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^{226}Ra uptake from soils into different plant species

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Accumulation of ^{226}Ra into different plant species from contaminated soils was measured on site within the area of an uranium mill. Marinelli beakers and NaI(Tl) spectrometer were used for measurement of dried and weighted samples. While the ^{226}Ra activity concentration in soil on site ranged from 7.12 to 25.60 Bqg^{-1} (1 SD<±10%), in the plant species tested it ranged from 0.66 to 5.70 Bq·g^{-1} (1 SD<±10%). No significant differences in ^{226}Ra accumulation were found after cultivation of selected plant species in a glasshouse in relation to the outdoor experiments.

Introduction

Uranium mining and milling contribute to radionuclide contamination of the environment.[1] During the last two decades, many such facilities were closed because of a decrease in the demand of uranium and the increase in the overall supply. Implementation of area restoration represents an important task of various countries.

A long-term contamination in the neighbourhood of uranium mines and mills remains in soils. The concentration of activity of ^{226}Ra is used as its measure. There is a limit of 200 Bq·kg^{-1} averaged over the first 15 cm below the ground surface.[2]

For remediation of man-made environmental radionuclides, phytoremediation[3,4] can be effectively applied. An environment-friendly and cost-effective uptake of radionuclides by root systems from contaminated soils and/or surface waters has enabled many applications.[15–12] Although these procedures were studied for different radionuclides, no systematic study for ^{226}Ra was published until now.

In this paper, heavily polluted soils in the area of a closed uranium mill were used to study the accumulation of ^{226}Ra in different plant species. The study was performed either on-site using simultaneous measurement of a mean ^{226}Ra volume activity of the soil surrounding roots or in a glasshouse using defined conditions. Plants growing naturally on-site were used as well as some of which had been transplanted.

Experimental

The analyzed plants and samples of soils surrounding their roots (to 15 cm below the ground surface) were collected at different sites of a previous uranium mill during May and June and transported in special bags to labs. The soil samples were dried for 3–4 hours using a temperature of about 110 °C and were filled into 1 dm^3 Marinelli beakers. Usually, whole dried plant species were used. Traces of soil were carefully mechanically removed from roots before treatment, if necessary. In the case of trees, leaves were measured. Stiff parts of plant samples (e.g., some stems) were taken for counting after crushing. No special homogenization was done. The mass of each sample was determined. Afterwards, covers of beakers were puttied in place using a silicon-binding agent. Approximately, the same quantity of each sample was prepared and measured three times and the mean value was calculated. The background to be subtracted for each soil or plant sample, respectively, was determined as a mean of the same quality and approximately the same quantity using five samples collected at different other sites outside the experimental area.

For plant cultivation in a glasshouse, being outside the contaminated area, samples of substrate originating from the uranium waste disposal site and gardener substrate were mixed (3 : 1). Concentration of activity of the substrate at the start of cultivation was established to be 15.49 Bq·g^{-1}. Five different plant species were cultivated in 4 dm^3 pots. The temperature in the glasshouse was set to be at least 15 °C, the glasshouse was vented, but not cooled. Plants were grown the photoperiod 12/12 L/D, additional light was supplied by sodium bulbs (400 W, Thorn Radbay) at an intensity of 72 μmol·m^{-2}·s^{-1}. Grown plants were harvested, immediately dried and subjected to analysis in the same way as plants species from the contaminated area.

The concentration of ^{226}Ra activity in soil on fifteen sites of the mill area ranged from 7.12 to 25.60 Bq·g^{-1} (1 SD<±10%). The resulting accumulation in the individual plant species was expressed as a ratio of ^{226}Ra Bq·g^{-1} of plant to ^{226}Ra Bq·g^{-1} of soil surrounding the plant roots.

The gamma-spectrometry was carried out using a scintillation spectrometer (Canberra-Packard, Model PCAP-NaI 2007, detector 802-3x3 W, lead screening 727 R). The channel width was set at 4.986 keV,

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the energy resolution was 9% at 662 keV peak of ^{137}Cs. The measurement of the ^{226}Ra spectrum was carried out by means of PC programme Genie 2000 (Canberra-Packard). All samples were measured at least 38 days after the sealing of the Marinelli beakers to allow them to reach the decay equilibrium.

Results and discussion

From the measured spectra (Fig. 1) of a ^{226}Ra standard in a sealed Marinelli beaker (Czech Institute of Metrology, type MBSS 5, 3.000 kBq) as well as that of a soil sample, it is evident that the ^{226}Ra peak (186.2 keV, 3%) enabled only low detection sensitivity in the relatively high background region. Therefore, for all measurements, the peak of ^{214}Bi (609.3 keV, 46.3%) was used for evaluation of the ^{226}Ra activity. Using individual measured points corresponding to this ^{214}Bi peak (Fig. 2), its shape was created by the programme Interactive Peak Fit (Canberra–Packard). The hatched area of the peak (Fig. 2), representing the measured ^{214}Bi peak counts of the ^{226}Ra standard, was compared with the corresponding ^{214}Bi peak area of each measured sample. The full part under the peak representing noise was not taken into consideration. The ^{226}Ra value of the sample was calculated from the ratio of the ^{214}Bi sample peak to that in the ^{226}Ra standard spectrum (Fig. 2). To obtain a standard deviation of ^{226}Ra activity determination lower than ±10%, a corresponding measuring time was chosen.[13]

The results obtained proved that the ^{226}Ra accumulation (Table 1) was rather different for the higher plant species tested. Some of them could be applied for effective large-scale and long-time decrease of ^{226}Ra activity concentration in highly contaminated soils. Moreover, using selected plant species could be considered for biomonitoring. The results obtained can be helpful in the choice of suitable plant species for ^{226}Ra phytoremediation and/or phytomonitoring within the areas of uranium facilities.

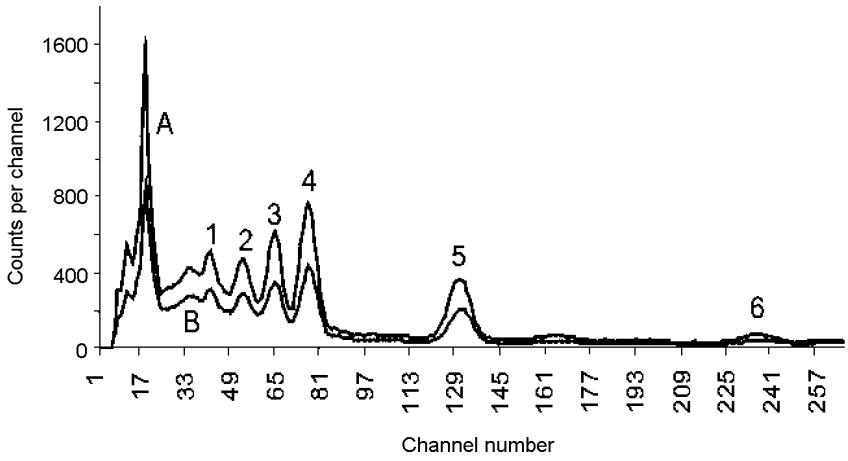

Fig. 1. Measured ^{226}Ra spectra in decay equilibrium. Curve A – standard, curve B – a soil sample; 1 – ^{226}Ra (186.2 keV), 2 – ^{214}Pb (238.6 keV), 3 – ^{214}Pb (295.2 keV), 4 – ^{214}Pb (351.9 keV), 5 – ^{214}Bi (609.3 keV), 6 – ^{214}Bi (1120.3 keV)

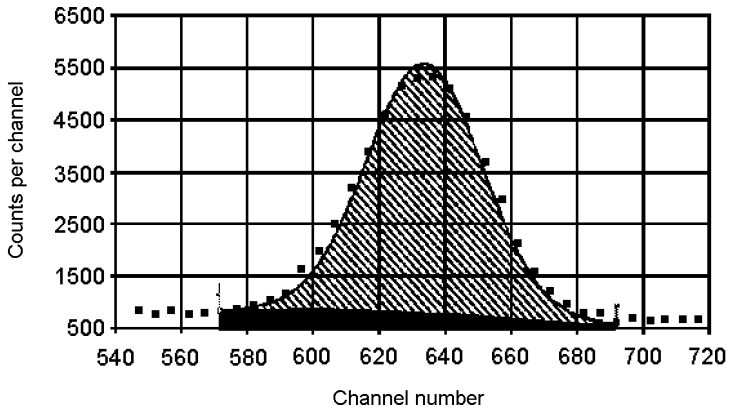

Fig. 2. Evaluation of ^{214}Bi peak of 609.3 keV for determination of the ^{226}Ra activity

Table 1. ^{226}Ra accumulation in different plant species*

Plant tested	Relative ^{226}Ra activity
Vetch *(Vicia tenuifolia)*	4.15
Reed *(Phragmites australis)*	4.21
Poplar *(Populus tremula)*	4.34
White sweet-clover *(Melilotus albus)*	6.29
Silver birch *(Betula pendula)*	8.55
Evening-primrose *(Oenothera biennis)*	9.12
Spurge *(Euphorbia esula)*	9.31
Strawberry *(Fragaria vesca)*	10.57
Perforate St Johnswort *(Hypericum perforatum)*	11.07
Blueweed *(Echium vulgare)*	11.26
Centaury *(Centaurium erythraea)*	11.95
Sunflower *(Helianthus annus)*	12.14
Creeping thistle *(Cirsium arvense)*	12.58
Bladder campion *(Silene vulgaris)*	13.33
Dewberry *(Rubus caesius)*	13.46
Corn *(Zea mays)*	13.84
Chee reedgrass *(Calamagrostis epigeios)*	17.36
Black medick *(Medicago lupulina)*	17.74
Lupin *(Lupinus polyphyllus)*	20.25
White mustard *(Sinapis alba)*	22.52
Wild carrot *(Daucus carota)*	23.27
Pea *(Pisum sativum)*	23.40
Corn mint *(Mentha arvensis)*	25.16
Silverweed *(Potentilla reptans)*	25.72
Hemp *(Cannabis sativa* cv. Beniko)	27.04
Sudan grass *(Sorghum bicolor)*	27.30
Amaranth *(Amaranthus caudatus)*	29.56
Mercury weed *(Mercurialis annua)*	35.85

* The relative ^{226}Ra activity in % expressed as 100× ratio of concentration of ^{226}Ra activity of the plant species (1 SD<±10%) to the mean value of concentration of ^{226}Ra activity in the soil surrounding the roots (1 SD<±10%).

Conclusions

The paper summarized measurements carried out during three years in an uranium mill area containing soils with ^{226}Ra activity concentration up to two orders of magnitude higher than the permissible limit. Using different plant species, the values of ^{226}Ra accumulation was found to be considerably different. For some of them, application for effective phytoremediation can be considered.

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