Vertical distribution of ^{90}Sr and ^{137}Cs in uncultivated soil in three localities of central Bohemia

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1. Introduction

^{90}Sr and ^{137}Cs represent the highly radiotoxic, long-lived and world-widely dispersed artificial radionuclides. They were mainly deposited in our environment in the 1960's as a result of global fallout from nuclear weapon tests and again in 1986 after the Chernobyl reactor accident. The radioactive contamination in the terrestrial environment due to atmospheric fallout varies with the geographic locations. The mobility of the radionuclides in the soil system depends not only on physicochemical properties of the radionuclides themselves but also on (a) climatic conditions, such as rainfall, temperature, humidity and biological activity of microorganisms in soil; (b) physicochemical properties of the soil, such as acidity, organic matter content and particle size; and (c) human activities, such as cultivation, irrigation and fertilization (Jia et al. 1999).

The purpose of this work was study the vertical distribution of ^{90}Sr and ^{137}Cs in uncultivated soils. The radionuclide contents in different soil layers and the cumulative deposition of radionuclides at the study sites were to be established with a view to possible identification of their sources.

The present paper investigated the vertical distribution of ^{90}Sr and ^{137}Cs in uncultivated soil samples in central Bohemia and inventory of these radionuclides at representative sampling sites. These data provide the information about the availability, movement and environmental behaviour of these important radionuclides in the study region.

2. Material and methods

2.1 Sites and soil properties

The samples were collected during the period 2003-2004 at three places in the central Bohemia. Sampling sites were mainly collected in areas believed to be mostly undisturbed for at least 45-50 years. Soil samples were taken at the foot of Oblík's hill (site 1) in the Louny district situated about 70 km north-west of Prague; at the village of Krusičany (site 2) in the district of Benešov situated about 25 km south of Prague; and at Kbely (site 3), which is a suburb in the northern part of Prague. Soil samples were collected from grass-covered areas. The mean annual precipitation at sites 1, 2 and 3 is 500, 600 and 490 mm. respectively. The mean annual temperature at these three sites is 9°C, 8-9°C and 9-10°C, respectively. In the WRBS and USDA classification systems the soil at site 1 is classified as Chernozems and Mollisols. According the same classification systems the soil at site 2 is classified as Cambisols and Inceptisols. In the FAO classification system the soil at site 3 is classified as Calgaro-calcic chernozem. Some physicochemical properties of the soil in the soil layers are given in Table 1a and Table 1b.

2.2 Sampling and sample preparation

A soil core was taken with an area of 30 cm x 30 cm and depth 50 cm. The core was subdivided into 8 sections. The first six sections were taken at 5 cm intervals to a depth of 30 cm while the other two had the intervals of 10 cm each down to 50 cm. Each section was prepared and analyzes separately. The air-dried soil samples were ground, sieved through a 1.0 mm screen for a removal of stones and fragments of plant roots, and carefully mixed; then the samples were dried to constant weight at 105°C for 24 h.

Table 1a Soil characterization

Location sample	Depth [cm]	Soil horizon	pH exch.	Clay <0.01mm [%]	Silt 0.01-0.05 mm [%]	Sand 0.05-2.0 mm [%]	Org.matter (SOM) [%]
Site 1 Oblík	0÷5	A1	5.09	44.9	36.4	18.7	11.0
	5÷30	A2	5.41	52.4	32.8	14.8	-
	30÷50	A3	5.74	57.5	37.1	5.4	7.0
Site 2 Krusičany	0÷5	A	4.98	21.4	22.4	56.2	2.5
	13÷20	Bv	4.42	13.7	13.3	73.0	-
	50÷60	BC	4.52	15.9	16.9	67.2	2.0
Site 3 Kbely	0÷50	A	7.10	35.1	41.3	23.6	3.52

Table 1b Chemical properties of soil samples

Location sample	Depth [cm]	C ox. [%]	T (CEC) [mmol+/100g]	H+ exchangeble [mmol+/100g]	K exchangeble [mmol+/100g]	Ca exchangeble [mmol+/100g]	Mg exchangeble [mmol+/100g]
Site 1 Oblík	0÷5	6.18	53.2	15.5	0.49	40.00	4.37
	5÷30	5.04	45.7	9.0	0.48	41.08	4.86
	30÷50	4.32	50.1	7.0	0.98	43.94	5.62
Average	**0÷50**	**5.18**	**49.7**	**10.5**	**0.65**	**41.67**	**4.95**
Site 2 Krusičany	0÷5	1.5	13.1	3.0	0.61	5.75	1.07
	13÷20	0.20	9.1	4.0	0.28	6.08	1.30
	50÷60	0.20	9.7	3.0	0.15	6.61	1.33
Average	**0÷60**	**0.63**	**10.6**	**3.3**	**0.35**	**6.15**	**1.23**
Site 3 Kbely	**0÷50**	**2.04**	**21.7**	**<0.5**	**0.70**	**22.3**	**1.83**

Table 2. International comparison test: ^{90}Sr measurement in mineral matrix

Organizer of comparison	Year	Matrix	Mark	Radionuclide	Activity (our results) [Bq/kg]	Activity (averrage of all participating laboratories) [Bq/kg]	Number of participating laboratories
IAEA. Vienna Austria	1998-1999	Mineral matrix	26A4	^{90}Sr	0.13±0.031	0.146±0.0019	Not mentioned
			26A5	^{90}Sr	0.17±0.031	0.197±0.0026	Not mentioned
			26A6	^{90}Sr	0.23±0.023	0.246±0.0032	Not mentioned
			26A7	^{90}Sr	5.32±0.38	5.37±0.070	Not mentioned
			26A9	^{90}Sr	18.26±1.06	19.46±0.26	Not mentioned

2.3 Method for ^{90}Sr and ^{137}Cs analysis in soil

^{90}Sr determination. For the analysis of ^{90}Sr 150 g sieved soil was used. ^{90}Sr in soil was determined using calcium oxalate method applied to soil (Volchok, 1984). Strontium from carbon free ash was leached using HCl and NaOH solutions. Repeated Ca oxalate precipitation, then co-precipitation with $Fe(OH)_3$ and $Ce(OH)_3$ were carried out. After secular radioactive equilibrium between ^{90}Sr and ^{90}Y was reached, the ^{90}Y was separated as $Y(OH)_3$ and transferred to oxalate before activity measurement, which was performed with a low background proportional alpha-beta counter IN20 (Eurisys, France). Chemical yields of ^{90}Sr and ^{90}Y were determined by gamma measurement of ^{85}Sr carrier and by titration method, respectively.

The procedure was checked by analyzing five certified samples supplied by IAEA (Analytical quality control service, IAEA Vienna, 2000) and the results given in Table 2 were in good agreement with recommended values.

^{137}Cs determination. For the analysis of ^{137}Cs was used approximately 500 g sieved soil. The soil samples were counted by gamma spectrometry before any radiochemical separation.

3. Results and discussion

3.1 Vertical distribution of ^{90}Sr and ^{137}Cs in soil

Table 3a presents the activity concentrations and the cumulative depositions of ^{90}Sr at all three sites. Similar results for activity concentrations and cumulative deposition of ^{137}Cs are presented in Table 3b.

Table 3a Vertical distributions of ^{90}Sr

	Locality	Site 1 - Oblík		Site 2 - Krusičany		Site 3 - Kbely	
	Depth [cm]	Activity	σ	Activity	σ	Activity	σ
Activity concentration ^{90}Sr [Bq/kg]	0÷5	4.29	0.17	3.15	0.49	2.08	0.10
	5÷10	3.95	0.21	2.21	0.31	2.41	0.43
	10÷15	2.11	0.18	2.23	0.39	1.80	0.06
	15÷20	0.81	0.08	2.07	0.08	1.72	0.05
	20÷25	0.40	0.07	1.79	0.25	1.76	0.32
	25÷30	0.19	0.09	1.04	0.47	1.43	0.16
	30÷40	0.13	0.05	0.43	0.19	1.01	0.12
	40÷50	0.10	0.07	0.20	0.11	0.38	0.03
Cumulative deposition [Bq/m^2]		700	60	1390	130	970	100

Table 3b Vertical distributions of ^{137}Cs

	Locality	Site 1 – Oblík	Site 2 - Krusičany	Site 3 - Kbely
	Depth [cm]	Activity	Activity	Activity
Activity concentration ^{137}Cs [Bq/kg]	0÷5	27.5	24.8	71.8
	5÷10	8.80	12.9	50.0
	10÷15	2.80	4.5	18.7
	15÷20	1.10	1.7	7.3
	20÷25	0.50	1.1	4.3
	25÷30	0.50	0.3	2.5
	30÷40	0.30	0.2	1.9
	40÷50	0.20	<0.3	0.9
Cumulative deposition [Bq/m^2]		260	510	10000

3.2 Discussion

From the data given in Tables 3a and 3b follows that sites 1, 2 and 3 had 36, 24 and 17% (i.e. about 28%) of activity of ^{90}Sr is present in the 0-5 cm layer, respectively. The percentage increases to about 65% activity at a depth of 0-15 cm. Sites 1, 2 and 3 had 66, 55 and 46% ^{137}Cs activity, respectively, in the 0-5 cm layer, and more than 92% activity can be detected in the 0-15 cm layer. However, at a depth of 30-50 cm were present 2, 13 and 11% activity of ^{90}Sr in these three sites, and less than 5% of ^{137}Cs activity was detected at all these sites. The largest affinity is seen for the surface layers, which suggests that surface deposited radionuclides will accumulate in these layers and not migrate to deeper horizons. That agree with data obtained from other studies which suggests that ^{90}Sr and ^{137}Cs are retained in the surface layer (0-30 cm) (Forsberg et al., 2000, Solecki, 2005). Qualitatively similar profiles were found at the other sites. Below that layer the activity concentrations decreased rapidly and were negligible for depths <30 cm.

The cumulative deposition of ^{90}Sr at sites 1, 2 and 3 were found of 700, 1390 and 970 Bq/m^2, respectively, The cumulative deposition of ^{137}Cs at sites 1, 2 and 3 were found of 260, 510 and 10000 Bq/m^2, respectively. The obtained values of ^{90}Sr are about 47% lower than the value of UNSCEAR inventories, but our average value of ^{137}Cs (3590 Bq/m^2) agree with the cumulative deposition of ^{137}Cs from the atmospheric nuclear tests in northern 40°-50° temperate zone according UNSCEAR inventories. The radioactive decay should have reduced the reported ^{90}Sr and ^{137}Cs inventories in the northern hemisphere to 2190 Bq/m^2 and 3580 Bg/m^2, respectively. In our earlier work we found the cumulative deposition of ^{137}Cs in value 5400 Bq/kg (Závěrečná zpráva IV SÚRO, 2005).

The role of soil organic matter (SOM), one of the characteristics of soil, in the distribution of these radionuclides has been studied (Kim et al., 1998). The concentration of ^{90}Sr and ^{137}Cs increased proportionally to the SOM content in soil, but the difference of the degree of correlation among ^{90}Sr and ^{137}Cs could be explained by the fact that SOM held ^{137}Cs more strongly than ^{90}Sr. ^{137}Cs showed very low mobility in soil profiles in undisturbed soils, therefore, it was concentrated in the upper 10 cm of the soils. Although the concentration of ^{90}Sr increased with organic content through an anion exchange reaction, which is enhanced by SOM keeping high cationic exchange capacity, the retention of Sr was relatively weak in contrast to Cs. It is related with high mobility of Sr that is a member of group II in the periodic table and acts like Ca. The fact that the peak ^{90}Sr concentration usually could be found below 10 cm of the soil profile is a proof, whereas some 85% of the ^{137}Cs was found within 10 cm from the surface soil layers. It is well known that calcium plays an important role in the adsorption process. For retention and migration of ^{90}Sr in the soil profile the surface layers soil tend to be most important (Jedináková-Křížová, 1998). All these properties of soil and our results in this work will be discussed.

Fig. 1

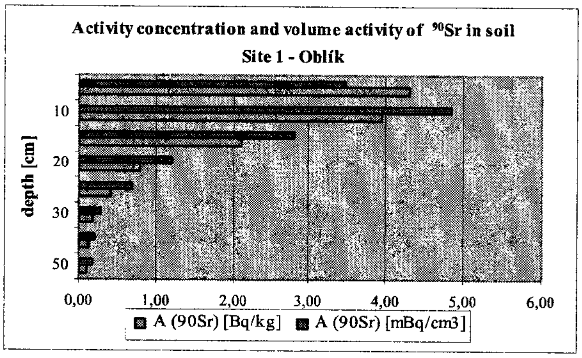


Fig. 2

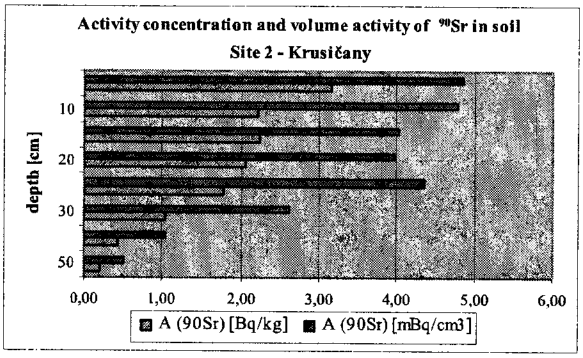


Fig. 3

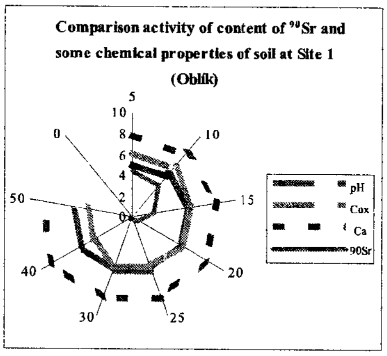


Fig. 4

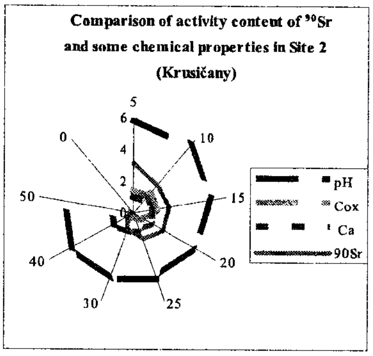
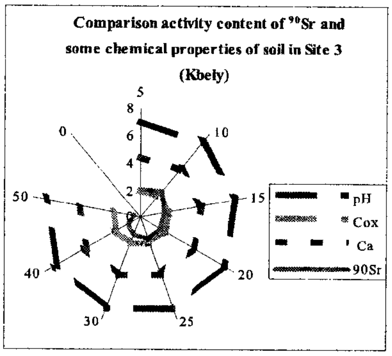


Fig. 5



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