Polish Journal of Environmental Studies Vol. 11, No. 2 (2002), 157-163

Studies on Horizontal and Vertical Migration of ^{90}Sr in Soil Systems

J. Solecki, S. Chibowski

Department of Chemistry and Colloid Chemistry, Faculty of Chemistry,
Maria Curie-Skłodowska University, 20-031 Lublin,
pi. M. Curie-Skłodowskiej 3, Poland

Received: October 8, 2001
Accepted: December 6, 2001

Abstract

Here are presented results of studies on vertical and horizontal migration of strontium ^{90}Sr isotope in soil systems of Bug river valleys. Mean radioactivity of strontium in these soils was 26.15 ± 22.2 Bq/kg. Atmospheric precipitation of strontium on this area was 0.35 kBq/m^2/cm. Radioactivity in soil profiles ranged from 17 to 130 Bq/m^2/cm and the isotope was detected even at 30 cm depth. Some tendency to vertical migration of strontium towards the river was noticed. Obtained results were compared with concentrations of calcium and strontium in the soil. Any clear influence of these elements on the migration of strontium ^{90}Sr was noticed.

Introduction

Nearly 99% of radioisotopes from atmosphere go to soil through precipitation. Adsorbed by soil radioactive elements may move with soil solutions horizontally and vertically or stay in root region of plants and then enter the food chain [1].

The process of vertical migration of radioactive fallout, downwards into soil is limited. Year velocity of migration ranges from a few to several cm and depends on the type of soil, its permeability, amount of rainfall, and type, as well as properties of the element. Among the many elements found in fallout the most active is ruthenium and the least is cesium. For this reason the greatest concentration of cesium is in surface 1 cm layer, two thirds of strontium is in 5 cm layer and ruthenium is in deeper layers. In samples of uncultivated soil most of these isotopes were found at 30 depth of the profile. Such behavior is probably caused by different sorption affinity of these elements to soil minerals. In samples collected from cultivated areas the concentration of radioactive isotopes does not depend on the depth of the profile. The whole radioactive contamination of the soil is closed within 30cm depth [2].

Correspondence to: Dr. J. Solecki

Strontium isotope ^{90}Sr is treated as one of the most dangerous products of nuclear fission for human beings. For this reason its properties and migration in the environment are widely studied [3-13]. This paper presents the results of studies on vertical and horizontal migration of strontium ^{90}Sr isotope in soil systems of the Bug River valleys.

Experimental

All measurements were done on soil samples collected from Bug River valleys. In ten sites (Fig 1.) surface soil samples (10 cm cube) were collected according to the scheme (Fig. 2, points B and C) and bottom sediments (point A) for studies of horizontal migration.

Surface soils were taken with 10 x 10 cm acid resistant steel, in each point seven samples (one in the middle and six on circumference of 2 m circle). From this point a bottom sediment was collected with Kajok's tube [14] in five to ten portions, 1-2 meters from the riverbank, depending on the bottom thickness. From sites numbered 1,2,3,4 and 10 soil profile samples were taken for vertical migration of the isotope. The procedure applied was as follows: A 100x50x50 cm pit was excavated, vegetation was removed and with special 30x20x5 cm shovel soil layers

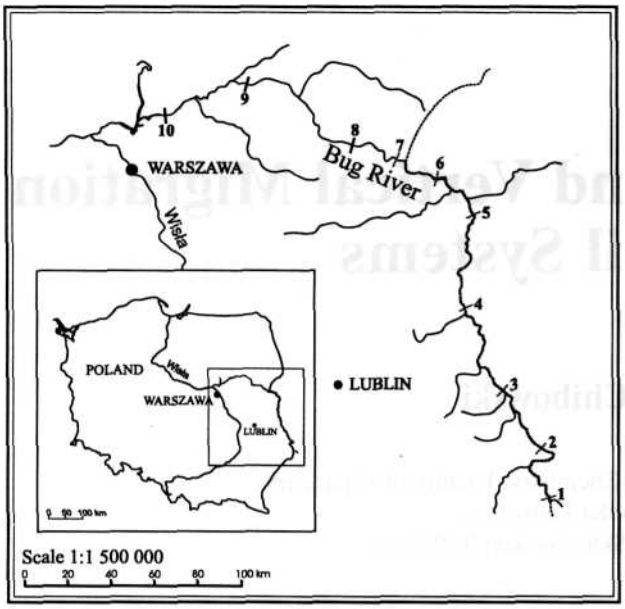


Fig. 1. Location of sampling points along Bug River.

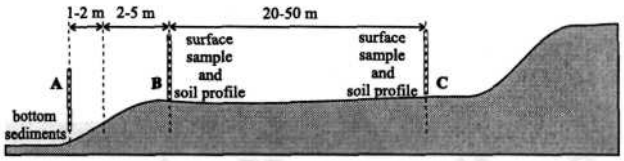


Fig. 2. Scheme of sediment and soil collection sites.

5 cm thin were collected down to 40 cm along one of the dig walls.

All samples were air dried in a laboratory, dried in 60°C, then ground and sieved through a 1 mm sieve. Averaging sample of 200 g was incinerated in 600°C to remove organic matter. Mineralization (ash percent) was calculated as a difference of mass before and after incineration. Each sample of the soil profile was weighted and these data were used for a calculation of isotope distribution.

Measurements of ^{90}Sr radioactivity were preceded by analytical procedure according to the scheme presented in Fig. 3 [13]. Extraction capacity of strontium from soil samples by concentrated HNO_3 was measured using the ED-XRF method.

It was assumed that isotopic exchange goes between stable strontium and its isotope ^{90}Sr in the soil. This process runs during migration of the strontium in the soil and while leaching the soil with concentrated HNO_3 (during two hours at 80°C). Previous experiments [7, 13], proved that the extraction process of strontium runs with various yields, depending on the type of soil sample, and must be controlled. For this purpose the activity of isotope ^{90}Sr was determined from activity of isotope ^{90}Y in yttrium oxalate (Fig. 3 in [13]) with Low Background Counting System by Silena. The appliance consists of two plastic detectors working in anticoincidence system, calibrated for ^{90}Sr source with surface beta emission with 4π geometry and activity 68.6Bq. The yield of β radiation counting was 27% with γ background equal to 2.6 cpm. To estimate efficiency of counting a surface calibration source (2u geometry) from the standard solution of ^{90}Sr delivered by Amersham was made. Its total activity was 428 Bq and allows estimated counting efficiency of the applied counter on 22-25% (for each measured sample) at the background 96-110 mcps. For all measurements of radioactivity determined MDA (Minimum Detectable Amount) values ranged 4.5-17 mcps and error was 4-10%. Each sample of yttrium oxalate was measured 10 times at 1000 s. Correctness of the analysis was verified by repeated measurements of every one sample, after two weeks, when short-lived ^{90}Y disappeared. Properly done analysis procedure gave activity of the sample at background level. Reliability of the above analysis was checked with sample Soil 6 from IAEA of mean radioactivity (^{90}Sr) 30.67 Bq/kg and permissible error 24.2-31.67 Bq/kg. The mean results from two 5 g samples was 27.5 ± 2.3 Bq/kg with strontium extraction yield 43 and 45% respectively.

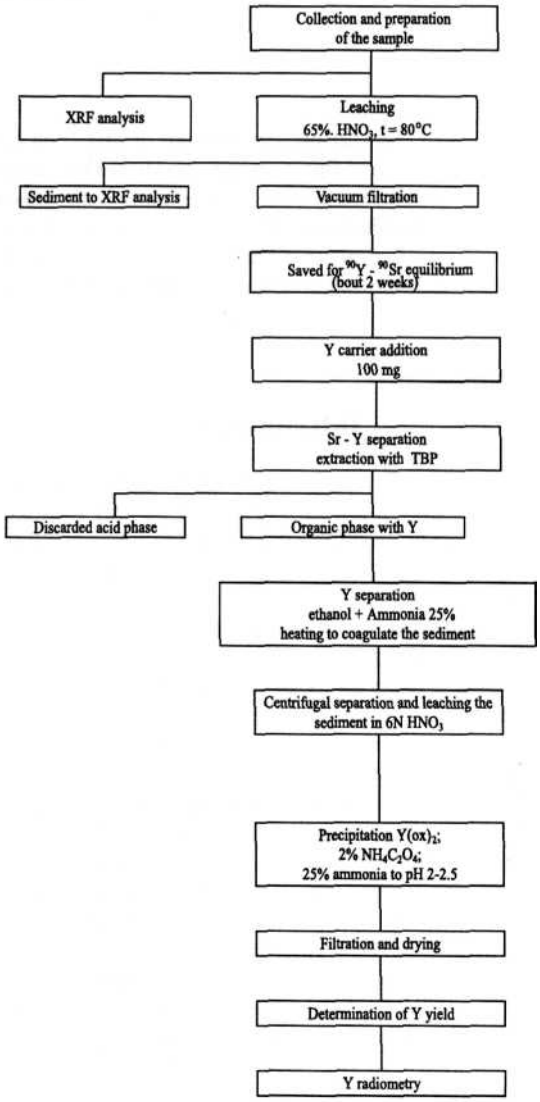


Fig. 3. Diagram for ^{90}Sr analysis in soil samples.

^{90}Sr radioactivity in studied soil samples was calculated according to the following formula:

$$C^{90}_{Sr} = \frac{(R - R_0) \cdot e^{\Delta t\lambda} \cdot 2}{E \cdot f \cdot W_E}$$

R - total counting rate of the sample [cps]
R_o - background counting rate [cps]
Δt - time between ^{90}Y separation and background measurement [h]
λ - decay coefficient of total ^{90}Y (equal 0.0108 h^{-1})
f - chemical yield of yttrium separation
E - counting efficiency
2 - geometry coefficient
W_E - extraction efficiency

The average scattering of the radioactivity measurements of ^{90}Sr ranged 1-5 Bq/kg. A distribution of radioactivity in subsequent layers of each examined profile was calculated according to the equation:

$$As = \frac{A \cdot m}{s \cdot l} \text{ [Bq/m}^2\text{/cm]}$$

Where:
A - radioactivity [Bq/kg]
m - total weight of profile sample [kg]
s - area of the profile (0.2x0.3 =0.06 [m^2])
1 - thickness of the profile layer [cm]

Results and Discussion

From The Geochemical Atlas of Poland and determined concentration of humus examined soils were classified as sandy and loamy soils. Radioactivity values of strontium ^{90}Sr in soil and bottom sediments are listed in Table 1. For analyzed points C, B and A measured radioactivity ranged from 5.51 to 69.78, from 6.77 to 85.3 and from 4.19 to 20.48 Bq/kg, respectively. These data demonstrate meaningful differences of the radioactivity measured in the samples collected in various sites along the river that was confirmed by calculated standard deviation of arithmetic means. The greatest concentration of the studied isotope was in point C, 20-50m away from the river bank (28.53 ± 21.40 Bq/kg), lower in point B 2-5 m away from the river bank (23.78 ± 23.00 Bq/kg), and the lowest in bottom sediments, point A (9.95 ± 5.13 Bq/kg). The diagram in Fig. 4 demonstrates measured radioactivity of discussed samples collected along the river. One should expect that run-off of surface water towards the river will also cause radioisotope transportation. That would be demonstrated by higher radioactivity of samples collected at the riverbank and in bottom sediments. Such behavior was observed in points 2, 4, 7 and 10. Additionally, some correlation between ^{90}Sr in soil and bottom sediments may be noticed. For higher concentration of the isotope in soil also the higher concentration in the respective sediment was measured for all samples except points 6, 7 and 9. A horizontal migration of ^{90}Sr in soil can be seen from the obtained data, although it has no general tendency. Besides the diffi-cult-to-define physicochemical properties of the soil system there are additional factors having influence on the migration of the isotopes:

- type of soil
- type of plants
- terrain slope
- precipitation
- agricultural activity
- composition of soil elements
- acidity of soil

All samples of soil were collected from the left bank of the Bug River. The slope of the terrain was mellow, covered with uniform vegetation. Mentioned factors as well as soil type and similar acidity values are not enough to formulate uniquivocal conclusions describing the phenomenon and mechanism of the horizontal migration of ^{90}Sr in studied systems.

Additionally, the concentration of ^{90}Sr was compared with concentrations of calcium and strontium obtained by XRF analysis of the collected samples. Above data are listed in Tables 2 and 3 and presented in Figures 5-8.

Table 1. Radioactivity of ^{90}Sr in soil samples and bottom sediments in Bq/kg (Bug river valley 1999).

No. sample	Activity ^{90}Sr in Bq/kg		
	C (soil)	B (soil)	A (sediment)
B1	15.51	15.87	8.57
B2	6.65	15.11	13.58
B3	21.63	16.03	12.57
B4	19.29	21.28	12.19
B5	54.3	36.39	20.48
B6	45.22	17.22	9.07
B7	69.78	85.33	4.85
B8	16.06	14.18	10.37
B9	31.53	6.77	3.65
B10	5.31	9.65	4.19
Arithmetic mean	28.53 ± 21.4	23.78 ± 23.00	9.95 ± 5.13

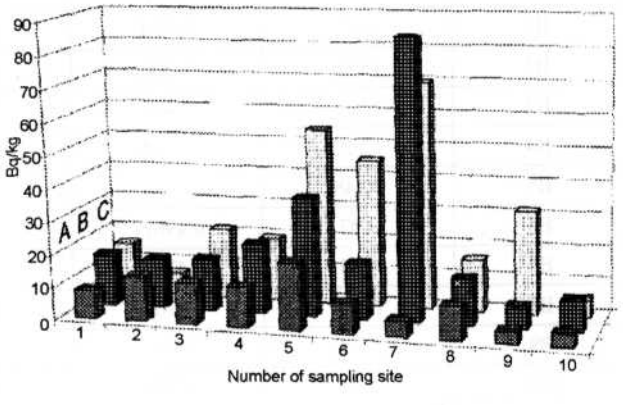


Fig. 4. Radioactivity of ^{90}Sr in bottom sediment and soil samples from Bug River valleys.

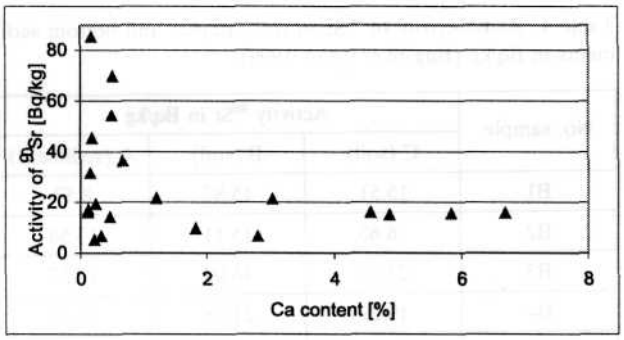


Fig. 5. Radioactivity of ^{90}Sr as a function of concentration of calcium in the soil samples.

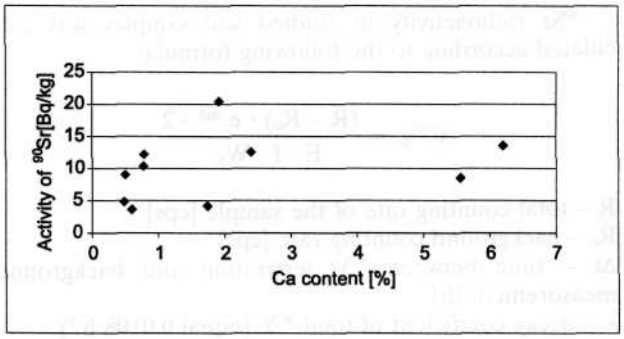


Fig. 6. Radioactivity of ^{90}Sr as a function of the concentration of calcium in the bottom sediment samples.

Table 2. Radioactivity of ^{90}Sr in examined soil samples and bottom sediments compared to total concentration of calcium in the samples (Bug River, 1999).

No. sample	A(sediment)		B(soil)		C(soil)	
	Activity of ^{90}Sr [Bq/kg]	Concentration of calcium [%]	Activity of ^{90}Sr [Bq/kg]	Concentration of calcium [%]	Activity of ^{90}Sr [Bq/kg]	Concentration of calcium [%]
B1	8.57	5.559	15.87	6.687	15.51	5.838
B2	13.58	6.206	15.11	4.862	6.65	2.802
B3	12.57	2.402	16.03	4.564	21.63	1.208
B4	12.19	0.7855	21.28	3.028	19.29	0.24543
B5	20.48	1.920	36.39	0.67177	54.3	0.49934
B6	9.07	0.49677	17.22	< 0.1269	45.22	0.18846
B7	4.85	0.47915	85.33	0.18056	69.78	0.5122
B8	10.37	0.77036	14.18	0.47054	16.06	< 0.12286
B9	0.65	0.59493	6.77	0.33008	31.53	0.15439
B10	4.19	1.738	9.65	1.829	5.31	0.22284

Table 3. Radioactivity of ^{90}Sr in examined soil samples and bottom sediments compared to total concentration of strontium samples (Bug River 1999).

No. sample	A(sediment)		B(soil)		C(soil)	
	Activity of ^{90}Sr [Bq/kg]	Total content of strontium [ppm]	Activity of ^{90}Sr [Bq/kg]	Total content of strontium [ppm]	Activity of ^{90}Sr [Bq/kg]	Total content of strontium [ppm]
B1	8.57	238.036	15.87	315.785	15.51	202.356
B2	13.58	263.627	15.11	226.224	6.65	120.655
B3	12.57	114.95	16.03	221.697	21.63	84.451
B4	12.19	61.573	21.28	155.926	19.29	26.393
B5	20.48	133.822	36.39	70.73	54.3	60.538
B6	9.07	43.132	17.22	24.037	45.22	26.729
B7	4.85	43.957	85.33	26.967	69.78	55.083
B8	10.37	56.638	14.18	60.66	16.06	27.021
B9	3.65	51.476	6.77	47.785	31.53	34.066
B10	4.19	77.456	9.65	88.408	5.31	37.191

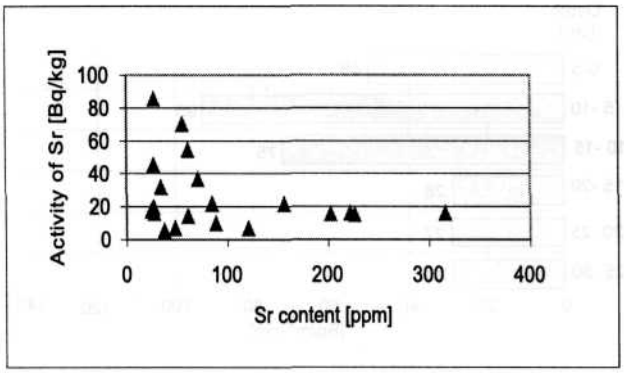


Fig. 7. Radioactivity ^{90}Sr in examined soil samples as a function of strontium concentration in the sample.

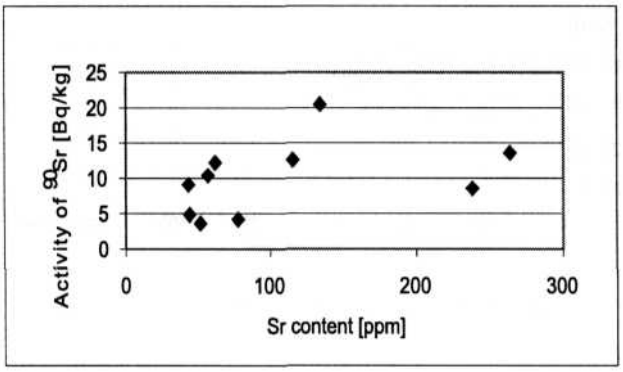


Fig. 8. Radioactivity of ^{90}Sr as a function of the concentration of strontium in the bottom sediment samples.

Table 4. Radioactivity of ^{90}Sr in soil samples calculated for the surface (kBq/m^2/10 cm).

No. sample	Activity ^{90}Sr (kBq/m^2/10 cm)	
	B	C
B1	1.188	1.331
B2	2.230	0.530
B3	2.375	3.477
B4	2.023	2.481
B5	5.565	6.199
B6	2.112	6.580
B7	12.313	10.330
B8	2.405	2.512
B9	0.364	2.244
B10	1.610	0.921
Arithmetic mean	3.22 ± 3.46	3.66 ± 3.1

The results presented in Table 2 and Fig. 5 reveal some complex dependency between concentrations of calcium and ^{90}Sr in examined samples. For total concentration of calcium lower than 1% the concentration of radioactive strontium is much higher. The lack of general tendency mentioned above may be caused by many factors, among them radioactive fallout. No influence of calcium concentration of ^{90}Sr content was observed for bottom sediment samples (See Fig. 6).

A dependence between ^{90}Sr and total strontium concentration as a geochemical background is presented in Table 3 and in Figs. 7 and 8. In this case, such as for calcium, the total concentration of strontium influences the concentration of its radioisotope, especially below 100 ppm where higher radioactivity was noticed. Also, no influence of the strontium concentration on amount of its radioisotope in sediment sample was observed.

In Table 4 the amount of ^{90}Sr is calculated for surface unit and 10 cm depth. The concentration of the studied isotope in 10 cm thick layer ranged from 0.364 to 12.313 kBq/m^2 at calculated mean radioactive precipitation on this area equal to 3.5 ± 3.3 kBq/m^2/10 cm.

Vertical migration of strontium in the same systems is presented in Figs. 9-18. Respective data concern sites 1-4, 10. As can be seen, the distribution of strontium ^{90}Sr in examined profiles is not uniform. Transportation of strontium downwards is more pronounced for sites located at, river bank, i.e. BIB (Fig. 9), B2B (Fig. 11), B3B (Fig. 13) and B4B (Fig. 15). An interesting correlation was noticed for samples B2B (Fig. 11, B3B (Fig. 13) B3C (Fig. 14) B4B (Fig. 15) and B10C (Fig. 18), where maximum radioactivity was measured at 0-15 cm layer

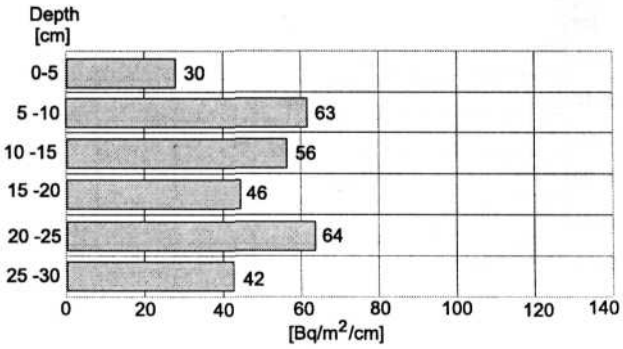
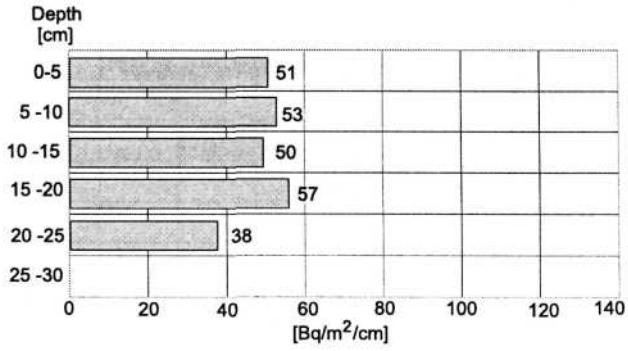


Fig. 9. Radioactivity of ^{90}Sr in sample BIB.

Fig. 10. Radioactivity of ^{90}Sr in sample B1C.



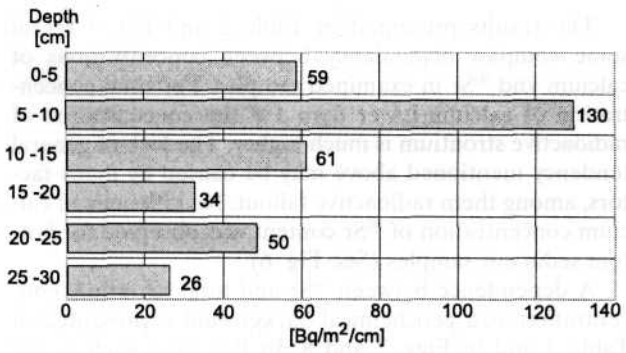


Fig. 11. Radioactivity of ^{90}Sr in sample B2B.

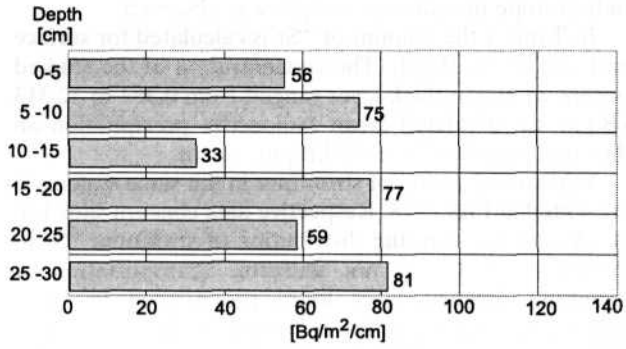


Fig. 12. Radioactivity of ^{90}Sr in sample B2C.

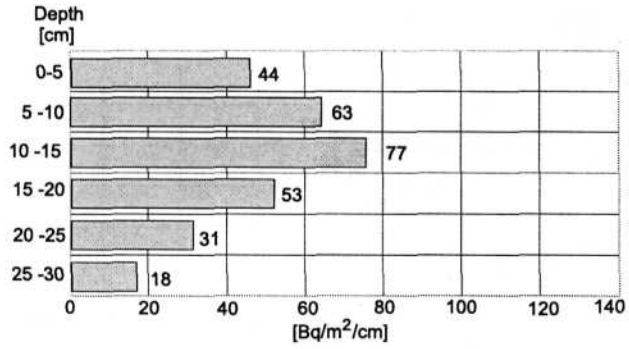


Fig. 13. Radioactivity of ^{90}Sr in sample B3B.

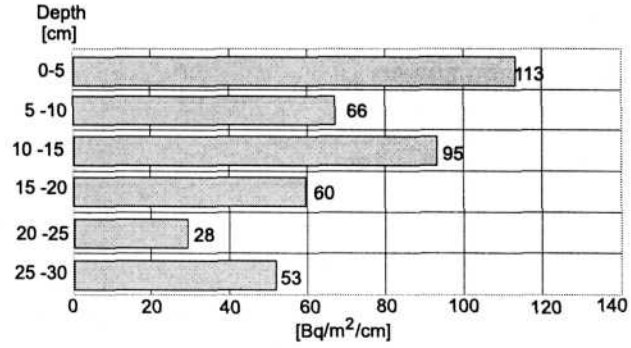


Fig. 14. Radioactivity of ^{90}Sr in sample B3C

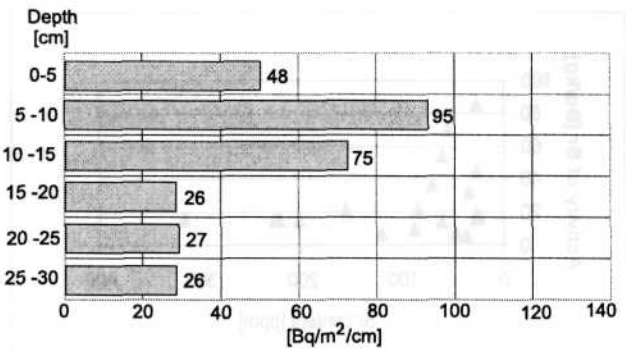


Fig. 15. Radioactivity of ^{90}Sr in sample B4B.

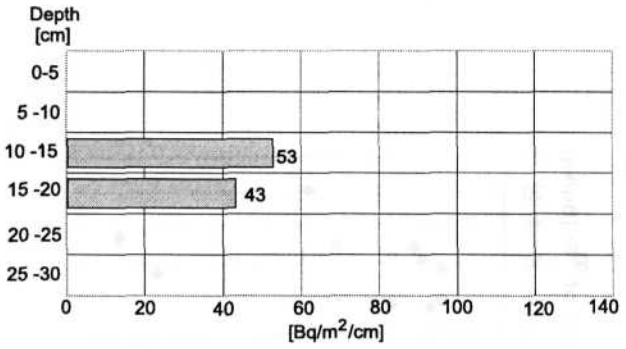


Fig. 16. Radioactivity of ^{90}Sr in sample B4C.

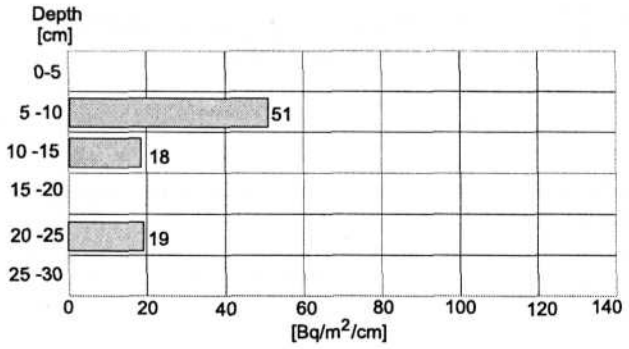


Fig. 17. Radioactivity of ^{90}Sr in sample B10B.

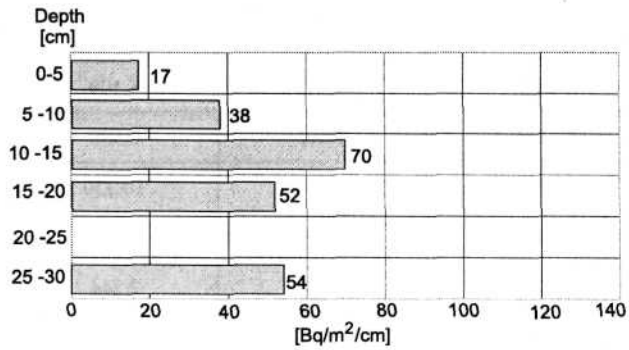


Fig. 18. Radioactivity of ^{90}Sr in sample B10C.

and deeper layers reveal lower radioactivity. This effect may be connected with different sorption properties of respective layers of the profile.

Generally one may say that ^{90}Sr isotope is kept in all studied layers of 0-30 cm profile with various degrees. The results confirm that most of the isotope is kept in level 0-15 cm. Sample B4B (Fig. 16) was an exception as maximum concentration of the isotope was noticed in the layer 10-20 cm.

Conclusions

1. Horizontal migration of ^{90}Sr towards the river was observed.
2. The influence of calcium and strontium ions on the presence of ^{90}Sr is unclear.
3. Mean radioactivity of ^{90}Sr in the soil was 26.15 ± 22.2, in bottom sediments 9.95 + 5.13. Mean precipitation of this isotope was 3.5 ± 3.3 kBq/m^2/10 cm (0.35kBq/m7cm)
4. The radioactivity of ^{90}Sr isotope in soil profiles ranged from 17 to 130 Bq/m^2/cm.
5. Strontium isotope ^{90}Sr was detected down to 30 cm. Its vertical and horizontal migration is a complex process, depending on many factors that are reflected by nonuni form distribution of ^{90}Sr radioactivity.
6. Obtained results confirmed literature data concern ing the range of the vertical migration of strontium [e.g. 4, 11, 12]

Acknowledgments

This research was supported by KBN Grant No. 3T09C 034 15.

References

1. BIESZCZADA ST., SOBOTA J., Zagrozenia, ochrona i ksztaltowanie srodowiska przyrodniczego, Wydawnictwo AR, Wroclaw, **1993.**
2. SZEPKE R., Promieniorworczosc jest wsrod nas, Wydaw nictwo MON, Warszawa, **1988.**
3. JEDINAHKOWA-KRIZKOWA V., Radionuclides Migra tion in the Geosphere and their Sorption on Natural Sorbents, Journal of Radioanalytical and Nuclear Chemistry, 208(2), 559, **1996.**
4. JEDINAHKOWA-KRIZKOWA V., Migration of Radionuclides in Environment, Journal of Radioanalytical and Nuclear Chemistry, **229** (1-2) ,13, **1998.**
5. KIRCHNER G., Modeling the Migration of Fallout Radionuclides in Soil Using a Transfer Function Model, Health Physics, **74** (1), **1998.**
6. KIRCHNER G., Modeling the migration of fallout radionuclides in soil using a transfer function model, Health Physics Society, **1998.**
7. FRIBERG I., Determination of ^{90}Sr and the Transuranium Elements in the Event of a Nuclear Accident, Department of Nuclear Chemistry Chalmers University of Technology Se - 412 96 Goteborg, Doctoral thesis, **1998.**
8. FILSS M., BOTSCH W., HANDL J., MICHEL R., A Fast Method for the Determination of Strontium-89 and Strontium-90 in Environmental Samples and its Application to the Analysis of Strontium-90 in Ukrainian Soils, Radiochimica Acta **83,** 81, **1998.**
9. PIMPL M., $^{89}Sr/^{90}Sr$ - Determination in Soils and Sediments Using Crown Ethers for Ca/Sr - Separation, Journal of Radioanalytical and Nuclear Chemistry Articles **194,** 311, **1995.**
10. IGARASHI Y., AOYAMA M, MIYAO T, HIROSE K, TOMITA M., Anomalous ^{90}Sr deposition during the fall, 1995 at MRI, Tsukuba, Japan, Journal of Radioanalytical and Nuclear Chemistry, **239,** (3), 539, **1998.**
11. GASTBERGER M., STEINHAUSLER F., GERZABEK M. H., LETTNER H., HUMBER A., Soil-to-plant transfer of fallout ceasium and strontium in Austrian lowland and Alpine pastures, Journal of Environmental Radioactivity **49,** 217, **2000.**
12. SMITH J. T., ELDER D. G., A comparison of models for characterizing the distribution of radionuclides with depth in soils, European Journal of Soil Science, **50,** 295, **1999.**
13. SOLECKI J., CHIBOWSKI S., Studies of soil samples min eralization conditions preceding the determination of ^{90}Sr, Journal of Radioanalytical and Nuclear Chemistry, **247,** (1), 165, **2001.**
14. ZABIN A., Metody badan hydrobiologicznych, PWN War szawa, Poland **1966.**