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Concentration of selected radionuclides in skulls of European badger *Meles meles* from eastern Poland

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Abstract: Concentration activity levels of selected radionuclides in skulls of European badger from eastern Poland were determined. It was found that studied skull bones contained on average 1.6 ± 1.7 Bq kg⁻¹ of ¹³⁷Cs (range: 0–6.4 Bq kg⁻¹), 2.2 ± 0.87 Bq kg⁻¹ of ⁹⁰Sr (range: 0.93–3.9 Bq kg⁻¹), and 8.4 ± 4.1 Bq kg⁻¹ of ²³⁹⁺²⁴⁰Pu (range: 0–53 Bq kg⁻¹). These values prove to be rather low in comparison with earlier literature data on anthropogenic radionuclides (being a result of the Chernobyl fallout) present in skulls of other mammals. This suggests that migration processes of anthropogenic radionuclides have caused a diminishing of contamination in the upper layer of soil.

Key words: Anthropogenic radionuclide, skull bone, European badger, eastern Poland

1. Introduction

Environment contamination with anthropogenic radionuclides, such as plutonium isotopes (²³⁸Pu and ²³⁹⁺²⁴⁰Pu), strontium (⁹⁰Sr), and cesium (¹³⁷Cs), has been present in Europe since the Second World War. Generally, there were two sources of these radionuclides: global fallout from nuclear tests and the Chernobyl disaster. Levels of radionuclide activity concentration (artificial as well as natural ones) in various components of the environment are rather well known (1). However, still interesting is the way of transportation of these radionuclides from soil into living organisms. Especially important are those radionuclides that are named bone-seekers: plutonium and strontium. Skeletons of higher vertebrates such as mammals and birds are good models for such studies. The investigation of the presence of ²³⁸Pu, ²³⁹⁺²⁴⁰Pu, ²⁴¹Am, and ⁹⁰Sr in bones of free-living mammals has been conducted since the late nineties of the last century. Carnivores and opportunistic omnivorous mammals of medium size are especially important groups of animals for such studies. On the other hand, concentration levels and the way of radionuclide accumulation in bodies of game animals are also important. These animals are valuable resources of

nourishment and have other benefits with the industrial utilization of their bodies (2–8).

Detailed study has been performed on bones and internal organs of red fox (*Vulpes vulpes*) (8,9). Unfortunately, data concerning other medium-sized mammals, including European badger (*Meles meles*), are missing. For this reason, the purpose of our study was to present the first determination of radionuclides in skulls of more than ten individuals of European badger, as well as to compare the results with other mammals such as foxes or ungulates. The latter are rather distant in taxonomy but can exist in similar habitats.

2. Materials and methods

Skulls of fifteen individuals of European badger were obtained from southeastern Poland (only two samples came from a northeastern part of Poland). In Poland, the European badger is a game animal. A part of the specimens (six individuals) were gained by local hunters in the hunting season (1 September to 30 November) in 2009–2011, while the others (nine individuals) were victims of road casualties found by the authors. Collecting points of analyzed material are presented in the Figure.

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Figure. Geographical distribution of sample collection points.

Collected material was mechanically cleaned from soft tissue and the remnants were removed by boiling in 10% NaOH solution. Skull bones were subjected to determination of anthropogenic radionuclides: ^{137}Cs , ^{238}Pu , $^{239+240}\text{Pu}$, and ^{90}Sr coming from atmospheric fallout, being a result of nuclear tests and the Chernobyl disaster in the last century. Before measurements the samples were prepared properly, according to the procedure's requirements.

2.1. Determination of gamma-emitting isotopes (including ^{137}Cs)

Skull bones were crushed with a hammer and ground with an electric grinder. Ground and homogenized bones (about 30 g) were placed in a flat container of 50 mm in diameter and about 5 mm in height and placed on top of a detector for gamma spectrometric measurement. Measurements were performed with the gamma spectrometer (Canberra/Silena) equipped with the HPGe detector of 87 cm³ active volume, 17.5% relative efficiency, and 1.8 keV FWHM resolution. The Canberra Genie-2000 software was used for determining the activity of natural and artificial radioactive isotopes.

2.2. Determination of alpha-emitting $^{239+240}\text{Pu}$ and ^{238}Pu

Determination of plutonium in environmental samples requires separation of its isotopes in a pure form and fixing them on a plate, which enables alpha spectrometric measurements (10,11). This was realized by a radiochemical multistage procedure. In the first stage a sample was ashed (450–500 °C) and dissolved in concentrated HNO_3 . Mass of ash taken for analysis was 3–7 g (8–20 g dry mass). After

dissolution (boiling, preceded by a few days of conditioning at room temperature), ferric ions were introduced and plutonium was coprecipitated with ferric hydroxide. The supernatant was retained for determination of radioactive strontium. The precipitate was dissolved in nitric acid and next coprecipitation with calcium oxalate was performed. After calcination of this precipitate at 450 °C the sample was dissolved in nitric acid, and sodium nitrite was added to change the oxidation state of plutonium to the Pu^{4+} form. Next, the solution was introduced on an anion-exchange column filled with Dowex 1×8 (50–100 mesh). Before sample introduction the column was washed with 8 M HNO_3 in order to exchange chloride onto nitrate ions. After the sample was passed through the column, the next solutions were introduced in order as follows: 8 M HNO_3 , 6 M HCl, and concentrated HCl, with the purpose of eliminating trace elements (Th, Am). In the final stage Pu was eluted from the column using concentrated HCl solution containing 0.1 M ammonium iodide. The obtained solution was evaporated to dryness. Finally, Pu was coprecipitated with neodymium fluoride and filtrated by membrane filters (Eichrom). After drying the filter was fixed to the aluminum plate and alpha spectrometric measurement was carried out. Measurements were performed using the 7401 Canberra Alpha Spectrometer with a 1520 mixer/router and S-100 multichannel analyzer equipped with a PIPS detector of 17 keV FWHM resolution. For quantitative analysis Canberra Genie-2000 software was used. Chemical yield was monitored by addition of a standard ^{242}Pu solution to the sample.

2.3. Determination of beta-emitting ^{90}Sr

Determination of beta radioactive ^{90}Sr in environmental samples demands isolating it from other, natural beta-emitting isotopes appearing in the sample matrix (12,13). To the supernatant kept during plutonium determination an yttrium carrier was added and yttrium oxalate was precipitated. After dissolution of the precipitate in concentrated HNO_3 the Y^{3+} was extracted with three portions of TBP (saturated with HNO_3). Next, the organic phase was diluted with ethyl alcohol and yttrium oxalate was then precipitated. Filtered and dried precipitate was weighted to calculate a radiochemical yield and submitted to radiation measurement after mixing with a water and scintillation cocktail (Insta-Gel Plus, PerkinElmer) in a low-potassium standard glass vial. Measurements were performed by the use of a Quantulus (Wallac, PerkinElmer) ultra-low-level spectrometer during 300 min. The applied procedure allowed for obtaining a minimum detectable amount of 0.02 Bq kg⁻¹ and mean radiochemical yield of about 80%.

3. Results

Results of determination of selected, important gamma-emitting nuclides (^{40}K , ^{137}Cs , ^{210}Pb , and ^{226}Ra) are presented in Table 1, and alpha-radiating nuclides (^{238}Pu and $^{239+240}\text{Pu}$) and beta ^{90}Sr are shown, respectively, in Table 2. All results were calculated taking into account a dry mass of skull bone sample. General ascertainment coming from our measurements is that measured values in many cases only slightly exceeded the minimum detectable activity (MDA) value. Therefore, we should realize that the uncertainty of measurement was rather large.

As seen from Table 1, average ^{137}Cs concentration in European badger skull samples was $1.6 \pm 1.7 \text{ Bq kg}^{-1}$ (range: $0\text{--}6.4 \text{ Bq kg}^{-1}$, median value: 1.6 Bq kg^{-1}). Presented results of ^{40}K concentration (Table 1) show mean activity equal to $21 \pm 11 \text{ Bq kg}^{-1}$ (median: 18 Bq kg^{-1}) and a rather large range of $6.9\text{--}39 \text{ Bq kg}^{-1}$. Mean concentration of ^{90}Sr ($2.2 \pm 0.87 \text{ Bq kg}^{-1}$, range: $0.93\text{--}3.9 \text{ Bq kg}^{-1}$), shown in Table 2, is rather low, but individual results exceeded the MDA values. Plutonium concentration in skull bones shown in

Table 2 was found to be on average $8.5 \pm 8.8 \text{ Bq kg}^{-1}$ d.m. (^{238}Pu) in the range of $0\text{--}30 \text{ Bq kg}^{-1}$, and $8.4 \pm 4.1 \text{ Bq kg}^{-1}$ ($^{239+240}\text{Pu}$) in the range of $0\text{--}53 \text{ Bq kg}^{-1}$.

Concerning our results for ^{40}K (presented in Table 1), amounting averagely to $21 \pm 11 \text{ Bq kg}^{-1}$, data for Lithuanian foxes were found to be higher: $46\text{--}111 \text{ Bq kg}^{-1}$. Comparing the results of ^{137}Cs and ^{40}K in skull bones, a lack of any important correlation between these two isotopes was found ($r = 0.172$, $n = 11$, $P = 0.687$). Four values from the set of results (for ^{137}Cs) were eliminated as they did not exceed a limit of detection (see Table 1). Additionally, an interesting fact was observed: increase in latitude correlated with decrease in concentration of ^{137}Cs (-0.857 , $n = 11$, $P = 0.03$) and ^{226}Ra (-0.945 , $n = 11$, $P = 0.004$). However, such a relation was observed neither in the case of ^{210}Pb (-0.577 , $n = 15$, $P = 0.23$) nor, especially, in the case of ^{90}Sr (-0.04 , $n = 15$, $P = 0.888$).

Surprisingly, the determined high level of ^{238}Pu concentration in our studies is rather a result of a large uncertainty of determination and overestimation arising.

Table 1. Gamma-emitting isotope contents in skull bones of European badger *Meles meles* from eastern Poland.

Sample no.	^{40}K	^{137}Cs	^{210}Pb	^{226}Ra
	[Bq kg ⁻¹ dry weight]			
1	27 ± 4.2	1.6 ± 0.25	6.9 ± 2.3	<5.5
2	12 ± 3.6	<0.26	11 ± 2.1	<4.8
3	7.5 ± 3.8	0.13 ± 0.20	14 ± 2.4	2.8 ± 3.0
4	37 ± 4.6	0.22 ± 0.23	1.0 ± 2.3	0.63 ± 3.2
5	21 ± 4.0	<0.39	0.78 ± 2.3	<5.7
6	37 ± 6.3	0.16 ± 0.34	16 ± 3.6	1.6 ± 4.7
7	39 ± 4.9	2.0 ± 0.31	6.8 ± 2.8	<7.2
8	12 ± 3.9	1.2 ± 0.25	9.8 ± 2.4	2.9 ± 3.1
9	18 ± 3.7	0.39 ± 0.21	9.8 ± 2.2	0.80 ± 2.8
10	14 ± 4.0	<1.2	2.9 ± 1.6	2.2 ± 3.1
11	28 ± 4.1	6.4 ± 0.65	10 ± 2.6	11 ± 3.6
12	17 ± 4.6	<2.0	16 ± 2.3	8.8 ± 3.3
13	6.9 ± 4.7	2.1 ± 0.57	16 ± 2.4	8.9 ± 3.3
14	13 ± 3.6	0.74 ± 0.51	19 ± 2.2	12 ± 3.0
15	31 ± 4.9	2.9 ± 0.56	15 ± 2.3	11 ± 3.2
Mean \pm SD	21 ± 11	1.2 ± 1.7	10 ± 5.9	3.9 ± 4.7
Median	18	0.56	10	1.9
Minimum	6.9	0	0.78	0
Maximum	39	6.4	19	12

Table 2. Plutonium isotopes and ^{90}Sr contents in skull bones of European badger *Meles meles* from eastern Poland (on dry mass basis).

Sample no.	^{238}Pu	$^{239+240}\text{Pu}$	^{90}Sr
	[mBq kg $^{-1}$]	[mBq kg $^{-1}$]	[Bq kg $^{-1}$]
1	7.0 ± 2.9	5.0 ± 3.3	3.1 ± 0.28
2	11 ± 4.7	4.0 ± 5.6	3.9 ± 0.34
3	18 ± 5.9	53 ± 10	1.9 ± 0.30
4	<76	14 ± 17	1.4 ± 0.23
5	7 ± 3	7.0 ± 4.8	2.0 ± 0.22
6	2.0 ± 1.4	<9	1.7 ± 0.21
7	4.0 ± 1.6	2.0 ± 1.9	2.0 ± 0.23
8	12 ± 3.0	4.0 ± 4.0	3.9 ± 0.22
9	21 ± 5.0	20 ± 6.4	1.4 ± 0.22
10	<6	<10	2.6 ± 0.23
11	30 ± 4.2	2.0 ± 1.2	1.8 ± 0.16
12	1 ± 0.64	4.0 ± 1.8	1.6 ± 0.09
13	10 ± 1.9	4.0 ± 2.0	0.93 ± 0.07
14	1 ± 0.67	1.0 ± 1.0	2.8 ± 0.16
15	4 ± 1.4	6.0 ± 2.3	2.6 ± 0.19
Mean ± SD	8.5 ± 8.8	8.4 ± 4.1	2.2 ± 0.87
Median	7.0	4.0	2.0
Minimum	0	0	0.93
Maximum	30	53	3.9

Such a high concentration is not likely, as the global fallout was mainly the source of soil contamination with plutonium (9,16). We do not find any correlations between ^{137}Cs and ^{40}K in the skull bones of badgers. Similar relationship between ^{137}Cs and ^{40}K was observed in the case of fox skulls (8).

4. Discussion

Similarly to our results of ^{137}Cs determination in skulls (see Table 1), five individuals of foxes from Lithuania revealed only a small accumulation of ^{137}Cs (from 2.6 to 5.2 Bq kg $^{-1}$ ash). Concentration in muscle tissue was a little higher and equal on average to 5.8 Bq kg $^{-1}$ (8). The concentration of ^{137}Cs determined in the bones of Polish foxes ranged from below 1.5 Bq kg $^{-1}$ to up to 41 ± 3.6 Bq kg $^{-1}$ (9). However, it is obvious that cesium is not a bone-seeking element and it accumulates rather in soft tissue of mammals (4,7,9). It was observed that large mammals for which mushrooms are an important component of their diet (for example,

boars *Sus scrofa*) accumulate more ^{137}Cs in muscles (4,7). A seasonal fluctuation in the activity concentration was also found: ^{137}Cs concentration varied from 0.4 to 612 Bq kg $^{-1}$ in boar muscles. The highest values were found in autumn. It was suggested that this is a result of higher mushroom intake by boars at that time (4).

Thus, skulls coming from northern Poland contain larger amounts of these radionuclides in comparison with southern part of the country. This claim is not very strong because of the fact that the measured values exceeded only slightly the limit of detection, while the correlation found was at a significance border. At the same time, ^{137}Cs concentration in soils of northeastern and southeastern Poland reveal quite the opposite relationship (11,14,15).

Mean concentration of ^{90}Sr , shown in Table 2, was much lower than the concentration of this radionuclide in bones of other mammals found in the literature. For example, values of ^{90}Sr concentration activity in all fox samples from a territory of eastern Poland ranged from

2.2 Bq kg⁻¹ to 41 Bq kg⁻¹ in ashed samples (9). Thus, the mean activity concentrations of ⁹⁰Sr in fox jaws from three different areas of eastern Poland were equal to 26, 24, and 21 Bq kg⁻¹ ash, respectively (9). It should be remembered that sample ashing causes a loss of more than one-third of the dry mass. This fact should be kept in mind when comparing the results. However, the data concerning foxes revealed concentration activities still several times higher than our results. Furthermore, other literature data on ungulates (2,3) showed the results being dramatically higher than our data, even taking into account a mass loss during ashing, and equal to 326 Bq kg⁻¹ ash (deer), 218 Bq kg⁻¹ ash (roe deer), 168 Bq kg⁻¹ ash (elk), and 100 Bq kg⁻¹ ash (wild boar). It is worth also noting that the lowest ⁹⁰Sr concentration activity level revealed the species with the largest diversity of food resources, like wild boar.

The very small concentration of plutonium found by us in skulls of European badger corresponds with the literature data concerning deer (3) and is in contrary to the foxes' results (9), which arises from the definite feeding habits of badgers. Badgers are considered as opportunistic omnivorous species that use the most available resources of feed in their environment. Generally, badgers can be viewed as species with the capacity to survive on different feeding resources (17,18). Visible feed opportunism allows badgers to feed on earthworms, snails, insects, small mammals and carrion, amphibians and reptiles, bird eggs and chicks, and mushrooms. Badgers also feed on edible plants such as fruits, cereals, grass, and clover (6,19). In Europe badgers inhabit the forests close to meadows and cultivated fields. However, recently a very pronounced increase in the number of badger individuals has been observed. This resulted in enlargement of their inhabit area by taking other territories, including even in cities (19–21). The above considerations on the feeding ecology of badgers allow us to explain the somewhat low concentrations of the radionuclides determined. Additionally, it is worth noting that radionuclides still migrate downward in the

soil. Therefore, after about 25 years from the Chernobyl disaster and 48 years from the maximum of global fallout, the concentration in the soil layer accessible for badgers became very low (16).

The performed radiochemical and spectrometric studies on European badger skull bones have shown that the concentration of selected anthropogenic radionuclides is rather low in comparison with bones of other mammals, especially those collected soon after the Chernobyl disaster. Median values of studied natural radionuclides were as follows: ⁴⁰K, 18 Bq kg⁻¹; ²¹⁰Pb, 10 Bq kg⁻¹; and ²²⁶Ra, 1.9 Bq kg⁻¹. Median anthropogenic radionuclide activity concentrations were found to be: ¹³⁷Cs, 0.56 Bq kg⁻¹; ²³⁸Pu, 7 mBq kg⁻¹; ²³⁹⁺²⁴⁰Pu 4, mBq kg⁻¹; and ⁹⁰Sr, 2 Bq kg⁻¹.

The correlation of the radionuclide concentration level with the geographical position of a sampling point has been proven poor. This was caused by rather high uncertainty of the measurements being a result of low radioactivity present in the sample. Low radionuclide concentrations in skull bones of European badger evidenced the low contamination of the soil layer accessible for the studied mammal. Presented studies have shown that anthropogenic radionuclide activity levels present in bodies of game mammals have diminished with time lapse (in comparison with the literature data concerning Chernobyl contamination). This is a positive finding because it reduces the risk of radionuclides entering human bodies, as many game animals are hunted for consumption. Simultaneously, it can be concluded that the radiological conditions of the environment in eastern Poland are at present sufficiently good, although the Chernobyl fallout in this area was rather intensive.

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