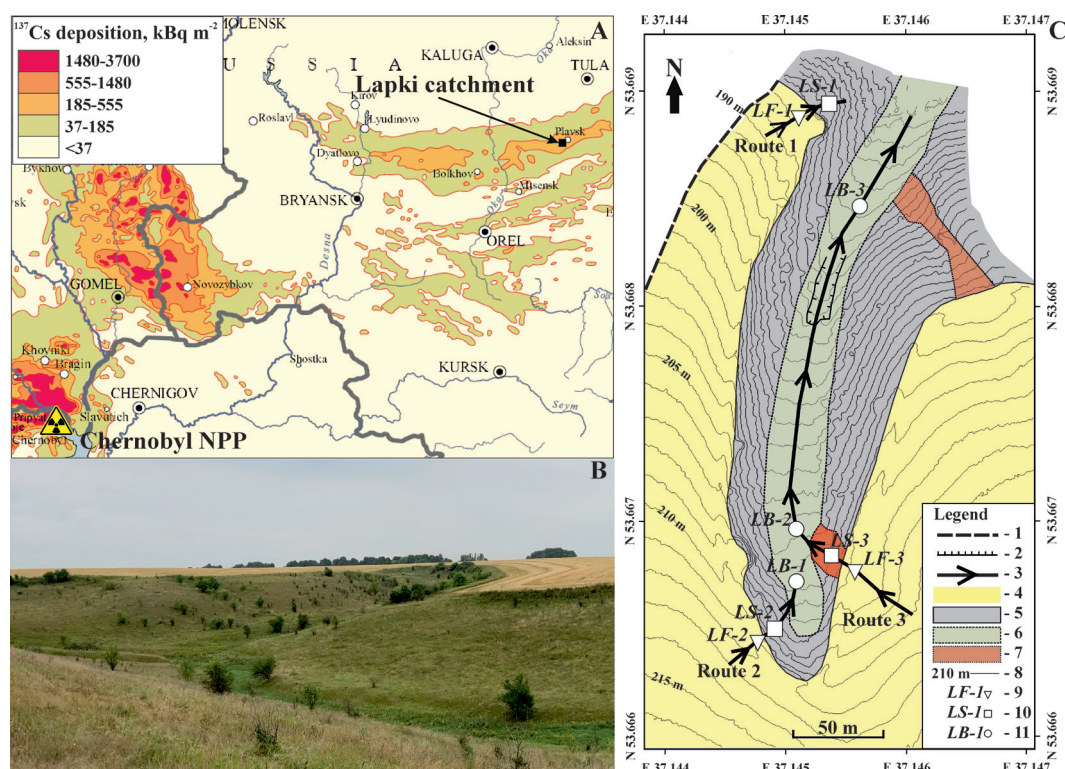


Fig. 1. The map of ^{137}Cs Chernobyl fallout (after Izrael et al., 1996) and location of the study area (A). The photo of the mouth of the Lapki catchment was made in 2021 (B). Observed routes of sediment transport (D): 1 – catchment boundary; 2 – steep eroded slopes; 3 – observed routes of sediment transport; 4 – arable slopes; 5 – dry valley's sides; 6 – dry valley's bottom; 7 – slope wash fans and covers; 8 – counter lines, a.s.l.; sampling points with depth incremental sampling on different geomorphic units: 9 – foot of arable slopes, 10 – dry valley sides and slope wash fans, 11 – dry valley bottom

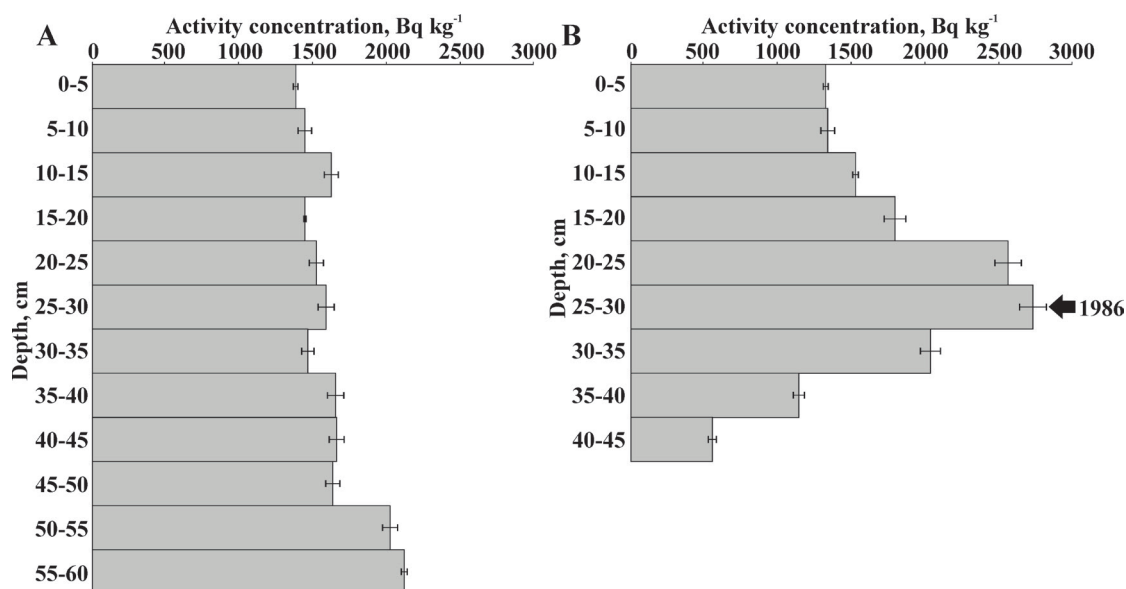


Fig. 2. The depth distribution of ^{137}Cs at points LF-1 (A) and LS-1 (B)

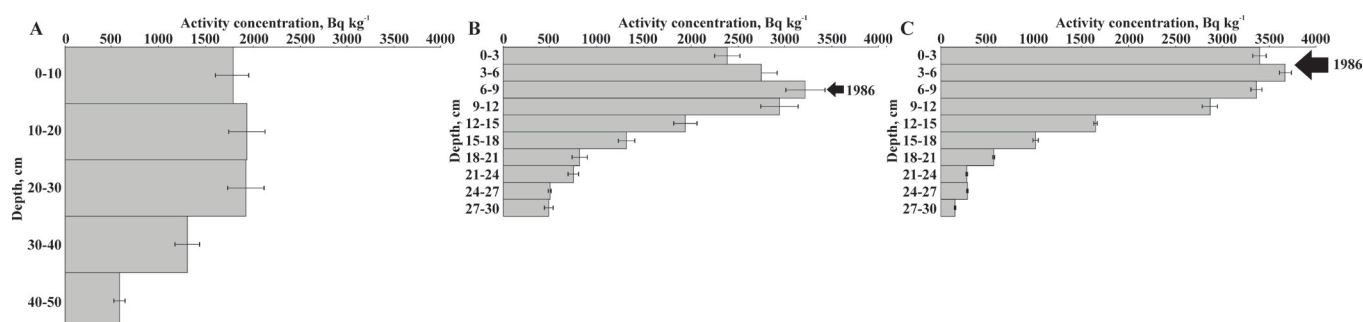


Fig. 3. The depth distribution of ^{137}Cs at points LF-2 (A), LS-2 (B) and LB-1 (C)

samples near the surface are subject to vertical migration of radionuclides, including that along plant roots, it is more likely that there was no accumulation in this location.

At the foot of the slope, at point LF-3, the distribution is similar to that seen in the previously described example at LF-1 (Fig. 2A), indicating intensive accumulation due to high values of slope runoff delivered through the slope hollow (see Table 1). The activity concentration varies between 1263 ± 118 and $1484 \pm 114 \text{ Bq kg}^{-1}$, and there is no obvious decreasing trend observed (Fig. 4A).

High accumulation is also seen on the surface of the slopewash fan at point LS-3, with more than 39 centimeters accumulated between 1986 and 2022 (Fig. 4B). The upper 24 cm of the sediment are characterized by a gradual increase in ^{137}Cs concentration moving down, with values that lie close to those observed in LF-3, ranging from 1188 ± 77 to $1437 \pm 98 \text{ Bq kg}^{-1}$. In the deeper part (24–39 cm), this growth becomes more intense: from 1580 ± 112 to $2651 \pm 172 \text{ Bq kg}^{-1}$ and indicates older material than observed at the point LF-3.

In upper reach of the valley bottom (LB-2), the thickness of post-Chernobyl accumulation drops to 27 cm. Here, there is a clear increase in ^{137}Cs concentration, starting at the surface. It is likely that most of the accumulation occurred during a short period after the fallout, when the concentration did not decrease to levels observed in LS-3 and the upper part of LF-3: from 3469 ± 173 to $1380 \pm 76 \text{ Bq kg}^{-1}$ (Fig. 4C).

Along the valley bottom, the rate of accumulation continues to decline, and near the mouth of the valley (LB-3), the accumulation is less than 18 cm over the period 1986–2024. The concentration of activity in the accumulated sediments ranges from 1664 ± 98 to $3416 \pm 159 \text{ Bq kg}^{-1}$ (Fig. 4D). This range is almost like that observed at the upper reach (LB-2), indicating the same age of the deposited material. It turns out that modern products of soil erosion are hardly represented here.

Grain-size analysis of the collected samples suggests that there has been a selective transfer of clay particles. The percentage of particles smaller than 2 microns gradually decreases from 12.48% at the foot of the arable slope to 9.66% near the dry valley's mouth. At the same time, the proportion of particles thicker than coarse silty (16 microns) shows an increase as they move downstream (Fig. 5).

The grain-size composition shows no clear trend in the vertical distribution. At the observed points, each fraction has random fluctuations which show (Fig. 6).

Comparison of ^{137}Cs depth distributions obtained in 1997 (Golosov et al., 1999a) and in 2024 showed that the depth of the peak of activity concentration has changed: in LB-2 from 12–15 cm in 1997 (Fig. 7A) to 27–30 cm in 2024 (Fig. 7B) and in LB-3 from 7–9 cm (Fig. 7C) to 15–18 cm in 2024 (Fig. 7D). Accordingly, in both locations, the rate of accumulation over 27 years decreased almost twice: from $1.1\text{--}1.4 \text{ cm year}^{-1}$ to $0.7\text{--}0.8 \text{ cm year}^{-1}$ in LB-2, and from $0.6\text{--}0.8 \text{ cm year}^{-1}$ to $0.4\text{--}0.5 \text{ cm year}^{-1}$ in LB-3. The deposition was still much higher in the valley's upper part (LB-2 compared to LB-3), but the ratio of the accumulation rates of LB-2 to LB-3 was stable, at 1.4–2.3 in 1997 and 1.4–2.0 in 2024. Also, the mean activity concentration in the upper samples, 1798 Bq kg^{-1} in LB-2 versus 2358 Bq kg^{-1} in LB-3, indicates a different age of sediment (Fig. 7 A, C).

DISCUSSION

Summarizing the results presented, the following points can be made. The transport of sediment and radioactive isotopes from agricultural slopes is primarily determined by the concentration of slope runoff. This, in turn, is influenced by both the topography of the slope and the microrelief at its foot. It is clearly indicated by the depth

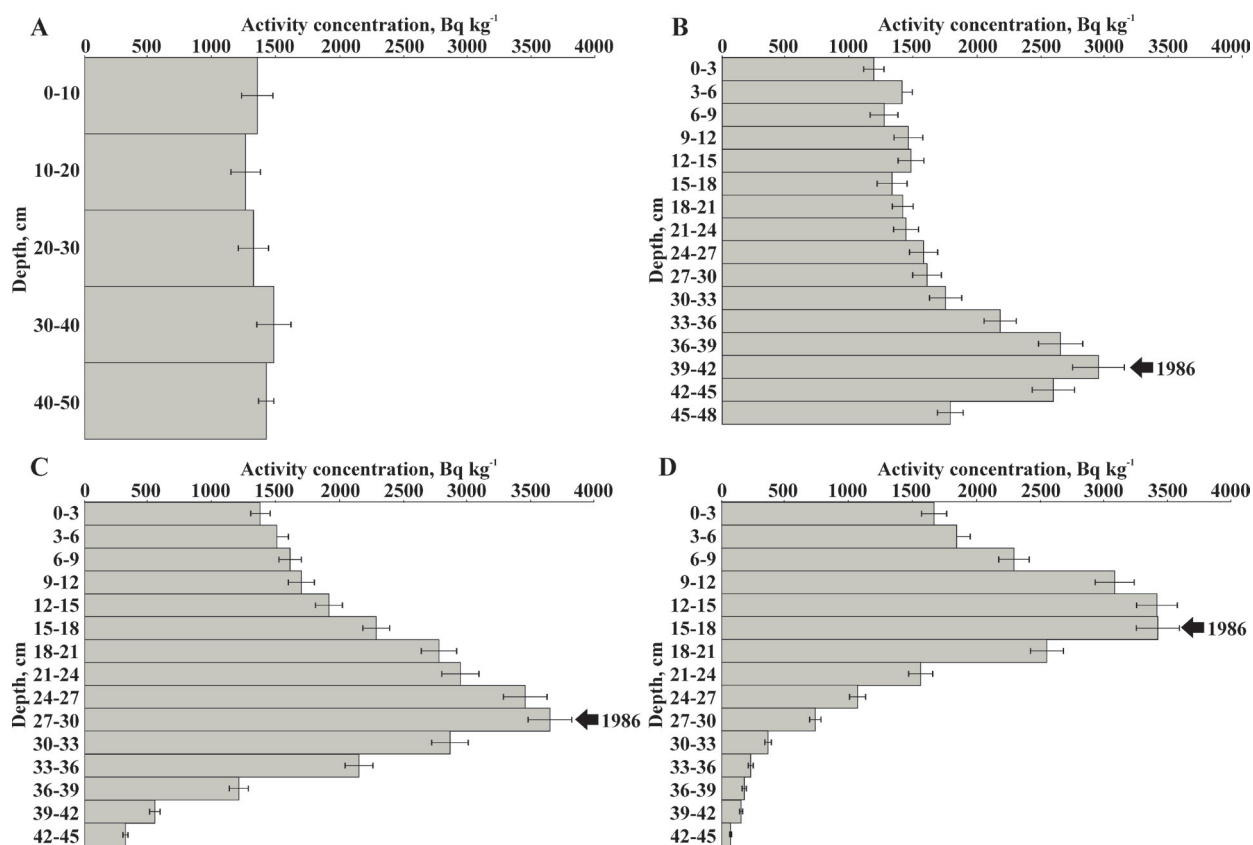


Fig. 4. The depth distribution of ^{137}Cs at points LF-3 (A), LS-3 (B), LB-2 (C) and LB-3 (D)

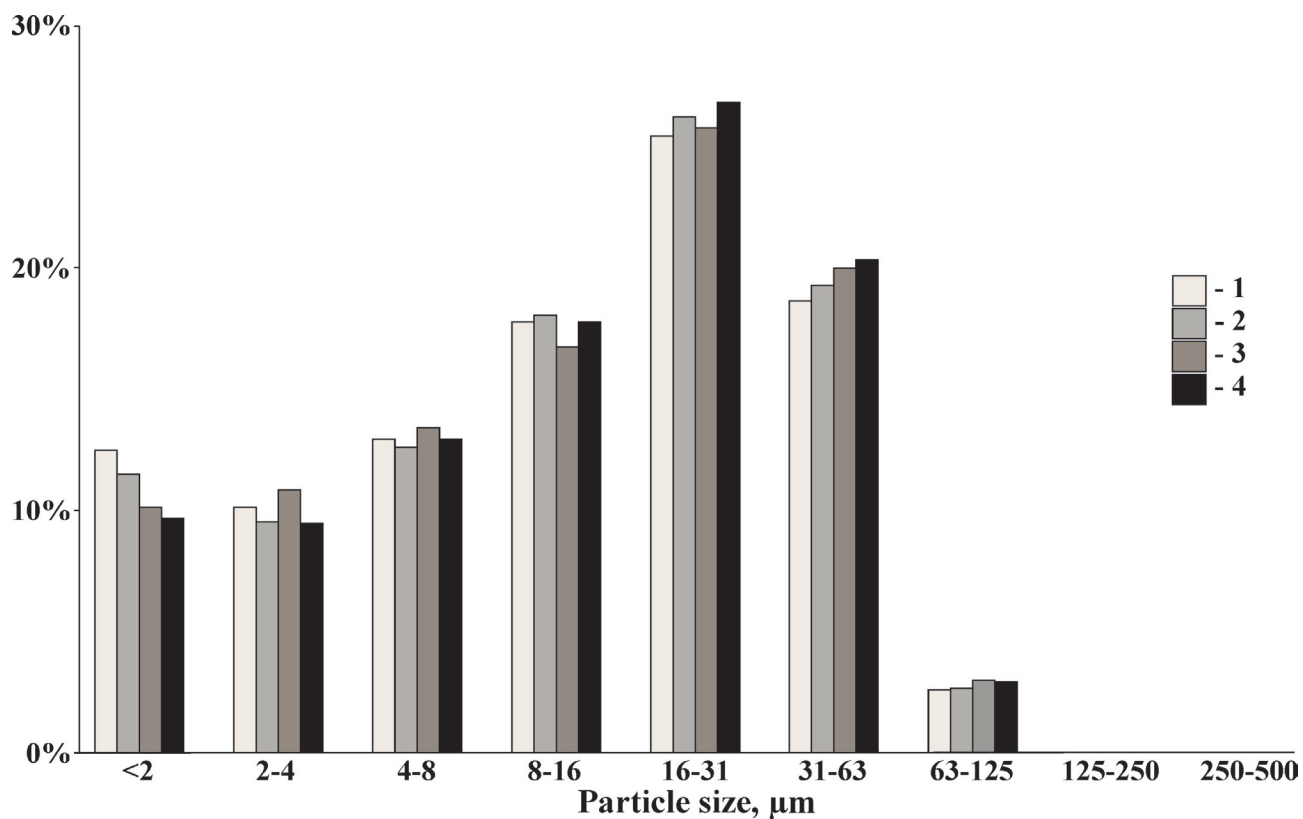


Fig. 5. Mean grain-size composition of sediments from Route 3: 1 – LF-3; 2 – LS-3; 3 – LB-2; 4 – LB-3

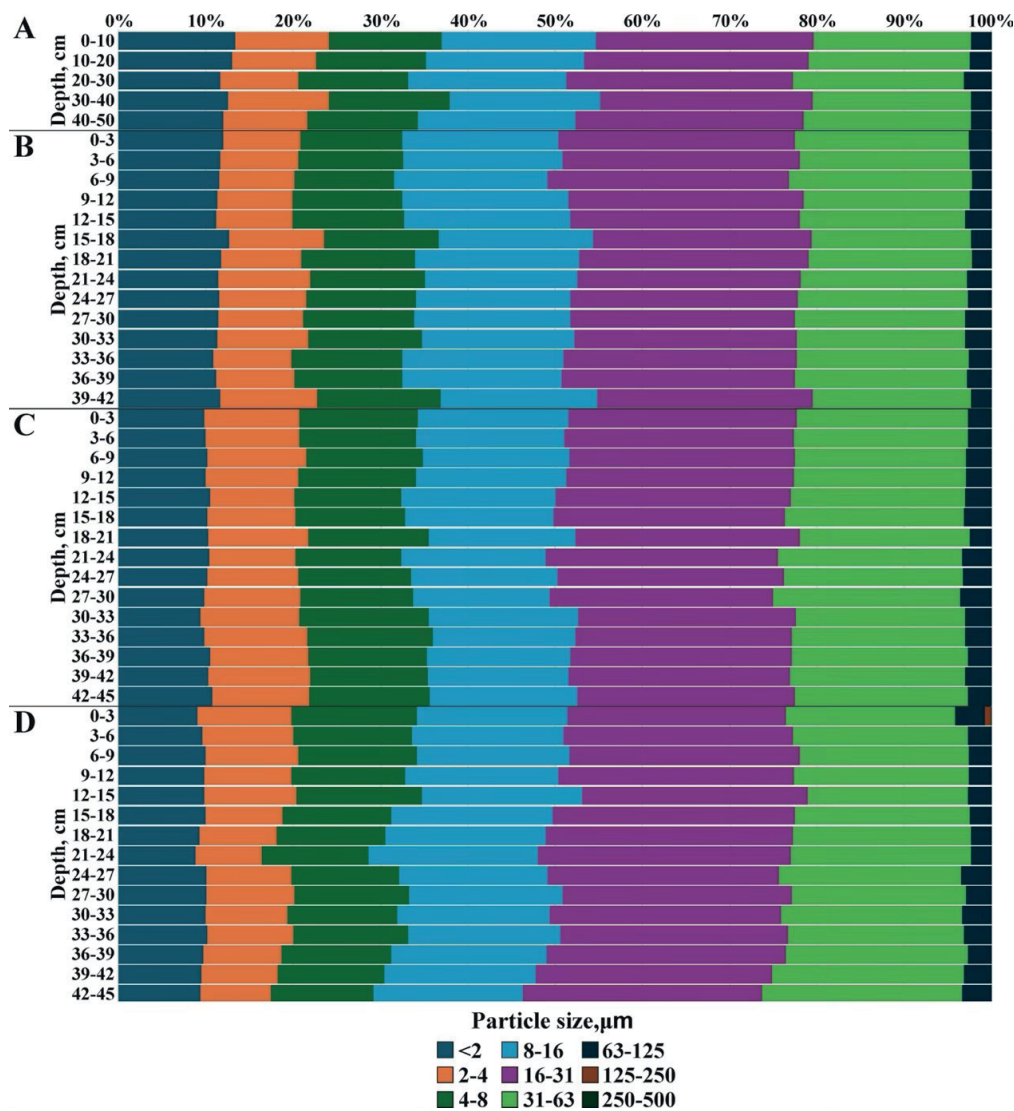


Fig. 6. Depth distribution of grain-size composition of sediments from Route 3: A – LF-3; B – LS-3; C – LB-2; D – LB-3

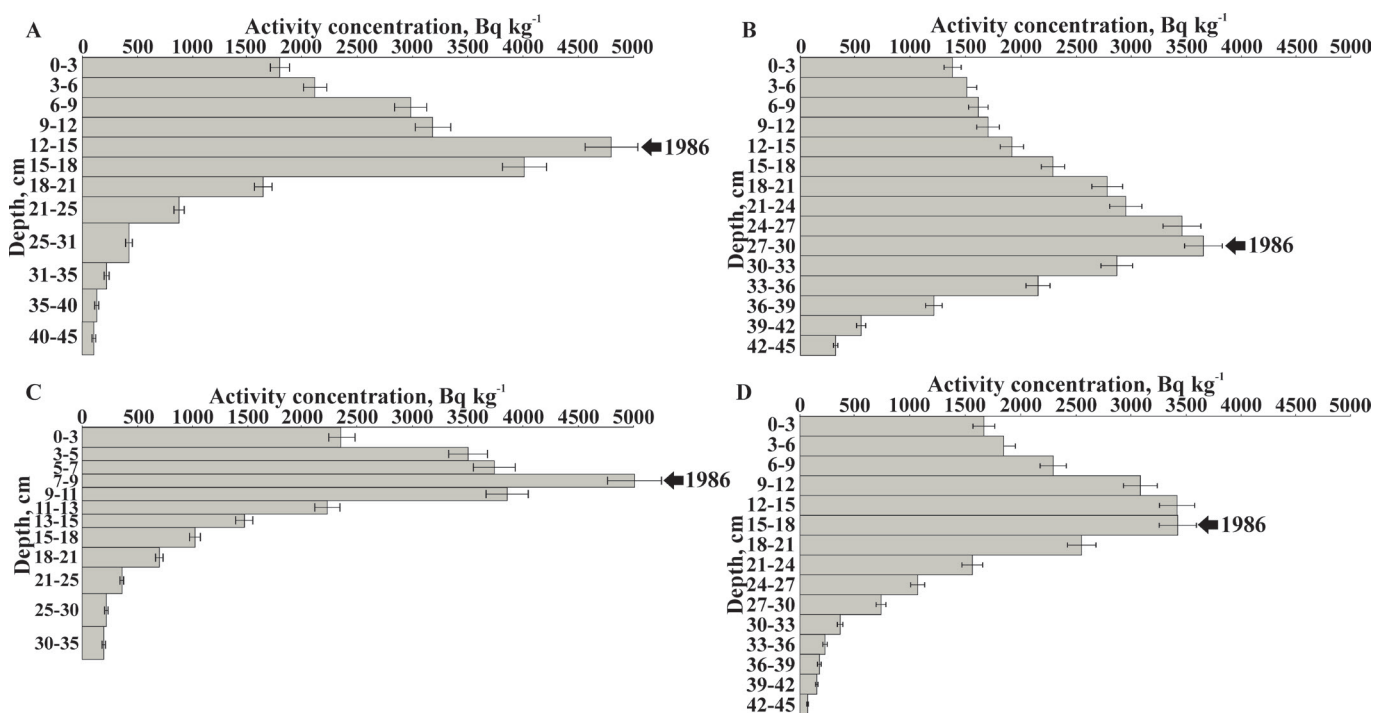


Fig. 7. The depth distribution of ^{137}Cs at points LB-2: in 1997 (A) (after Golosov et al., 1999a), in 2024 (B), and LB-3: in 1997 (C) (after Golosov et al., 1999a), in 2024 (D)

distribution of ^{137}Cs in sediments on both sides of the lower boundary of the cultivated field. The decrease in ^{137}Cs activity concentrations in sediments mobilized on arable slopes and redeposited downstream is typical for all cases observed. This decrease was not linear, with a rapid decline shortly after fallout, becoming smoother over decades until relatively stable values recently. The selective transport of clay particles could affect activity concentration during transportation. The depth distribution of ^{137}Cs suggests an increase in accumulation in the buffer zone on the slopes of the dry valley and in the upper reach of the dry valley. This pattern is consistent with observed climate trends: decreasing snowmelt runoff and no increase in intense rainfall.

The obtained picture is consistent with the current understanding of the lateral migration of particulate ^{137}Cs in areas with intense fallout. The Chernobyl incident was followed by a sharp increase in the contamination of the subsurface soil layer. Within the arable slopes, activity concentration dropped shortly after plowing, which depth was recommended to increase for remediation purposes (Alexakhin et al. 1992). A similar situation was observed in the affected areas of Fukushima, where the activity concentration of ^{137}Cs in terrestrial environments decreased rapidly relative to expectations due to active land use and decontamination efforts (Onda et al., 2020). Afterwards, it was expected that concentration would decrease due to a number of factors.

There would be a loss of upper, highly contaminated soil layers due to erosion and harvesting, which would result in the involvement of deeper and cleaner material during plowing. Freeze-thaw processes can lead to unstable soil surfaces and the development of intense rill erosion in springtime, which in turn causes a decrease in activity concentrations in mobilized sediment (Wakiyama et al. 2019; Igarashi et al. 2021). However, given the increasing average temperature and the reduction of snowmelt runoff in the beginning of the XXI century (Baranov et al. 2018), this factor does not seem to play a significant role. Also, activity concentration values would decline as a result of the complex migration of radionuclides primarily down through the soil profile. However, the latter effect was expected to be negligible (Golosov et al. 2013).

The sorting of material occurs along the entire transportation pathway and can potentially affect the concentration of ^{137}Cs in sediments. Shamshurina et al. (2011) found that activity concentration correlates with the share of soil aggregates. In the soils of the upper and middle slopes, approximately 50% of the total ^{137}Cs inventory is associated with aggregates larger than 2 mm. In the lower part of slopes, this share rises to about 70%. As the material moves and aggregates break down, sorting occurs primarily based on the size of individual particles. The selective deposition of larger particles leads to the enrichment of the sediment load with clay and fine silt (Golosov et al. 2000). In turn, the selective transport of clay and silt particles may lead to the intensive migration of bound radionuclides (Evrard et al. 2015; Konoplev et al. 2016). However, over a short delivery distance, it is unlikely that the sorting process will significantly alter the downward trend of ^{137}Cs concentrations. Given a single sediment source, the activity concentration of ^{137}Cs can be used as an indicator for the

relative age of the deposited sediment.

As the number of sediment sources increases, the picture of contamination is likely to become more complicated, but changes in activity concentrations may be used for fingerprinting tasks (Schuller et al. 2013; Evrard et al. 2020). If the radiocesium content from different sources is varying, it is possible to understand their contributions by comparing the ^{137}Cs depth distribution in deposits before and after the confluence of sediment fluxes.

Panin et al. (2001) reported that the long profile of the valley gradually decreases from its upper reaches to its mouth but has some slight convexities along the way, indicating separate episodes during the period of cultivation. As it has been declared, for the valley that receives sediment load from the explored catchment, the main way that deposits can be mobilized is through the incision of bottom gullies. Otherwise, the valley bottom provides long-term storage for eroded sediment and radionuclides. This statement may be supported by the fact that the activity concentrations in the upper samples in soil sections along the slope and at the bottom of the selected valley have different values, and consequently, sediments are of different ages. Sediments downstream are found to be older than those upstream, as can be seen in the example of LB-2 and LB-3 (Fig. 4C, D). This pattern has been consistent over decades (Fig. 5). Thus, distribution of the ^{137}Cs activity concentration may act as a geochemical indicator of geomorphic disconnectivity.

Using the distribution of activity concentration as a proxy for the age of sediment mobilization may help us to better understand sediment accumulation by correlating it with specific time periods. Sediment budget studies on small catchments are a useful way to validate estimations of soil erosion and sediment delivery from cultivated slopes to dry valleys and further along fluvial networks (Walling et al. 2002; Reid and Dunne 2016; Zhidkin et al. 2023) obtained results may be used to calibrate existing models. As accelerated erosion is a major source of sediment-associated contaminants, including radioactive ones (Lal 1994; Quinton and Catt 2007; Konoplev et al. 2021; Rashmi et al. 2022), the rate at which eroded material is delivered to watercourses is critical for assessing current environmental quality and forecasting future scenarios.

CONCLUSION

Since intrabasin sediment deposition constitutes a significant part of the sediment budget in river catchments with intensive anthropogenic influence, any additional time markers to explore sedimentation would be instrumental and should be included in the toolbox. The study conducted has shown that the pattern of Chernobyl-derived ^{137}Cs contamination has a close relationship to sediment redistribution in almost all decades after the fallout. The decrease of activity concentration during the post-Chernobyl period demonstrates high potential as a surrogate of relative age. This finding is consistent with previous research and sheds light on the potential use of ^{137}Cs depth distribution as a proxy of the sediment age during post-Chernobyl accumulation. However, the proposed approach requires a clear understanding of the long-term variation in the radionuclide content in material eroded from slopes and transported into the fluvial system. ■

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