# Radionuclides in Bones of Diurnal Birds of Prey and Owls from the Eastern Poland\*

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The paper presents the analysis method and determination results of <sup>137</sup>Cs, <sup>40</sup>K, <sup>90</sup>Sr, <sup>238</sup>Pu, <sup>239+240</sup>Pu and <sup>241</sup>Am concentrations in bone samples of 7 species of diurnal birds of prey and owls from the eastern Poland. The samples consisted of mostly complete skeletons separated mechanically from the soft tissues. The analytical method consisted of two main steps preceded by ignition in 400°C: gamma spectrometric measurements and radiochemical analyses. Radiochemical analysis required dissolution of the samples in hot acids and subsequent separation of Pu, Am, and Sr by co-precipitation with oxalates, followed by anion exchange and extraction chromatography. No correlation between Sr. Cs, Am, and Pu activities for the whole set of samples was observed. Even <sup>238</sup>Pu and <sup>239+240</sup>Pu showed no clear correlation. However, some results indicated higher <sup>238</sup>Pu/<sup>239+240</sup>Pu activity ratio than that observed for the global fallout. In this case, the influence of the Chernobyl fallout was suggested. Significant correlation between 90Sr and 239+240Pu for Tawny owl Strix aluco and between <sup>241</sup>Am and <sup>239+240</sup>Pu for three subgroups was observed. Although some results for Pu isotopes might indicate that the traces of the Chernobyl fallout are measurable in birds from the eastern part of Poland, no clear geographical pattern related to the known distribution of the Chernobyl fallout has been observed yet.

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<sup>\*</sup> Dedicated to Professor Rajmund Dybczyński on the occasion of his 75th birthday.

Przedstawiono metode analityczna oraz wyniki oznaczeń steżeń aktywności 137Cs, 40K, 90Sr, <sup>238</sup>Pu, <sup>239+240</sup>Pu i <sup>241</sup>Am w kościach ptaków drapieżnych i sów należących do 7 gatunków pochodzacych ze wschodniej Polski. W wiekszości przypadków były to kompletne szkielety. Kości były oczyszczone mechanicznie z tkanek miękkich. Zastosowana metoda składała się z dwóch zasadniczych etapów poprzedzonych spopieleniem w 400°C: pomiaru gammaspektrometrycznego i analizy radiochemicznej, gdzie próbki były roztwarzane w goracych kwasach po czym następowało sekwencyjne oddzielenie frakcji zawierającej Pu i Am od frakcji zawierającej Sr polegające na współstrąceniu wraz ze szczawianem wapnia przy dwóch różnych wartościach pH, po czym przeprowadzano zasadnicze wydzielenie z zastosowaniem chromatografii jono-wymiennej (Pu i Am) lub chromatografii ekstrakcyjnej (Sr). Nie stwierdzono korelacji między stężeniami aktywności izotopów Sr, Cs, Am i Pu dla pełnego zestawu analizowanych próbek. Co więcej, nie stwierdzono korelacji nawet między <sup>238</sup>Pu oraz <sup>239+240</sup>Pu. Niektóre próbki wykazały wyższe niż typowe dla opadu globalnego wartości ilorazu aktywności 238Pu do 239+240Pu, co zostało zinterpretowane jako dowód obecności śladów Pu pochodzenia czarnobylskiego. Nie stwierdzono związków między obserwowanymi skażeniami a miejscem pochodzenia próbek. Zaobserwowano jedynie korelacje między stężeniami aktywności 90Sr oraz 239+240Pu w obrębie podgrupy zawierającej wszystkie analizowane kości puszczyka Strix aluco oraz między 241 Am i 239+240 Pu dla trzech innych podgrup.

The first massive sources of the radioactive contamination of the environment were nuclear weapon tests conducted in the open atmosphere from 1945 till 1980 [1]. The majority of explosions occurred before 1963, when the atmospheric nuclear test ban treaty was signed by the main nuclear powers. After this date only some Chinese and French tests were conducted. The last Chinese test took place in 1980 [1]. The tests resulted in the so-called global fallout of radionuclides. Then, in April 1986, the release of radionuclides from the damaged Chernobyl Nuclear Power Plant became a source of a serious contamination of a large area, mainly in the former Soviet Union. The area of Poland was contaminated both by nuclear explosions and Chernobyl fallout at the moderate scale. Geographical distribution of contamination and other properties, such as isotopic ratios characteristic for each of the sources, were the subject of investigation in the past and therefore are well known [2, 3], also from our papers [4–8].

Out of many artificial long-lived isotopes still present in the environment, these of plutonium (<sup>238</sup>Pu,<sup>239</sup>Pu,<sup>240</sup>Pu), americium (<sup>241</sup>Am), and <sup>90</sup>Sr are considered "bone-seekers", while <sup>137</sup>Cs tends to concentrate rather in soft tissues. Plutonium and americium are also known to accumulate in liver [9, 10].

Radioactive contamination in wild animals has been the subject of our concern for many years. For example, bones of rodents (*Rodentia*), ungulates (deer *Cervus elaphus*, roe-deer *Capreolus capreolus*, elk *Alces alces*, boar *Sus scrofa*), and insectivore (*Insectivora*) such as shrews (*Sorex* sp.) were analyzed in the past [11–17]. Recently, bones of different groups of carnivore mammals: red fox (*Vulpes vulpes*)

[18], two species of eagles [19] and Barn owl (*Tyto alba*) [20] have been the subject of similar analyses. The present paper shows the data obtained for other raptors: diurnal birds of prey (*Falconiformes*) and some owls (*Strigiformes*) breeding in the eastern part of Poland. Recently, in other studies the determination of natural <sup>210</sup>Po, <sup>234</sup>U and <sup>238</sup>U as well as artificial <sup>238</sup>Pu, <sup>239+240</sup>Pu alpha radionuclides in different organs and tissues (also skeleton) in marine birds from northern Poland were analyzed by Fabisiak and Skwarzec [21, 22].

## EXPERIMENTAL

#### Materials, method, and study area

Since the contaminants are expected to accumulate at the higher level of the food chain, therefore it seemed crucial to study the radioactive contamination for diurnal and nocturnal raptors. In the present study, bones of Goshawk (*Accipiter gentilis*), Sparrowhawk (*Accipiter nisus*), Osprey (*Pandion haliaetus*), Eurasian Buzzard (*Buteo buteo*), Common Kestrel (*Falco tinnunculus*), Tawny owl (*Strix aluco*), and Long-eared owl (*Asio otus*) were analyzed for the presence of all above-mentioned artificial isotopes. The details on sample size and sampling sites locations are given in Table 1. Presently analyzed samples are the continuation of the previous study on bone samples of White-tailed eagle (*Haliaeetus albicilla*), Lesser-spotted eagle (*Aquila pomarina*), and Common Barn owl (*Tyto alba*) [17, 19, 20].

Code	Name of species	Location	Part of Poland	Ash weight of available bones [g]
PW 1	Osprey Pandion haliaetus	Chełm	CE	39.0
PW 2	Long-eared owl Asio otus	Łabunie near Zamość	CE	5.42
PW 3	Tawny owl Strix aluco	Dzieraznia near Zamość	CE	7.21
PW 4	Tawny owl Strix aluco	Białystok	NE	15.78
PW 5	Tawny owl Strix aluco	Olsztyn	NE	5.15
PW 6	Long-eared owl Asio otus	Dojlidy near Białystok	NE	4.17
PW 7	Tawny owl Strix aluco	Kamień near Chełm	CE	17.1
PW 8	Tawny owl Strix aluco	Wola Wereszczyńska	CE	16.85

Table 1. Basic characteristics of the analyzed samples (NE – North-Eastern, CE – Central-Eastern)

(Continuation on the next page)

Code	Name of species	Location	Part of Poland	Ash weight of available bones [g]
PW 9	Tawny owl Strix aluco	Niedrzwica Duża	CE	2.81
PW 10	Tawny owl Strix aluco	Chełm	CE	15.62
PW 11	Eurasian Buzzard Buteo buteo	Łaziska near Zamość	CE	18.81
PW 12	Goshawk Accipiter gentilis	Łapy near Białystok	NE	8.81
PW 13	Common Kestrel Falco tinnunculus	Białystok	NE	2.01
PW 14	Sparrowhawk Accipiter nisus	Białystok	NE	5.89
PW 15	Sparrowhawk Accipiter nisus	Siemiatycze	NE	2.89
PW 16	Common Kestrel Falco tinnunculus	Czołki near Zamość	CE	5.47
PW 17	Goshawk Accipiter gentilis	Sejny	NE	10.12
PW 18	Goshawk Accipiter gentilis	Siemiatycze	NE	10.27
PW 19	Eurasian Buzzard Buteo buteo	Sitaniec near Zamość	CE	12.67
PW 20	Goshawk Accipiter gentilis	Olsztyn	NE	11.03
PW 21	Eurasian Buzzard Buteo buteo	Olsztyn	NE	8.82
PW 22	Eurasian Buzzard Buteo buteo	Czarna Białostocka	NE	9.05
PW 23	Common Kestrel Falco tinnunculus	Topornica near Zamość	CE	4.06
PW 24	Eurasian Buzzard Buteo buteo	Łapy near Białystok	NE	35.7

#### Table 1.(Continuation)

The study area covers the eastern part of Poland, from the northern to the central part of the country. This area was affected by the Chernobyl fallout, especially in the northeastern part, where the presence of the important fallout of the "fuel-like hot particles" was observed [2, 6]. No animal was killed purposely for the project. All birds, which skeletons were used, had been found dead because of different reasons, mainly collisions with the high-voltage electric wires, cars, trains, TV or mobile telephone towers, etc. All the birds, except for one osprey (sample PW 1) were local breeders in the areas where they were found dead. The osprey specimen was found electrocuted near Chełm. It originated from the nest located near Narpio, Vasa, (West Finland, ornithological ring: M 41339).

Bones were cleaned mechanically from soft tissues and air-dried. At the beginning of the analyses, all the samples were weighed and then ignited at the temperature of 400°C using the electric furnace. The temperature was set low to prevent cesium evaporation. The remaining ashes of bones were ground and mixed to obtain the homogenous distribution of the analyzed nuclides within the sample, and re-weighed. All available material was used as a sample. Activity concentrations of gamma emitters present in the incine-rated bone samples (in practice: <sup>40</sup>K and <sup>137</sup>Cs) were measured using a low background gamma-ray spectrometer with HPGe detector calibrated for different sample volumes. The calibration method was described elsewhere [23].

After gamma spectrometric measurements, bone samples were re-ignited at 600°C. This step was necessary to remove any traces of organic matter and especially carbon components. To all the samples <sup>85</sup>Sr, <sup>243</sup>Am, and <sup>242</sup>Pu tracers were added and ignited bone samples were dissolved using *ca* 6 mol L<sup>-1</sup> hot HCl with a few drops of H<sub>2</sub>O<sub>2</sub> added until dark traces of carbon vanished. The solutions containing mineralized bones were evaporated to about 30–50 cm<sup>3</sup>, diluted, and calcium oxalates were precipitated at pH = 3. Plutonium (III, IV) was expected to co-precipitate with calcium oxalate. The supernatant solutions were diluted and strontium oxalates were precipitated at pH = 4.5. Centrifuged oxalates from both fractions were decomposed separately using hot concentrated HNO<sub>3</sub>. Afterwards, they were diluted with 150 cm<sup>3</sup> of ~1 mol L<sup>-1</sup> HNO<sub>3</sub>, or with 100 cm<sup>3</sup> of ~3 mol L<sup>-1</sup> HNO<sub>3</sub> for "Pu" fractions (co-precipitation with oxalates at pH = 3) and "Sr" fractions (co-precipitation with oxalates at pH = 4.5), respectively [19].

Prior to Pu separation from "Pu" fraction on a Dowex–1 anion exchange column using 8 mol  $L^{-1}$  HNO<sub>3</sub>, the oxidation step adjustment procedure was applied [24]. The procedure started from adding of 1 cm<sup>3</sup> of hydrazine to the hot solution. After 10 min of boiling, 30 cm<sup>3</sup> of concentrated HNO<sub>3</sub> were added to destroy hydrazine and, after cooling down to the room temperature, 5 g of NaNO<sub>2</sub> were added. The solution was re-boiled for about 0.5 h and its volume was completed to 200 cm<sup>3</sup> with concentrated HNO<sub>3</sub> to obtain final concentration of the acid of 8 mol L<sup>-1</sup>.

Solution was passed through the Dowex–1 anion-exchange column of 8 cm in length and 3 mm in the inner diameter. The effluent from the column was used for Am analyses. Th was eluted from the column using 50 cm<sup>3</sup> of 12 mol L<sup>-1</sup> HCl. Pu was eluted using 0.1 mol L<sup>-1</sup> HCl–0.1 mol L<sup>-1</sup> HF mixture. Americium was pre-concentrated from the evaporated and diluted solution by two subsequent co-precipitations (again with oxalates and with iron hydroxide); later, the remaining traces of Th were removed from this fraction with TEVA-resin (by Eichrom Inc.) by passing the sample in the form of solution in 4 mol L<sup>-1</sup> HNO<sub>3</sub> through the TEVA-resin column; finally, Am was separated from rare earth elements using methanol–acid mixed solutions.

<sup>90</sup>Sr was separated from the Sr-fraction on the Sr-resin column (Eichrom, Inc.) with 3 mol  $L^{-1}$  HNO<sub>3</sub> [25]. Strontium was eluted from the column with 50 cm<sup>3</sup> of 0.01 mol  $L^{-1}$  HNO<sub>3</sub> and later separated from any possible traces of <sup>210</sup>Pb by precipitation of lead iodide [17].

Pu and Am alpha spectrometric sources were prepared using the NdF<sub>3</sub> co-precipitation method [26] and measured using a Silena AlphaQuattro spectrometer with Canberra PIPS (passivated implanted planar silicon) detectors. The activity of <sup>90</sup>Sr was determined using a Wallac 1414–003 Guardian liquid scintillation spectrometer, and <sup>85</sup>Sr recovery was determined applying gamma spectrometry.

The activity ratio of plutonium isotopes could be used to distinguish between Chernobyl and global fallouts as the source of contamination [4]. For the global fallout the <sup>238</sup>Pu/<sup>239+240</sup>Pu activity ratio is 0.03 [17], whereas for Chernobyl it is 0.50 [19].

As the minimum detectable concentration we considered the activity calculated from the peak counts exceeding 3-fold the standard deviation of the mean background – the mean result for the blank sample [11]. The reason for such approach was discussed in that paper. Namely, if the mean count rate for the blank in <sup>239+240</sup>Pu ROI (region of interest) was 14 counts in 400000 s, the detection limit was calculated from

the background – corrected peak count rate exceeding  $3\sqrt{14} \approx 11$  counts in 400000 s (using actual mass of the sample). In practice, for Pu or Am it was a few mBq/kg (depending on the sample mass).

The quality of radiochemical analyses was ensured by the analysis of the reference material – IAEA Soil 375 and blank samples. For <sup>90</sup>Sr and <sup>239+240</sup>Pu the results were  $125 \pm 19$  Bq/kg (101–114 Bq/kg is the IAEA 95% certified interval) and  $0.268 \pm 0.034$  Bq/kg (IAEA 95% certified interval is 0.26-0.34 Bq/kg), respectively. Relatively high uncertainty of the result for strontium was caused by low statistics for the <sup>85</sup>Sr tracer – low activities were added not to disturb the LSC spectra by secondary effects.

Average chemical recovery and the corresponding standard deviation (SD) for strontium, plutonium, and americium were, respectively: 58.3% and SD = 22.9%, 83.9% and SD = 14.8%, and 91.3% and SD = 19%.

## **RESULTS AND DISCUSSION**

The measured activity concentrations for each sample are presented in Table 2. For consistency with all previous results obtained in our laboratory for animal bones results are presented as concentration with respect to ash weight. No correlation between Sr, Cs, Pu, and Am activities was observed for the whole set of samples. Even between <sup>238</sup>Pu and <sup>239+240</sup>Pu, or between americium and plutonium no clear correlation could be observed for the whole set of samples. However, in many cases relatively high <sup>238</sup>Pu content compared to that of <sup>239+240</sup>Pu was detected. <sup>238</sup>Pu content was higher than that expected for the global fallout and it was suggested to be related to the Chernobyl fallout. Unfortunately, the uncertainties were too large to draw stronger conclusion on plutonium origin proportions.

In order to find any regularity among the obtained results, the samples were divided into different subsets. The most obvious subset was a group of all the samples, which belonged to a given species. To check if feeding ecology or hunting habits play any role in the radionuclides accumulation in the raptors' bones, the samples were again divided into different subgroups comprising more than one species. The obtained statistical parameters are presented in Tables 3. The comparison between diurnal raptors (diurnal birds of prey apart from osprey) and nocturnal raptors (owls) showed no significant correlation. Moreover, a comparison between raptors, which are rodenteaters and these, which feed mainly on birds, did not show any significant differences in terms of the applied statistical test (ANOVA). The comparison between diurnal rodent eaters (Eurasian Buzzard and Common Kestrel) and diurnal bird eaters (Goshawk and Sparrowhawk) gave similar results – no difference between mean or median activities were found.

otopes, and <sup>241</sup> Am
Pu is
<sup>90</sup> Sr,
<sup>137</sup> Cs,
r concentrations of <sup>40</sup> K,
The measured activity
Table 2.

	<sup>241</sup> Am	< 0.003	$0.013\pm0.010$	$0.005\pm0.006$	$0.003 \pm 0.004$	$0.019\pm0.007$	< 0.006	$0.006\pm0.003$	$0.009\pm0.002$	< 0.040	$0.007\pm0.003$	< 0.010	$0.010 \pm 0.006$
	$^{238}\mathrm{Pu}$	$0.0010 \pm 0.0004$	$0.02\pm0.006$	$0.011\pm0.003$	$0.004\pm0.003$	$0.024\pm0.01$	$0.023\pm0.013$	$0.006\pm0.003$	$0.001\pm0.001$	$0.030\pm0.013$	$0.002\pm0.001$	$0.005\pm0.002$	$0.009 \pm 0.003$
10 unc. [Bq/kg ash weight]	<sup>239+240</sup> Pu	$0.007\pm0.001$	$0.012\pm0.003$	$0.008\pm0.002$	$0.003\pm0.001$	$0.022\pm0.005$	$0.034\pm0.006$	$0.027\pm0.012$	$0.009\pm0.001$	$0.027\pm0.002$	$0.033\pm0.004$	$0.063\pm0.011$	$0.038 \pm 0.006$
Activity ±	<sup>90</sup> Sr	$3.3\pm0.3$	88.7 ± 7.6	$13.8 \pm 1.4$	$13.4 \pm 0.7$	$23.8 \pm 2.0$	$12.7 \pm 1.5$	$38.6 \pm 1.8$	$11.9 \pm 0.7$	$18.6 \pm 4.5$	$61.4 \pm 6.0$	$19.1 \pm 2.0$	$12.5 \pm 1.3$
	$\rm M^{40}K$	170 ± 17	$269 \pm 84$	$298 \pm 108$	451 ± 51	$953 \pm 122$	$693 \pm 58$	367 ± 37	166 ± 35	258 ± 54	$400 \pm 22$	$69 \pm 16$	<b>3</b> 97 ± 86
	<sup>137</sup> Cs	$3.0\pm0.8$	9 >	$48 \pm 10$	$108 \pm 5$	$13 \pm 7$	$2 \pm 3$	$18 \pm 3$	$5\pm 2$	$1 \pm 4$	$6 \pm 1$	$1 \pm 1$	9 >
Codo	Cone	PW 1	PW 2	PW 3	PW 4	PW 5	PW 6	PW 7	PW 8	6 M	PW 10	PW 11	PW 12

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Table 2.	

			Activity	± 1σ unc. [Bq/kg ash weight]		
0	$^{137}Cs$	$^{40}$ K	$^{1}\mathrm{S}^{06}$	<sup>239+240</sup> Pu	<sup>238</sup> Pu	<sup>241</sup> Am
[3	3 ± 6	$344 \pm 75$	$18.1 \pm 2.7$	$0.159\pm0.035$	$0.016\pm0.018$	$0.026\pm0.017$
14	26 ± 7	392 ± 96	$11.6 \pm 1.3$	$0.069\pm0.008$	$0.021\pm0.005$	< 0.004
15	$54 \pm 18$	$850 \pm 267$	$14.3 \pm 2.1$	$0.175\pm0.022$	$0.02 \pm 0.01$	< 0.014
16	9 >	317 ± 118	$9.0 \pm 1.0$	$0.077\pm0.01$	$0.012\pm0.004$	$0.034\pm0.009$
17	$3\pm 2$	269 ± 36	$11.6 \pm 1.4$	$0.040\pm0.005$	$0.009\pm0.003$	$0.009\pm0.005$
18	$6\pm3$	$399 \pm 47$	$18.7 \pm 1.8$	$0.044\pm0.005$	$0.011\pm0.003$	$0.005\pm0.006$
19	< 3	$323 \pm 46$	$18.0 \pm 2.3$	$0.042\pm0.005$	$0.008\pm0.002$	$0.007\pm0.005$
20	$3\pm 2$	385 ± 29	$13.4 \pm 0.9$	$0.003\pm0.001$	$0.009\pm0.003$	$0.004 \pm 0.004$
21	$5 \pm 4$	$185 \pm 57$	$27.2 \pm 1.5$	$0.006\pm0.002$	$0.009\pm0.003$	$0.007\pm0.003$
22	$4\pm 2$	279 ± 26	$14.6\pm0.9$	$0.002\pm0.001$	$0.014\pm0.007$	$0.010 \pm 0.010$
23	$23 \pm 9$	248 ± 128	$37.8 \pm 2.6$	$0.057\pm0.009$	$0.014\pm0.007$	$0.018 \pm 0.013$
24	$2 \pm 1$	L ∓ 86	$14.3 \pm 1.3$	$0.010\pm0.002$	< 0.002	< 0.002

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	Table 5.

				Mean/ Median/ Stand	lard Deviation [Bq/kg a	sh weight]	
No.	Species (a subgroup)				Nuclides		
		$^{40}\mathrm{K}$	<sup>137</sup> Cs	$^{90}$ Sr	<sup>238</sup> Pu	<sup>239,240</sup> Pu	<sup>241</sup> Am
1	Tawny owl	393/298/263	27.7/13.0/39.0	25.9/18.6/18.2	0.011/0.006/0.011	0.018/0.022/0.012	0.010/0.007/0.007
2	Tawny owl + Long- eared owl	413/298/254	21.8/5.0/35.7	31.4/18.6/27.0	0.013/0.011/0.011	0.019/0.022/0.012	0.009/0.007/0.007
3	Goshawk	363/391/63	3.0/3.0/2.4	14.0/12.9/3.2	0.010/0.009/0.001	0.031/0.039/0.019	0.007/0.007/0.003
4	Eurasian Buzzard	212/185/138	2.2/2.0/1.9	18.6/18.0/5.2	0.007/0.008/0.005	0.025/0.010/0.027	0.005/0.007/0.0044
5	Common Kestrel+ Eurasian Buzzard	246/283/118	4.6/2.5/7.6	19.8/18.1/8.9	0.010/0.011/0.005	0.052/0.050/0.052	0.012/0.007/0.013
9	Common Kestrel + Eurasian Buzzard + Long-eared owl	293/293/175	3.9/2.0/6.9	26.0/18.1/23.5	0.011/0.013/0.007	0.046/0.038/0.047	0.011/0.007/0.012
7	Goshawk+ Sparrowhawk	449/395/203	15.3/4.5/21.1	13.7/12.9/2.7	0.013/0.010/0.006	0.062/0.042/0.059	0.005/0.004/0.004

 Table 4.
 The results of the statistical analysis of samples grouped in geographical regions of origin (CE, NE – Tab. 1)

			Me	an/ Median/ Standard	Deviation [Bq/kg ash	weight]	
Region (subgroup)				N	uclides		
<sup>40</sup> K <sup>137</sup> Cs	<sup>40</sup> K 1 <sup>37</sup> Cs	<sup>137</sup> Cs		$^{90}\mathrm{Sr}$	<sup>238</sup> Pu	<sup>239,240</sup> Pu	$^{241}\mathrm{Am}$
All species, Zamość region 272/284/97 10.2/3.0/15.6 (CE)	272/284/97 10.2/3.0/15.6	10.2/3.0/15.6		31.7/18.8/25.7	0.011/0.010/0.009	0.036/0.030/0.024	0.011/0.007/0.010
All species, NE 438/392/250 17.6/4.0/31.0	438/392/250 17.6/4.0/31.0	17.6/4.0/31.0		15.9/14.3/4.8	0.013/0.011/0.007	0.047/0.034/0.057	0.007/0.005/0.008

Although all the samples were collected in the eastern Poland, they could be subdivided into two other groups according to their geographical origin. The first group originated from the northeastern part of the country (NE in Tab. 1), in the vicinity of Białystok (53° 08'N, 23° 10'E) or Olsztyn (53°45' N, 20°31'E), where the Chernobyl influence was expected to be more pronounced. Another group originated from the central part of eastern Poland (CE in Tab. 1), from the vicinity of Zamość (50° 43'N, 23° 16'E), where only a tiny part of the total radioactive fallout (Pu, Am or <sup>90</sup>Sr) could be of the Chernobyl origin. However, such division did not lead to any statistically important differences between the samples in terms of unpaired t-test with Welch correction, or t-test for mean or median (all distributions were Gaussian, but standard deviations for Am, 239+240Pu and 90Sr were different for both groups). This result is a little bit surprising, since the Chernobyl actinides and <sup>90</sup>Sr deposition in Poland was found mostly in the north-eastern part of the country, as it was discussed for example in a review [6] and confirmed by the following studies [8]. Mean and median values and standard deviation were calculated for both geographical subgroups of the samples and are presented in Table 4.

Correlations were checked for all the analyzed radionuclides, for whole sets of samples, as well as for all subgroups. Significant correlation (significance level p < 0.05) was found in four cases. The details concerning these four correlations are presented in Table 5.

Subgroup name	Radionuclides	Pearson's correlation factor	Signifcance level
Kestrels, Buzzards, Long eared owls	<sup>241</sup> Am and <sup>239+240</sup> Pu	$r^2 = 0.494$	p = 0.035
NE region (all species)	<sup>241</sup> Am and <sup>239+240</sup> Pu	$r^2 = 0.397$	p = 0.020
All diurnal raptors	<sup>241</sup> Am and <sup>239+240</sup> Pu	$r^2 = 0.345$	p = 0.021
Tawny Owl	<sup>239+240</sup> Pu and <sup>90</sup> Sr	$r^2 = 0.641$	p = 0.021

**Table 5.** Parameters of the observed significant correlations (p < 0.05) between the activities of different<br/>subgroups

In all cases correlation was moderate. There were three cases of correlation between <sup>241</sup>Am and <sup>239+240</sup>Pu (for all diurnal raptors, for all raptors from NE subgroup, and for all these, which feed on small mammals - buzzard, kestrel and long eared owl). The fourth and most significant correlation (Fig. 1) was observed between <sup>90</sup>Sr and <sup>239+240</sup>Pu for the samples of Tawny owl. This correlation might be important because this owl species is widespread in Poland, breeds moderately, is strictly sedentary with a wide food niche based on small mammals and birds [27, 28]. This result opens

future perspectives for the use of Tawny owl skeleton in biomonitoring of radionuclides because of easy-to-obtain bone samples (especially after severe winters).



**Figure 1.** The most significant correlation found between <sup>90</sup>Sr and <sup>239+240</sup>Pu for the samples of Tawny owl *Strix aluco*. Pearson correlation factor  $R^2 = 0.641$ , level of significance p = 0.03

Geographical distribution of radionuclides based on their accumulation in the bones of studied raptors does not correspond to the data on radionuclide distributions obtained in the studies of other samples (soil samples, plant sample, insects samples) [8, 14, 16, 29]. Bio-accumulation of nuclides does not take place in the bones of raptors.

# CONCLUSIONS

The applied radiochemical method is appropriate for determination of trace amount of radionuclides in bones of wild animals. The results for Pu isotopes ratios suggest that the traces of the Chernobyl fallout occur at the measurable level in birds from the eastern Poland, but no clear geographical pattern related to the known distribution of the Chernobyl fallout was observed. A comparison with previously obtained results for other animals' bones suggests that no accumulation of the contaminants can be observed, as it has been found recently for a carnivore – Red fox [18].

No high accumulation of <sup>90</sup>Sr, <sup>241</sup>Am, and Pu isotopes in the bones of the analyzed species of diurnal birds of prey in comparison to the bones of their potential preys was found. Also, one observed no significant differences in nuclides concentrations between the samples originating from the area of Poland affected by the Chernobyl radioactive fallout and the region where the influence of Chernobyl was only a minor part of the total radioactive contamination. In general, there is no correlation between

the observed activity concentrations of the analyzed radionuclides. Moderate correlations between plutonium and americium were found in three cases. A significant correlation between <sup>90</sup>Sr and <sup>239+240</sup>Pu isotopes was observed ( $R^2 = 0.641$ , p = 0.03) only in one case (Tawny owl).

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