Study of radiophytoremediation on heavily polluted area in South Bohemia

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Abstract. A phytoremediation study was performed on the area of the old uranium mill tailings waste depot of a former uranium ore reprocessing factory in South Bohemia. The distribution of ^{226}Ra in contaminated soil was found to be extremely variable (from 7 to 32 Bq ^{226}Ra/g of DW). No direct relation was proved between plant species characteristics and their radioactivity content. The results showed a great range of variation in the accumulation of ^{226}Ra by the plant species found. The highest activity of ^{226}Ra was found in *Potentilla reptans* (4.09 Bq ^{226}Ra/g of DW), *Mentha arvensis* (4.00 Bq ^{226}Ra/g of DW), and *Daucus carota* (3.70 Bq ^{226}Ra/g of DW). The greenhouse and small scale field experiments show *Cannabis sativa* “Beniko” as a good potential accumulator of activity ^{226}Ra.

Introduction

Soil contaminated with radionuclides poses a long-term radiation hazard to human health through exposure *via* the food-chain and other pathways. Phytoremediation of radionuclide-contaminated soils has become increasingly important. The understanding of the mechanism of radionuclide uptake and accumulation is necessary prerequisite for the application of radiophytoremediation in “real” scale.

The objectives of our work were to: (i) perform a geobotanical study of the wild plants growing in the area of uranium mill tailings; (ii) determine the distribution of ^{226}Ra among selected plant species; (iii) carry out the biomonitoring of recultivated areas; (iv) test the low contaminated sited for production of non-food plants and (v) propose the mechanism of clean-up of water contained uranium and ^{226}Ra. The goal of this work was also to select appropriate plant candidates for potential utilization in radiophytoremediation processes.

Material and Methods

Collection and preparation of substrate samples for measurement

To determine the basic chemical properties of the mill tailings substrate, the samples (three duplications) were collected at five sampling points on the top of dump K1. The samples were dried at 80 °C for 72 h. and afterwards weighed and measured in Marinelli beakers. The average characteristics of mill tailings deposited in waste dump K1 are presented in Table 1.

Table 1. Substrate properties of mill tailings deposited in waste dump K1.

soil characteristic	
SiO_2	600 g/kg
gypsum	100 - 200 g/kg
Fe, Al hydroxide	20 - 100 g/kg
grain size	< 0.5 mm
soluble compounds	30 - 53 g/kg
^{226}Ra	5 - 32 kBq/kg
Mn	1050 mg/kg
NH_4^+	750 - 1050 mg/kg
U	< 1.5 mg/kg
SO_4^{2-}	18 - 30 mg/kg
heavy metals (Zn,Ni,Co,Cd)	≈ 3 mg/kg
pH	5.0 - 7.0

Greenhouse experiment

The technical crops, food crops, and fodder crops were cultivated in control conditions in mixture of uranium mill mine tailings (32 Bq ^{226}Ra/g DW) and soil in ratio 1:3. The total activity of soil mixture was about 13 Bq ^{226}Ra/g DW.

Small scale field experiment

The technical crops, food crops, medicinal plants, grasses and fodder crops were cultivated from June to October 2004 on field with the activity about 9 Bq ^{226}Ra/g DW.

Collection and preparation of plant samples for measurement

Plant samples were collected in the autumn of 2004. The whole plants were rinsed in a high pressure water stream, dried at 80 °C for 72 h., homogenized and then weighed and measured in Marinelli beakers. Only one sample was prepared from each plant species which represented an average of many whole plants, because the Marinelli beaker used for the measurement of radioactivity had to contain about 100 g of dry material.

Sample measurement

The ^{226}Ra radioactivity in the sample was determined after reaching the decay equilibrium in sealed Marinelli beakers by means of a gamma scintillation spectrometer (Canberra – Packard, model PCAP-Nal 2007, channel width 4.986 keV, energy resolution 9% at 662 keV) relatively to ^{226}Ra standard of 3.000 kBq (Czech Institute of Metrology, type MBSS 5). Using PC programme Genie 2000 (Canberra – Packard), the comparison of a sample and standard peaks of ^{214}Bi (609.3 keV) was applied for the evaluation of ^{226}Ra activity. For Quality Assurance and Control two standards were also used: ^{60}Co (Czech Institute of Metrology, type MBSS 7) for checking of spectrometer stability and ^{137}Cs (Czech Institute of Metrology, type MBSS 4) for energy resolution measurement. All three standards were used for energy calibration of the spectrometer (Soudek et al. 2004a).

Table 2. The plant species with the most high and lowest activity, which was collected on the top of dump K1.

Plant species	**Activity ± S.D. [Bq ^{226}Ra/g DW]**
Potentilla reptans	4.09 ± 0.043
Mentha arvensis	4.00 ± 0.077
Calamagrostis epigeios	3.40 ± 0.033
Daucus carota	3.70 ± 0.035
Rubus caesius	2.65 ± 0.025
Silene vulgaris	2.60 ± 0.028
Cirsium arvense	2.46 ± 0.013
Hypericum perforatum	2.13 ± 0.033
Echinum vulgare	1.79 ± 0.047
Sphagnum fallax	1.76 ± 0.042
Artemisia vulgaris	0.19 ± 0.006
Urtica dioica	0.11 ± 0.008
Sisymbrium loesselli	0.10 ± 0.003
Tanacetum vulgare	0.08 ± 0.006
Melilotus officinalis	0.06 ± 0.004
Melilotus alba	0.02 ± 0.003
Amanita phalloides	0.00 ± 0.000
Trifolium repens	0.00 ± 0.000
Polygonum amfibium	0.00 ± 0.000

Results and Discussion

Concentrations of radionuclides in plant samples collected in the surrounding areas of the uranium ore processing factory were studied. Radionuclides detected both in soil and plants were ^{226}Ra, ^{214}Pb and ^{214}Bi. We found the best accumulation ability (cca. 4 Bq ^{226}Ra/g of dry weight (DW) to 1.8 Bq/g DW) in *Calamagrostis epigeios, Hypericum perforatum, Silene vulgaris, Cirsium arvense* and *Rubus caesius* etc. All this plants was collected on sampling point with activity 32 Bq/g of dry soil.

The greenhouse experiments were in progress from March to July 2004. The first results show differences between crops plant species, which was cultivated in flowerpots. The counting rate of soil was about 13 Bq ^{226}Ra/g DW. The best accumulation we detected for *Amaranthus tricolor* „Early Splendor“ and *Lupinus polyphyllus* (2.16 or 2.20 Bq ^{226}Ra/g DW). The significant differences between cultivars of same plant species were not found. But we found differences in accumulation of ^{226}Ra between plants from the same genus (*Lupinus* sp.).

The small field experiments were done on experimental are which was situated on lakeside of sludge bed IV/R (activity 9 Bq ^{226}Ra/g DW). The best activity accumulation was determined for *Mentha pipericum* (about 3.05 Bq ^{226}Ra/g DW). We not found again differences in accumulation of ^{226}Ra for different cultivars of all tested plant species and between plants from same genus, except *Lupinus* sp. The high activity was determined also in grasses *Bromus lanceolatus* and *Festuca glauca* (2.17 or 1.70 Bq ^{226}Ra/g DW).

Table 3. The activity of plants cultivated in greenhouse on soil mixture with activity 13 Bq ^{226}Ra/g DW and on small field on the lakeside of sludge bed IV/R on soil with activity 9 Bq ^{226}Ra/g DW. (n/g = not growth).

Plant species	Activity ± S.D. [Bq ^{226}Ra/g DW] greenhouse	small field
Linum usitatissimum „Atalante“	0.45 ± 0.013	0.15 ± 0.006
Linum usitatissimum „Jitka“	0.35 ± 0.018	0.26 ± 0.012
Cannabis sativa “Beniko“	0.42 ± 0.008	0.28 ± 0.006
Cannabis sativa „Juso-11“	0.40 ± 0.016	0.41 ± 0.010
Cannabis sativa „Silesia“	0.52 ± 0.010	0.38 ± 0.024
Amaranthus hypochondriacus „Pygmy Torch“	0.74 ± 0.020	0.18 ± 0.004
Amaranthus tricolor „Early Splendor“	2.16 ± 0.071	n/g
Amaranthus tricolor	0.59 ± 0.022	0.27 ± 0.005
Amaranthus caudatus „Atropurpureus“	0.76 ± 0.020	0.44 ± 0.006
Phaseolus vulgare „Bobis Nano“	0.95 ± 0.039	0.30 ± 0.006
Phaseolus vulgare „Aida Gold“	0.55 ± 0.031	0.57 ± 0.013
Pisum sativum „Ambrosia“	0.32 ± 0.030	n/g
Pisum sativum „Gloriosa“	0.43 ± 0.019	n/g
Capsicum annuum „Berta“	0.40 ± 0.026	0.20 ± 0.020
Capsicum annuum „Drákula“	0.46 ± 0.025	0.94 ± 0.081
Capsicum annuum „Maryša“	0.56 ± 0.021	0.20 ± 0.035
Lycopersicon lycopersicum „Albertovské“	0.86 ± 0.027	0.47 ± 0.008
Lycopersicon lycopersicum „Stupické“	0.57 ± 0.023	0.28 ±0.005
Lycopersicon lycopersicum „Start F1“	0.32 ± 0.021	0.26 ± 0.010
Lupinus albus	0.72 ± 0.039	0.89 ± 0.042
Lupinus luteolus	1.24 ± 0.093	0.66 ± 0.014
Lupinus polyphyllus	2.20 ± 0.097	1.29 ± 0.031
Daucus carota	1.10 ± 0.016	-
Sinapis alba	0.55 ± 0.012	0.31 ± 0.008
Helianthus annuus	0.41 ± 0.016	n/g
Brassica oleracea	0.75 ± 0.030	0.61 ± 0.008
Zea mays	n/g	n/g
Panicum miliaceum	-	0.38 ± 0.005
Achillea millefolium	-	0.55 ± 0.008
Achillea filipendulina	-	0.78 ± 0.051
Sorghum bicolor	-	1.03 ± 0.014
Sorghum nigrum	-	1.55 ± 0.020
Euphorbia marginata	-	0.08 ± 0.005
Hypericum perforatum	-	1.02 ± 0.062
Lepidium sativa	-	0.14 ± 0.006
Festuca glauca	-	1.70 ± 0.041
Agrostis nebulosa	-	0.67 ± 0.017
Bromus lanceolatus	-	2.17 ± 0.021
Iberis umbellata	-	0.25 ± 0.011
Solidago canadiensis	-	1.60 ± 0.025
Mentha pipericum	-	3.05 ± 0.032

Conclusion

The obtained results prove the possibility of utilization of radiophytoremediation for practical application, at least in the case of wastewater treatment, where the conditions of contaminants uptake can be similar to hydroponic arrangement. Of course, for the soil-cleaning purposes, the solubility of contaminant and its mobility in soil will be the most limiting factor as well as extend of root-zone of selected plant species.

The second problem of practical application is after-harvest treatment of contaminated plant material. Its storage and composting (as radioactive waste), or its incineration under strictly controlled conditions, respectively, can be considered.

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