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Background concentrations of ^{226}Ra in terrestrial animals

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Abstract. Radium (^{226}Ra) accumulation by terrestrial animals was studied in the territory of the former USSR. Concentrations of ^{226}Ra in different soils, plants and animals are similar to the background level. For soils it fluctuates in range 0.5–1.5 $\times$ 10^{-12} g g^{-1} (18.3–55 Bq/kg), for plants between 0.1–1.7 $\times$ 10^{-12} g g^{-1} (3.7–62.1 Bq/kg), and for animals – 0.02–1.5 $\times$ 10^{-12} g g^{-1} (0.73–55 Bq/kg). There were no significant differences in ^{226}Ra concentration between animals of different groups. The maximal ^{226}Ra concentration is marked for the woodlouse *Hemilepistus aphganicus* from the Badkhyz reserve in Turkmenistan – 1.49 $\times$ 10^{-12} g g^{-1} of dwt (54.4 Bq/kg), the minimal one – for Colorado beetle *Leptinotarsa decemlineata* from the vicinity of Moscow – 0.02 $\times$ 10^{-12} g g^{-1} of dwt (0.73 Bq/kg). Concentration ratios for links soil – plant, plant – animal and prey – carnivore are usually close to 1. A hypothesis on the similarity of ^{226}Ra concentration in different animals is discussed.

Introduction

Radium was among the first elements of which accumulation in biota had been studied by biogeochemists. Vernadsky published already in 1929 the first work on Ra accumulation by four *Lemna* species and Kunasheva issued in 1944 the first review on Ra content in plant and animal organisms. Research on Ra accumulation by terrestrial plants was continued after World War II (Kovalevskii 1966; Titaeva & Taskaev 1983; Alexakhin 1990) but terrestrial animals were not included in these studies. Papers on Ra content in wild animals started to appear (Maslov 1972; Maslov & Maslova 1972a, b; Krivolutzkii et al. 1981; Cloutier et al. 1985, 1986; Clulow et al. 1988, 1991, 1992) only during the last decade. This is in connection with interest Ra as a contaminant and a model element for estimations of ultramicroelement migration in the biosphere.

However, background levels of ^{226}Ra in animals are practically unknown. The objectives of this paper was to demonstrate levels of its accumulation in animals from different groups and biomes.

Figure 1. Approximate location of sample sites. 1 – Ceske Budejovice, Czech Republic; 2 – Syktyvkar, Komi Republic, Russian Federation; 3 – Zvenigorod, Moscow Region, Russian Federation; 4 – Central-Chernozem Reserve, Kursk Region, Russian Federation; 5 – Lenkoran Reserve, Azerbaijan; 6 – Firjuza, Turkmenistan; 7 – Badkhyz Reserve, Turkmenistan; 8 – Repetek Reserve, Turkmenistan.

Material and methods

All samples were collected in 1968–1988 during field studies in different parts of the former USSR and Czech Republic (Figure 1). All plots were state reserves or areas of biological stations. The Station of Institute of Biology of Komi Center of the Russian Academy of Sciences is situated in a taiga forest biome close to Syktyvkar (Komi Republic, Russian Federation). The Zvenigorod Station of Moscow University is situated in a mixed forest biome near Moscow (Russian Federation). The Central-Chernozem Reserve is in a forest-steppe biome near Kursk (Russian Federation). The Lenkoran Reserve is in the moist subtropical zone near Lenkoran (Azerbaijan). The Firjuza Station is in the dry subtropical zone near Ashgabat (Turkmenistan). The Repetek Reserve is in the sand desert region near Chardzhou (Turkmenistan) and the Badkhyz Reserve is in the dry subtropic/clay desert zone near Kushka (Turkmenistan). Some samples were collected together with Czech colleagues in deciduous forest or agricultural landscapes near Ceske Budejovice, Czech Republic (CR).

Millipedes *Ambliulus continentalis* collected in the Lenkoran Reserve were kept in plastic 10-L pots for 30 days (50 specimens per pot) at 25 °C. Litter of *Quercus castanofolia* was used as a feed. Faeces were removed every day to prevent their consumption by animals. Loss of litter mass in pots was considered as consumption. Data on consumption and defecation of this species (Striganova & Rakhmanov 1973) were used for the evaluation of the radionuclide balance in the animal body.

Soil was collected from the top 20-cm soil layer. Sample weights were up to 10 kg. Plants and plant litter (up to 10 kg of fresh weight) were gathered around a soil sampling site. All animals collected for ^{226}Ra analyses were dominant or pest species with high population numbers and biomass. Weight of one sample was up to 2–4 kg of wet weight. All samples were dried at 105 °C and ground with mortar and pestle.

Ground samples of soil, plants and animals were ashed in a muffle furnace at 400 °C. Sample sizes included 10–20 g of soil or ash of plants or animals for one ^{226}Ra analysis. The weight requirement was the main limitation for a number of samples of plants or animals because every analysis was duplicated. Samples were stored in ash form.

The analytical procedure was as follows: 16 g of Na_2CO_3 and 120 g NaOH were fused for 40 min in nickel pots at 800 °C. After cooling of the fused mixture, 10 g of soil or 10–20 g of ash was placed in a pot and the pots were heated for 1 hour in a furnace at 800–800 °C. Then every pot was put on a thermoglass (1000 mL) and the glass was brought to volume with boiling super-demineralized water. The solution was neutralized with 200–250 mL of concentrated HNO_3 up to the end of CO_2 discharge. Then the solution pH was adjusted to 1–2 with HNO_3. Precipitates of MnO_2 were dissolved with NH_4Cl. The cooled solution was filtered through dense filter paper and put into a sparger for collection of ^{222}Rn evolved during decay of ^{226}Ra. Then ^{222}Rn was determined with α-spectrometer 'Alpha-I' with a sensitivity of 2 $\times$ 10^{-12} g/g (73 Bq per kg) and error $\pm$ 15% (Titaeva & Taskaev 1984).

Results

Radium-226 contents were similar in all soils from the investigated plots (Table 1), except for a soil with a high Ra level from the Komi Republic.

There were no significant differences between plant and plant litter samples from different investigated plots (Table 2). Plants do not accumulate Ra and a comparison of data from Table 1 and Table 2 indicates that the concentration ratio in soil:plant is somewhat less than 1 for leaves and significantly lower than 1 for wood.

The same picture is shown for ^{226}Ra in terrestrial and soil animals from various investigated plots (Table 3). Concentrations of the element do not contrast sharply for animals. The highest ^{226}Ra concentration was detected for the woodlouse *Hemilepistus aphganicus* (the Badkhyz reserve, Turkmenistan) – 1.49 $\times$ 10^{-12} g g^{-1} of dwt (54.4 Bq/kg). It was only twice as high as the

Table 1. Level of ^{226}Ra in soils of research area.

Soil and plot names	^{226}Ra (10^{12} g g^{-1} dwt)	^{226}Ra Bq kg^{-1}
Podzol gley soil (Komi Republic)*	0.6–4.2	21.9–153.3
Meadow alluvial soil (Komi Republic)*	2.1–2.7	76.7–98.6
Podzol soil (Zvenigorod)	1.0	35.4
Chernozem soil (Central Chernozem Reserve)	1.5	54.8
Serozem sandy soil (Repetek Reserve)	0.7	25.6
Serozem clay soil (Badkhyz Reserve)	0.8	29.2
Yellow clay soil (Lenkoran Reserve)	0.8	29.2

* From Titaeva & Taskaev 1984.

Table 2. ^{226}Ra concentration in plant and plant litter samples from the research plots.

Sample and plot names	^{226}Ra (10^{-12} g g^{-1} dwt)	^{226}Ra Bq kg^{-1}
Pinus sibirica wood (Komi Republic)*	0.16	5.8
Pinus silvestris wood (Ceske Budejovice, CR)	0.01	0.37
Pinus silvestris wood (Zvenigorod)	0.20	7.3
Picea abies wood (Ceske Budejovice, CR)	0.04	1.5
Betula sp. leaves (Komi Republic)*	0.71	25.9
Betula sp. wood (Komi Republic)*	0.09	3.3
Gramineae stems (Komi Republic)*	0.11–0.16	4.0–5.8
Carex spp. stems (Komi Republic)*	0.14–0.21	5.1–7.7
Gramineae litter (Central Chernozem Reserve)	1.00	36.5
Grass and herb mixed litter (Central Chernozem Reserve)	1.00	36.5
Mixed forest litter (Ceske Budejovice, CR)	0.32	11.7
Quercus robur leaves (Central Chernozem Reserve)	1.50	54.8
Quercus robur wood (Zvenigorod)	0.30	10.9
Populus tremuloides wood (Zvenigorod)	0.10	3.7
Haloxylon aphyllum branches (Repetek Reserve)	0.34	12.4
Haloxylon persicum (Repetek Reserve)	1.70	62.1
Carex arenaria (Repetek Reserve)	0.29	10.6
Bromus tectorum (Repetek Reserve)	0.54	19.7
Poa sp. (Ceske Budejovice, CR)	0.04	1.5
Ammodendron karelini branches (Repetek Reserve)	0.14	5.1
Quercus castanofolia litter (Lenkoran)	0.78	28.5
Parrotia persica litter (Lenkoran)	0.78	28.5

* Titaeva & Taskaev 1984.

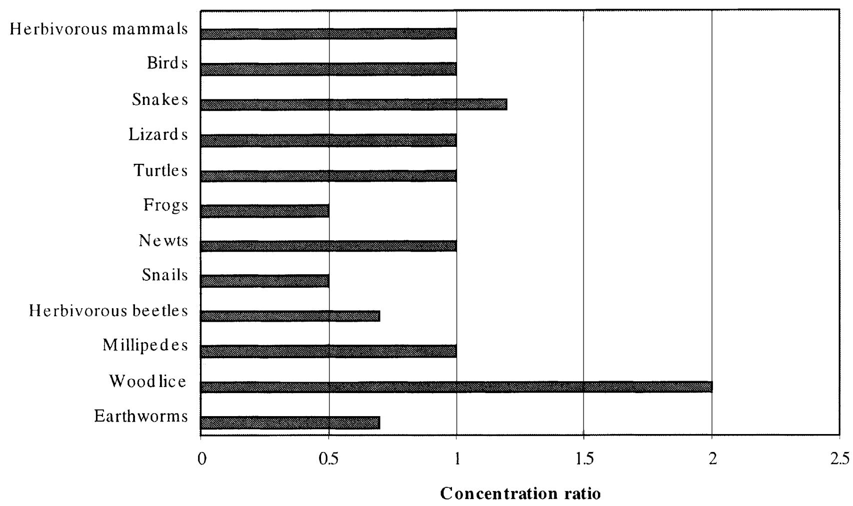

Figure 2. ^{226}Ra Concentration ratio (Ra in animal/Ra in food substrate) for different animal groups.

concentration in the majority of species under study. The lowest concentration was found in Colorado beetle *Leptinotarsa decemlineata* [0.02×10^{-12} g g^{-1} of dwt (0.73 Bq/kg)] from the vicinity of Moscow. Only animals from regions with high content of natural radionuclides (the Komi Republic) have relatively high Ra concentrations in their bodies.

There were no noticeable differences in Ra content in vertebrate and invertebrate animals. Concentration ratios for animals from different groups relative to that substrate were slightly less than 1 almost in all cases (Figure 2). The Ra concentration in the diplopod *Ambliulus continentalis* is slightly lower than that in the litter fed and the faeces excreted. This is quite different from the concentration ratios observed from uranium (U) and thorium (Th) in the same species (Figure 3). Amounts of Ra consumed and excreted by the millipedes were very similar (Figure 4) but the ratio of Ra content in the whole animal body to that of daily intake of Ra (Intake Ratio) was less than 1 (Figure 5). The concentration ratio for Ra was almost equal to 1.

Discussion

Soil and plant data obtained in this study are similar to data have been published by other authors (Baranov & Zeitlin 1941; Kovalevskii 1966; Titaeva & Taskaev 1984; Kabata-Pendias & Pendias 1984; Alexakhin 1990). Radium concentrations in soils fluctuated in the ranges $0.5–1.5 \times 10^{-12}$ g

Table 3. ^{226}Ra concentration in terrestrial animals from the research plots.

Species and plot names	^{226}Ra (10^{-12} g g^{-1} dwt)	^{226}Ra Bq kg^{-1}
Earthworms		
Lumbricus terrestris (Central Chernozem Reserve)	0.74	27.1
Lumbricus rubellus (Zvenigorod)	0.20	7.3
Octolasium lacteum (Central Chernozem Reserve)	0.70	25.6
Nicodrilus caliginosus (Badkhyz Reserve)	0.60	21.9
Woodlice		
Protracheoniscus orientalis (Badkhyz Reserve)	0.94	34.3
Protracheoniscus sp. (Firjusa)	0.79	28.8
Protracheoniscus sp. (Badkhyz Reserve)	0.83	30.3
Hemilepistus aphganicus (Badkhyz Reserve)	1.49	54.4
Diplopods		
Schizophillum caspium (Lenkoran Reserve)	0.97	35.4
Ambliulus continentalis (Lenkoran Reserve)	0.66	24.1
Beetles		
Scarabaeus sp. (Repetek Reserve)	0.29	10.6
Copris hispanus (Badkhyz Reserve)	0.29	10.6
Copris lunaris (Badkhyz Reserve)	0.23	8.4
Curculionidae larvae (Repetek Reserve)	0.63	23.0
Blaps fausti (Repetek Reserve)	0.57	20.8
Blaps sp. (Repetek Reserve)	0.14	5.1
Leptinotarsa decemlineata (Zvenigorod)	0.02	0.73
Molluscs		
Succinea pfeifferi (Zvenigorod)	0.08	2.9
Bradybaena fruticum (Central Chernozem Reserve)	0.15	5.5
Arianta arbustorum (Ceske Budejovice, CR)	0.24	8.8
Deroceras reticulatus (Zvenigorod)	0.63	23.0
Amphibians		
Triturus vulgaris (Central Chernozem Reserve)	0.63	23.0
Rana temporaria (Komi Republic)	0.88	32.1
Rana temporaria (Komi Republic)[a]	0.06	2.2
Rana temporaria (Komi Republic)[b]	0.96	35.0
Rana temporaria (Zvenigorod)	0.06	2.2
Rana ridibunda (Badkhyz Reserve)	0.49	17.9
Bufo bufo (Zvenigorod)	0.11	4.0

Table 3. (continued).

Species and plot names	^{226}Ra (10^{-12} g g^{-1} dwt)	^{226}Ra Bq kg^{-1}
Reptilians		
Testudo horsfieldi (Badkhyz Reserve)	0.39	14.2
Lacerta vivipara (Komi Republic)[a]	0.05	1.9
Teratoscincus scincus (Repetek Reserve)	1.00	36.5
Agama sanguinolenta (Badkhyz Reserve)	0.54	19.7
Phrynocephalus interscapularis (Repetek Reserve)	0.03	1.1
Phrynocephalus mystaceus (Badkhyz Reserve)	0.31	11.3
Eumeces taeniolatus (Badkhyz Reserve)	0.11	4.0
Eremias guttulata (Badkhyz Reserve)	0.40	14.6
Eremias strauchi (Badkhyz Reserve)	0.09	3.3
Eremias grammica (Badkhyz Reserve)	0.40	14.6
Coluber jugularis (Badkhyz Reserve)	0.51	18.6
Birds		
Anas crecca (Komi Republic)[a]	0.15	5.6
Anas crecca (Komi Republic)[b]	1.77	65
Anas penelope (Komi Republic)[a]	0.12	4.6
Mergus merganser (Komi Republic)[a]	0.11	3.9
Mergus merganser (Komi Republic)[b]	1.87	68.5
Accipiter gentilis (Komi Republic)[a]	0.04	1.5
Accipiter gentilis (Komi Republic)[b]	0.17	6.1
Accipiter nisus (Komi Republic)[a]	0.05	1.9
Accipiter nisus (Komi Republic)[b]	0.19	7.1
Bubo bubo (Komi Republic)[a]	0.15	5.6
Bubo bubo (Komi Republic)[b]	1.52	55.5
Strix uralensis (Komi Republic)[a]	0.15	5.4
Strix uralensis (Komi Republic)[b]	1.57	57.5
Dendocopos major (Komi Republic)[a]	0.05	1.9
Dendocopos major (Komi Republic)[b]	0.45	16.5
Dryocopus martius (Komi Republic)[a]	0.05	1.9
Dryocopus martius (Komi Republic)[b]	0.61	22.0
Perisoreus infaustus (Komi Republic)[a]	0.03	0.95
Perisoreus infaustus (Komi Republic)[b]	0.10	3.5
Loxia curvirostra (Komi Republic)[a]	0.03	0.95
Loxia curvirostra (Komi Republic)[b]	0.06	2.0
Loxia pytyopsittacus (Komi Republic)[a]	0.03	0.95
Loxia pytyopsittacus (Komi Republic)[b]	0.06	2.0
Pyrrhula pyrrhula (Komi Republic)[a]	0.02	0.75
Parus major (Komi Republic)[a]	0.02	0.75
Parus major (Komi Republic)[b]	0.04	1.5
Motacilla alba (Komi Republic)[a]	0.03	0.95

Table 3. (continued).

Species and plot names	^{226}Ra (10^{-12} g g^{-1} dwt)	^{226}Ra Bq kg^{-1}
Motacilla alba (Komi Republic)[b]	0.04	1.5
Turdus philomelos (Central Chernozem Reserve)	0.06	2.2
Cinclus cinclus (Komi Republic)[a]	0.08	2.8
Cinclus cinclus (Komi Republic)[b]	1.31	48.0
Phasianus colchicus (Ceske Budejovice, CR) bones	1.04	38.0
Phasianus colchicus (Ceske Budejovice, CR) head	0.57	20.8
Phasianus colchicus (Ceske Budejovice, CR) carcass	0.004	0.15
Phasianus colchicus (Ceske Budejovice, CR) stomach	0.02	0.73
Phasianus colchicus (Ceske Budejovice, CR) wings	0.95	34.7
Lirurus tetrix (Komi Republic)[a]	0.13	4.8
Lirurus tetrix (Komi Republic)[b]	0.94	34.5
Tetrastes bonasia (Komi Republic)[a]	0.05	1.9
Tetrastes bonasia (Komi Republic)[b]	0.58	21.5
Lagopus lagopus (Komi Republic)[a]	0.09	3.2
Lagopus lagopus (Komi Republic)[b]	0.45	16.0
Tetrao urogallus (Komi Republic)[a]	0.01	3.7
Tetrao urogallus (Komi Republic)[b]	0.89	32.5
Mammals		
Talpa europaea (Komi Republic)[a]	0.24	9.1
Talpa europaea (Komi Republic)[b]	1.66	61
Sorex araneus (Komi Republic)[a]	0.08	2.95
Sorex araneus (Komi Republic)[b]	1.26	46.0
Lepus timidus (Komi Republic)[a]	0.06	2.1
Lepus timidus (Komi Republic)[b]	0.76	28.0
Sciurus vulgaris (Komi Republic)[a]	0.02	0.75
Sciurus vulgaris (Komi Republic)[b]	0.30	11.0
Eutamias sibiricus (Komi Republic)[a]	0.05	1.65
Eutamias sibiricus (Komi Republic)[b]	0.61	22.0
Apodemus sylvaticus (Ceske Budejovice, CR)	0.04	1.46
Microtus oeconomus (Komi Republic)[a]	0.18	6.5
Microtus oeconomus (Komi Republic)[b]	1.16	42.5
Microtus agrestis (Komi Republic)[a]	0.18	6.5
Microtus agrestis (Komi Republic)[b]	1.01	37.0
Microtus arvalis (Ceske Budejovice, CR)	0.03	1.1
Clethrionomys glareolus (Komi Republic)[a]	0.13	4.8
Clethrionomys glareolus (Komi Republic)[b]	1.22	44.5
Clethrionomys rutilus (Komi Republic)[a]	0.13	4.6
Clethrionomys rutilus (Komi Republic)[b]	1.07	39.0
Arvicola terrestris (Komi Republic)[a]	0.13	4.5
Arvicola terrestris (Komi Republic)[b]	1.01	37.0

Table 3. (continued).

Species and plot names	^{226}Ra (10^{-12} g g^{-1} dwt)	^{226}Ra Bq kg^{-1}
Martes martes (Komi Republic)[a]	0.06	2.1
Martes martes (Komi Republic)[b]	0.93	33.5
Mustela erminea (Komi Republic)[a]	0.10	3.5
Mustela erminea (Komi Republic)[b]	0.81	29.5
Mustela nivalis (Komi Republic)[a]	0.10	3.5
Mustela nivalis (Komi Republic)[b]	0.90	33.0
Mustela lutreola (Komi Republic)[a]	0.08	2.8
Mustela lutreola (Komi Republic)[b]	0.45	16.5
Lutra lutra (Komi Republic)[a]	0.07	2.6
Lutra lutra (Komi Republic)[b]	0.45	16.5
Vulpes vulpes (Komi Republic)[a]	0.04	1.5
Vulpes vulpes (Komi Republic)[b]	0.25	9.0
Capreolus capreolus (Central Chernozem Reserve) femur	0.08–0.28*	2.9–10.2
Alces alces (Komi Republic)[a]	0.03	0.95
Alces alces (Komi Republic)[b]	0.30	11.0
Alces alces (Central Chernozem Reserve) femur	0.21–0.35*	7.7–12.8
Rangifer tarandus (Komi Republic)[a]	0.03	1.1
Rangifer tarandus (Komi Republic)[b]	0.30	11.0
Sus scrofa (Central Chernozem Reserve) femur	0.09–0.13*	3.3–4.8
Sus scrofa (Ceske Budejovice, CR) femur	0.13–0.24*	4.8–8.8
Bovine meat (Ceske Budejovice, CR)	0.01	3.65
Bovine bones (Ceske Budejovice, CR)	0.08	2.9
Bovine liver (Ceske Budejovice, CR)	0.007	0.26
Bovine spleen (Ceske Budejovice, CR)	0.005	0.18
Bovine kidney (Ceske Budejovice, CR)	0.012	0.44

[a] Recalculated data for background sites (from Maslov & Maslova 1972a, b; Alexakin 1990).
[b] Recalculated data for sites with a high level of natural radionuclides (from Maslov & Maslova 1972a, b; Alexakin 1990).
* Data from two animals.

g^{-1} (18.3–55 Bq/kg), excluding a soil with high Ra content, and in plants from 0.1–1 $\times$ 10^{-12} g g^{-1} (3.7–62.1 Bq/kg). The concentration ratio on a dry weight basis in the link soil – plant was always < 1, but was sometimes higher than 1 (up to 8.5) on ash basis (Titaeva & Taskaev 1984; Alexakhin 1990).

Published data on Ra content in animals and concentration ratios in the link plant – animals and prey – carnivore also are close to our data. Only animals from ecosystems with a high level of natural radionuclides in soil have relatively high Ra concentrations in their bodies (Maslov 1972; Maslov

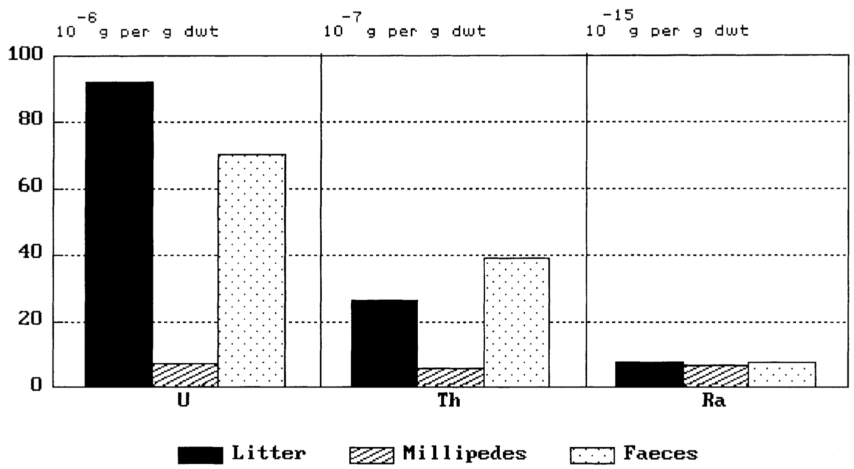

Figure 3. Relations of natural radionuclide concentrations in millipedes *Ambliulus continentalis*, their food and faces during a 30 day experiment (after Krivolutzkii et al. 1981)

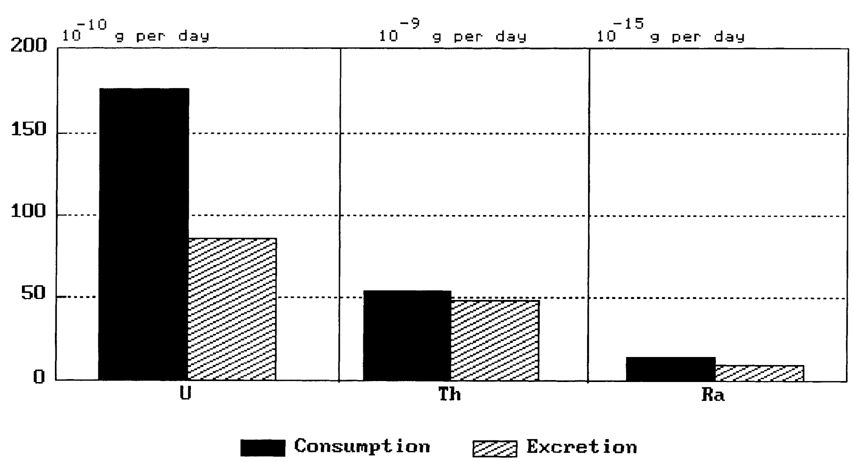

Figure 4. Daily balance of natural radionuclides in millipedes *Ambliulus continentalis* during 30 day experiment (after Krivolutzkii et al. 1981).

& Maslova 1972a, b). The concentration ratio for most animals studied was less than or close to 1 (Maslov & Maslova 1972a, b; Krivolutzkii et al. 1981; Cloutier et al. 1985, 1986; Clulow et al. 1988, 1991, 1992). Geochemists consider Ra as an element migrating in landscapes and food chains like calcium (Perelman 1972), but our data show that species which accumulate Ca (diplopods, woodlice, molluscs, vertebrates) are similar in Ra body concentration to other terrestrial animals. Our data on natural radionuclide balance in the millipede *Ambliulus continentals* indicated a very low rate of

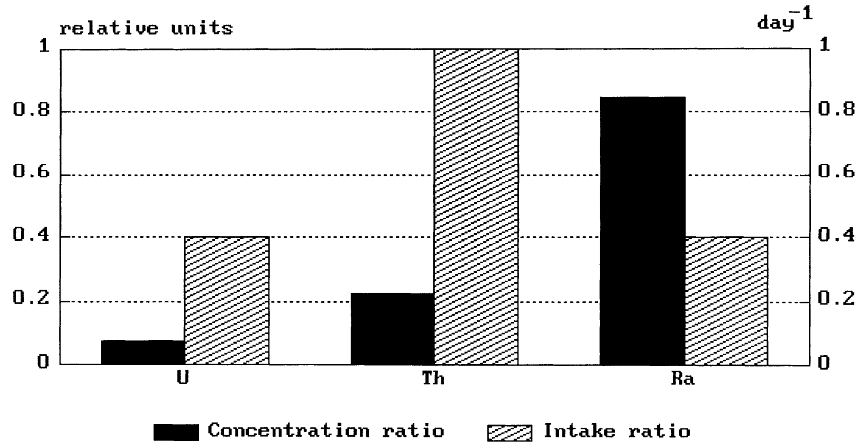

Figure 5. Concentration ratio and intake ratio of natural radionuclides for milipedes *Ambliulus continentalis*.

Ra accumulation in this Ca accumulating species. Although concentration ratios for Th and U for this species are much lower than that for Ra, the intake ratio for uranium is almost equal and that of thorium three times higher than that for radium. It is possible that processes of body accumulation of U, Th and Ra are quite different and not connected with Ca.

Why does radium-226 not accumulate in food chains?

Geochemists noted that radium is strongly sorbed by soil and has not formed own minerals in soil (Perelman 1972; Pavlotskaya 1974). In their opinion, Ra is included in the migration of its element-carrier (Ca) due to its very low concentration in soil. At such ultralow concentrations (for comparison concentration of Ra is 10^{-12} g g^{-1} and of gold – 4×10^{-9} g g^{-1} (Perelman 1972; Fortescue 1980) other geochemical relations dominate. Low accumulation of Ra in Ca-containing tissues may be related to the inability of Ra to form its own minerals. Strong sorption of Ra by organo-mineral particles of soil and low solubility of Ra salts prevent its transfer through biological barriers both in animals and plants (Perelman 1972; Alexakhin 1990). Thus it is difficult to explain Ra migration through food chains by its geochemical connection with Ca. However water-soluble forms of Ra were accumulated in plants 2–10 times higher than forms of Ra applied with labelled fertilizers. Regular water-spraying increased Ra accumulation in plants in comparison with plants without regular water-spraying (Alexakhin 1990).

Our hypothesis on Ra migration in terrestrial food chains is that all available Ra forms are water-soluble at ultra low concentrations. In these forms radium enters via biological barriers in plants. In plants it is bound by biological molecules and accumulates in such organic forms. After animal consumption of plants only water-soluble radium forms transfer through biological barriers in the animal gut. In animals the element is bound by biological molecules as in plants, not forming its own minerals. This may explain why Ra concentrations in different animals are so close to each other. Chemical, not biological processes, may possibly be responsible for Ra fate in food chains and perhaps water is the real carrier of the element in food chains.

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