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Caesium-137 in the muscles of game animals in 2015-2022 – levels and time trend

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Abstract

Introduction: Radioactive caesium-137 occurring in the environment may be taken up by plants and animals and pose a trophic threat to humans. Game animals living in forest ecosystems are very good bioindicators of the level of environmental contamination by ionising radiation. The main species measurably exposed to caesium-137 are the wild boar (*Sus scrofa*), the roe deer (*Capreolus capreolus*), and the red deer (*Cervus elaphus*). The study determined the levels of Cs-137 in muscle samples of these game animals in 2015–2022. **Material and Methods:** Using gamma radiation spectrometry, 858 samples of game animal muscle tissue were examined: 508 wild boar, 145 roe deer and 205 red deer samples. **Results:** Concentrations of Cs-137 varied widely (from minimum detectable activity (MDA) values to over 4,000 Bq/kg). In 63.4% of cases, the obtained concentrations exceeded the MDA. The permissible limit (600 Bq/kg for food) was exceeded in nine wild boar muscle samples, whereas it was not even exceeded once in roe or red deer muscle. The average concentration in wild boar was three times higher than in roe and red deer and amounted to 42.84 Bq/kg. The highest concentration of Cs-137 in wild boar muscle was 4,195 ± 372.0 Bq/kg, in roe deer muscle it was 111.5 ± 12.50 Bq/kg, and in red deer muscle was 86.70 ± 3.470 Bq/kg. **Conclusion:** The seven years' data indicate that wild boar absorb the most caesium-137 among game animals. The concentrations of Cs-137 in the muscle of game animals in the years 2015–2022 were at a nearly constant level, a very slow diminution being noticeable over time in roe and red deer muscle.

Keywords: Cs-137, contamination, game animals, concentration factors, effective equivalent dose.

Introduction

Nuclear accidents may have long-lasting consequences for biocenoses (21). Some radionuclides have the propensity to move from the abiotic environment to biotic components and accumulate there, an example of such a radionuclide being Cs-137, the main one formed after Chernobyl (8). The affinity of the radioactive isotope caesium-137 for potassium (K-40) facilitates the element's penetration of tissues and muscles. The consequence of this phenomenon is a chain of secondary biological reactions, which include the occurrence of cancer (4, 18, 22).

The results of regular control tests of animal products conducted in Poland have shown that the most contaminated group of animals are game species, and the most notably contaminated of them is the wild boar (13). It is well described in the literature that some mushrooms eagerly eaten by animals have a strong propensity to accumulate radioisotopes of caesium (7). Examples of such mushrooms in Poland are the brown bolete (*Xerocomus badius*) and deer truffle (*Elaphomyces granulatus*), which may be a source of radionuclides for wild boar (9). Moreover, animals eating deer truffles may ingest contaminated soil components (26, 27). The concentration of radioactive isotopes in food products should at most match the values specified in the Decree by the Council of Ministers of the Government of the Republic of Poland of April 27, 2004, which states that the maximum permissible levels in food products cannot exceed 1,250 Bq/kg (14). This level refers to isotopes with a half-life longer than 10 days and mainly concerns Cs-134 and Cs-137. Additionally, in accordance with European law, the determined activity of radioactive isotopes in foodstuffs and food products should fall below the values specified in the European Commission Implementing Regulation (EU) 2020/1158 of 5 August 2020 (11). This states that the concentration of the Cs-137 isotope must not exceed 600 Bq/kg in all foodstuffs and products except milk and dairy products, where its concentration must not exceed 370 Bq/kg.

The geopolitical situation in Eastern Europe elevates the importance of conducting an assessment of the current state of environmental contamination. The armed conflict in Ukraine spans territory where a nuclear power plant operates and carries the risk of release and emission of ionising radiation; in the event of a serious radiation emergency, knowledge of the pre-emergency level of radioactive contamination provides better accuracy in an assessment of the environmental radiation situation post radioactive release.

The observed environmental pollution in Poland is mainly related to the explosion at the Chernobyl nuclear power plant (31). The accident occurred as a result of a catastrophic power surge in reactor unit 4 during a planned test procedure, which caused an explosion and fire in the reactor. The result was the emission of huge amounts of radioactive elements from the reactor fuel and core materials into the atmosphere (12). Although this event took place 37 years ago, the effects of the environmental pollution are still felt today. This is due to the long half-life of the radioactive contaminants, which in the case of Cs-137 is 30.2 years.

Numerous studies indicate that wild animals such as wild boar, roe deer and red deer easily absorb the radioactive isotope caesium-137 from food. Therefore, when tested, these game species play very useful roles as bioindicators in determining radioactive environmental contamination (30). The aim of the study was to determine the levels of Cs-137 in muscle samples of game animals sent to the National Veterinary Research Institute as part of monitoring tests in the period 2015–2022, to analyse time trends in their radioactivity, and then to assess the current state of radioactive contamination of the environment in Poland.

Material and Methods

Material for research. The research material consisted of muscle tissue of game animals: the wild boar (*Sus scrofa*), the roe deer (*Capreolus capreolus*) and the red deer (*Cervus elaphus*). Samples for testing were collected by the veterinary inspectorate from processing plants in Poland. Figure 1 shows the number of samples tested in each year. A total of 858 game animal muscle samples were tested: 508 from wild boar, 145 from roe deer and 205 from red deer.

Instrumental analysis. An accredited test method (AB 95; Polish Centre for Accreditation, Warsaw, Poland) using gamma-ray spectrometry with a solid-state detector (high-purity germanium HPGe detector) and a scintillation detector (NaI (TI) crystal) placed in a lead shield was used to determine the radioactive concentration of caesium-137 (25). The laboratory at the Department of Radiobiology of the National Veterinary Research Institute has participated in tests of its proficiency in this method several times and has passed on each occasion. Crushed, ground and homogenised muscle samples were transferred to Marinelli measuring containers (450 cm^3). The geometry of the multi-nuclide MBSS 2 calibration source (The Czech Metrology Institute, Jihlava, Czech Republic), which was used to calibrate the detectors, was preserved. The measurement time was 72,000 s (20 h). The collected gamma-ray spectra were analysed using Genie 2000 software (Mirion Technologies, Atlanta, GA, USA) (Fig. 2).

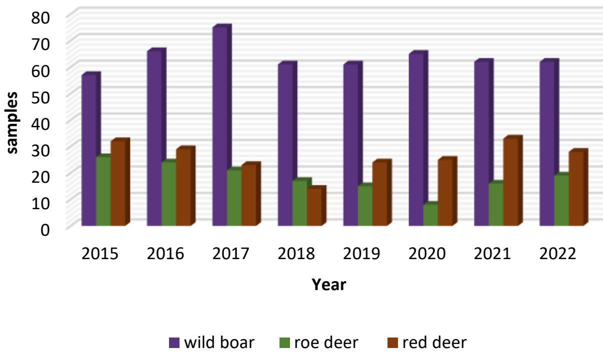

Fig. 1. Number of muscle samples tested in particular years

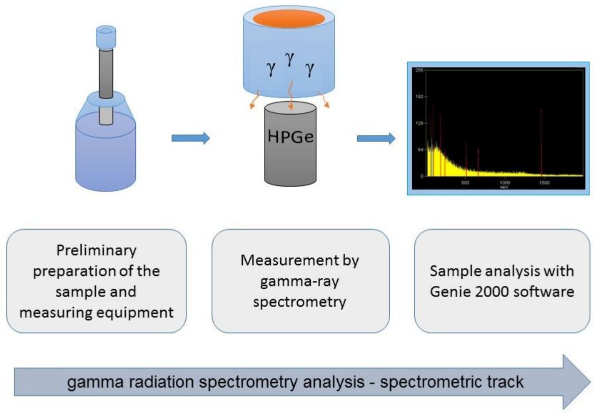

Fig. 2. Scheme of the analysis HPGe – high-purity germanium radiation detection

Statistical analysis. A summary of the analyses performed was made for the matrix data, and then the mean concentration, standard deviation and median were calculated for the obtained data using Microsoft Excel (Redmond, WA, USA). The minimum and maximum measured concentrations and the number of samples above and below the minimum detectable activity (MDA) were summarised in the way described by Done and Ioan (6). Slope coefficients were determined for the calculated mean and median values for the years 2015–2022 using the linear regression method in Microsoft Excel. The fit of the function to the data was verified using the value of the coefficient of determination R^2 with the same software. It was judged against the generally accepted criterion for a satisfactory or very good fit of $0.6 \leq R2 \leq 1$. Concentrations over time were additionally checked using the nonparametric Mann–Kendall (M-K) test published by Salmi *et al.* (28). The normality of distribution for each of the three studied groups was evaluated with the Kolmogorov–Smirnov (K-S) statistical test in Statistica v. 13.1 (Dell, now TIBCO, Palo Alto, CA, USA).

Determination of the concentration coefficient. A concentration factor is commonly used to quantify the transfer of radionuclides into meat (23). The transfer of radionuclides from the environment to the bodies of wild animals has been described in the literature several times (1, 15). It was assumed to be a dependence on the concentration ratio (CR) and is described by the following formula:

$$\mathrm{CR} = \frac{\text{Activity concentration in whole organism (Bq/kg fresh weight)}}{\text{Activity concentration in soil (Bq/kg dry weight)}}$$

To calculate the CR, making the same assumption as Beresford *et al.* (3), Kapała *et al.* (20) and Yankovich *et al.* (32), the concentration of muscle activity was assumed to be the concentration of activity of the whole organism. Data from the Central Laboratory for Radiological Protection (CLOR) published in annual monitoring reports on the concentration of Cs-137 in soil were used to calculate the CR in the individual years 2016, 2019 and 2020 (5, 17).

Estimation of effective equivalent dose. Estimating the dose of Cs-137 absorbed by an adult through consumed game is extremely difficult because data is lacking on the amount of game consumed in Poland. Therefore, 1 kg was assumed for the calculations as the annual intake for an adult. The literature contains information obtained from hunters from north-eastern Poland. Kapała *et al.* (20) stated that the annual consumption of game by an adult is 20–50 kg. The effective dose E_{ff} consumed in contaminated meat by the reference adult resulting from radionuclide uptake was calculated using the following formula:

$$E_{ff} = AM \times k \times m$$

where E_{ff} is the effective dose (Sv), AM is the arithmetic mean Cs-137 concentration in muscles in Bq/kg, k is the adult dose conversion factor of 1.3×10^{-8} Sv Bq^{-1} used for Cs-137 (16) and m is the annual intake by adults (kg).

Results

Measurement of Cs-137 activity in game animal muscle samples. The research conducted over the stated 2015–2022 timeframe provided valuable information and enabled the creation of a large database. Every year in the studied period, a certain number of determinations were made in wild boar, roe deer and red deer muscle samples (Fig. 1). The measurements of these samples facilitated the determination of average concentrations of caesium-137 as well as the determination of its minimum and maximum concentration in each tested matrix (Table 1).

Statistical analysis and calculations for Cs-137 in the muscles of game animals. The K-S statistical test showed the lack of normal distribution for each of the three studied groups. For wild boar muscles, the K-S test parameters were d = 0.42594 and P-value < 0.01, for roe deer muscles they were d = 0.292960 and P-value < 0.01, and for deer muscles they were d = 0.27901 and P-value < 0.01. Figure 3 shows the average concentrations of caesium-137 in samples of wild boar, roe deer and red deer muscle and time trends covering the period from 2015 to 2022.

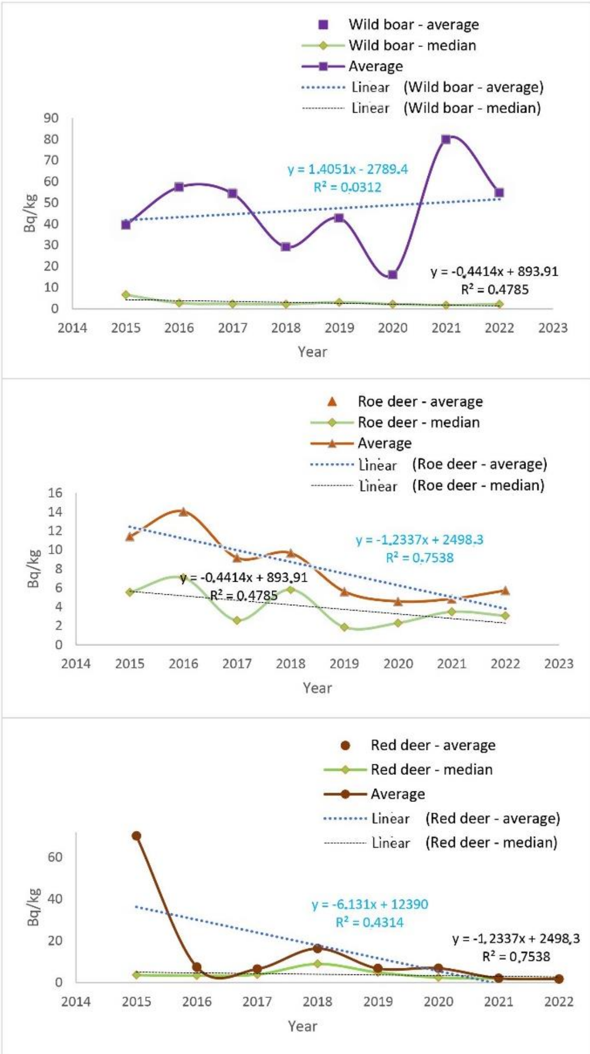

Fig. 3. Average concentrations of caesium-137 in wild boar, roe deer and red deer muscle samples and time trends covering the period from 2015 to 2022

Statistical calculations for Cs-137 in wild boar muscles. The regression analysis performed for wild boar muscles indicated a slow decreasing trend for the medians (slope coefficient −0.441 Bq/kg), which was also confirmed by the M-K test, which indicated a negative trend. However, regression analysis for average values apparently indicated an increase in concentrations over time (slope coefficient 1.405 Bq/kg), and the M-K test did not confirm statistically significant changes and indicated no trend over time. These discrepancies may have resulted from the occurrence of individual high concentrations in individual samples, which would have significantly affected the arithmetic mean. This also caused the coefficient of determination for the mean to be very low (3%) and indicated unsatisfactory fit. The coefficient of determination for the median was higher and amounted to (48%), but it was also not satisfactory in fit.

Statistical calculations for Cs-137 in roe deer muscles. In roe deer muscles, the mean and median showed a downward trend (respectively −1.234 and −0.441 Bq/kg), while the M-K test conducted in both cases did not confirm statistically significant changes. The coefficient of determination for the means indicated satisfactory agreement (75%), while for the median it was not satisfactory and amounted to 48%.

Statistical calculations for Cs-137 in red deer muscles. The regression analysis performed for deer muscles indicated a slow decreasing trend for the medians (−1.234 Bq/kg) and mean values (−6.131 Bq/kg). The M-K test for mean values showed a negative trend, while the regression coefficient was unsatisfactory (43%). For the median, the M-K test did not confirm any significant statistical changes, but the regression coefficient was satisfactory and amounted to 75%.

Figure 4 shows two maps of Poland. The map on the left (Fig. 4A) is reproduced from the 2011 edition of the Radiation Atlas of Poland (17). It presents the distribution of Cs-137 concentration in the surface layer (0–10 cm) of soil in Poland and shows that Poland is contaminated with this radionuclide in an uneven manner. The illustration on the right (Fig. 4B) shows the highest recorded concentrations of caesium-137 in the muscles of game animals in 2015–2022. Even though almost 40 years have passed since the Chernobyl accident, there are similarities between the areas contaminated after the disaster and the areas where the highest concentrations of caesium-137 were recorded in the muscles of game animals in the years 2015–2022. Extensive literature data confirm the levels of Cs-137 concentrations in soil occurring in and characteristic of specific regions of the country (20, 24).

Measurement of Cs-137 activity in wild boar muscles. Caesium-137 activity was determined in 508 wild boar muscle samples. In 321 samples (63.2%), the results exceeded the MDA value, and in 9 (1.8%) the measured concentrations exceeded the applicable limit (600 Bq/kg). The highest measured wild boar muscle concentration was 4,195 ± 372.0 Bq/kg. The average concentration in this matrix was 42.84 Bq/kg, the median was 2.370 Bq/kg and the calculated standard deviation showed a value of 251.4 Bq/kg.

Table 1. Average and ranges of caesium-137 concentrations in wild boar, roe deer and red deer muscle in individual years

Year	Wild boar muscle				Roe deer muscle				Red deer muscle			
	n	Average (Bq/kg)	Median (Bq/kg)	Range	n	Average (Bq/kg)	Median (Bq/kg)	Range	n	Average (Bq/kg)	Median (Bq/kg)	Range
2015	58	50.39	6.620	0.340–591.7	26	11.41	5.545	0.590–111.5	32	70.33	3.676	0.630–46.71
2016	66	57.26	2.715	0.400–1,089	24	14.06	7.085	0.500–110.5	29	7.519	3.400	0.570–37.61
2017	75	54.44	2.330	0.500–2,851	21	9.172	2.590	0.600–48.60	23	6.559	3.940	0.680–53.78
2018	61	29.19	2.170	0.130–693.0	17	9.671	5.840	1.150–73 84	14	16.36	8.985	0.340–86.67
2019	61	42.82	3.040	0.230–806.8	15	5.612	1.870	0.320–49.98	24	6.788	4.967	0.280–31.39
2020	65	16.09	2.160	0.320–203.6	8	4.583	2.310	0.620–14.40	22	6.930	2.480	0.500–38.58
2021	62	80.05	1.750	0.500–4,195	16	4.847	3.490	0.310–31.30	33	2.149	2.000	0.360–6.798
2022	60	54.80	2.165	0.500–988.3	18	5.732	3.070	0.500–31.30	28	1.774	2.000	0.490–6.000

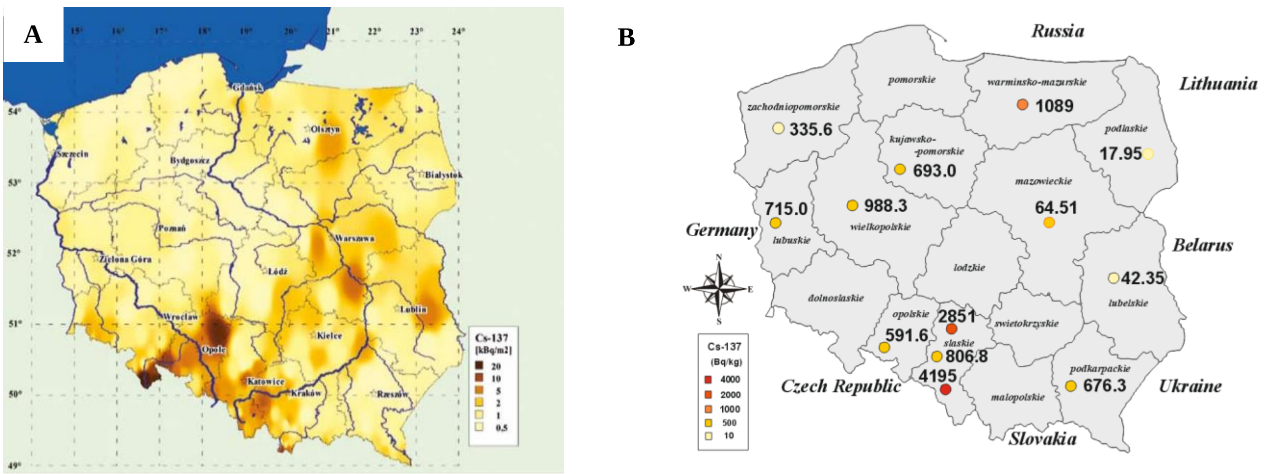

Fig. 4. A – The distribution of radioactive fallout in Poland (Isajenko *et al.* (17)); B – The highest recorded concentrations of caesium-137 in the muscles of game animals in 2015–2022

Table 2. Caesium-137 in the muscle tissue of the studied animals

Parameter	Matrix		
	Boar muscle (*Sus scrofa*)	Roe deer muscle (*Capreolus capreolus*)	Red deer muscle (*Cervus elaphus*)
Number of samples (n)	508	145	205
Minimum concentration (Bq/kg)	0.130	0.310	0.280
Maximum concentration (Bq/kg)	4,195	111.5	86.67
Average concentration (Bq/kg)	42.84	9.909	5.265
Standard deviation	251.4	16.11	9.952
Median	2.370	3.680	2.570
n > MDA	321	96	127
n < MDA	187	49	78

Table 3. Concentration ratios (CR) in the muscle tissue of the studied animals and effective equivalent doses of caesium-137 from their consumption

	AM (Bq/kg)	CR		
Year	Soil at depth up to 10 cm	Boar muscle (*Sus scrofa*)	Roe deer muscle (*Capreolus capreolus*)	Red deer muscle (*Cervus elaphus*)
2016	13.16	4.365	1.068	0.571
2019	12.50	3.425	0.450	0.545
2020	15.20	1.059	0.301	0.456
		Effective equivalent dose (µSv)		
2015	–	0.655	0.148	0.914
2016	–	0.744	0.183	0.097
2017	–	0.707	0.119	0.085
2018	–	0.379	0.126	0.213
2019	–	0.556	0.073	0.088
2020	–	0.209	0.060	0.090
2021	–	1.040	0.063	0.028
2022	–	0.712	0.075	0.023

AM – arithmetic mean

Measurement of Cs-137 activity in roe deer muscles. In the 145 roe deer muscle samples tested, 96 samples (66.2%) showed caesium-137 activity above the MDA value. However, in no case was the foodstuff limit of 600 Bq/kg exceeded. The highest concentration recorded in roe deer muscles was 111.5 ± 12.50 Bq/kg, and the average measured concentration was 9.909 Bq/kg, while the median was 3.680 Bq/kg. The standard deviation calculated for roe deer muscle samples was 16.11 Bq/kg.

Measurement of Cs-137 activity in red deer muscles. The last group of game animals examined were European red deer. A total of 205 muscle samples were tested. Measurement was possible in 127 samples (62%) of their caesium-137 concentrations, which, as in the case of the concentrations in roe deer muscles, did not exceed the MDA and averaged 5.265 Bq/kg during the period examined. The median was 2.570 Bq/kg. The highest concentration recorded was 86.67 ± 3.470 Bq/kg. The calculated standard deviation value for the tested red deer muscle samples was 9.952 Bq/kg.

Table 2 presents a summary of the analyses performed and the data obtained for the indicated matrices, *i.e.* average concentration, minimum measured concentration, maximum measured concentration, standard deviation, median and the number of samples above and below the MDA.

Determination of concentration factors and effective equivalent dose. The collected data on the concentration of Cs-137 in the soil for the years 2016, 2019 and 2020 sufficed for the determination of the CR for each tested matrix (Table 3). For the tested samples of wild boar muscles, the determined ratio was always above 1.000 Bq/kg, with the highest value (4.365 Bq/kg) recorded in 2016. However, for the tested muscle samples of both roe and red deer, the determined ratio was considerably lower at 0.500 Bq/kg, except in 2016, when it was 1.068 Bq/kg in roe deer tissue. With the collected data on the average concentration of Cs-137 in the years 2015–2022 in the muscles of wild boar, roe deer and red deer, it was possible to determine the effective equivalent doses. The estimated doses of Cs-137 absorbed by an adult through consumed game ranged from 0.209 to 1.040 µSv. For the roe deer and red deer muscle samples tested, the estimated dose did not exceed 0.213 µSv except in one case: in red deer tissue in 2015 (Table 3).

Discussion

Radiochemical tests carried out on the muscles of game animals indicate that the caesium-137 isotope is still present in the natural environment. Having a long half-life, caesium-137 is constantly present in the environment. This was confirmed by the tests carried out on the muscles of game animals. Over 60% of all the analyses performed for each tested matrix were results with measured activity of caesium-137. The data amassed in the experiment were extensive and made possible an attempt to estimate the current level of radioactive contamination of the Polish environment. The obtained results and calculations of the average concentration and standard deviation were used to determine the level of contamination. Additionally, the effective equivalent dose was estimated, and the CR was determined for each game matrix. The measured activity in muscle samples of game animals confirm that this group of organisms is prone to absorbing radioactive caesium-137. This is supported by two main factors: animals populating areas contaminated by radioactive fallout (mainly forest areas) and the way wild animals eat and feed (10). The feeding behaviour of wild animals and its effect on the uptake of radioactive caesium-137 was similarly described in a paper by Kapała *et al.* (20).

Among game animals, wild boar are the ones that have shown the greatest susceptibility to absorbing caesium-137. The average concentration for wild boar was three times higher than those for roe deer and red deer and amounted to 42.84 Bq/kg. Similar results were obtained by Kapała *et al.* (20) in their work, in which samples of meat from game animals from the Podlaskie voivodeship (Knyszyńska Forest) were analysed. Every year of the study period, significant concentrations exceeding the permissible limits were recorded in wild boar muscle samples. These values ranged from 591.7 Bq/kg to as much as 4,195 Bq/kg. Such high concentrations of caesium-137 were not recorded in the tested deer muscle samples of either species. The highest concentrations measured in roe deer tissue ranged from 14.40 to 111.5 Bq/kg. In the case of red deer tissue, the measured concentrations did not exceed 100 Bq/kg and values recorded in the studied period ranged from 6.000 to 86.67 Bq/kg (Table 2). The levels of Cs-137 in the muscles of game animals were similar to those in the work by Ołoś *et al.* (24) regarding game in the Opole Anomaly area in Poland in 2012–2020.

The presented results point to some difficulty in accurately estimating the level of Polish environmental contamination with the radionuclide caesium-137. The difficulty stems from the element's migration, which occurs to varying degrees into the soil and from the soil to the plants and fungi which constitute the basal nutrition of game animals (19). Another significant factor frustrating estimation attempts is the uneven distribution of radioactive fallout across the country. Radioactive fallout is often even point-like, causing so-called "hot spots" (2). Figure 4 compares data showing the distribution of radioactive fallout over Poland with data showing the distribution of the highest measured concentrations of caesium-137. The juxtaposed data confirm the similarity of the part of Poland affected by post-Chernobyl contamination with the geographical locations of the highest concentrations of caesium-137 measured in the study period.

Besides a geographical pattern, time trends may be noticeable in the average concentrations and medians of caesium-137 in the muscles of wild boar, roe deer and red deer, as Fig. 3 shows. Nevertheless, analysis based on statistical tests does not clearly indicate the existence of such time trends. Time trends are well described by Strebl and Tataruch (29) for roe deer and wild boar in Austria over two post-Chernobyl decades. In roe deer and red deer muscle samples, a very slow decay of caesium-137 over time can be noted in the present research data. Fluctuations in average concentrations were at a similar level. The current radioactive pollution of the environment can be estimated based on the 5.000–10.00 Bq/kg concentrations in these matrices. Wild boar muscles have a greater sorption capacity of caesium-137 from the environment, and correspondingly the estimate of environmental pollution was calculated at a higher concentration level when this matrix was considered than it was using roe deer and deer muscle sample concentrations. This was due to sporadic results in which the concentration of caesium-137 significantly deviated from the determined average. Based on the wild boar muscle tissue sample concentration of 42.84 Bq/kg, radioactive environmental pollution would be estimated to be considerably higher than when estimated based on the deer muscle concentrations. This suggests that environmental radionuclide occurrence should be estimated from tissue samples of more than one wild animal species.

Over the 40 years since the introduction of the transfer coefficient concept, much research has been conducted to determine values for a range of radionuclide–animal product combinations. To accurately estimate intake, both dietary composition and relative environmental pollution must be taken into account. The determined CR indicator shows and confirms the ongoing transfer of the Cs-137 radionuclide from the environment to the bodies of game animals. The largest uptake of Cs-137 occurred in wild boar, and the calculated CR was several times lower for roe and red deer. The effective equivalent dose based on the consumption of wild boar, roe deer and red deer muscle tissue and as determined in this investigation confirms the literature data on the highest possible exposure resulting from the consumption of game. In this case, there is also a noticeable correlation between the determined CR and the estimated effective equivalent dose. This confirms that the higher the transfer of caesium-137 is from the environment to living organisms, the greater the human exposure is to ionising radiation and the higher the dose is of that radiation. However, the research conducted and the results obtained do not currently indicate any significant threat from radioactive caesium-137 to humans.

Conclusion

Continuing measurements in game muscle samples will afford the right conditions for a fairly accurate assessment of any newly formed contamination in the event of an uncontrolled introduction of the radionuclide caesium-137 into the environment in the future. In light of the results collected and presented in this article, estimating the current level of pollution was an important undertaking, especially in the current geopolitical situation in Eastern Europe.

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