THE EFFECT OF PARTICLE SIZE COMPOSITION AND STABILITY OF SOIL AGGREGATES ON ^{137}CS VERTICAL MIGRATION IN SANDY SOILS WITHIN THE 50-KM CHERNOBYL NPP ZONE.

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Abstract
The results of long-term investigation of ^{137}Cs vertical migration in sandy soils within the 50-km zone around Chernobyl NPP have been compared with data on radiocaesium distribution among water-stable aggregates and particles of various size in the studied soils. To evaluate quantitative distinctions of ^{137}Cs vertical migration in the examined soils, it was significant to take into account the portion of ^{137}Cs migration just below the first 10 centimeters of topsoil ranging from 1 to 10% depending on soil type. On the basis of particle size analysis and aggregate soil composition, the size of soil components with vertical migration potential and the amount of ^{137}Cs potentially tending to migrate with the soil components along soil profile have been assessed. The following correlations were found for the tested soils: a decrease in the content of organic matter and highly dispersed soil clay component results in an increase in the ratio of radiocaesium associated with clay fraction, with simultaneous reduction of soil aggregate stability. Both factors promote ^{137}Cs vertical migration with fine, non-aggregated and prone-to-vertical-transport soil particles. Based on findings showing a ^{137}Cs partitioning among water-stable soil aggregates of diverse size and pattern of the radionuclide vertical distribution in the first 10 centimeters of topsoil, it was assumed that the so-called slow constituent of ^{137}Cs vertical migration (in terms of the quasidiffusion description of ^{137}Cs profile in soil) could not be explained by self-motion of soil aggregates and particles with associated radiocaesium. A hypothesis of root intermixing as principal mechanism responsible for ^{137}Cs vertical transport in the topsoil was postulated.

Introduction
In studies on radionuclide migration it is essential to elucidate qualitative factors controlling the phenomenon, find out what soil processes affect radionuclide translocation in soil profile and which of them play a key role. Such research is vital for further quantitative description of the process to forecast radionuclide behaviour in the course of time.

Radiocaesium was chosen for further studies since this radionuclide is soil-bound and its migration is determined by migration of soil matter allowing to use it as a tracer in monitoring soil processes (1, 2).
It was reported (3) that the portion of Chernobyl radiocaesium in exchangeable form tended to rise with increase in site radius off the nuclear power plant because precipitation-derived contribution into fall-out was growing. Yet, irrespective of fall-out origin, the bulk of radiocaesium is retained in the upper soil layer for a long period, building up dose loads (4). Even 40 years after nuclear bombing of Japan, behaviour of weapon-derived radiocaesium in soil would not change, similar to behaviour of Chernobyl radiocaesium.

So far several models have been developed to describe migration of radiocaesium in soil (5, 6). The defect of many migration models is analysis of radionuclide transfer in liquid phase only (7). The authors point out necessity to take into account migration of radionuclides due to bioturbation. It is claimed (1) that main factors responsible for distribution of radionuclides in soil after fall-out are diffusion and convection processes. Relocation of high radionuclide amounts is observed namely in the area surrounding plant root system (8).

Earlier the studies were carried out to investigate distribution of radiocaesium in different particle size fractions and water-stable aggregates of diverse soil types (9). Study on partitioning of radiocaesium in water-stable aggregates and particles of different size in mineral soils demonstrated that 100 to

250 μm fraction constituted the bulk of tested soils (from 50 to 70%). Nevertheless, only minor part of radiocaesium (7 to 9%) is associated with this fraction. Principal carriers of radioactive fallout in soil are particles under 10 μm collecting 70 to 80% of total radionuclide. Percentage of these particles in examined mineral soils is relatively small ranging from 2 to 5%. The tiny fractions show the highest specific activity. In natural environment fine particles with adsorbed radiocaesium are components of water-stable soil aggregates. Aggregate complexes may be defined as the basis of soil structural pattern.

We made an attempt in this investigation to find out the impact of distinctions in soil grain size composition, soil aggregate stability and differences in radiocaesium soil particle distribution on vertical migration of the radionuclide. Such goal required absolute ruling out of other factors affecting caesium behaviour.

Methods

Mineral soils with similar characteristics differing in such parameters as grain size composition and aggregate stability were chosen as objects for further studies.

To study vertical migration, soil samples were taken from at least 5 sites, each located several meters one from another at the same plot where sampling had been performed to evaluate distribution of radiocaesium in soil aggregates and particles of different size. The following sampling procedure was used: the 10 centimeter samples were collected in the dug vertical soil profile starting from the lowest layers with minimal activity upwards. Sampling was carried out down to 50 cm depth. The samples were air-dried during several days at room temperature and homogenized. Radiocaesium in soils was measured by gamma-spectrometry, using “Ortec” spectrometer with high-pure germanium detector.

Aggregate structure was quantitatively evaluated by Kachinsky method. According to Kachinsky, structural factor (Fstr) characterizing stability of soil aggregates is defined as the ratio of under 1 mkm size fraction in aggregate analysis to that in grain size analysis. The higher the value of this factor, the more vulnerable is the soil texture.

Since specific activity of soil fraction depends on initial radionuclide level in soil, this parameter should not be applied in comparative soil studies. Radionuclide percentage in soil fraction is related to the ratio of the fraction in soil. To assess enrichment of soil fractions with radionuclides, radionuclide concentration coefficient was employed (Cc). Concentration coefficient is defined as the ratio of specific activity of soil fraction to specific activity of intact soil. This value demonstrates relative superiority of specific activity in soil fraction over specific activity of original soil.

Results

The soils were numbered in the order reflecting increase of fraction < 10 μm (Table 1).

Table 1.

Mineral soil types investigated

Soil number	Soil classification (FAO1990/Belarus)	^{241}Am/^{137}Cs ratio (for top 10cm layer)	Particle size fraction < 10μ content, %
N 1 (“Lomachi”)	Dystric podzoluvisols / Soddy-podzolic sandy soils (for all)	1 : 800	1.8
N 2 (“Lesok”)		1 : 300	3.5
N 3 (“Kruki”)		1 : 1000	4.5
N 4 (“Masany”)		1 : 100	4.6

Mineral soils N3 and N4 bear close resemblance in main properties, with the only exception of the fallout source. Specimen N3 was dominated by precipitated radionuclide while specimen N4 contained fuel derived radioactive fallout. Despite the difference in the origin of fallout, we found no correlation between this factor and movement of radiocaesium in the soil profile. Further on this parameter was not taken into account.

Main chemical properties of tested soils are illustrated in the table 2.

Table 2.

Basic soil properties

Soil	pH (aq.)	pH (1M KCl)	CEC, meq/100g	Total organic matter content, %
N 1	4.7	4.1	3.1	0.77
N 2	4.8	4.0	6.7	2.1
N 3	4.8	3.9	4.5	1.5
N 4	4.8	4.0	4.9	1.7

The data indicate close chemical proximity of samples. Elevated value of cation exchange capacity for soil N2 may be interpreted by higher ratio of organic matter.

Based on findings showing a ^{137}Cs partitioning among water-stable soil aggregates of diverse size and pattern of the radionuclide vertical distribution in the first 10 centimeters of topsoil, it was assumed that the so-called slow constituent of ^{137}Cs vertical migration (in terms of the quasidiffusion description of ^{137}Cs profile in soil) could not be explained by self-motion of soil aggregates and particles with associated radiocaesium. A hypothesis of root intermixing as principal mechanism responsible for ^{137}Cs vertical transport in the topsoil was postulated.

The results of investigation of ^{137}Cs vertical migration in sandy soils (table 3) unexpectedly demonstrated relatively high reproducibility of caesium levels in 10 cm soil samples. A portion of radiocaesium translocated below top 10 cm soil layer was assumed as a quantitative measure of vertical migration.

Table 3.

Radiocaesium portion transferred below top 10cm soil layer

Soil	N 1	N 2	N 3	N 4
Radiocaesium amount, %	9.6±1.5	4.5±1.5	0.8±0.5	0.4±0.2

Interpreting variations in migration behaviour of radiocaesium in studied soils by different distance from Chernobyl Nuclear Power Plant is not grounded. Ten year record of research focused on vertical migration of radiocaesium in numerous sampling sites unequivocally refute this statement.

Fig. 1 represents correlation between the ratio of < 10µm fraction in the soils and percentage of radiocaesium removed beyond 10cm top soil layer.

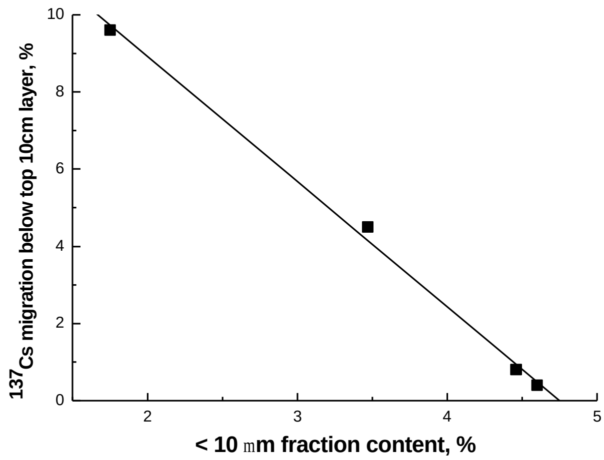

Fig. 1. Correlation between the ratio of < 10µm fraction in the soils and percentage of radiocaesium removed beyond 10cm top soil layer.

The results shows very good correlation between ratio of the grain size fraction < 10 μm and the amounts of radiocaesium migrated below top 10 cm layer for the mineral soils investigated. It may be seen that increased ratio of fine-sized fraction results in decline of caesium vertical migration caused by enhanced stability of soil aggregates. Kachinsky structural coefficient for soil N1 is 26 and that corresponding value for soil N3 is 15 that is stability of soil aggregates in soil N1 is twice lower.

Fig. 2 demonstrate that the radiocaesium concentration coefficient for <1μm fraction increases when the ratio of this fraction decreases in studied soils.

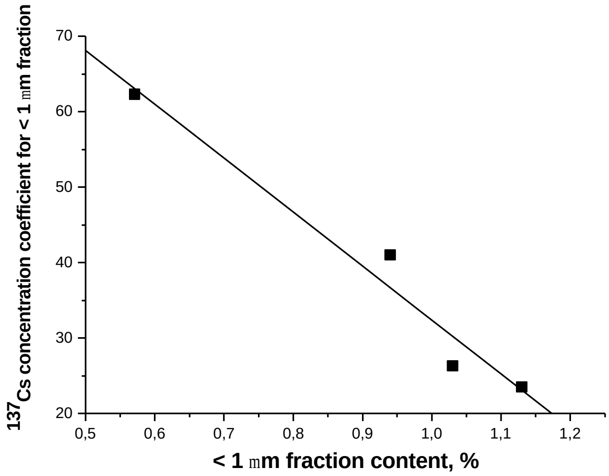

Fig. 2. Correlation between radiocaesium concentration coefficient for <1μm fraction and the ratio of this fraction.

It may be seen that correlation between radiocaesium concentration coefficient for <1μm fraction and the ratio of this fraction is not so evident.

Fig. 3 shows that radiocaesium concentration coefficient for < 10μm fraction and percentage of the radionuclide migration below top 10cm layer are freely associated.

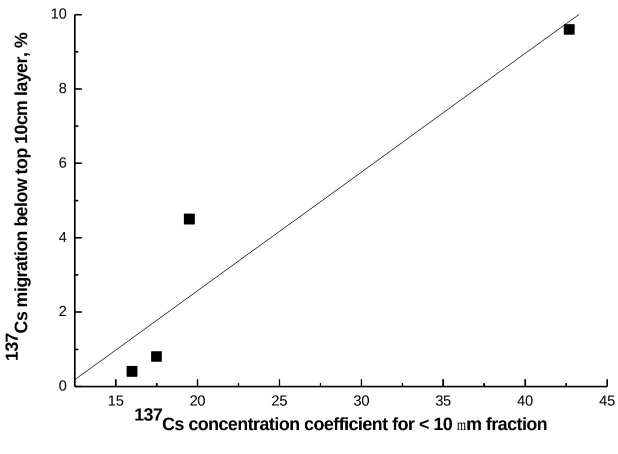

Fig. 3. Correlation between radiocaesium concentration coefficient for < 10μm fraction and percentage of the radionuclide migration below top 10cm layer.

Thus, result of our investigations testify to existing relationship between such parameters of studied mineral soils as the ratio of fine size fraction, distribution of radiocaesium in soil particle size fractions and vertical migration of the radionuclide.

In our opinion, more detailed correlations could be deduced only in laboratory model experiments where impact of adverse factors on vertical migration of radionuclides in soil profile may be minimized (or controlled).

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