# Vertical Distribution of Cs-137 in the Catena Soils of Agricultural Landscapes in the Bryansk Region

V. G. Linnik<sup>a, \*</sup>, A. P. Borisov<sup>a</sup>, O. M. Ivanitsky<sup>a</sup>, I. V. Mironenko<sup>b</sup>, and A. V. Sokolov<sup>a, c</sup>

<sup>a</sup>Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, Moscow, 119991 Russia <sup>b</sup>Faculty of Geography, Lomonosov Moscow State University, Moscow, 119991 Russia

<sup>c</sup>Kharkevitch Institute For Information Transmission Problems, Russian Academy of Sciences, Moscow, 127051 Russia

\*e-mail: linnik@geokhi.ru

Received February 1, 2020; revised April 9, 2020; accepted April 10, 2020

Abstract—The paper presents data on the vertical distribution of <sup>137</sup>Cs of Chernobyl origin in arable and meadow soils to a depth of 40 cm in typical catenary junctions of predpolessie, predopolje, and opolje land-scapes with a different density of <sup>137</sup>Cs contamination 30 years after the accident at the Chernobyl nuclear power plant. Newly obtained data on the removal of <sup>137</sup>Cs beyond the arable horizon presented in the paper indicate that this parameter varies depending on the landscape conditions: it varies in the range of 2.7–17.7% in predpolessie landscapes and in the range of 0.5 to 9% in opolje landscapes. In opolje landscapes, the coefficient of <sup>137</sup>Cs removal from the arable horizon was proved to systematically decrease depending on the landscape catenary position: it amounts to 8.5% in the transit zone and is 1.4% in the accumulation zone. The vertical distribution of <sup>137</sup>Cs and <sup>210</sup>Pb<sub>ex</sub> (excess) in the sod and floodplain soils in the basin of the Kostitsa River was studied to determine the sedimentation rate. Based on the analysis of the distribution of <sup>137</sup>Cs in the soil, the sedimentation rate over the past 30 years in various parts of the floodplain was estimated at 0.42 ± 0.08 to 0.58 ± 0.11 cm/year. The average estimated sedimentation rate in the floodplain during approximately 50 past years was, according to the <sup>210</sup>Pb<sub>ex</sub> dating was equal to 0.7 ± 0.18 cm/year. The sedimentation rate in the bottom of a ravine was evaluated for sod soils based on the distribution of <sup>210</sup>Pb<sub>ex</sub> according to the CRS model for 1961–2016 y. It was revealed that the minimum sedimentation rate of 0.24 cm/year occurred in 1961–1988, and the maximal (0.7 cm/year) in 2000 to 2007.

*Keywords:* soils, catena, vertical migration, floodplain, geochemical tracers (<sup>137</sup>Cs, <sup>210</sup>Pb), Chernobyl **DOI:** 10.1134/S0016702920100092

#### INTRODUCTION

After the 1986 accident at the Chernobyl nuclear power plant, predicting the vertical migration of anthropogenic radionuclides with the use of computer simulations was a top-priority avenue of radioecological studies (Velasco et al., 1993; Mamikhin, 1995; Mamikhin et al., 2016; Ivanov et al., 1997; Bunzl et al., 2000; Almgren and Isaksson, 2006). The spatiotemporal estimation of depths reached by radionuclides in the soils is of crucial importance for predicting how radionuclides are taken up from the soil to plant (Lukšienė et al., 2013; Beresford et al., 2016); how radiation dozes from external sources vary with time (Arapis et al., 1999; Ramzaev and Barkovsky, 2018); how the groundwaters can be potentially contaminated (Bugai et al., 1996), and how the soils are eroded (Owens and Walling, 1996). This is also important for understanding how floodplains of the rivers are contaminated from facilities of nuclear power plants (Standring et al., 2009).

The real distribution of artificial radionuclides in soil profiles is obtained using lay by lay sampling of the soil in field or by conducting laboratory experiments, when radionuclide tracers are introduced into monoliths (Bunzl et al., 2000). The data obtained are used to verify mathematical models of vertical migration rates of artificial radionuclides in soils.

The character of the vertical distribution of radionuclide in soil varies with time. Initially, all radionuclides were deposited on the soil surface as "thin films" shortly after the Chernobyl accident but started to penetrate deeper into the soils already a few months after the rains. The rapid vertical migration of <sup>137</sup>Cs shortly after an accident was also detected in the vicinities of the Fukushima nuclear power plant. After raining two months after the Fukushima accident, the storage of <sup>134</sup>Cs + <sup>137</sup>Cs in the 0–3 cm layer of the virgin soil was 88%, whereas that in the layer 0–5 cm was 96% (Shiozawa, 2013).

In 1987–1987, the soils contaminated after the Chernobyl accident were characterized by particularly

rapid migration of radionuclides with infiltration flows in the form of solutes or colloids (Bulgakov et al., 1990; Konoplev, 2016). The drastic decrease in the <sup>137</sup>Cs activity with depth is well fitted with an exponential function as a solution of the diffusion equation, if the convection is neglected (Bulgakov et al., 1990; Isaksson and Erlandsson, 1995).

However, over the following years <sup>137</sup>Cs adsorption on clay minerals in soils (Cremers et al., 1988) led to that <sup>137</sup>Cs amounts migrating with soil moisture significantly decreased. As a consequence, the <sup>137</sup>Cs distribution in the upper and lower parts of the most strongly contaminated upper 20-cm layer became less contrasting, and the vertical profile of the <sup>137</sup>Cs concentration became notably different from an exponential distribution.

In addition to transfer with solutions, the vertical migration of <sup>137</sup>Cs is also facilitated by lessivage (Shcheglov, 2000) and the activities of the soil biota. The concerted action of biogenic and abiogenic processes enhances the migration of radiogenic isotopes and, hence, their dissipation throughout the soil profile.

The formation of radionuclide contamination can be viewed as a stochastic process: the spatial distribution of <sup>137</sup>Cs is characterized by heterogeneity of the contamination levels, hence, the contamination field is analyzed with the application of stochastic analysis methods (Arutyunyan et al., 1994).

Along with soil–geochemical conditions that control <sup>137</sup>Cs adsorption in soil, the heterogeneity of <sup>137</sup>Cs penetration to various depth levels is influenced by landscape factors, such as topographic position, the angles of the slopes and their exposures, the thickness of the humus-accumulated horizon, soil hydromorphism, etc.

The view of the vertical <sup>137</sup>Cs distribution obtained by analyzing layer by layer soil sampling is controlled by the local landscape-geochemical conditions affecting radiocesium migration.

Studies of the migration of radionuclides in the soils in the 30-km zone around the Chernobyl nuclear power plant during a few years after the accident have revealed more rapid <sup>137</sup>Cs migration in hydromorphic soils than in automorphic ones (Ivanov et al., 1997; Shcheglov, 2000).

The dependence of <sup>137</sup>Cs penetration into different depth levels related to microlandscape conditions was studied at a monitoring area of the Russian Research Expert Center, in the valley of the Kamenka River (at the village of Barki in the Bryansk Region) in 1992 (Linnik, 2018). A vertical profile of depth penetration of <sup>137</sup>Cs was studied in five soil profiles in a catena 50 m long, with elevations varying within 2.2 m (Korobova et al., 1998). Systematic changes in the <sup>137</sup>Cs concentration were detected in soil profiles in the catenary sequence of automorphic to hydromorphic settings. In an automorphic one, the maximum  $^{137}$ Cs activity was detected in the layer 0–3 cm, with a subsequent nonlinear decrease in the activity with depth in the soil profile. In the hydromorphic soil, the  $^{137}$ Cs activity linearly decreased to a depth of 20 cm (Linnik, 2018).

Studies of the penetration of <sup>137</sup>Cs to various depths in virgin soils at predpolessie landscapes eight years after the Chernobyl accident (1994) at 16 experimental plots of the Taifun Research and Development Association showed that the maximum of the <sup>137</sup>Cs concentration in automorphic conditions had shifted from the surface down the soil profile. In hydromorphic settings, the <sup>137</sup>Cs activity decreased more evenly with a diffused activity peak in the layer 0–10 cm (Martynenko et al., 2003; Linnik, 2018).

Studies of the vertical <sup>137</sup>Cs distribution 15–20 years after the Chernobyl accident confirmed that the vertical <sup>137</sup>Cs distribution is markedly different in automorphic and hydromorphic settings (Kvasnikova et al., 2005, 2006).

Studies of <sup>137</sup>Cs migration in noncultivated and arable soils in the Tula region have reproduced the longterm regime of vertical <sup>137</sup>Cs migration. A few years after the accident, radiocesium actively migrated from the soil surface to the layer 2–6 cm. During the next five to ten years, the vertical migration of <sup>137</sup>Cs maximally slowed down, its concentration practically did not vary within the layer 2–13 cm, and its total activity increased to 25% in deeper soil layers. Ten to fifteen years later, the vertical penetration of <sup>137</sup>Cs in the old arable soil stabilized at an average rate of 1 mm/year, which is five- to tenfold lower than within a few years after the accident (Lipatov et al., 2003).

For long time spans, analysis of the vertical distribution of <sup>137</sup>Cs in the soils of agrocenoses shall be conducted with regard for the effects of sheet erosion (which amounts to a few millimeters a year; Konoplev et al., 2016), whose order is comparable to the vertical <sup>137</sup>Cs penetration (1 mm/vear) (Lipatov et al., 2003).

Erosion processes in the accumulation zones (in the bottom parts of the slopes, in floodplains, and in ravines) increase the thickness of the soil layer proportionally to the amount of the deposited diluvium. The soil layer contaminated in 1986 is afterwards overlain by younger sediment. The identification of such a horizon in a soil profile is used to date the sediments and thus calculate the sedimentation rate (Golosov, 2006).

Anthropogenic and natural radionuclides (<sup>137</sup>Cs and <sup>210</sup>Pb) have long been used as geochemical tracers in estimating the balance of erosional debris (He and Walling, 1997; Walling et al., 2000; Mabit et al., 2014). Unlike anthropogenic <sup>137</sup>Cs, whose atmospheric fallout vary with time (as a result of some minor radiation accidents or severe accidents, such as those at Chernobyl and Fukushima), the flux of <sup>210</sup>Pb is commonly



Fig. 1. Location map of agrolandscape catenas in Bryansk oblast: (1) Dubrovka, (2) Kostitsa, (3) Vyshkov, (4) Brakhlov.

believed not to vary with time, and this warrants its usage in dating recently accumulated sediments (Appleby, 2008).

In this paper, we study the patterns of the vertical distribution of rediocesium in the soils of agrocenoses. The plots selected for this purpose form catenary junctions and thus make it possible to study how radiocesium reaches various penetration depths of soil depending on their catena position in the following landscapes: (1) predpolessie (with sod—podzol sandy loam soils) and (2) opolje landscapes (with gray forest medium loam soils) at the variable density of radioactive contamination. In the alluvial soils, we studied both <sup>137</sup>Cs and <sup>210</sup>Pb distributions to evaluate the sedimentation rates of the alluvial and diluvial sediments.

#### MATERIALS AND METHODS

The studies were carried out at four model slope plots of argocatenas (Fig. 1) in the western part of the Bryansk Region. They are characterized by different landscape conditions of <sup>137</sup>Cs migration. Two of them occur at predpolessie landscapes (Vyshkov and Dubrovka), and the other two belong to opolje landscapes (Brakhlov and Kostitsa).

To measure the activity of radionuclides in the laboratory, the soils were sampled in 2016 and 2017. Because  $^{137}$ Cs was evenly distributed in the arable layer (20–25 cm) of the agrocenoses, the soil samples were taken from the soil profiles at 5 cm spacing, with outlining the area to a depth of 30 cm. The layer 30–40 cm was characterized by a single sample. Samples from virgin soils at the Kostitsa test site (two floodplain sampling sites, 427 and 432, and one sample site, 427, in a ravine) were collected in 2016. In the virgin-soil areas (floodplain and the bottom of a ravine), samples were gathered with regard for the lithologies, and hence, the sampling sites were spaced 3 to 5 cm apart. The samples were collected to a depth of 30–40 cm (to the perched seasonal water level). The total numbers of samples from a single profile were 7 for agrosoils and 7–9 for hydromorphic soils.

The numbers of the sampling sites at the study areas were as follows: (1) four soil pickets at Dubrovka (29 samples), (2) six soil pickets at Vyshkov (43 samples), (3) four soil pickets at Brakhlov (28 samples), (4) four soil pickets at Kostitsa (21 samples), and (5) three soil pickets - two at the floodplain of the Kostitsa River plus one ravine (22 samples).

#### GAMMA-RAY SPECTROMETRY

The radionuclide composition of some soil samples taken from individual soil layers was analyzed at the Laboratory for Environmental Geochemistry of the Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, by a low-background gamma-spectrometric system with multilayer passive protection (lead, old iron, copper, and acrylic plastic). The recording system was a BEGe3825 (Canberra, USA) broadband detector with a thin (0.6 mm) carbon composite inlet glass (crystal diameter 70 mm, area 380 mm<sup>2</sup>, thickness 25 mm), which allows registering gamma quanta 5 keV to 3 MeV. The energy resolution on the line 1332 keV Co-60 was no worse than 0.6 keV. Information was acquired and processed on a DSA-1000 (USA) spectrum analyzer. The raw spectra were processed with the Genie 2000 ver. 3.2.1 software. The measuring system was certified on a regular basis at the All-Russia Scientific Research Institute for Physical Engineering and Radiotechnical Metrology (VNIFTRI). According to the latest checkup, the minimum measurable activity over a time span of 3600 s is not worse than 0.1 Bq/sample. The standard sample weight for activity measuring was 100 g.

#### EVALUATION OF SEDIMENTATION RATE USING THE <sup>137</sup>Cs AND <sup>210</sup>Pb ISOTOPES

Data on the vertical distribution of the radionuclides were acquired by measuring the <sup>137</sup>Cs and <sup>210</sup>Pb isotopes, whose half-lives are 22.3 years for <sup>210</sup>Pb and 30.17 years for <sup>137</sup>Cs. The <sup>137</sup>Cs and <sup>210</sup>Pb isotopes were measured with precision to 10 and 12%, respectively.

To calculate the average sedimentation rate in the soil profile  $^{210}$ Pb activity was measured in individual layers. In addition, we also analyzed  $^{226}$ Ra to calculate what fraction of the  $^{210}$ Pb was in equilibrium with  $^{226}$ Ra.

The accumulation rate of alluvium in the floodplain of the Kostitsa River was quantified by analyzing the distribution of <sup>137</sup>Cs and <sup>210</sup>Pb<sub>ex</sub>. The average sedimentation rates by the method of unequilibrated <sup>210</sup>Pb is based on a technique described in (Sapozhnikov et al., 2006; Rusakov et al., 2019). Measurements of the average sedimentation rates by the method of nonequilibrium <sup>210</sup>Pb are based on a technique described in (Sapozhnikov et al., 2006; Rusakov et al., 2019).

The age of sediments from the measured <sup>210</sup>Pb data was calculated by the CRS (Constant Rate of Supply) model (Appleby and Oldfield, 1978; 1983; Sikorski and Bluszcz, 2003; Bonotto and García-Tenorio, 2014). The CRS model assumes that the flux of atmospheric <sup>210</sup>Pb does not vary with time, whereas the sed-iment rate can vary over the modeled time span. The age of sediment layers was calculated by the formula

# $\mathbf{t} = (1/\lambda) \times \ln[A_{\rm c}(0)/A_{\rm c}(x)],$

where  $A_c(0)$  is the specific activity of excess <sup>210</sup>Pb for the whole profile,  $A_c(x)$  is the integral activity in a layer below a depth (x), and  $\lambda = \text{year}^{-1}$  is the exponential decay constant, which is 22.3 years for <sup>210</sup>Pb.

## **RESULTS AND DISCUSSION**

The "global" <sup>137</sup>Cs fallout in 1985 in the Bryansk Region showed roughly equal contamination level with <sup>137</sup>Cs at a density of about 1.3 kBq/m<sup>2</sup> (Vorob'ev, 2013). For the four model plots, the contamination levels with <sup>137</sup>Cs at that time varied from 0.74 to 1.5 kBq/m<sup>2</sup>. The 1986 Chernobyl accident has manyfold increased the "global" <sup>137</sup>Cs fallouts: by approximately 1000 times for the Novozybkov district (where

the Vyshkov test site is located), 100 times for the Brakhlov test site, and 10 times for the Kostitsa and Dubrovka test sites (Linnik et al., 2017).

Aerial gamma radiation survey (AG) data acquired in 1993 (Linnik, 2018) indicate that the level of contamination with <sup>137</sup>Cs (with regard for decay as of 2017, when the samples were collected) at the other two agrocatenas (Kostitsa and Dubrovka), 280 and 250 km away from the Chernobyl nuclear power plant, was 5– 12 and 1.7–9 kBq/m<sup>2</sup>, respectively. The contamination levels at two other agrocatenas, Vyshkov and Brakhlov, at 175 and 195 km away from the plant, were 265–907 and 35–120 kBq/m<sup>2</sup>, respectively.

Detailed landscape—biogeochemical analysis of these model plots is presented in (Linnik et al., 2017). Agrolandscape catena soils are characterized by a vertical <sup>137</sup>Cs distribution pattern in the arable layer to a depth of 40 cm (Borisov et al., 2019). Elevated levels in the profiles were determined using the SRTM database of elevations (Jarvis et al., 2008).

The spatial heterogeneity of the <sup>137</sup>Cs distribution in the agricultural soils is much less significant than that in the old arable soils and those in forest landscapes (Lipatov et al., 2007). The <sup>137</sup>Cs vertical distribution in the agrocenoses varies depending on the soil management (arable land or pasture) and on the character of the earlier soil-protection measures. To diminish the contamination of the agricultural produce in the compulsory evacuation zone (Vyshkov), the soil was deeply replowed in 1990, and the contaminated layer was displaced to deeper soil levels.

Radiocesium penetrates into the sub-arable horizon of the arable soils by diffusion and convective transfer with soil moisture. Annual soil treatment makes the <sup>137</sup>Cs contamination of the arable layer more even both laterally and vertically. Under certain conditions, if the specific soil density drastically changes at the boundary of the plow pan, which decreases its permeability to water, the specific <sup>137</sup>Cs concentration may sometimes increase (Lipatov et al., 2007).

#### DUBROVKA AGROLANDSCAPE CATENA

The Dubrovka agrolandscape catena (Fig. 2) is characterized by elevations of 165-185 m and includes two autonomous segments with different directions of lateral transfer: (1) a segment at the interfluvial plain is perpendicular to the flow line of the lateral transfer (sampling sites 136, 138, and 139) and (2) a catena line across a convex slope, along the erosion wash-off direction (sampling sites 140 and 142). The thickness of the arable layer  $A_{\rm ara}$  reaches 20 cm. Agricultural sod– podzol gleyic diluvial loam soils are found in local depressions, with the arable layer  $A_{\rm ara}$  25 cm thick.

In the bottom part of the slope (Site 142, arable soil, transit zone), sheet erosion exposes a loamy moraine with small shingles from beneath the soil.

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Fig. 2. Dubrovka agrolandscape catena. (1) Line of elevations; (2) sampling sites; (3) vertical <sup>137</sup>Cs distribution in the soil.

The soil profile is as follows:  $A_{ara}$  is loam with small shingles (to a depth of 22 cm) and dense loam below (to a depth of 40 cm). As a consequence of the intense removal of the finest <sup>137</sup>Cs-enriched fractions, Site 142 is characterized by the lowest specific activity of <sup>137</sup>Cs (as low as 7–8 Bq/kg), and the <sup>137</sup>Cs concentrations in the uppermost 5-cm layer are the lowest: 5 Bq/kg. Because of the barrier effect of the plow pan for <sup>137</sup>Cs migration, <sup>137</sup>Cs penetrates to lower layers in a manner analogous to that at Site 136 (vertical migration is 17.7%).

Site 136 is located in the zone of the most intense soil wash-off from the plowing land. The average specific  $^{137}$ Cs activity in the arable layer 0–20 cm is 10.4 Bq/kg. The radial migration removal of the arable laver (0-25 cm) is 5.6%. In the accumulation zone (Site 138) in the bottom part of the slope in the treebelt area (which serves as a biogeochemical barrier) outside the field, the thickness of the contaminated soils increases. The vertical <sup>137</sup>Cs distribution in the layer 0-30 cm (the average specific activity is equal to 11.2 Bq/kg) is practically homogeneous. The increase in the average <sup>137</sup>Cs specific concentration due to the accumulation of fine fractions is 8%. In the lower layer 30–40 cm at Site 138, <sup>137</sup>Cs activity is almost twofold lower (6.5 Bg/kg), which is much higher than the  $^{137}$ Cs contamination level of the subarable layer (<1 Bg/kg) at the field. The highest specific concentration of <sup>137</sup>Cs was detected in a closed ravine in the interfluvial plain (Site 139), where the specific concentration in the layer 0-5 cm increases to 29 Bq/kg due to the accumulation of the clay fraction.

At the convex segment of the agrocatena (elevations 177-183 m, Sites 140 and 142), with  $^{137}$ Cs the soil contamination density varies from 1.7 to 2.6 kBq/m<sup>2</sup> (AG data). The minimum  $^{137}$ Cs contamination density (1.7 kBq/m<sup>2</sup>) was found at Site 140 (plowing land). Unlike the character of the vertical <sup>137</sup>Cs distribution at Site 136, where the subarable layer is contaminated (<sup>137</sup>Cs activity in the layer 25–30 cm is  $1.6 \pm 0.3$  Bq/kg), a geochemical barrier was detected at Site 140, at the boundary of the plow pan (20 cm): the specific <sup>137</sup>Cs activity decreases from 10.8 ± 0.7 Bq/kg in the arable layer to 0.3 ± 0.13 Bq/kg in the layer 20–25 cm. The radial migration for this part of the catena soil is 2.7%.

#### VYSHKOV AGROLANDSCAPE CATENA

The Vyshkov agrolandscape catena (Fig. 3) occurs at elevations of 132-160 m, with predpolessie landscapes characterized by a light granulometric composition of the soil-forming rocks. The tops of the moraine hills are covered with agricultural sodmedium podzol sandy-loam soils with permanent grasses (Site 80). The soil typically has a thick arable humus horizon (to 25 cm), which is underlain by sandy material, and this ensures intense water filtration and potentially intense <sup>137</sup>Cs vertical migration. In the transit zone of the agrolandscape catena (Site 81), the thickness of the humus horizon reaches 28–30 cm. In the bottom part of the slope, on the diluvial blanket with thick sod buried soils (Site 83), the thickness of the humus horizon reaches 30 cm, and it is underlain (to a depth of 40 cm) by sandy sediments.

In the Vyshkov agrocatena, six soil profiles were studied: three of them were on a slope and the other three were in the floodplain of the Buldynka River. At the top of the agrocatena (Site 80), the arable layer to a depth of 25 cm shows an uneven <sup>137</sup>Cs distribution (1450–1740 Bq/kg). In the subarable layer at a depth of 30–40 cm, the <sup>137</sup>Cs activity decreases to 7 Bq/kg. The specific <sup>137</sup>Cs concentration at the boundary of

Vyshkov. Agrolandscape catena



Fig. 3. Vyshkov agrolandscape catena. (1) Line of elevations; (2) sampling sites; (3) vertical <sup>137</sup>Cs distribution in the soil.

the arable layer  $(20-25 \text{ cm}, 1452 \pm 81 \text{ Bq/kg})$  and the subarable layer  $(25-30 \text{ cm}, 41 \pm 2.3 \text{ Bq/kg})$  differs by a factor of 35, which corresponds to the rate of <sup>137</sup>Cs vertical migration in the Dubrovka agrocatena (Site 140), where the analogous parameter is 36. The removal of <sup>137</sup>Cs outside the arable layer is evaluated at 0.74%.

In the transit zone of the agrocatena (Site 81), no boundary of the plow pan was found to a depth of 40 cm. The vertical distribution of <sup>137</sup>Cs is less homogeneous: the soil profile can be subdivided into two parts with different specific activity of <sup>137</sup>Cs. The <sup>137</sup>Cs activity in the layer 0–20 cm varies within the range of 1595–1810 Bq/kg (the average <sup>137</sup>Cs value is 1700  $\pm$  96 Bq/kg), whereas the specific <sup>137</sup>Cs concentration in the layer 20–40 cm increases to 2160–2312 Bq/kg (the average <sup>137</sup>Cs value is 2260  $\pm$  126 Bq/kg).

The <sup>137</sup>Cs concentration in the lower layer 20– 40 cm is 1.32, which is atypical of the natural distribution of <sup>137</sup>Cs in soil as a result of the diffusion and diffusion–convection migration of radionuclides. It very likely occurs due to significant rehabilitation conducted in 1990, when the uppermost contaminated soil layer of the soils was buried at greater depths.

An elevated <sup>137</sup>Cs concentration was detected at Site 81 (layer 0–5 cm, 1810  $\pm$  102 Bq/kg), compared to lower horizons (5–20 cm, average <sup>137</sup>Cs specific activity of 1660  $\pm$  92 Bq/kg). The increase in <sup>137</sup>Cs specific activity in the layer 0–5 cm compared to the layer 5– 20 cm is 9%. A plausible explanation of <sup>137</sup>Cs concentrating in the upper layer 0–5 cm is that the soil was brought there as a result of erosion from the upper part of the agrocatena (Site 80), where the uppermost layer 0–5 cm contains similar specific <sup>137</sup>Cs concentration (1743  $\pm$  91 Bq/kg). The erosion-driven removal of material at Site 81 is, thus, combined with the simultaneous accumulation of a soil layer, i.e., the geochemical migration is of the transit—accumulated type.

In the lower part of the agrocatena, which is located on a diluvium blanket (Site 83), the specific <sup>137</sup>Cs activity is notably higher in all soil layers compared to the removal zone (Site 80) and the transit-accumulated zone (Site 81). The specific <sup>137</sup>Cs activity in the layer 0–20 cm reaches 2700–2900 Bq/kg, starts increasing at a depth of 25 cm, and increases to  $3570 \pm 200$  Bq/kg as its maximum in the layer 30–40 cm. The anomalously high specific <sup>137</sup>Cs activity in the lower part of the soil profile would be a consequence of soil remediation in 1990.

In general, the Vyshkov agrocatena is characterized by significant changes in the contamination density that occurred in the zone of slope junction over the three decades after the accident. The differences in the contamination density with <sup>137</sup>Cs at Sites 80, 81, and 83, which is 610, 1170, and 1760 kBq/m<sup>2,</sup> respectively, are explained by lateral transfer.

According to estimates in (Kuznetsov et al., 2018), predpolessie landscapes in the Novozybkov district are characterized by weak soil wash-off of approximately 2 tonnes/ha per year. With regard to the slope part of the Vyshkov agrocatena, it can be stated that lateral migration in the lower part of this agrocatena leads to <sup>137</sup>Cs accumulation, and this results in the deposition of in diluvial material with high specific <sup>137</sup>Cs concentrations. The accumulation of <sup>137</sup>Cs to the highest concentrations in diluvial material was previously detected in many parts of Russia, e.g., in the Stavropol krai (Belyaev et al., 2005), the Tula Region (Golosov, 2006; Kuznetsov et al., 2018), and the Bryansk region (Linnik, 2018).

Sites 70, 91, and 92 are situated in the floodplain of the Vyshkov agrocatena. The profile of the vertical distribution of radionuclides in floodplain soils largely depends on the hydrological regime during the high water period, when suspended material is deposited onto the soil (Linnik, 2001).

The floodplain of the Buldynka River has a surface topography undulating within 2–2.5 m, with numerous dry channels. To study the local variations in the vertical <sup>137</sup>Cs distribution samples were taken at the elevated part of the floodplain (Site 91) and at a nearby swampy lowland on the floodplain (Site 92). The high parts of the floodplain (Sites 70 and 91) were rehabilitated in 1990. These floodplain parts typically had an old arable horizon  $A_{ara}$  up to 28 cm thick. The soil is soddy floodplain, medium loamy, gleyish at Site 70 and light loamy gleyish at Site 91. The floodplain has likely never been plowed and remained meadows. In the hydromorphic part of the soil (Site 92), alluvial soddy granular gleyish loamy soils with humus horizons were formed. The upper part of the profile contains much organic matter.

The contamination density of the floodplain part of the Buldynka River is remarkably variable depending on the elevation. The maximum <sup>137</sup>Cs contamination density is found in the high part of the floodplain (Site 70, <sup>137</sup>Cs = 740 kBq/m<sup>2</sup>). In the lower part of the floodplain at Sites 91 and 92, the <sup>137</sup>Cs contamination density is 455 and 346 kBq/m<sup>2</sup>, respectively. Thus, the maximum <sup>137</sup>Cs contamination density in the floodplain of the Buldynka River is insignificantly higher than that in the upper part of the agrocatena (Site 80) and 2.4 times lower than that in the zone with diluvium depositions (Site 83).

Owing to rehabilitation measures in the floodplain, the vertical distribution of <sup>137</sup>Cs has a complicated character, and a conventional method of <sup>210</sup>Pb<sub>ex</sub> sediments dating is there inapplicable. At Site 70, a buried horizon with a <sup>137</sup>Cs concentration of  $3150\pm174$  Bq/kg (an effect of land rehabilitation) was found at a depth of 15–20 cm, whereas the upper part of the profile shows an inverse distribution of <sup>137</sup>Cs specific concentration displaying a monotonouse increase from 1720 ± 96 Bq/kg in the layer 0–5 cm to 2564 ± 142 Bq/kg in the layer 10–15 cm.

In the soil profile at Site 91 (old arable soil),  $^{137}$ Cs activity in the upper part of the soil profile at the high floodplain (to a depth of 20 cm) reaches 1360 Bq/kg, which is significantly lower than that in the arable soils of this agrocatena. The activity of  $^{137}$ Cs at a depth of 32–40 cm decreases to 27 Bq/kg. The radial migration beyond the boundaries of the old arable layer is thus 4.9%.

The soil profile at Site 92 shows a complicated <sup>137</sup>Cs distribution: its low specific concentrations to a depth of 4 cm characterize the contamination of the upper layer of the ground litter, and the layer 4–7 cm also

contains partly decomposed organic matter. The maximum <sup>137</sup>Cs activity was detected at a depth of 22–27 cm (1180 Bq/kg) which may be attributed to a buried layer of sediments of the Chernobyl-driven fallout in 1986, later overlain by alluvial sediment. In the profile at Site 70, the maximum <sup>137</sup>Cs activity occurs at a depth of 15–20 cm, which likely results from the 1986 fallout.

### BRAKHLOV AGROLANDSCAPE CATENA

The Brakhlov agrolandscape catena (Fig. 4) crosses a plowed interfluvial plain of rolling topography, a terrace of the Snov River, and terminates at the floodplain. The watershed area is made up of gray forest soils (Site 100), and the terrace has soddy–podzol powdery sandy soils (Sites 123 and 125). The floodplain is dominated by gleyish soils with a meadow vegetation. The significant range of the elevations (from 167 to 140 m) is favorable for erosion processes (Linnik et al., 2017). The <sup>137</sup>Cs contamination density down the profile is as follows: 77 kBq/m<sup>2</sup> at Site 100, 45 kBq/m<sup>2</sup> at Site 123, 260 kBq/m<sup>2</sup> at Site 125, and 148 kBq/m<sup>2</sup> at Site 128.

The distribution of  $^{137}$ Cs activity in the arable layer is practically homogeneous to a depth of 0–20 cm (to a depth of 25 cm in the watershed, Site 100). No barrier effect of the plow pan is detected in this part of the landscape, and  $^{137}$ Cs activity monotonously decreases from 168 to 36 Bq/kg to a depth of 40 cm. The  $^{137}$ Cs vertical removal to the layer 30–40 cm is at a maximum in the catena: 9%.

The average <sup>137</sup>Cs concentration in the arable layer of the lateral transfer zone at Site 100 (180 Bq/kg) is higher than that in the transit zone (<sup>137</sup>Cs activity 146 Bq/kg at Site 123). The thickness of the arable layer in the transit zone decreases to 20 cm because of erosion. The <sup>137</sup>Cs removal to the arable sublayer drastically decreases to 1.2 Bq/kg in the layer 30–40 cm. The intensity of <sup>137</sup>Cs vertical migration out of the layer 0-30 cm is minimal 0.5%.

In the accumulation zone (Site 125), the arable layer is as thick as 25 cm and the average specific activity of <sup>137</sup>Cs in it increases to 186 Bq/kg, which is comparable to this parameter value on the watershed (Site 100). The vertical <sup>137</sup>Cs migration of is moderate, with <sup>137</sup>Cs removal outside the arable layer 0–25 cm equal to 3%.

In the valley of the Snov River, in the flattened waterlogged floodplain, <sup>137</sup>Cs was deposited on the floodplain (Site 128). The floodplain is characterized by the maximum specific activity of <sup>137</sup>Cs across the whole agrocatena: 1300 Bq/kg in the layer 0-5 cm. The decrease in the radiocesium activity in the lower layers is nearly exponential (to 6 Bq/kg in the layer 30-40 cm), which is absolutely atypical of alluvial soils. Presumably, the predominantly stagnant regime during the high-water period, with the floodplain



Fig. 4. Brakhlov agrolandscape catena. (1) Line of elevations; (2) sampling sites; (3) vertical <sup>137</sup>Cs distribution in the soil.

turning into a still water body, whose sedimentation resembling that in lakes and lagoons, and being responsible for the situation. The <sup>137</sup>Cs contamination density in the floodplain part is 150 kBq/m<sup>2</sup>. The dominant <sup>137</sup>Cs source for the floodplain is the upper part of the Snov River basin (Linnik et al., 2017).

Consequently, the Brakhlov agrocatena is characterized by intense lateral transfer, reflected in the increase in the <sup>137</sup>Cs contamination density and an increase in the thickness of the arable layer and in the specific radiocesium concentration down the slope.

#### KOSTITSA AGROLANDSCAPE CATENA

The Kostitsa agrocatena (elevations 165-150 m) is approximately 500 m long (Fig. 5) and runs across a plowed slope of the northern exposure of the Kostitsa River, with gray forest soils. The thickness of the humus horizon is controlled by the boundary of the plow pan: it is not flat, and its depth varies from 20 to 35 cm at the top of the catena, and  $A_{ara}$  increases to 28–32 cm in its transit–accumulation part (Site 156). At the boundary of the field (accumulation part of the agrocatena), a buried arable horizon 25 (28) cm thick is detected. The granulometric composition of soils in the arable horizon systematically varies from loamy (Site 157) to powdery sandy clay (Site 156) and to light loam in the lower part of the agrocatena (Site 153).

The vertical distribution of <sup>137</sup>Cs at the three soil pickets in the agrocatena is practically even within the arable layer, and the maximum of the specific concentration is 14–16 Bq/kg. The average <sup>137</sup>Cs concentration in the arable layer increases down the slope and is 14.4 Bq/kg at Site 157, 15.4 Bq/kg at Site 156, and 15.8 Bq/kg at Site 153. However, because of the spatial variability in the average density of the soil (1.72, 1.67, and 1.82 g/cm<sup>3</sup>, respectively), the <sup>137</sup>Cs contamination density is 9.94, 8.4, and 8.55 kBq/m<sup>2</sup>, respectively.

The soil lateral transfer is thus relatively weak and local. The reason for the weakness of the lateral transfer is a combination of factors such as the northern exposure and the shortness of the slope. A combination of these features is unfavorable for erosion (Golosov, 2006; Linnik et al., 2017; Kuznetsov et al., 2018).

The intensity of the  $^{137}$ Cs radial migration (transfer outside the arable layer) significantly varies over the agrocatena. The intensity of  $^{137}$ Cs removal outside the arable layer (below 30 cm) in this agrocatena predictably decreases down the slope: it is 26.3% in the autonomous setting, 8.5% in the transit zone, and 1.4% in the accumulation zone.

# ESTIMATION OF SEDIMENTATION RATE AND DYNAMICS

The network of ravines and river floodplains is the final link in the chain of migration processes of the eroded material, which is brought from the watershed area and from the slopes of river basins. Much of the eroded material is deposited in the upper parts of the drainage network (Walling, 1983; Golosov, 2005). Studies of the radial distribution of <sup>137</sup>Cs in floodplain soils and those in the network of ravines are an efficient tool for evaluating the sedimentation rate.

Among the four analyzed agrocatena test sites, only the floodplain the Kostitsa River has not been plowed. The sampling sites were chosen in the Kostitsa landscape—radiation catena of the Pochep opolje (Linnik et al., 2017). Two sites are located in the floodplain the Kostitsa River (Sites 422 and 427), and one is in the bottom part of a ravine (Site 432). The wet grass meadow vegetation operates as a powerful biogeochemical barrier, accumulating 24% <sup>137</sup>Cs (Linnik et al., 2019).

The soil profiles are described in (Linnik et al., 2019). Site 422 (Fig. 6) is located on the right-hand floodplain, whereas Site 427 is in the central part of



Fig. 5. Kostitsa agrolandscape catena. (1) Line of elevations; (2) sampling sites; (3) vertical <sup>137</sup>Cs distribution in the soil.



Fig. 6. Radial distribution of <sup>137</sup>Cs and <sup>210</sup>Pb in the floodplain soils on the Kostitsa River.

the left-hand floodplain, and they are characterized by principally different hydrological conditions during high-water periods. The right-hand floodplain belongs to the transit zone of suspended riverine material, whereas the left-hand floodplain is a deposition zone of the suspended material, which is brought from the upper reaches of the river, from the ravine network, and from adjacent slopes.

Site 422 (Fig. 6) has a floodplain soddy loamy soil with a buried gleyed heavy-loam horizon. No obvious maximum in <sup>137</sup>Cs activity corresponding to the 1986

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contamination was detected in this soil profile. The specific <sup>137</sup>Cs activity remains uniform (44 Bq/kg) to a depth of 15 cm, while it decreases (to 32 Bq/kg) in the layer 15–20 cm, and reaches 19 Bq/kg in the layer 25–30 cm. A buried humus horizon consisting of loam is found at a depth of 24 cm. Starting at a depth of 25–30 cm, the <sup>137</sup>Cs concentration slightly increases (to 25 Bq/kg), which may be explained by the occurrence of a geochemical barrier (gley horizon).

The vertical distribution of  $^{210}Pb_{ex}$  (Fig. 6) in the transit zone of riverine water at turbulent flow (soil



Fig. 7. Distribution of the specific  $^{210}$ Pb and  $^{137}$ Cs activity and calculated sedimentation rate in 1960–2016. The marker indicates the lower time boundary of the time span.

profile at Site 422) differs from the theoretical distribution for stationary sedimentation of suspended material (an exponential decrease in the specific activity with depth), which gives way to an increase in the activity with depth, starting from a depth of 30 cm. The correlation of the decrease in  $^{210}$ Pb<sub>ex</sub> with depth is weak ( $R^2 = 0.64$ ), complicating a reasonably accurate application of  $^{210}$ Pb<sub>ex</sub> sediments dating. By neglecting the lowest layer 30–35 cm, we obtained a sedimentation rate of 0.7 ± 0.18 cm/year for the rest of the soil profile.

For the upper part of the soil profile, the sedimentation rate can be evaluated using the <sup>137</sup>Cs tracer. Assuming that the "Chernobyl" layer corresponds to a depth of 15–20 cm (the uppermost layer 0–15 cm is mixed), we expect the accumulation rate of alluvium over the past 30 years at this site of the floodplain to be up to  $0.58 \pm 0.11$  cm/year.

Because the eroded material was transported from the plowed basin of the Kostitsa River, unusual conditions were produced at the central part of the left-hand floodplain (Site 427) (Fig. 6). In contrast to the profile at Site 422, where it was problematic to distinguish the "Chernobyl horizon", the profile at Site 427 shows a clearly discernible layer (12–14 cm) with the maximum <sup>137</sup>Cs concentration (75 Bq/kg), which can date back to 1986.

The upper soil horizons (to 8 cm) are clearly discernible in the soil profile by both color and granulometric composition (silt with an anomalously low <sup>137</sup>Cs concentration of 6.5 Bq/kg). It can comprise deposited eroded material carried from the plowed slopes. This presumption is favored by the fact that the uppermost 8-cm layer of the profile at Site 428 has a <sup>210</sup>Pb concentration (77 Bq/kg) which belongs to <sup>210</sup>Pb range concentration (60–100 Bq/kg) in the arable soils (Linnik et al., 2019).

If the uppermost mixed layers to a depth of 12 cm and the use of  $^{210}$ Pb<sub>ex</sub> for the rest of the cores are not

taken into account, we arrive at an estimate of 0.7  $\pm$  17 cm/year for the sedimentation rate.

According to data on the <sup>137</sup>Cs distribution in the soil profile, this estimate yields the following result. Inasmuch as the "Chernobyl horizon" corresponds to a depth of 12–14 cm, we obtain that the alluvium sed-imentation rate over the past three decades in the upper horizons of the soil profile at Site 427 was likely up to  $0.42 \pm 0.08$  cm/year.

The bottom part of a ravine (Site 432, Fig. 7) is characterized by virgin-soil conditions of the sedimentation of eroded material. This undoubtedly follows from the character of the <sup>137</sup>Cs and <sup>210</sup>Pb<sub>ex</sub> distribution in the soil profiles (the distributions are satisfactorily fitted by an exponential distribution model) and also from the visual examination of the lithological structure of the soil in the course of sampling. Since autochthonous <sup>210</sup>Pb did not reach minimal values due to its decay (the layer 35–40 cm shows an increase in the specific <sup>210</sup>Pb<sub>ex</sub> activity that is difficult to interpret), the results presented below shall be viewed as provisional.

Using the CRS model (Sikorski and Bluszcz, 2003; Bonotto and García-Tenorio, 2014), we have evaluated the intensity of sedimentation. The modeling results demonstrate remarkable differences between accumulation episodes of the suspended material. The sod and humus horizons of the soil (0-15 cm) were formed starting from 1992. This soil layer is readily discernible by morphological features and by density  $(0.7-0.87 \text{ g/cm}^3)$ . The maximum erosion of soil from the fields took place in 2000-2007: the sedimentation rate was 0.7 cm/year. The intensity of soil wash-off from the fields in 2007-2016 was lower: the sedimentation rate in the ravine decreased to 0.55 cm/year, which is comparable to this parameter value during the economic recession in 1992-2000, when the sedimentation rate was 0.62 cm/year. This erosion rate is comparable to that when farming was conducted in

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the Soviet Union in 1982-1992, when the diluvium sedimentation rate was 0.5 cm/year. The sedimentation rate of eroded material in this ravine was at a minimum (0.24 cm/year) in 1961-1982.

The low erosion rates may be explained by the application of principally different, less economically efficient farming techniques and less powerful machinery. It is worth mentioning that lower soil horizons are compacted in the course of sedimentation of deposited sediments under gley conditions, which is confirmed by the higher density  $(1.6-1.8 \text{ g/cm}^3)$  of the soils than that of the humus horizon.

#### CONCLUSIONS

Data presented above on the vertical distribution of  $^{137}$ Cs of "Chernobyl origin" in four agrolandscape catenas in the Bryansk Region (the data were acquired in 2016–2017) characterize  $^{137}$ Cs migration 30 years after the accident at the Chernobyl nuclear power plant. Experimental data on the  $^{137}$ Cs distribution confirm that the radiation conditions at the radionuclide-contaminated territory have stabilized.  $^{137}$ Cs is contained mostly in the humus arable layer, and its removal outside the arable layer depends on landscape conditions: this parameter is evaluated at 2.7–17.7% for the Dubrovka agrocatena at a predpolesie landscape and at 0.5–9% for the Brakhlov agrolandscape catena at an opolje landscape.

In addition to the natural migration of radiocesium into the subarable layer, <sup>137</sup>Cs was sometimes plowburied up to a depth of 40 m, i.e., outside the arable layer, when the depth of the plow pan varied. This situation was found in an opolje landscape (Kostitsa agrocatena), in an automorphous setting of the agrocatena, where the mechanical transfer of <sup>137</sup>Cs outside the 25-cm arable layer was 26.3%. In the transit position of the same agrocatena, the vertical transfer from the arable layer was 8.5, and 1.4% in the accumulation zone.

The vertical migration in the catenary junctions generally varies in the intensity of <sup>137</sup>Cs removal outside the arable layer because of the heterogeneity of the soil–physical conditions of water migration in the subarable layer, as was detected by analyzing the vertical radiocesium distribution at the model plots.

In landscapes with a high <sup>137</sup>Cs contamination density (Vyshkov agrocatena ), soil rehabilitation operations were carried out in 1990, with the soil layer contaminated with radionuclides was overturned and buried. The radionuclides were carried outside the soilroot zone to diminish the contamination of the plantgrowing produce. Lateral migration facilitates the natural burial of <sup>137</sup>Cs in the bottom parts of the slopes (Vyshkov agrocatena), whereas <sup>137</sup>Cs concentration in an automorphous position tends to further decrease due to lateral <sup>137</sup>Cs removal.

Three decades after the Chernobyl accident, the alluvial soils are the hardest to study their radial <sup>137</sup>Cs distribution. The reason for this is both the rehabilitation operations (Vyshkov agrocatena) and the spatiotemporal heterogeneity in the development of soilforming process on the floodplain. The balance of eroded material in the floodplains and ravines is evaluated using geochemical tracers <sup>137</sup>Cs and <sup>210</sup>Pb<sub>ev</sub>. However, their application in practice faces several methodical limitations, first and foremost, the violation of the stationary nature of the accumulation of the buried sediments, as well as their mixing. We encountered this situation when analyzing the <sup>210</sup>Pb distribution in the floodplain of the Kostitsa River with the application of the CRS model. In view of this, the results obtained on the sedimentation rates should be regarded as fairly approximate. However, even such approximate evaluations are important for evaluating sedimentation rates in the floodplains of minor streams as an important stabilizing factor of the ecological conditions.

According to the analysis of <sup>137</sup>Cs distribution in the soils, the sedimentation rate over the past three decades in various parts of the floodplain was  $0.42 \pm 0.08$  to  $0.58 \pm 0.11$  cm/year. The average calculated sedimentation rate in the floodplain over the past five decades is, according to data on the <sup>210</sup>Pb<sub>ex</sub> distribution,  $0.7 \pm 0.18$  cm/year.

For the sod soil in the bottom part of a ravine, we used data on the  $^{210}$ Pb<sub>ex</sub> distribution and the CRS model to evaluate the sedimentation rate in 1961–2016: the minimum sedimentation rate of 0.24 cm/year was detected in 1961–1988, and the maximum value of 0.7 cm/year was obtained for 2000–2007.

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Translated by E. Kurdyukov