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Studies of Radiocaesium Migration in Soils with High Organic Matter Content

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Abstract

This paper encloses results of field tests carried out on two types of organic soils. Activities of caesium, originating from global fallout as well as from the Chernobyl power plant breakdown, were measured for these two soils. Radioactive caesium is present to a depth of 40 and 70 cm, in black soil and in peat, respectively, although the highest activity was found in the topmost layers. Caesium originating from the Chernobyl accident constitutes about 70% of the total activity in the 0-10 cm layers of the soils. The vertical migration rates of caesium from both sources were also measured. In the case of peat soil the migration rate of the Chernobyl caesium as well as that from global fallout were found to be similar and amounted to 0.2 cm/year. In the case of black soil the differences were more pronounced (0.34 and 0.14 cm/year, respectively).

Results of the physicochemical analysis of both tested soils are also included in this work.

Keywords: caesium, radioactivity, soil, migration, Chernobyl fallout.

Introduction

Caesium introduced into soil with a high content of organic matter behaves in a different way than in soil rich with mineral components (sand, loam, etc.) [1]. An exact mechanism and factors that cause the retention of caesium and result in its vertical migration have not yet been fully understood. However, knowledge of these processes is important for several reasons. The amount of ^{137}Cs and its chemical form in which it occurs in soil, especially in the layer containing plant roots, and the change of its radioactivity with time are essential for determining caesium concentration assimilated by plants. Secondly, the vertical migration rate is related directly to the migration time of caesium from soil to groundwater. Thirdly, the contribution of ^{137}Cs to the external dose of radiation received by humans and its change with time are also strongly related to the vertical migration rate.

In a previous work the authors discussed the behaviour of caesium in several types of soils, namely: in silty brown soil, loamy lessive soil underlined by silty and sandy deposits, and in sandy podzolic soil [2]. In the present paper tests were carried out on two organic soils: low peat-muck soil and mucky black earth.

Materials and Methods

Soils

The two soil profiles were taken in April 1995, ten years after the Chernobyl nuclear power plant breakdown. Sampling spots of both profiles were localized in the Leczynsko-Wlodawskie lake district in the central-eastern part of Poland. The low peat-muck soil was sampled from a widespread barren while the black earth samples were taken from a wet meadow overgrown with a few bushes in the Sumin lake vicinity. Both sampling stations were overgrown with short grass and the sampling areas have not been farmed for over 40 years.

Sampling Procedure

20 × 30 cm blocks were cut out from soil. The sampling procedure involved removal of consecutive soil layers, 5-10 cm thick. The thickness of the soil horizons was chosen arbitrarily and did not correspond with the distribution of the morphological strata. The horizons were separated from one

another by using specially designed, 20 × 30 cm scoop, sharpened on one side. After sampling, the soil horizons were kept in polyethylene bags and then taken to our lab. All overground parts of plants were removed from the soil surface just before sampling. Profile depth was limited by the groundwater level and amounted to 70 cm for the low peat-muck soil and up to 35 cm in black mucky soil.

Soil Properties

The soil profile samples were dried out at 95°C. In order to remove any stones and plant parts rarely present in the samples, the black soil samples were sieved on a 1 mm opening screen according to a procedure recommended by IAEA [3]. The low peat-muck soil samples were first pulverized in a Fritsch pulverizer and then screened on a 1 mm opening screen. Plant roots were removed manually from the 0-5 and 5-10 soil horizons before pulverization.

In order to determine the physicochemical properties of the soils the following tests were performed: pH was measured using a N-510 Mera-Elwro pH-meter with a ERH-110 combined electrode, organic substance content was assessed with the roasting method [4], and organic carbon content was determined by Tiurin's method modified by Simakow [4]. The analytical procedure recommends the determination of $C_{org.}$ instead of the percent content of organic matter, especially for soils with organic matter content below 20%. Exchangeable cations were analyzed in a buffered 1N ammonium acetate solution (pH=7 in case of the peat-muck soil, pH=8 for the mucky black one) according to Schollenberg's method [4], exchangeable hydrogen was assessed with Kappen's method [4]. Analysis of granulometric composition was performed only on the black soil using Bouyoucos's method modified by Casagrand and Proszynski [4]. This type of analysis is not performed on peat soils. The experimental results are presented in Table 1.

Gamma-ray Measurements

The soil profile samples were weighed and then placed in 0.5 dm^3 Marinelli beakers. Measurements of caesium activity were carried out using a Silena gamma ray spectrometer with a 4096-channel amplitude analyzer coupled with a germanium detector produced by Princetton GammaTech. Detector resolution was 1.7 keV for the 1.33 MeV peak and its efficiency was 15%. Quantitative analysis was made with the use of a SIMCAS ver. 4.11 computer program. The spectrometric measurements were conducted for 20 hours for each sample.

In order to eliminate any density and thickness differences between particular layers, activities of ^{137}Cs and ^{134}Cs were expressed in $Bq/m^2/cm$. The radiocaesium activity expressed in such a way was calculated from the following formula:

$$A_T = A \cdot m / s \cdot l$$

where A_T – activity in $[Bq/m^2/cm]$, A – activity in [Bq/kg], m – total mass of a layer [kg], s – cross-sectional area $[m^2]$, l – thickness of a layer [cm] [2]. The percent content of ^{137}Cs originating from the Chernobyl accident was calculated based on the activity of ^{134}Cs. The sampling stations are localized in an area where no potential sources of caesium exist. Hence, caesium present in soil comes either from nuclear test explosions or from the damaged reactor in Chernobyl. Because of the short half-life of ^{134}Cs (2.06 y [5]), it can be assumed that ^{134}Cs in soil comes from Chernobyl only. At the moment of a nuclear explosion a ^{134}Cs /^{137}Cs ratio is 0.528 [6]. Taking into account the law of radioactive decay, this ratio at the time of sampling, 9 years after the accident, was 0.03148. Using this value the activities of ^{137}Cs liberated from Chernobyl were simply calculated as follows:

$$A_{Cs\text{-}137} = A_{Cs\text{-}134} / 0.03148 \quad [7].$$

Table 1. Chemical and physical properties of the examined soil profiles.

Profile Depth [cm]	Yields of Size Fractions [%]				pH (KCl)	C_{org}	OM %	Exchangeable Cations [meq/100 g of soil]						Base Saturation
	1–0.1 mm	0.1–0.02 mm	< 0.02 mm	< 0.002 mm				Ca^{++}	Mg^{++}	K^+	Na^+	Hh	Sorption Capacity	
low peat-muck soil														
0– 5	–	–	–	–	4.6	–	88	87	2.9	0.8	0.5	85	176	58
5–10	–	–	–	–	5.4	–	88	128	0.7	0.5	0.3	33	163	79
10–15	–	–	–	–	5.7	–	77	167	0.6	0.5	0.2	25	194	87
15–20	–	–	–	–	5.7	–	83	140	0.5	0.3	0.2	24	165	85
20–30	–	–	–	–	5.7	–	87	176	0.4	0.4	0.2	24	201	88
30–40	–	–	–	–	5.6	–	99	102	0.3	0.3	0.2	27	130	79
40–50	–	–	–	–	5.4	–	97	128	0.6	0.2	0.2	31	161	80
50–60	–	–	–	–	5.4	–	74	107	0.7	0.4	0.1	30	138	78
60–70	–	–	–	–	5.4	–	90	140	1.1	0.1	0.2	30	172	82
mucky black earth														
0– 5	–	–	–	–	6.9	–	38	67	1.1	0.5	0.3	8.4	78	89
5–10	–	–	–	–	7.0	–	27	61	0.5	0.2	0.2	6.6	69	90
10–15	–	–	–	–	6.3	–	36	55	0.5	0.1	0.2	7.5	64	99
15–20	63	33	4	2	6.4	7.3	–	32	0.3	0.04	0.1	4.8	38	98
20–25	–	–	–	–	6.7	–	43	18	0.3	0.04	0.1	4.1	22	81
25–30	72	19	9	4	6.7	2.0	–	18	0.3	0.03	0.1	2.4	21	88
30–35	76	14	10	4	6.7	1.3	–	14	0.3	0.03	0.1	1.8	16	88

Table 2. Activities of ^{137}Cs and ^{134}Cs in particular soil layers.

Profile Depth [cm]	^{137}Cs Total activity	^{134}Cs Chernobyl fallout	^{137}Cs Chernobyl fallout	^{137}Cs Global fallout	$\%^{137}Cs$ Chernobyl fallout
	[Bq/m^{2}cm]				
Mucky black earth					
0– 5	508.8	10.62	337.4	171.4	66
5–10	160.1	2.69	85.42	74.68	53
10–15	30.5	–	–	30.5	–
15–20	16.0	–	–	16.0	–
20–25	6.9	–	–	6.9	–
25–30	2.3	–	–	2.3	–
30–40	1.4	–	–	1.4	–
Low peat-muck soil					
0– 5	2050.2	50.5	1604.2	446.0	78
5–10	185.7	3.76	119.6	61.1	64
10–15	111.1	–	–	111.1	–
15–20	156.6	–	–	156.6	–
20–30	49.5	–	–	49.5	–
30–40	10.2	–	–	10.2	–
40–50	8.7	–	–	8.7	–
50–60	5.1	–	–	5.1	–
60–70	4.6	–	–	4.6	–

Obtained results are summarized in Table 2.

Values of average and maximum vertical migration rates of caesium were calculted as follows:

$$V_{mean} = d_{1/2}/t \qquad \text{and} \qquad V_{max.} = d_{max.}/t$$

where: $d_{1/2}$ – thickness of the top soil layer containing half of the total activity of caesium from Chernobyl or global fallout in soil profile [cm], $d_{max.}$ – maximum radiocaesium detection range [cm], t – time of migration.

The total activity of caesium was calculated by summing up the activity values of all measured layers taking into account their thickness.

Results and Discussion

The performed tests took place 9 years after the Chernobyl accident. After such a long time 95.16% of ^{134}Cs had already decayed. Its low activity and slow vertical migration in the deeper soil layers (below 10 cm) make the determination of the exact percent contribution of the Chernobyl caesium almost impossible. Since caesium tends to cumulate in the upper soil layers (0-10 cm) the experimental error of the spectrometric measurements is not large (about 5%). Table 2 also presents total ^{137}Cs activities in particular layers of the soil profiles, the percent contribution of the Chernobyl caesium and the activities of ^{134}Cs. The results indicate that prevailing activity of Cs comes from the topmost horizon (0-10 cm). In the peat-muck soil this contribution is 85%, while in the black mucky soil the 0-10 cm layer contains 92% of total caesium activity.

Values of the vertical migration rate of caesium in the tested profiles are presented in Table 3. In case of the black soil containing 60-70% of organic matter, Chernobyl caesium migrates substantially faster than caesium originating from global fallout. The migration rates are 0.34 cm/y and 0.14 cm/y for the Chernobyl and the global fallout respectively. One may suppose that such a large difference between the migration rates results from a high content of mineral substances. Similar trends were observed in earlier studies carried out on a typical mineral soil, i.e. silty brown soil, in which C_{org} was about 1% [2]. Loamy minerals reveal high sorption capacities of monovalent ions, i.e. Cs^+, Rb^+, K^+, Na^+ ($Cs^+ > Rb^+ > K^+ > Na^+$) [8], that are bonded in a less or more reversible manner depending on type and composition of the minerals. Loamy minerals exhibit a stratified structure and ions can be bonded between the strata. The immobilization process of ions inside a mineral occurs as a result of slow diffusion and it is probably responsible for the difference in the migration rates of the Chernobyl and the global caesium [9, 10]. For the dozen or so years that have passed since global fallout, ^{137}Cs originating from test nuclear explosions has probably been bonded permanently, to some extent, in the interlayer spaces of the loamy minerals. In Chernobyl caesium, most of this isotope is weakly bonded and therefore still able to move freely through soil horizons.

We did not observe this type of behaviour in the peat-muck soil in which the organic matter content is about 90%. The average vertical migration rates of caesium originating from Chernobyl and from global fallout are 0.29 and 0.2 cm/y, respectively. The retention of caesium in soil, (at very low content of mineral substances), has to be caused by other processes that do not lead to differences in the caesium migration rates originating from different sources. It can be inferred that caesium in peat soil occurs at present in similar chemical forms. Dialysis tests indicate that the interaction of caesium with humic acids does not result in the formation of permanent and stable chemical associations [11]. The extraction of humic acids with an NaOH solution from a soil containing large amounts of radiocaesium also confirms the lack of strong interactions. The extracted fraction of humic acids contained very small quantities of radiocaesium. Our field experiments indicate that a substantial amount of caesium is present in the deeper layers of the peat soil reaching the groundwater level. This results from the weak interaction of caesium with organic matter. Such a phenomenon can be observed in peat soil where the maximum depth of the ceasium migration probably exceeds 70 cm. This trend was not found in soils with a low content of organic matter. In these types of soil, caesium can migrate up to about 30 cm [2]. High content of exchangeable cations, especially calcium, is possibly one of the factors that accelerate the migration of

Table 3. Vertical migration rates of radiocaesium in the examined organic soil profiles.

	[cm/y]	Mucky black earth	Low peat-muck soil
Global fallout	V_{mean}	0.14	0.20
	$V_{max.}$	> 1.25	> 2.18
Chernobyl accident	V_{mean}	0.34	0.29

caesium in the peat soil. This can be clearly seen from Table 1. The exchangeable cations compete with caesium in sorption on soil material. The competition results in an increase of the content of the easily migrating caesium in soil solution at the expense of the form strongly adsorbed on soil material.

Strong retention of caesium in the surface layer of the peat soil can be explained in terms of the effect of a small amount of mineral matter frequently present in this type of soil. As a result, such soil always contains organo-mineral compounds. Humates, by forming complexes with soil minerals, change their properties and therefore are capable of, to some extent, affecting the retention of caesium in soil [1]. Some authors suggest that organic matter itself is able to interact with caesium very strongly [12]. This conclusion, however, was not confirmed by our experiments.

In the case of a low mineral matter content, it is very likely that the recycling process of matter can affect the migration rates of caesium. The effect of the circulation process should be pronounced in the upper soil horizons, rich with plants and organic remains, resulting in an increase of caesium activity in the 0-5 cm layer of peat soil (Table 2) [13]. One of the important processes that may affect the retention of caesium is the ability of mycelial cells, commonly occurring in organic soils, to assimilate substantial quantities of this element [13, 14]. These processes may be of particular importance in tested peat soil. It was found in the physicochemical analysis that the surface horizon of this soil revealed the lowest pH value (4.6). As pH decreases, more and more caesium exists as free Cs^+ ions – a form that is easily assimilated by plant roots and mycelial cells [13].

Conclusions

These results confirm the fact that substantial amounts of caesium are present in the surface soil layer regardless of soil type. Although, generally, the mechanisms of caesium retention are different and depend on many factors. Stronger retention takes place in mineral soils with a low content of organic matter. This confirms the critical role of mineral fractions. In the absence of the mineral fraction, biological processes and the sorption of caesium by organic matter start to dominate as it is clearly observed in organic soils.

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