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Biological availability of ^{238}U, ^{234}U and ^{226}Ra for wild berries and meadow grasses in natural ecosystems of Belarus

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ABSTRACT

This work is devoted to investigation of behavior of ^{234}U, ^{238}U and ^{226}Ra by determining the soil to plant transfer under different natural conditions such as forest or swamped areas and meadow lands with different soil types. The paper summarizes the data on investigation of uranium and radium uptake by wild berries and natural meadow grasses in the typical conditions of Belarus. Parameters characterizing the biological availability of ^{234}U, ^{238}U and ^{226}Ra for bilberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium viti-idaea*), blueberry (*Vaccinium iliginosum*) and cranberry (*Vaccinium oxycoccus palustris*) as well as for widely occurring mixed meadow vegetation, which belongs to the sedge-grass or grass-sedge associations and forbs, have been established. In the sites under investigation, the deposition levels of $^{238+239+240}Pu$ were less than 0.37 kBq m^{-2} and ^{137}Cs deposition ranged between less than 0.37 and 37 kBq m^{-2}. It was found that activity concentrations of radionuclides in berries varied in the ranges of 0.037–0.11 for ^{234}U, 0.036–0.10 for ^{238}U and 0.11–0.43 Bq kg^{-1} for ^{226}Ra, but in the mixed meadow grasses they were 0.32–4.4, 0.24–3.9 and 0.14–6.9 Bq kg^{-1} accordingly. The $^{234}U/^{238}U$ activity ratios were 1.02 $\pm$ 0.01 for wild berries, 1.20 $\pm$ 0.09 for underground meadow grasses and 1.02 $\pm$ 0.02 for proper soils. The concentration ratios (CRs, dry weight basis) of ^{234}U and ^{238}U for mixed meadow grasses were 0.036–0.42 and 0.041–0.46 respectively. The correspondent geometric means (GM) were 0.13 and 0.15 with geometric standard deviations (GSD) of 2.4. The CRs of ^{226}Ra for meadow grasses were 0.031–1.0 with GM 0.20 and GSD 2.6. The CRs of ^{234}U, ^{238}U and ^{226}Ra for wild berries ranged within 0.0018–0.008 (GM is 0.0034, GSD is 1.8), 0.0018–0.008 (GM is 0.0035, GSD is 1.8) and 0.005–0.033 (GM is 0.016, GSD is 2.1) accordingly. The highest CR values of uranium for mixed meadow grasses were found in the sites with high-moor peat and sandy soils. The lowest CRs for grasses were common to loamy and peat-gley soils. The CRs for the same berry species in the sites with sandy soils exceeded the appropriate CR values in the sites with loamy soils by factors of 3–4 for uranium and 4–6 for radium. The data obtained on radionuclide accumulation by plants were used to estimate the average annual effective doses to the population from radionuclide intake through the “soil – wild berries – man” and “soil – meadow vegetation – animal – cow milk-beef – man” trophic chains. The effective doses resulting from ^{234}U, ^{238}U and ^{226}Ra intake with the wild berries for adults were estimated as 0.02–0.09 μSv y^{-1} (GM is 0.044, GSD is 1.6). It was established that only in the territory with ^{137}Cs deposition of ~1.0–1.5 kBq m^{-2} the doses resulting from ^{234}U, ^{238}U and ^{226}Ra intake with wild berries can be comparable with corresponding doses from ^{137}Cs. In the territories with higher levels of ^{137}Cs deposition the doses resulting from its intake with the wild berries are usually over the summarized doses of uranium and ^{226}Ra. The total doses for adults resulting from uranium and ^{226}Ra intake with cow milk and beef ranged between 0.2 and 7.2 μSv y^{-1} (GM is 2.0; GSD is 2.9) and the doses from ^{226}Ra usually exceeded the appropriate doses of uranium with a factor of 3–37. In the sites with ^{137}Cs deposition less than 3.7 kBq m^{-2}, the doses from ^{234}U, ^{238}U and ^{226}Ra intake with cow milk and beef were assessing as 1.1–7.2 μSv y^{-1} and they were usually higher than the doses from ^{137}Cs, which were assessing as 0.4–3.2 μSv y^{-1} for its deposition 2 kBq m^{-2}. In the territory with ^{137}Cs deposition 10–20 kBq m^{-2} and higher, the internal doses resulting from ^{137}Cs intake with cow milk and beef (10–50 μSv y^{-1}) exceeded the proper doses from natural ^{234}U, ^{238}U and ^{226}Ra.

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1. Introduction

Natural background concentrations of the primordial radionuclides in the environment differ between various countries. Natural sources of ionizing radiation constitute an essential contribution to radiation dose for the population of Belarus. A significant fraction of the radiation dose to the population arises from long exposure to the natural radionuclides. The internal dose is dominated by ^{40}K followed by radionuclides of the ^{238}U and ^{232}Th decay series (Hughers and O'Riordan, 1993). Alpha-emitting radionuclides are the most hazardous among them. They are characterized by alpha-radiation energies ranging from 4.2 to 7.5 MeV, and they can produce the serious irreversible disorders in micro-volumes of biological tissues (Jonson and De Rossa, 1997).

Irradiation of the population from alpha-emitting radionuclides is mainly characterized by inhalation intake, but in some cases irradiation of the human body as a result of peroral intake of natural radionuclides with foodstuffs and drinking water can be important (Chen et al., 2005; Gaso et al., 2000; Santos et al., 2002). The annual effective dose of the adult inhabitants of Rio de Janeiro city from the long-lived natural radionuclides (^{232}Th, ^{238}U, ^{210}Pb, ^{226}Ra and ^{228}Ra) due to consumption of the vegetables and their derived products reached 14.5 μSv (Santos et al., 2002). Both, natural and man made alpha-emitting radionuclides in our environment can present a risk to human health (Van Kaick et al., 1995).

Uranium is a great interest as a potentially hazardous pollutant that can be released to the soil from fertilizers and processes related to nuclear fuel cycle. Its radiotoxicity is low, but its chemical toxicity in dissolved form is high and should not be ignored (ATSDR, 1999).

The most important radium isotope is ^{226}Ra that is a part of ^{238}U decay series. In the man body, radium is a calcium analog, and it goes primarily to the bone. In addition to its own radiological properties (alpha and gamma decay mode), it decays into ^{222}Rn that has now been classified as a human carcinogen by the International Agency for Research of Cancer.

In the Republic of Belarus, the Annual Limits of Intake (ALI) of uranium and radium intake to the human body by consumption of foodstuffs and drinking water have been set for critical groups of population, which are 1–2 and 12–17 years old for appropriate radionuclides. These ALI are 7700, 8400 and 670 Bq per year for ^{234}U, ^{238}U and ^{226}Ra respectively (Standards, 2001).

In connection with setting limits on radionuclide intake to humans, there is a requirement for information on radionuclide activity concentrations in environmental components from which radionuclides transfer into the human body arose. Because the "soil – plant" system is one of the main radionuclide exposure pathways for human foodchains, information about the radionuclide content in soils and their bioavailability for plants is a subject of current interest. The concentration ratio (CR) of the radionuclide from soil to plant is one of the most significant parameters characterizing the radionuclide accumulation by plant needed for environmental safety estimation (Chibowski, 2000; Yanagisawa et al., 2000).

^{234}U, ^{238}U and ^{226}Ra are among the most common environmental alpha-emitting radionuclides. Bioavailability of uranium for leaves and stems of blueberry (*Vaccinium pallidum Aiton)* in native habitat was studied by Morton et al. (2002). They established that at uranium mean concentrations in soils 21.1 mg kg^{-1} the mean uranium concentrations in leaf and stem tissues were $8.65 \cdot 10^{-3}$ and $7.95 \cdot 10^{-3}$ mg kg^{-1} respectively. The corresponding mean CRs were 0.0041 and 0.0038. In the majority of plants, the non edible parts accumulate more radionuclides than the edible parts (Shanthi et al., 2011).

The data on ^{234}U, ^{238}U and ^{226}Ra content in Belarusian soils and other components of surrounding as well as information on the biological availability of uranium and radium to plants growing in the Belarusian climate are limited or practically absent. The present work was devoted to investigation of ^{234}U, ^{238}U and ^{226}Ra soil to plant transfer for often used wild berries and widely occurring meadow grasses in typical natural conditions to access of their biological availability. A further aim was to estimate the average annual effective doses of the population by ^{234}U, ^{238}U and ^{226}Ra intake through the "soil – wild berries – man" and "soil – meadow vegetation – animal – cow milk-beef – man" trophic chains and to compare them with appropriate doses by ^{137}Cs intake in the sites under investigation.

2. Materials and methods

2.1. Sampling and sample preparation

The soil and vegetation were sampled outside the zones contaminated by transuranium elements (the level of $^{238+239+240}$Pu deposition was less than 0.37 kBq m^{-2}). One group of sampling sites represented the plots with most abundant wild berries taken in usually as food by the population of Belarus. Seven plots with wild berries (bilberry, lingonberry, blueberry, and cranberry) in typical sites of their growing were chosen for investigation. Wild berries and corresponding soils were sampled in the harvesting season of 2010. The soils were sampled to a depth of 20 cm from the soil surface by a metal drill. The sub-samples of soil were sampled in the corners and in the center of plots of about 100 m^2 where the berries were taken.

Another group of sampling sites represented the main varieties of meadows widespread in different regions of Belarus with typical grasses for each type of meadow. The meadow plots were located in regions with predominant types of soil. In the south of Belarus (Gomel and Brest regions), the soils of light granulometric composition (sandy and sandy loam) as well as peaty soils (predominantly dried) are widespread, but in the north of the country (Grodno, Vitebsk region), the loamy and clay loam soils are the more frequently occurring in nature and they are enriched by natural uranium and radium to a greater extent than soils in the south part of Belarus (Shagalova, 1986). Minsk and Mogilev regions represented zone of transitional type with sandy loam, loamy sand and loamy soils occurring in comparable extents (National Atlas, 2002). Eleven meadow plots were chosen for investigation. Vegetation and soils were sampled in the harvesting season of 2008–2010. One plot was sampled at each location. The meadow grasses were cut at a height of 3–4 cm above the soil surface over a total area of 1 m^2 in the corners and in the center of plot 10 × 10 m. Every grass sub-sample was cut over an area of 0.2 m^2. All vegetation sub-samples were combined. The meadow soils were sampled to a depth of 10 cm from the soil surface by a metal drill. The sub-samples of soil were sampled in the corners and in the center of 10 × 10 m plots where the grass was cut.

The forest litter for each plot was separated from the soil sub-samples, cleaned from plant impurities and combined. The residues of soil sub-samples were also combined. They were sieved with a mesh size of 2 mm. The forest litter and sieved soil residues dried at a temperature no higher than 40 °C. The air dried samples of soil were treated in a drying chamber at 100 °C to constant weight and ignited in a muffle at 600 °C. After ignition the forest litter samples were combined with corresponding soil residues and ground into the fine powders in an agate mortar.

The samples of wild berries were washed out by running and deionized water and paper-towel dried. Then the samples of berries and grasses were dried in a drying chamber at a temperature of 100 °C to constant weight. Finally, the plant samples were separately ground and ignited in a muffle at 600 °C.