



SPECIATION OF RADIOCESIUM IN ATMOSPHERIC AEROSOL AFTER THE CHERNOBYL ACCIDENT

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Physico-chemical forms of radiocesium released into the atmosphere during the nuclear power plant accident in Chernobyl can significantly influence its behaviour, above all resuspension and transport to other compartments of the environment, including pedosphere and hydrosphere.

Hilton et al. [1], analyzing ^{137}Cs forms in atmospheric aerosol collected in May, 1986 at Oxfordshire location (U.K.), found out marked differences as compared with the results of the analysis of samples collected in 1959 which contained ^{137}Cs from nuclear weapon tests. A difference in water-soluble ^{137}Cs fraction was the most distinct, amounting to 72% as compared to only 8% in the stored samples.

Bobovnikova et al. [2], however, found out in the analysis of fallout material collected at a distance of 18 km from the NPP Chernobyl during the period of the accident 26 April - 3 May 1986 that for ^{137}Cs insoluble form was mainly typical. The content of water-soluble radiocesium fraction was about 9% and in the studied period ranged from 2.6 to 19%. Jansta [3] found in the analysis of the fallout material collected in Košice (Slovak Republic) in the period 29 April - 21 May 1986 the soluble fraction to be less than 10% of the total ^{137}Cs activity in the fallout.

On the basis of the mentioned different results we decided to analyze forms of radiocesium on aerosol filters exposed in Prague in the period after the Chernobyl accident [4]. The aim of this analysis was to verify the hypothesis that physico-chemical forms of radiocesium in the fallout after the accident could depend on the transport conditions, including the distance of a sampling location from Chernobyl.

Experimental

Four aerosol filters (marked A-D) exposed in the period of direct contamination (A-30/4/86, B-1/5/86, C-4/5/86, D-6/5/86) were analyzed. The set was completed with filter E which was exposed only after the direct contamination (12/5/86 to 19/5/86) had finished. The used procedure of sequential analysis was based on the method proposed by Tessier [5] for the study of soils and modified for the analysis of aerosol filters according to Hilton et al. [1]. The ^{137}Cs activity in the samples of individual steps of sequential analysis was determined by gamma spectrometry using GeLi detector. Experimental procedure and measurement technique are described in detail elsewhere [6].

Results

The results of the sequential analysis of ^{137}Cs forms in the samples of atmospheric aerosol collected in Prague in the period of direct contamination by Chernobyl plume are summarized in Table 1. For comparison also results are given of the sample E from a later period and the results adopted from [1] concerning atmospheric aerosol collected at Oxfordshire (U.K.) in May, 1986. The ^{137}Cs activity of individual sequential analysis fractions is expressed as a percentage of the total ^{137}Cs activity in the analysed sample.

From the results it is obvious that the prevailing form in all samples taken in the period of direct contamination was water-soluble radiocesium. The observed values, ranging from 40 to

44%, are, however, lower when compared with the results given by Hilton et al. who found 72% of soluble ^{137}Cs .

Tab. 1 Results of a sequential extraction analysis of radiocesium in atmospheric aerosol samples (percentage of the total activity).

Step	Extractant	A	B	C	D	E	UK*
1	H ₂ O	43	44	49	40	30	72
2	1 M MgCl ₂ (pH 7)	14	21	16	26	16	0.5
3	1 M NH ₄ Cl	3	4	4	7	3	15
4	1 M NaOAc (HOAc, pH 5)	N	2	N	6	1	3
5	0.04 M NH ₂ OH.HCl in 25% HOAc	N	2	N	8	2	2
6	0.1 M HCl	2	N	3	N	N	N
7	3 vol. 0.02 M HNO ₃ + 5 vol. 30% H ₂ O ₂	12	2	13	11	6	2.5
8	Residue	26	25	15	2	43	5

* taken from the publication by Hilton et al.

N the solution was not applied

The second most important fraction was formed by undissolved residues in which the content of ^{137}Cs ranged from 2 to 26% and gradually decreased with time from the accident. Another significant fraction was found after leaching with MgCl₂ solution, i.e. in the fraction subject to ion exchange. This fraction ranged from 14 to 25% and probably did not depend on the time of sample collection and also differs markedly from the results published by Hilton et al.

The results of analysis of sample E confirmed the expected increase in radiocesium content in the undissolved residue and the decrease of water-soluble fraction because after direct contamination had finished, resuspension became the main contamination source of ground-layer atmosphere. In resuspension, however, insoluble cesium forms are preferred because they stay on the surface and do not penetrate into deeper soil layers. In addition, the increase of insoluble ^{137}Cs fraction in resuspended material is influenced by interaction of soluble radiocesium with soil complex which results in firm bond of radiocesium with soil particles and its immobilization in a surface soil layer [7].

From the presented results it can be concluded that physico-chemical forms of radiocesium in atmospheric aerosol and fallout after the NPP accident at Chernobyl as well as particulate size distribution can depend on the distance or the conditions of transport from a contamination source to a sampling location. The influence of the conditions of radiocesium transport could result in the observed differences in the ^{137}Cs penetration into soil profile in different locations and also in the found dependence of the resuspension factor for ^{137}Cs on the level of its fallout in the period of NPP accident for different locations in Europe [8].

References

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