NUKLEONIKA 2005;50(Supplement 1):S25–S27

Bioaccumulation of ^{226}Ra in the plants growing near uranium facilities

Richard Tykva,
Eva Podracká

Abstract Radiopollution of soils in the close neighbourhood of a uranium mine and a uranium mill was characterized using measurement of specific ^{226}Ra activity by gamma spectrometry. It was found that it ranged from values lower than the admissible limit of 0.2 Bq·g^{-1} of soil (averaged over the first 15 cm below the ground surface) to values more than one hundred times higher. Bioaccumulation of ^{226}Ra from the measured soils in plant species which were naturally growing on the analysed sites was analysed. The specific ^{226}Ra activity of aboveground dry plant parts ranged within two orders of magnitude. Currently occurring and simply collected *Mentha piperita* was found to be useful for biomonitoring of soils.

Key words uranium mine • uranium mill • contaminated soils • plant accumulation

R. Tykva✉, E. Podracká
Institute of Organic Chemistry and Biochemistry,
Academy of Sciences of the Czech Republic,
2 Flemingovo Str., CZ-166 10 Prague 6, Czech Republic,
Tel.: +420-220 183269, Fax: +420-220 183565,
E-mail: tykva@uochb.cas.cz

Received: 10 July 2005
Accepted: 15 August 2005

Introduction

Implementation of restoration of the environment polluted by radionuclides [1, 5] represents an important task of various countries. One of the most important source of radiopollution has been uranium mining and/or milling. In their neighbourhood still persits radionuclide contamination even though such facilities have been closed.

Uranium consisting of ^{238}U (99.275%), ^{235}U (0.720%) and ^{234}U can be found in minor but still detectable quantities in nearly all soil and rock samples of different origin [6]. Primarily occurring ^{238}U generates, during its decay, further radioactive atomic nuclei and, in this way, naturally existing uranium/radium decay series is turned out [5]. Activity concentration of ^{226}Ra is used as a measure of environmental pollution by the radionuclides from this decay series. Its acceptable limit in soils is 0.2 Bq·g^{-1} averaged over the first 15 cm below the ground surface [2].

In this paper, specific ^{226}Ra activity in soil samples is measured in two types of areas: with an existing uranium mine and with a closed uranium mill. The ^{226}Ra bioaccumulation from such soils in different plant species growing there was compared.

Experimental

Two different uranium facilities were selected: an existing mine in Crucea, the North-East of Romania, and a mill in the South of Bohemia closed approxima-

Table 1. Specific ^{226}Ra activity of different soil samples collected near nuclear facilities (SD for all values less than ±10%)

Nuclear facility	Site of sample collection		Specific ^{226}Ra activity
	Quality	Approximate distance	[Bq·g^{-1}]
Mine (m.)	forest	200 m from m.	0.34
	forest	1 km from m.	0.08
	field (in the same direction)	1.5 km from m.	0.60
Mill	settling tank	covered surface	9.3 to 29.8
	settling tank	less than 50 m	0.05 to 1.65
	settling tank	more than 100 m	0.06 to 0.08
	sludge bed	covered surface	5.5 to 8.7

tely twenty years ago. The analysed plants and samples of soils surrounding their roots (to 15 cm below the ground surface) were collected in the selected sites during the same vegetation period (June 2003, 2004 and 2005).

After transport to the laboratory (in special bags), the soil samples were dried at about 110°C for 3–4 hours and then filled into Marinelli beakers (1 L). The beakers were carefully cemented using silicon binding agent to prevent release of ^{222}Rn, a member of the uranium/radium decay series. The measurement was made after at least 38 days after sealing the beakers, what allowed to reach transient equilibrium [5].

The amount of collected plants was large enough to reach high counting rates which contributed to relatively low statistical deviation within a reasonable counting time. Moreover, different counting geometries could also influence counting efficiency. Therefore, we tried to fill the beakers completely. The plant samples were dried, weighed and placed in the beakers. Stiff parts, e.g. stems, were crushed before further treatment.

The gamma spectrometric measurements were 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, the energy resolution was 9% at 662 keV peak of ^{137}Cs. Evaluation of spectra was carried out by means of PC Genie 2000 software (Canberra-Packard). As a reference, ^{226}Ra standard in a sealed Marinelli beaker (Czech Institute of Metrology; type MBSS 5; 3.000 kBq) was used. All the samples were measured three times and the mean values determined. The peak of ^{214}Bi (609.3 keV, abundancy 46.3%) was used for evaluation of the ^{226}Ra activity as previously described by us [3]. The standard deviations (SD) were determined using square root of the sum of squares of the corresponding two ^{214}Bi standard deviations. To obtain a final SD lower than ±10%, measurement time was chosen.

The bioaccumulation of ^{226}Ra from soils in different plant species was expressed as a ratio of Bq·g^{-1} of measured dry plant sample to Bq·g^{-1} of dry soil collected around the plant roots.

Results and discussion

It is evident that the main source of long-term radiopollution of soils (Table 1) is radioactive waste. The highest activities were found in the soil covering the top of the aboveground setting tank and slightly lower at the top of an underground sludge bed. However, it was proved (Table 1) that the specific ^{226}Ra activity of soils is decreased with increasing distance from the waste collector. However, it is under the permissible level in the distance of more than 10 meters.

Although the mine is active, the measured pollution of soils was quite lower (Table 1). A decreasing activity with an increasing distance was found as well as higher pollution of the opened field in relation to forest placed in the same direction. It can be assumed that the radioactive dust is predominantly responsible for the pollution.

In Table 2, ^{226}Ra bioaccumulation is compared in the selected plant species which occur naturally in neighbourhood of the both nuclear facilities. Among the analysed species, the bioaccumulation level differs twenty times approximately. In request to other previously measured plant species [4], *Mentha piperita*, especially roots, seems to be an excellent tool for bioaccumulation. Since it is a very common plant species, it can be considered for phytoremediation or biomonitoring.

The results of ^{226}Ra phytoaccumulation presented in this paper were calculated as means from two different sites in Europe and three years. Several different influences, e.g., soil type, heterogeneous distribution of radionuclides, meteorogical conditions, biomass of perennial plants etc., were not considered. Nevertheless, the differences in comparison of bioaccumulation are apparent. However, selected plant species are now treated in glasshouses to compare the bioaccumulation under defined conditions. The results of such study will be published elsewhere.

Table 2. Comparison of ^{226}Ra bioaccumulation in naturally occuring plant species (SD for all samples less than ±10%)

Plant	Relative ^{226}Ra activity [%][a]
Mentha piperita	41.25
Fragaria vesca	15.68
Cirsium arvense	5.10
Urtica dioica	2.74

[a] Expressed as 100x ratio of specific ^{226}Ra activity of a dried plant species to specific ^{226}Ra activity of the dry soil surrounding its roots.

Acknowledgment The paper was made in frame of the research plan No. Z 40550506. The authors thank to Dr. Karin Popa from Al. I. Cuza University, Iasi, Romania and Dr. Petr Soudek from their Institute for some samples.

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