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Features of ^{137}Cs distribution and dynamics in the main soils of the steppe zone in the southern European Russia

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ABSTRACT

Data are presented on the concentration and vertical distribution of the artificial isotope ^{137}Cs in soils (chestnut soils, chernozems, solonetzs, solonchaks, meadow soils and alluvial soils) of the steppe zone in the southern Russia. The work has been focused on the study of radiocesium distribution in undisturbed virgin lands. It has been shown that the mean concentration of the radionuclide in the upper (0 to 15 cm) soil layer is 20.5 Bq/kg. The proportions of adsorbed ^{137}Cs in the soils increase with increasing humus content and decreasing pH level. Two types of the profile distribution of radiocesium were distinguished: (1) with the maximum concentration in the upper soil layer and a relatively abrupt decrease with depth (the majority of ^{137}Cs is concentrated in the upper 0 to 15 cm soil layer) and (2) with the maximum concentration of ^{137}Cs shifted into the soil profile down to a depth of 45–55 cm. This difference is related to the soil type, humus content, pH value, and regional climatic conditions. Long-term (2000–2013) studies of the vertical distribution of ^{137}Cs in soil profiles have revealed a decrease in its total activity by 1.5 to 2.0 times. It has been shown that the migration of radiocesium in soil profiles depends on the soil type and the diffusion of the radionuclide itself, as well as on convective transfer, transpiration, infiltration, and colmatage.

1. Introduction

The assessment of the vertical distribution of ^{137}Cs concentration in soils is necessary for the understanding and simulation of different soil processes, primarily migration processes. In addition, the content of this isotope in the environment plays a significant role in the estimation of population radiation doses on the areas contaminated by nuclear accidents. A significant body of data has been accumulated by now on the contamination levels of different soils with the artificial radionuclide ^{137}Cs. The vertical distribution of ^{137}Cs in soils is analyzed not only during the long-term and reconnaissance surveys of contaminated (Isaksson et al., 2001; Hamarneh et al., 2003; Park et al., 2013) and background (Nabyvanets et al., 2000; Al-Masri, 2006; Doering et al., 2006; Turhan et al., 2012; Gaspar and Navas, 2013) areas, but also at the determination of population radiation doses caused by artificial radionuclides (Fujiwara et al., 2012), the estimation of soil erosion (Collins et al., 2001; Chappell and Warren, 2003; Li et al., 2010; Parsons and Foster, 2011), and the study of sources of artificial radionuclides (Lee et al., 2013). The environmental behavior of this radionuclide is also studied in model experiments for the determination of ^{137}Cs migration mechanisms in soil profiles (Shinonaga et al., 2005; Almgren and Isaksson, 2006; Matisoff et al., 2011; Nakanishi et al., 2014), the diffusion rate of the radionuclide (Krstic et al., 2004; Iurian et al., 2014), and its accumulation features in soils (Bai et al., 2002).

^{137}Cs with a half-life period of 30 years was emitted into the environment and trans located into soils, bottom sediments, plants, etc. during tests of nuclear weapons in 1950–1970 (global ^{137}Cs) and after the Chernobyl accident in 1986 (Chernobyl ^{137}Cs). Separate regions were also locally contaminated with this radionuclide because of incidents at enterprises of the nuclear fuel cycle: the East-Ural radioactive trace in the Russian Federation in 1957, the Three Mile Island accident in the USA in 1979 and the Fukushima nuclear disaster in Japan in 2011.

The pollution of terrestrial ecosystems (including soils) with ^{137}Cs due to nuclear weapon testing and accidents at enterprises of the nuclear fuel cycle was irregular and formed spots and bands of contamination of different shapes and sizes. After falling on the soil, ^{137}Cs migrated in the horizontal and vertical directions. The horizontal migration of the radionuclide is mainly affected by the climatic conditions and soil relief. ^{137}Cs reaches the soil with wet and dry fallout from the atmosphere and sorbs to clay and organic components of the soil cover (Tamura, 1964; Staunton et al., 2002). Later on, along with the natural decay of radiocesium, different processes, including plant uptake and

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bioturbation, affect the content and migration of this radionuclide in soil profiles (Müller-Lemans and Van Dorp, 1996; Gastberger et al., 2000; Staunton et al., 2002). The vertical migration of ^{137}Cs is affected by the physicochemical properties of soil (humus content, chemical composition, particle-size distribution), acidity (Askbrant et al., 1996; Matsunaga et al., 2013; Koarashi et al., 2012; Gaspar and Navas, 2013), relief (Korobova and Romanov, 2011), regional climatic features (Legarda et al., 2011) and plant uptake (Gastberger et al., 2000), etc.

The explicit dependence of ^{137}Cs concentration on the physicochemical properties of soil, including the correlation between the radionuclide content and the particle-size fractions in the topsoil (He and Walling, 1996; Askbrant et al., 1996; Navas et al., 2007; Koarashi et al., 2012; Matsunaga et al., 2013; Gaspar and Navas, 2013), allows this radionuclide to be used as a marker (or a tracer) of different soil processes. At the same time, the climatic features of the regions under study, their relief, and tillage practices should be taken into consideration in the study of ^{137}Cs vertical (and horizontal) migration in terrestrial ecosystems (Forsberg and Strandmark, 2001).

Researchers assign a significant role in the accumulation and migration of ^{137}Cs to the content of humus (organic component) in soil profiles (Ritchie and McHenry, 1973; McHenry and Ritchie, 1977; Rigol et al., 2002; Martinez et al., 2010; Navas et al., 2011). The mobility of ^{137}Cs is also related to the fact that ^{137}Cs is an isotope of an alkali metal, a chemical analogue of the essential biogenic element potassium (Sanzharova et al., 2005). The selective and unexchangeable sorption by the soil solid phase is characteristic for ^{137}Cs. The capacity of soils to fix Cs^+ is largely determined by their content of clay minerals where by illite-type hydromicas have the highest capacity of fixing K^+, NH_4^+, and Cs^+ (Dumat and Staunton, 1999; Rosén et al., 1999; Sanzharova et al., 2005; Ziembik et al., 2009, 2010).

It should be noted that almost no data are available on the dynamics of ^{137}Cs or its vertical migration in zonal and intrazonal soils of the steppe zone of Russian Federation over a long time period. Separate studies were performed during the additional radiation survey in 1993–2000 for compiling schematic maps of radioactive contamination in the Russian Federation as a consequence of the Chernobyl accident (Atlas, 1998), as well as for assessing the content of this radionuclide in soils of Rostov oblast within the framework of the prestart monitoring of the Rostov NPP observation zone (Report, 2000).

Presently, the radiation situation (mainly the equivalent dose rate of gamma radiation and the surface air radioactivity) in separate settlements is assessed by some state organizations and agencies (Hygienic and Epidemiological Center in Rostov oblast, 2015; Rosatom, 2014; Typhoon Scientific and Production Association, 2014). However, these works include neither observations in deep soil horizons nor analysis of the relationship between the profile distribution of radiocesium and the physicochemical properties of soils.

The aim of this work was to study the dynamics and distribution of the artificial radionuclide ^{137}Cs in zonal and intrazonal soils of the southern European Russia (Rostov oblast) under consideration for their humus contents and pH values. A distinctive aspect of the study is the use of unique data set on the vertical distribution and migration of radiocesium in undisturbed virgin soils. The determination of equivalent gamma dose rate on the areas under consideration was also an object of the work.

2. Materials and methods

2.1. Study area

The Rostov oblast is located in the Lower Don River basin of the southern European Russia. The oblast is within the steppe zone whereby only its extreme southeastern part is the transitional region between steppes and semideserts. The majority of the area is occupied by agricultural lands, predominantly on high-fertility chernozems.

The Rostov oblast has a moderate continental climate with frosty winters and hot droughty summers. The air temperature has a pronounced annual cycle. January is the coldest month with a mean air temperature of −5 to −9 °C and July is the warmest month with a mean air temperature of +22 to +24 °C. The minimum mean monthly air temperature is periodically observed in February and, more rarely, in December and the maximum air temperature is observed in August and, sometimes, in June or September (Hydrometcenter of Russia, 2015).

Wind erosion, which favors the spread of contamination plumes, is widely distributed in the southeast of the oblast and is favored by flat topography, the high degree of land plowing, the absence of forests, the poor state of forest belts, and dry climate. As a result, fine earth particles can be transferred over 3 to 5 thousand km (Svisyuk, 1989).

In the region of study, the zonal soils included chernozems and chestnut soils (Chernozems and Kastanozems, respectively, according to IUSS Working Group WRB, 2014). The instrazonal soils included solonchaks, solonetzs, and meadow-chernozemic soils (Solonchaks, Solonetzs, and Phaeozems, respectively, according to IUSS Working Group WRB, 2014). The river valleys are composed by alluvial-meadows and alluvial-layered soils (Fluvisols according to IUSS Working Group WRB, 2014). The peculiarity of chernozems is determined by their occurrence on high-carbonate loess-like rocks, clays and loams. Therefore, they are classified as calcareous ordinary chernozems and are characterized by a high content of carbonates. $CaCO_3$ content of was 0.4 to 0.7% in the surface horizons and increased with depth up to 16 to 19% in the parent rock (Bezuglova and Hyirhyirova, 2007). Chestnut soils have a similar calcareous profile. Another feature of the studied soils is the relatively high humus content. Although these chernozems are classified as low-humus soils, their humus content is usually in the range from 4.3 to 5.0% (Bezuglova et al., 1996).

These two components (carbonates and humus) play the deciding role in the fixation and accumulation of cesium. Particle size distribution is another important soil property. The vast majority of chernozems and chestnut soils have clay loamy texture, although heavier (light clayey) varieties can also be detected. On the contrary, lighter (medium and light loamy) varieties are the predominant chestnut soils. A correlation is observed between the particle size distribution and the content of humus in the soils (Bezuglova and Yudina, 2006). Thus, the particle size distribution in the soils also favors the fixation of cesium into low-mobile forms. The soils have a neutral or a weakly alkaline reaction, which also affects the mobility of cesium compounds in the soil profile.

2.2. Selection and preparation of soil samples

The numbers of soil sampling sites and the soil names are given in Table 1 and Fig. 1. Soil samples were taken from virgin lands of the Rostov oblast during the years 2000–2013. Before soil sampling, the equivalent dose rate of gamma radiation (EDR, μSv/h) was measured with a portable MKS-AT1117M dosimeters–radiometers on all of the control plots at a height of 100 cm from the soil surface. In total more than 3500 EDR measurements were performed.

The MKS-AT1117M dosimeter–radiometer with a BDKG-05 gamma radiation detector based on a NaI(Tl) scintillator is designed for measuring equivalent gamma dose rates within the range of 0.03–100 μSv/h. The range of recorded X-ray and gamma radiation is 50–3000 keV its sensitivity to gamma radiation is 760 (pulses·s^{-1})/(μSv·h^{-1}) and the intrinsic relative error of gamma-radiation EDR measurements is no more than ±20% (Atomtekh, 2014).

For studying the profile distribution of ^{137}Cs and assessing the effect of microrelief features on the radionuclide distribution, soil samples were taken from profiles, test pits, and trenches 30 to 150 cm deep, depending on the soil depth, on different plots by the envelope method (on a 10-m grid). The envelope method involves the selection of a leveled square area on the test plot. Soil profiles were established in the corners of the square and in its center. For the