

Plant uptake of radiocesium from contaminated soil

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Abstract Phytoextraction field experiments were conducted on soil contaminated with radiocesium to determine the capacity of autochthonous grasses and weeds to accumulate ^{137}Cs . The aim of the study was to evaluate the potential of spontaneously growing vegetation as a tool for decontamination of non-agricultural contaminated land. As a test field, the closed monitored area of the radioactive wastewater treatment plant of the Nuclear Power Plant in Jaslovské Bohunice, Slovakia was used. Contamination was irregularly distributed from the level of background to spots with maximal activity up to 900 Bq/g soil. Sequential extraction analysis of soil samples showed the following extractability of radiocesium (as percent of the total): water < 0.01%; 1 M MgCl_2 = 0.3–1.1%; 1 M CH_3COONa = 0.3–0.9%; 0.04 M NH_4Cl (in 25% CH_3COOH) = 0.9–1.4%; and 30% H_2O_2 – 0.02 M HNO_3 = 4.5–9.0%. Specific radioactivity of the most efficiently bioaccumulating plant species did not exceed 4.0 kBq kg^{-1} (dry weight biomass). These correspond to the soil-to-plant transfer factor (TF) values up to 44.4×10^{-4} (Bq kg^{-1} crop, d.w.)/(Bq kg^{-1} soil, d.w.). Aggregated transfer factor (T_{ag}) of the average sample of the whole crop harvested from defined area was 0.5×10^{-5} (Bq kg^{-1} d.w. crop)/(Bq m^{-2} soil). It can be concluded that low mobility of radiocesium in analysed soil type, confirmed by sequential extraction analyses, is the main hindrance for practical application of autochthonous plants as a phytoremediation tool for aged contaminated area of non-cultivated sites. Plant cover can efficiently serve only as a soil surface-stabilising layer, mitigating the migration of radiocesium into the surrounding environment.

Key words radiocesium • soil contamination • plant uptake • sequential extraction • case study

Introduction

Cesium is an alkali metal with chemical properties similar to potassium. It has unknown role in plant nutrition and it is not toxic to plants at the micromolar concentrations occurring naturally in soil solution. However, ^{137}Cs and ^{134}Cs radioisotopes are of environmental concern due to their relatively long half-life, emission of gamma radiation during decay and rapid incorporation into living organisms. There is considerably interest in remediation of sites contaminated by these isotopes using extraction by plants (phytoextraction) that do not enter the human food chain. Many original papers such as [4, 7, 10], reviews [2, 14] and books [8, 11] describing these topics were published. Our paper presents data characterising bioaccumulation activity of autochthonous plants spontaneously growing on non-cultivated and non-fertilised field contaminated with radiocesium originating from technology of radioactive wastewater treatment plant in the area of nuclear power plant.

Methods

Soil and plant samples were collected from monitored contaminated area inside the Nuclear Power Plant, Jaslovské Bohunice, Slovakia. Soil samples were taken from the vertical profile 0–5 and 5–20 cm. The lumps were

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crushed, dried, separated from plant roots, sieved to pass through 2-mm sieve (10 mesh). Plants in the middle of spring season (May) from the contaminated area (0.45×0.25 m) showing high specific radioactivity were harvested by cutting 10 cm above ground level and weight. Aliquots were air-dried, homogenised by knife blender and radioactivity was measured. For radiometric analysis gamma spectrometer scintillation detector 54BP54/2-X with well type crystal NaI(Tl), Scionix, Netherlands and data processing software Scintivision 32, Ortec, U.S.A. were used. Standard $^{137}\text{Cs}^+$ solution (obtained from Research Institute of Nuclear Energy, Trnava, Slovakia) was used as a standard for efficiency calibration in above-mentioned counting system.

To obtain data quantifying extractability of cesium in soil matrix, sequential extraction of soil samples according to the procedure of Tessier [12] and BCR method [5, 13] was used.

For the studied plants, ^{137}Cs uptake was expressed as transfer factors (TF) and aggregated transfer factor (T_{ag}). The soil-to-plant transfer factor (TF) [1, 3] and aggregated transfer factor (T_{ag}) [6] were defined as follows:

TF = activity concentration in plant (Bq kg^{-1} d.w.)/activity concentration in soil (Bq kg^{-1} dry weight) and
 T_{ag} = activity concentration in plant (Bq kg^{-1} d.w.)/total deposition of soil (Bq m^{-2}).

Results and discussion

Soil contaminated with higher than allowable level of radionuclides gives an exclusive chance for the study of radionuclides movement in environment influenced by biotic and abiotic factors both of natural and artificial

origin. From the agricultural point of view the contaminated soil under study can be characterised as non-cultivated, non-fertilised with low organic content and typical for areas influenced by the neighbouring building activities. Soil pH was in the range 7.66–7.78 (in 0.01 M CaCl_2) and soil cation exchange capacity (barium acetate method): 9.9–10.3 mEq/100 g.

Gamma spectrometric analysis of soil samples showed that ^{137}Cs represent more than 99.5% of the total gamma activity of contaminated soil (data not shown). Specific radioactivity of soil decreased with depth. Surprisingly, in certain cases the decrease was rather irregular. However, in these cases, building activities connected with transfer of the soil, were confirmed. The surface of the area under study was randomly covered with autochthonous grasses mainly with *Dactylis glomerata*, *Achillea millefolium* and members of the *Equisetaceae* family and with small amount of other not taxonomically classified plants. We found that grasses covering contaminated area accumulated significant amount of radiocesium. Other gamma emitters in the grass were not detected by the radiometric method which was used. The average specific radioactivity of the biomass harvested from the highly contaminated area was 1000 Bq kg^{-1} air-dried biomass. Only some plant species showed approximately four-time higher capacity for radiocesium bioaccumulation. Soil-to-plant transfer factor values TF (Bq kg^{-1} crop, d.w.)/(Bq kg^{-1} soil, d.w.) for *Dactylis glomerata*, *Achillea millefolium* and *Plantago officinalis* 0.59×10^{-3} , 1.29×10^{-3} and 4.44×10^{-3} , respectively were obtained. Aggregated transfer factor (T_{ag}) of the average sample of the whole crop harvested from defined area was 0.5×10^{-5} (Bq kg^{-1} d.w. crop)/(Bq m^{-2} soil).

Our data presented in Tables 1 and 2 shows very low cesium extractability from the given soil samples, obtained

Table 1. Sequential extraction of radiocesium from contaminated soil according to Tessier [12].

Fraction	Reagent	Time [h]	Temperature [°C]	Extracted [%] ^a
f1	water	6	20	0
f2	1 M MgCl_2 (pH 7.0)	1	20	0.3–1.1
f3	1 M CH_3COONa (pH 5.0 with CH_3COOH)	5	20	0.3–0.9
f4	0.04 M $\text{NH}_4\text{OH}\cdot\text{HCl}$ (in 25% CH_3COOH)	6	95	0.9–1.4
f5	a. 0.02 M HNO_3 b. 30% H_2O_2 (pH 2.0 with HNO_3)	5	85	4.5–9.0

^a Range of extracted radioactivity in percent of the total found in individual soil samples taken from 0 to 20 cm under ground level.

Table 2. Sequential extraction of radiocesium from contaminated soil according to BCR method [5, 13].

Fraction	Reagent	Time [h]	Temperature [°C]	Extracted [%] ^a
f1	0.1 M CH_3COOH	16	20	0.7–2.9
f2	0.1 M $\text{NH}_4\text{OH}\cdot\text{HCl}$ (pH 2.0 with HNO_3)	16	20	1.5–2.9
f3	a. 30% H_2O_2	1	20	
		2	85	
	b. 1 M $\text{CH}_3\text{COONH}_4$ (pH 2.0 with HNO_3)	16	20	5.6–10.3

^a Range of extracted radioactivity in percent of the total found in individual soil samples taken from 0 to 20 cm under ground level.

by two different extraction methods [5, 12, 13], even in the cases where very drastic extraction reagents, such as nitric acid and hydrogen peroxide at elevated temperatures for several hours were used. Very low extractability within the range of 0.3–2.9 percent of total was obtained, after mild reagents as sodium acetate or magnesium chloride or weak organic acids such as acetic acid were used. Practically no radioactivity was extracted by distilled water. This is in agreement with previously published papers, stressing the decisive role of soil clay materials in irreversible binding of cesium into inorganic crystal structures [9]. It can be concluded that extremely low concentration of cesium in soil solution is the factor limiting cesium uptake rate by the root systems.

It is generally known that bioaccumulation of metal ions by plants from soil solution depends on genetically determined biochemical properties, total metabolic activity and plant growth rate. There is a common tendency to improve bioaccumulation rates by fertilizers and soil conditioners and by changing biochemical properties of plants on the basis of molecular genetics. However, these tools can not improve the bioavailability of radiocesium incorporated into soil micaceous clays.

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