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NATURAL-RESTORATION OF RADIOPOLLUTED ECOSYSTEMS AND IMPACT ON HUMAN HEALTH

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ABSTRACT
The vertical migration velocity of radionuclides and the ability of soils components to immobilise them, as the most important parameters of natural-restoration, was studied. The Dose Equivalent Rate (DER) reduction of external γ-radiation was studied in order to assess its impact on human health. The vertical migration velocities of ^{137}Cs and ^{90}Sr in typical soils of contaminated regions in Ukraine (Chernobyl 30-km zone) and Belarus (Gomel region) have been evaluated annually during the last 8 or 10 years since the accident. In most of these soils the migration rate of ^{90}Sr was found to be higher than this of ^{137}Cs and ranged from 0.71 to 1.54 cm year^{-1} and 0 to 1.16 cm year^{-1} respectively. At present the main part of radionuclides is located in the upper 10 cm soil layer. The ability of the soil components to immobilise the radionuclides was also investigated from 1989 to 1994 and was found that approximately 57% of ^{137}Cs was converted in fixed forms. It is expected that this percentage will increase to 80% in the next years. Finally, we studied how the DER of γ-radiation, which changes with the migration of radionuclides in the soil, affects the human health. In comparison with 1986, when 100% of ^{137}Cs was distributed on the soil surface, a significant reduction of DER occurred in the studied areas and about ten years after the Chernobyl accident, it ranges from 17.5% to 45%, depending mainly on the level of initial contamination of soils and its migration velocity.

KEY WORDS: Radionuclides, radioactive pollution, natural rehabilitation, self-restoration, Dose Equivalent Rate reduction.

INTRODUCTION
Prior to recommending a large-scale application of any rehabilitation technique at radiopolluted ecosystems, it is important to know the medium- and long-term intensity of natural- or self-restoration for most of the affected territories. Three main ways express the process of self-restoration: 1) The natural radioactive decay; 2) the transfer of radionuclides out of natural ecosystems; and 3) the ability of some pedological components to fix-

ate the contaminants. The first way is a real decontamination process resulting in the removal from the biosphere of significant quantities of radionuclides. Indeed, during the last years the total activity of short-life-isotopes was decreased by a factor of some thousand and actually, in the Chernobyl area, the main contaminants (^{137}Cs and ^{90}Sr) are decreasing according to their half-life. The two other ways of self-restoration are closely connected with radionuclides solubility and migration (vertical or/and horizontal) in soils and are studied in this work. Moreover, it was evaluated how the DER of γ-radiation changes with the migration of radionuclides deep in different types of soil.

This paper illustrates the experience gained in the field of natural-restoration of radiopolluted ecosystems and the consequent reduction of external radiation, which can have an important impact on human health. This study was developed through the participation of our Laboratory in the Experimental Project Nr. 4 of the International Scientific Collaboration on the Consequences of the Chernobyl Accident, financed by the European Commission.

METHODS

The natural behaviour of ^{137}Cs and ^{90}Sr in typical soils of contaminated regions in Ukraine and Belarus was studied for 8 or 10 consequent years since the accident. Based on the concept that self-restoration of natural ecosystems is any removal of radionuclides from the active geochemical cycle of the affected areas due to the active migration and/or durable fixation into the soils, we considered three main lines of study. The first one was based on the evaluation of the intensity of radionuclides migration (vertical and horizontal). A study of redistribution of radionuclides in different soil types was conducted in Ukraine and Belarus. An important soil ablation may be occurring in sloping landscapes thus, they can be considered as a good topographical model, which may soon give indications of the long-term behaviour of radionuclides. Therefore, we studied the efficiency of self-restoration on four sloping sites with different incline, soil types, vegetation characteristics, density of contamination and form of radionuclides.

The second investigation line was based on the dynamics of distribution of ^{90}Sr and ^{137}Cs and their forms of occurrence in different types of soils and the concurrent changes of phytomass contamination.

The third line of our study was to evaluate how the DER of γ-radiation changes with the migration of radionuclides in the soil.

Therefore, one of the main aims of this study was to relate the modification of DER according to the ^{137}Cs penetration into the soil, as a result of the natural restoration of the affected territories.

A brief description of the methodology, in connection with the main steps of the above three lines, follows.

Vertical migration of radionuclides

Vertical migration of ^{137}Cs and ^{90}Sr was studied in soil profiles from non disturbed lands. The soil-samples were taken as below: Ten samples of 1 cm, up to 10 cm depth and one sample per 5 cm from 10 to 70-100 cm depth. ^{137}Cs and ^{90}Sr content of the samples was assayed in laboratory by gamma spectrometry and radiochemical analysis respectively (Arapis *et al*., 1997; Sadolko *et al*., 1995).

Migration of ^{137}Cs on slopes

The study of natural migration of radionuclides in sloping soils from the exclusion 30-km zone of Chernobyl (Ukraine) began systematically in September 1993. Soils from four "new" sampling areas were analysed and measured. These areas differ in slope declivity, in distance from Chernobyl NPP, in density and forms of deposition and in some bio-geochemical characteristics. In the present study previous data obtained in 1986-1993 from some "older" slopes were also used (Arapis *et al*., 1996).

Self-restoration was expressed by the Intensity (I) and Efficiency (E) of the process of ^{137}Cs migration, which were calculated by the simple equations:

$$E = \left(A_0 - A_T\right)/A_0 \quad (1)$$

$$I = dA/dt \quad (2)$$

where:

A is the measured radioactivity,

A_0 is the initial activity of radionuclides, and

A_T is the activity at the time of sampling.

The processes of physical decay of ^{137}Cs were also taken into account.

Occurrence form of radionuclides

In order to determine the form of radionuclides in soils, we investigated the site of Shepelichi, which is located at the first over-valley terrace of Prypiat river (Ukraine). The sampling point was located on a shallow-wavy and relatively dry terrace, where sandy soddy-podzolic soils are in lane on aged-alluvial sands. A humus layer of soils reaches 25-31 cm in depth. This site was ploughed before 1986.

A selective leaching of radioactive elements was achieved by consecutive treatment of soils using extractive solutions with different composition. For the extraction of the water-soluble forms distilled water was used; for the exchange forms 1M ammonium acetate solution; and for the acid-soluble forms 1M nitric acid solution. Leaching occurred at a ratio of solid to liquid phases equal to 1:5, without intensive mixing, at 20 ^{o}C temperature.

Measurements of EDR and of ^{137}Cs soil contamination *in-situ*

To verify the homogeneity of the surface soil contamination at the selected experimental areas, we used the Collimated Radiometer (CORAD) device (Chesnokov *et al*., 1997). This device measures in-situ the ^{137}Cs deposition and evaluates its penetration depth in the soil.

In brief, the "CORAD" device consists of a measuring head and a control unit. The measuring head is a collimated scintillated detector (NaI(Tl) crystal sized 50 x 50 mm) and a photomultiplier placed in a leaded shield. The control unit of "CORAD" is a portable 256-channel analyser with a preamplifier, a unit of high-voltage and of low-voltage supply, a battery, a spectrometric amplifier with programmed amplification, a 256-channel digital converter and a microprocessor (type 80C31). This portable version of "CORAD" weighs about 15 kg (the thickness of the lead shield of γ-ray detector is 30 mm) and carries out 150 measurements per day. The threshold of the device determination is 20 kBq m^{-2} and the measurement accuracy 20%. The exposition time of measurement did not exceed 10 minutes for ^{137}Cs contamination exceeding the device threshold. The measuring head of the device was placed at a height of 80 cm over the ground surface and measured gamma radiation from the area of two square meters. Except from the gamma-radiation measurement in each measuring point, we obtained also: 1) the quantity of ^{137}Cs deposition on soils, 2) the thickness of the soil layer containing more than 80% of the total deposition; and 3) the thickness of the clean soil layer covering the above content of radiopollution.

Table 1. Mean vertical velocity (cm year^{-1}) of ^{137}Cs in different types of soil and ecosystem of Chernobyl area, between 1987 and 1994.

Soil	Ecosystem	^{137}Cs vertical velocity	
		Average	Stand. Dev.
Soddy-semi-podzolic-sandy	pine forest	0.116	0.052
Soddy-podzolic dusty-sandy	pine, oak pine forest	0.117	0.097
Soddy-podzolic sandy, dusty-sandy	long-fallow land	0.226	0.090
Peat-gley, peat-bog	alder marshy forest	0.267	0.102
Primitive alluvial sandy	pioneer phytocoenosis	0.444	0.104
Alluvial semi-podzolic semi-gley sandy	meadow	0.633	0.215
Soddy-podzolic-gley dusty sandy	meadow drained	0.274	0.089
Soddy-podzolic-gley loamy-sandy	meadow non-drained	0.357	0.183
Alluvial gley loamy	meadow non-drained	0.471	0.112
Soddy-gley	Meadow	0.497	0.153
Peat-bog		0.659	0.317

Notes: * = error of definition is not more than 10%
( ) = data are obtained on the base of 2-3 profiles

RESULTS

Vertical migration of radionuclides

The results of the vertical distribution of ^{137}Cs in soils profiles from the Chernobyl area are presented in Table 1, as the mean vertical velocity of ^{137}Cs (cm year^{-1}). These results are based on experimental data collected since 1987, i.e. one year after the accident, for 8 consecutive years.

Table 1 shows that the slower rate of the vertical migration of ^{137}Cs (0.1-0.2 cm year^{-1}) was observed in areas with soddy-podzolic-sandy, dusty-sandy and sandy-loamy soils of eluvial areas, covered by pine and oak-pine forests. Such slow rate is probably due to the presence of litter, where the main part of radionuclides reserve is located. This litter works as an organic barrier against the active migration of ^{137}Cs. Areas represented by long-fallow lands and soddy-podzolic semi-gley dusty-sandy and alluvial dusty-sandy soils were characterised by higher rate of vertical migration of radiocaesium (approx. 0.27 cm year^{-1}). The velocity of migration was higher (0.36-0.50 cm year^{-1}) for soddy-podzolic gley loamy-sandy areas. The primitive alluvial sandy soils of river beaches showed same velocity of vertical migration. The highest velocity, approximately 0.7 cm year^{-1}, was observed on areas with soddy gley and peat-bog eutrophic soils, as well as in alluvial ones. Finally, it is of interest to note that the vertical velocity of ^{137}Cs for soils of the same type was not constant from year to year. In fact, the difference of depth of ^{137}Cs-reserve's centre was 40% higher between 1989 and 1990, compared to the period between 1992 and 1993.

The vertical distribution of ^{137}Cs in Belarus was studied in the topsoil of Khoiniki (Gomel region), which is characteristic of the whole department of Polessye that was highly contaminated by the Chernobyl accident. The following dominant soil types were investigated during the 10 years that followed the accident:

1) Soddy-podzolic soils (sandy, sandy-loam).
2) Soddy-podzolic soils with redundant moisture.
3) Soddy-podzolic-gley soils.
4) Soddy-gley soils.
5) Peat-marsh soils of a lowland type, as well as alluvial ones.

Table 2 shows the vertical migration velocity of ^{137}Cs and of ^{90}Sr in the above different types of soil in Belarus. The migration velocity in soddy-podzolic automorphous soil (sandy, sandy-loam) did not differ from that in soils with redundant moisture. The average rate of migration in these soil types was approx. 0.48 cm year^{-1} for ^{137}Cs and 0.74 cm year^{-1} for ^{90}Sr. In soddy-podzolic-gley soils, the average migration of ^{137}Cs was 1.16 cm year^{-1} in soils with constant high moisture (the so-called "wet" soils), whereas in "drier" soils of this type the migration rate of ^{137}Cs was considerably lower (0.43 cm year^{-1}). Analogous observations were made concerning the migration of ^{90}Sr. In "wet" soddy-podzolic-gley soils the migration rate was about 1.30 cm year^{-1} and in "dry" soils 0.65 cm year^{-1}.

Table 2. Velocity of ^{137}Cs and ^{90}Sr vertical migration in different soil-types in Belarus (Gomel-region).

Soil	Velocity of radionuclide (cm year^{-1})		
		^{137}Cs	^{90}Sr
Soddy-podzolic sandy and sandy-loam (automorphous)	*Average*:	0.45	0.71
	St. Dev.:	± 0.24	± 0.13
Soddy-podzolic sandy and sandy -loam with moisture	*Average*:	0.50	0.76
	St. Dev.:	± 0.14	± 0.21
Soddy-podzolic-gley (wet)	*Average*:	1.16	1.30
	St. Dev.:	± 0.26	± 0.19
Soddy-podzolic-gley (dry)	*Average*:	0.43	0.65
	St. Dev.:	± 0.01	± 0.08
Soddy-gley	*Average*:	1.07	1.03
	St. Dev.:	± 0.39	± 0.11
Peat-marsh (non improved)	*Average*:	0.92	1.54
	St. Dev.:	± 0.26	± 0.72
Peat-marsh (improved)	*Average*:	0.39	0.52
	St. Dev.:	± 0.10	± 0.06

Table 3. Distribution of ^{137}Cs in soil profiles from a sloping site in Savichi (Chernobyl-30 km zone, 1994)

Sampling point on the slope	Upper soil-layer activity (A_{up}) Ci km^{-2}	Total activity (A_{tot}) Ci km^{-2}	A_{tot}/A_{up}
1 (upper)	28.7	32.1	1.12
2	24.0	24.0	1.00
3	35.7	40.5	1.13
4	29.6	35.6	1.20
5	25.7	31.0	1.21
6	16.6	24.1	1.45
7	18.4	26.0	1.41
8	25.3	29.1	1.15
9	19.4	24.4	1.26
10	24.8	31.2	1.26
11 (down)	14.6	20.9	1.43

In soddy-gley soil, the migration rates for both radionuclides did not seem to differ and were found to be approximately 1 cm year^{-1}.

Finally, the migration characteristics of radionuclides in the peat-marsh soil of the lowland type varied. The speed of ^{137}Cs transfer in improved peat bogs was approximately 0.4 cm year^{-1} and in non-improved ones about 0.9 cm year^{-1}. The migration rate of ^{90}Sr was found to be 0.52 and 1.54 cm year^{-1} for the improved and non improved peat-marsh soils respectively.

Migration of ^{137}Cs on slopes

During the 9 years that followed the accident, the efficiency of self-decontamination (eq. 1) for the studied areas ranged from 7.5-8% (for the site of

Table 4. Content (%) of mobile form of radionuclides in vertical profile of soil from the sampling site of Chystogalov (Chernobyl-30 km zone).

Layer	Form	^{144}Ce %	^{106}Ru %	^{134}Cs %	^{137}Cs %	^{90}Sr %
0-1	water-soluble	0.40	0.61	0.22	0.15	2.19
	exchangeable	1.39	–	4.5	3.86	6.25
1-2	water-soluble	–	1.09	0.18	0.13	3.23
	exchangeable	1.65	1.21	6.07	4.38	41.3
2-3	water-soluble	–	1.2	0.31	0.11	1.46
	exchangeable	2.8	3.6	5.0	5.56	27.7
3-4	water-soluble	–	1.79	0.31	0.14	1.73
	exchangeable	1.95	1.27	5.6	4.0	30.0
4-5	water-soluble	-	4.24	0.96	0.24	3.02
	exchangeable	3.05	2.42	12.5	9.04	53.5
5-6	water-soluble	–	5.71	–	0.29	1.67
	exchangeable	5.35	4.43	8.92	5.11	45.9
6-7	water-soluble	7.19	10.0	1.11	0.38	7.22
	exchangeable	5.16	13.3	8.22	5.64	58.3
8-9	water-soluble	23	18.9	7.80	4.67	1.19
	exchangeable	–	45.7	71.2	70.8	50.0
10-11	water-soluble	–	–	17.4	8.89	2.13
	exchangeable	75	–	17.8	16.0	87.0
12-14	water-soluble	–	–	–	6.30	28.9
	exchangeable	–	–	–	9.26	45.6

Table 5. Mobile forms of ^{137}Cs and ^{90}Sr in soddy-podzolic soil.

Year	Mobile forms in soil		
	^{137}Cs (%)	^{90}Sr (%)	$^{137}Cs/^{90}Sr$
1987	8	15	0.53
1988	7.2	24	0.3
1989	8.1	30	0.27
1991	4.9	37	0.13
1992	3.5	40	0.09
1993	-	-	-
1994	2.2	49	0.036

Kopachy, Chernobyl-30 km zone) to about 20% (for the site of Novoselki, Chernobyl-30 km zone), or from 1% to 2.5-3% per year. In all cases, a tendency of redistribution of ^{137}Cs on the slopes was observed. In 1994, the maximum activity had the tendency to be concentrated in the middle part of the slope. A typical example of the above described behaviour of ^{137}Cs from the experimental sloping site of Savichi (Chernobyl-30 km zone) is presented in Table 3.

Occurrence form of radionuclides

The distribution of the occurring forms of radionuclides in the soil profile from the 30-km zone of Chernobyl is presented in Table 4.
Finally, in the investigated soddy-podzolic soil, the content of ^{137}Cs and ^{90}Sr varied from 10 to 200 Bq kg^{-1}. The mobile forms contents (sum of water-soluble and of exchangeable forms) of ^{137}Cs and ^{90}Sr in soils and their ratio are presented in Table 5.

Influence of the migration of ^{137}Cs in soil on the DER reduction

As a result of the above long term observations, the vertical migration- velocities of ^{137}Cs were classified in five main groups according to the type of soil in which they have been measured, as follows:
I. <0.25 cm year^{-1}: Soddy-podzolic sandy non-processed.
II. 0.25-0.5 cm year^{-1}: Soddy-podzolic sandy-loam (automorphous) and meliorated peat-mars.
III. 0.5-0.7 cm year^{-1}: Soddy-podzolic with redundant moisture.
IV. 0.7-1.2 cm year^{-1}: Soddy-podzolic-gley and peat-marsh.
V. >1.2 cm year^{-1}: Soddy-gley.
It should be noted that the above values of vertical migration velocities were the average values for the ten year term in the region of Gomel-Belarus. The DER values (average, nSv h^{-1}) as a function of ^{137}Cs vertical migration for the above five groups and for nine different levels of contamination are presented in Table 6. These values show that about ten years after the accident, the reduction of the dose rate due to the vertical migration of ^{137}Cs, compared to the initial period when 100% of ^{137}Cs was distributed on soil surface, ranged between 3.5% in the soils where the migration speed was low (< 0.25 cm y^{-1}), and were less contaminated (185 kBq m^{-2}) and 37% in the soils where the migration speed was high (> 1.2 cm y^{-1}), and were very contaminated (7.4×10^3 kBq m^{-2}). Furthermore, when the physical decay of ^{137}Cs is taken into account, the range of the reduction becomes 17.5% and 45% respectively. To

Table 6. DER values calculated on the basis of the different ^{137}Cs migration velocities in five classified groups of soils (for real soil density) and for nine levels of contamination.

		Soil deposition of ^{137}Cs, kBq m^{-2}								
		185	370	555	740	925	1110	1850	3700	7400
Group of soil	Vertical velocity, cm y^{-1}	DER values, nSv h^{-1}								
	0 (Initial)	350	556	763	969	1176	1382	2209	4275	8408
I	<0.25	338	534	730	925	1121	1317	2100	4057	7971
II	0.25-0.5	327	511	695	880	1064	1248	1986	3829	7515
III	0.5-0.7	301	459	618	775	934	1092	1724	3306	6470
IV	0.7-1.2	276	410	544	678	812	946	1482	2822	5500
V	>1.2	271	399	527	656	784	912	1425	2708	5273

validate the theoretical computations, a comparison between measured in-situ and calculated DER was carried out. A good correlation was found between measured and calculated DER (Arapis *et al*., 1999).

CONCLUSIONS AND DISCUSSION

Related to the vertical migration and distribution of radionuclides into the soil profiles, the study showed that the main part (up to 90%) of radionuclides is located in the upper 10 cm layer. The rate of vertical migration essentially depends on the type of soil and the composition of the natural ecosystem. The rate of penetration of the ^{137}Cs-reserve is found to be within 0.1 and 0.15 cm per year for grass-podzol sandy and sandy loam soils covered by pine forests and 0.7 cm (or more) per year for grass-gley and peat-bog soils.

For the sloping areas, the intensity of self-restoration depends on the type of soils, the characteristics of the cover-vegetation and the inclination of the slopes. For soils of the same type, the factor of acceleration of self-restoration is estimated to range from 1.5 to 3, when the slope incline increases from 8-10^{o} to 30-40^{o} respectively. However, in future, the intensity and efficacy of self-restoration will decrease, due to the increasing long-term fixation of radionuclides by different soil components. Moreover, the damages of the surface soil layer, caused by erosion processes or removal of vegetation, will become of great importance.

Since the accident more than 57% of ^{137}Cs was converted in fixed forms and for the next years it is expected that this percentage will rise to 80%. The distribution of the activity of mobile forms of ^{137}Cs and ^{90}Sr in vertical sections of soil coincides with that of the activity of radionuclides' vertical migration. However, 94-97% of the total ^{137}Cs activity is still accumulated in the upper 5-10 cm soil layer. From 1987 till 1994, a steady increase of ^{90}Sr mobile form content in soils and a decrease of that of ^{137}Cs were observed. Such effects can be explained by the strong immobilisation of ^{137}Cs in soil. Dynamics of the mobile forms' ratio of ^{137}Cs/^{90}Sr reflect the rate of ^{137}Cs immobilisation. The time of transformation of 50% of mobile ^{137}Cs into immobilised form was evaluated to be 3-5 years for the soils of Chernobyl-30 km zone.

Finally, the data obtained regarding the external dose of gamma-irradiation show that about ten years after the accident in the studied territory, a significant reduction of DER occurs in natural conditions and it ranged - in comparison with 1986 when 100% of ^{137}Cs was distributed on soil surface - between 17.5% in the less contaminated soils with low ^{137}Cs migration velocity and 45% in the most contaminated soils with high ^{137}Cs migration velocity. Since it is a fact that DER is reduced with time, these calculations must be performed in all studies, as they are essential for the determination of the future radioecological status of the affected areas and their impact to the human health.

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