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Levels of caesium-137 in food of animal origin in Poland

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Abstract

Introduction: Radioactive contamination of the environment is one of the greatest threats after a nuclear accident due to released radionuclides. From a radiotoxicological point of view, the most important radionuclide is caesium-137. Formed mainly during nuclear explosions, caesium-137 can persist in the soil for many years, from where it constantly enters the food chain. One of the elements of ensuring food safety is the monitoring of its radioactive contamination, mainly with radioactive caesium isotopes. The aim of the study was to determine the content of caesium-137 in food of animal origin. **Material and Methods:** A total of 1,416 muscle samples from cattle, sheep, pigs, game and fish, as well as chicken eggs and dairy products were examined using gamma-ray spectrometry. **Results:** Caesium-137 activities ranged from below the minimum detectable activity concentration (MDC) to over 4,000 Bq/kg wet weight (w.w.). Most often, the values did not exceed the MDC or were in a range below 100 Bq/kg. The exception was the muscle tissue of game animals, especially wild boar, where a significant activity of caesium-137 was recorded, the highest of which was 4,136.8 ± 238 Bq/kg w.w. Committed effective doses determined for each matrix ranged from 0.01 to 0.83 μSv/kg, with the highest value determined for wild boar. **Conclusion:** The calculated exposure doses with values well below the accepted low radiation dose (100 mSv) did not indicate any significant amounts of ionising radiation from the food consumed.

Keywords: ^{137}Cs, contamination, animal food, Chernobyl, effective equivalent dose.

Introduction

Advances in science are making it possible to discover dangers in many areas of life which have long existed, and at the same time are furnishing new opportunities for their negation. One of the current main dangers to which special attention should be paid is environmental pollution. Caring for the environment involves taking extensive measures to prevent its continued pollution. An example of pollution that can negatively affect the quality of the environment quite significantly and for many years is radioactive contamination. The main source of radioactive pollution occurring in Poland was the 1986 Chernobyl nuclear power plant accident. As a result of this accident, a radioactive cloud spread across Poland causing very uneven contamination. A number of health- and life-threatening radionuclides were released into the environment, including caesium-137, caesium-134, strontium-90 and iodine-131. The radionuclides released could quite easily enter living organisms, causing increased morbidity of individual tissues. Thirty-six years after the event, today the two radionuclides caesium-137 and strontium-90 play the largest role in contamination. This is due to the relatively long half-lives of these radioactive elements, which are 30.2 years for caesium-137 and 28.8 years for strontium-90. Owing to its similarities to potassium, caesium-137 is a major contaminant in food products, so it has also become an indicator of radioactive environmental pollution (12). In areas where there was rainfall and high density of the Chernobyl accident radioactive cloud, there was greater contamination. Because of wet deposition in the first days after the accident, ^{137}Cs flowed with rainwater from leaves to the surface of the soil litter (21). Literature data indicate that in this way radioactive caesium-137 entered the soil-plant-carnivore chain (5, 15, 16, 22). The vast majority of caesium-137 accumulates in the topsoil, which is rich in organic and mineral compounds. It has been suggested that the transfer of radioactive caesium from the soil by plants to animals is much higher in forest ecosystems than in agricultural environments, and the fall in caesium concentration is very slow (11, 13, 25).

The consequence of air and soil contamination with radionuclides has been and continues to be the contamination of food in Poland, especially that which forms the basis of the food basket. These food contaminants are the main source of radioactivity entering the human body. Exposure to ^{137}Cs by ingestion or inhalation distributes radioactive material to soft tissues, especially muscle tissue, increasing the risk of cancer (1).

Systematic surveillance of radioactive contamination of food of animal origin makes it possible to assess the radiological situation in the country reliably (4). These studies are carried out under the Regulation of the Polish Minister of Agriculture and Rural Development of June 21, 2017 (7). The activity of radioactive isotopes in food products should be evaluated against the values specified in the Regulation of the Council of Ministers of April 27, 2004 (6), stipulating that the maximum permitted levels in food products must not exceed 1,250 Bq/kg. This level refers to isotopes with a half-life greater than 10 days and mainly applies to ^{134}Cs and ^{137}Cs. Additionally, in accordance with European law, the activities of radioactive isotopes in foodstuffs and food products should be set against the values set out in Commission Implementing Regulation (EU) of August 5, 2020 (3). This document specifies that the concentration of the ^{137}Cs isotope may not exceed 370 Bq/kg in milk and milk products and 600 Bq/kg in all other foodstuffs and products.

Material and Methods

Research area. Material from different widely dispersed areas of Poland was used to determine the levels of ^{137}Cs in food of animal origin sent for testing as part of official monitoring tests of the state of radiation safety of food of animal origin in 2021 or for equivalent testing as part of a commercial service in 2017–2021. In both cases, the Radiobiology Department of the Polish National Veterinary Research Institute performed the tests. Random sampling was carried out on farms or at processing plants in the area shown in Fig. 1.

Research material. A total of 1,416 samples of food of animal origin were tested: 793 land animal muscle samples (from cattle, sheep, pigs, poultry and game), 195 fish muscle samples, 193 chicken eggs, and 235 milk and dairy product samples (including raw milk, cheese, milk powder and whey), as summarised in Table 1.

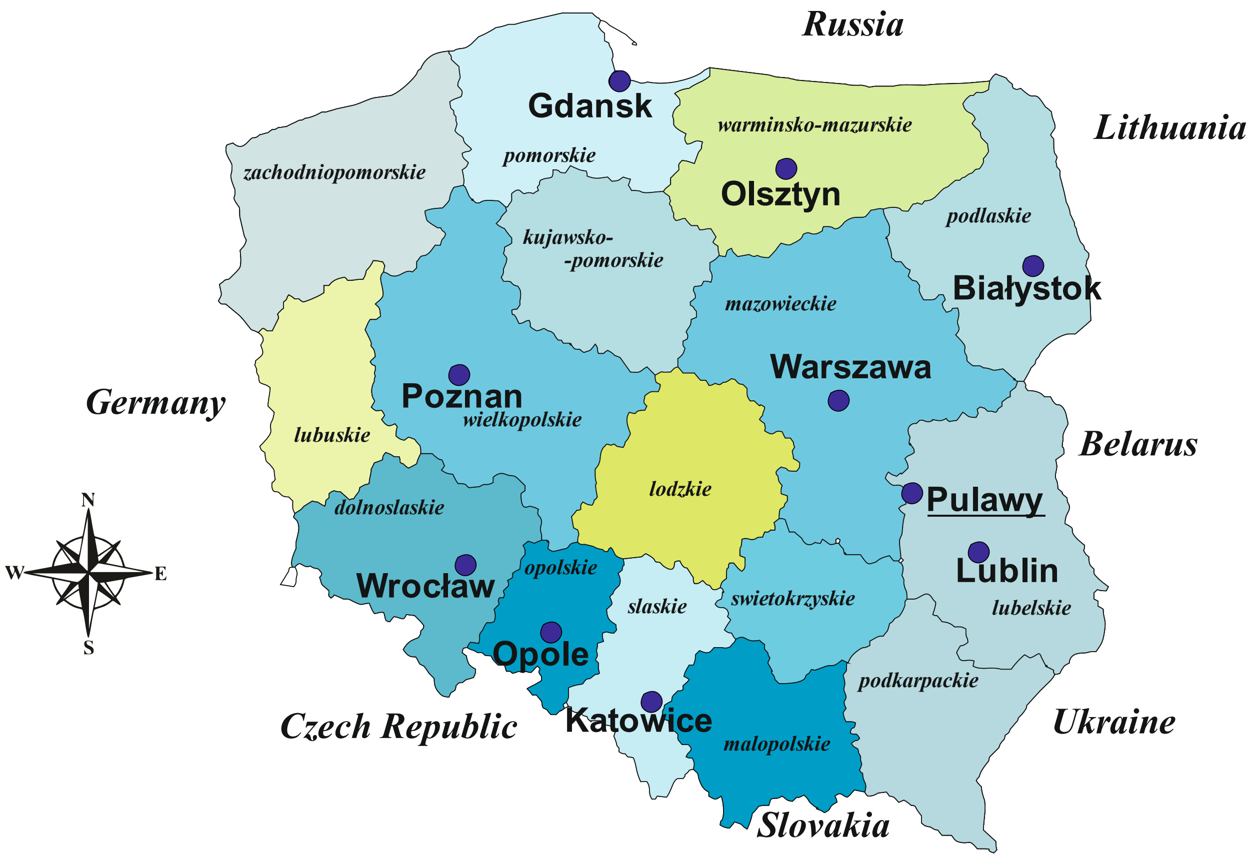

Fig. 1. Map showing the sampling area

Table 1. Summary of samples

	Official monitoring tests in 2021	Commercial sample tests 2017–2021	Total
Matrix		Number of samples	
Cattle – muscle tissue	202	3	205
Sheep – muscle tissue	79	-	79
Pigs – muscle tissue	180	3	183
Poultry – muscle tissue	192	20	212
Game animals – muscle tissue	110	4	114
Fish – muscle tissue	192	3	195
Chicken eggs	193	-	193
Raw milk	188	15	203
Cheese	-	26	26
Milk and whey powder	-	6	6
Total	1,336	80	1,416

Sample preparation. The initial sample preparation (*e.g.* of fish and carcass fragments of various animal species) consisted of separating inedible parts using a knife or scalpel. A Waring blender–type homogeniser, an electric grinding machine or a laboratory grinder was used to prepare the sample. The samples were not dried. Crushed, ground and standardised samples were transferred to Marinelli-type measuring containers of 450 cm^3 capacity.

Instrumental analysis. To determine the concentration of caesium-137, a gamma-ray spectrometry method was applied using a solid-state detector (high-purity germanium detector) manufactured and supplied by Mirion Technologies (Atlanta, GA, USA) and a lead-shielded NaI(TI) crystal scintillation detector manufactured by Scionix (Bunnik, the Netherlands). The full measurement set-up with the germanium detector consisted of the following components: a high-purity coaxial germanium detector of the GC1820-750SL type with a vertical dipstick cryostat and preamplifier, an IGS4/E lead shield, a miniNIM PPK Donor II power supply cartridge, a 3106D high-voltage power supply, a 2020 amplifier, a Multiport II MP2-1U multi-channel analyser, and a computer with peripherals. The scintillation detection was undertaken with the following equipment: a NaI(TI) type 51B51/2-E2 scintillation detector, a lead shield, a multi-channel analyser integrated with a high-voltage power supply, a preamplifier and an ASA-100 amplifier. The detectors prepared in this way determined radionuclides emitting gamma radiation in the energy range from 60 to 1836 KeV including caesium-137 (661.6 KeV). The following parameters were considered for sample activity calculations: net peak area, sample mass, attenuation-corrected yield, radioactive decay branching ratio, measurement time, and correction factors to account for nuclide decay. It was anticipated that samples would have ^{137}Cs activity at levels differing little from environmental samples and falling near the minima. Therefore, in each measurement it was important to determine the detection limit, the equivalent of which in radiometric studies is the accepted value of the MDA (minimum detectable activity) which is determined by the software for each sample individually. In order to determine the detection limit, Currie's definition of the limit was used (2).

The method used is one certified for competent performance by the National Veterinary Research Institute by the national accreditation body (17), and this competence has been repeatedly validated in proficiency tests. The geometry of the multi-nuclide source which was used to calibrate the detectors in respect of energy and efficiency was maintained. The energies of the emitted gamma quanta from the source were distributed over the entire energy range under study in such a way that the counting efficiencies of the measuring device could be determined with sufficient accuracy as a function of energy. The measurement time was 72,000 seconds (20 h). The collected gamma-ray spectra were analysed using Genie 2000 Version 3.1 software (Mirion Technologies) (Fig. 2).

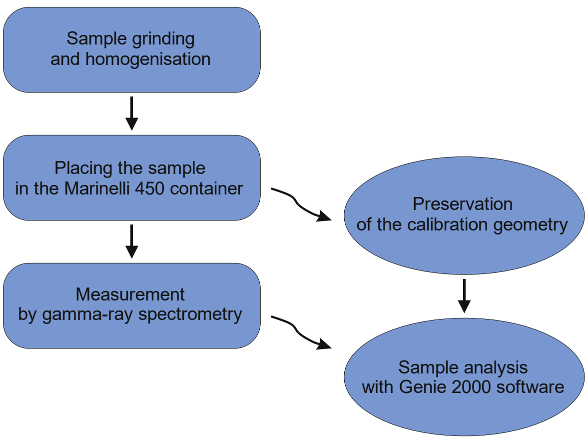

Fig. 2. Schematic diagram for gamma-ray spectrometry analysis

Statistical analysis and calculations. Caesium-137 activities for the studied species were presented with minimum, maximum, and arithmetic mean. The high percentage of results below the MDC (91%) prevented the use of more accurate statistics.

Assessment of radiation dose. In addition, calculations were performed to assess the radiation dose from ^{137}Cs for food consumers in Poland. A factor of 1.3×10^{-8} Sv Bq^{-1} was used to convert the dose per kg of body weight (9, 18). The committed effective dose coefficient for ^{137}Cs was defined by the equation defined by the equation:

$$H_E = A_k m D_{kf(k)}$$

where H_E is the committed effective dose (Sv), A_k is the concentration of radionuclide k (Bq kg^{-1}), m is the amount of food consumed (kg) and $D_{kf(k)}$ is the dose conversion factor for radionuclide k (27).

Results

The collected test results by food type are presented in Fig. 3. In most of the results obtained, minimal activity was recorded, which, as a rule, is characteristic for environmental samples. The MDA values obtained ranged from 0.30 to 2.50 Bq/kg.

Measurement of ^{137}Cs activity in cattle muscle tissue. In the study conducted, 205 muscle samples from cattle were measured. In 177 samples, values below the MDC were obtained, and in the remaining 28 samples, the values were above the MDC, the highest value being 23.5 ± 1.47 Bq/kg w.w. The average of the measured concentrations in cattle muscle samples was 3.08 ± 0.48 Bq/kg w.w.

Measurement of ^{137}Cs activity in sheep muscle tissue. Another type of meat tested was mutton. A total of 79 samples were tested, in 20 of which the results obtained were higher than the MDC; in the remaining 59 samples the results were below this value. The highest recorded activity among the sheep muscle samples tested was 48.4 ± 4.50 Bq/kg w.w. The average concentration of measured activities for this muscle group was 4.05 ± 1.40 Bq/kg w.w.

Measurement of ^{137}Cs activity in pig muscle tissue. Pig muscles were the next group of meat products tested. Results from 183 samples were reported. Only one sample recorded a value exceeding the detectable minimum, the results falling below this limit for 182 samples. The measured activity in the test sample was 3.18 ± 0.32 Bq/kg w.w.

Measurement of ^{137}Cs activity in poultry muscle tissue. A total of 212 samples of poultry muscle were tested. No activities higher than the MDC were found in this group of products and the typical results oscillated around 1.21 Bq/kg w.w.

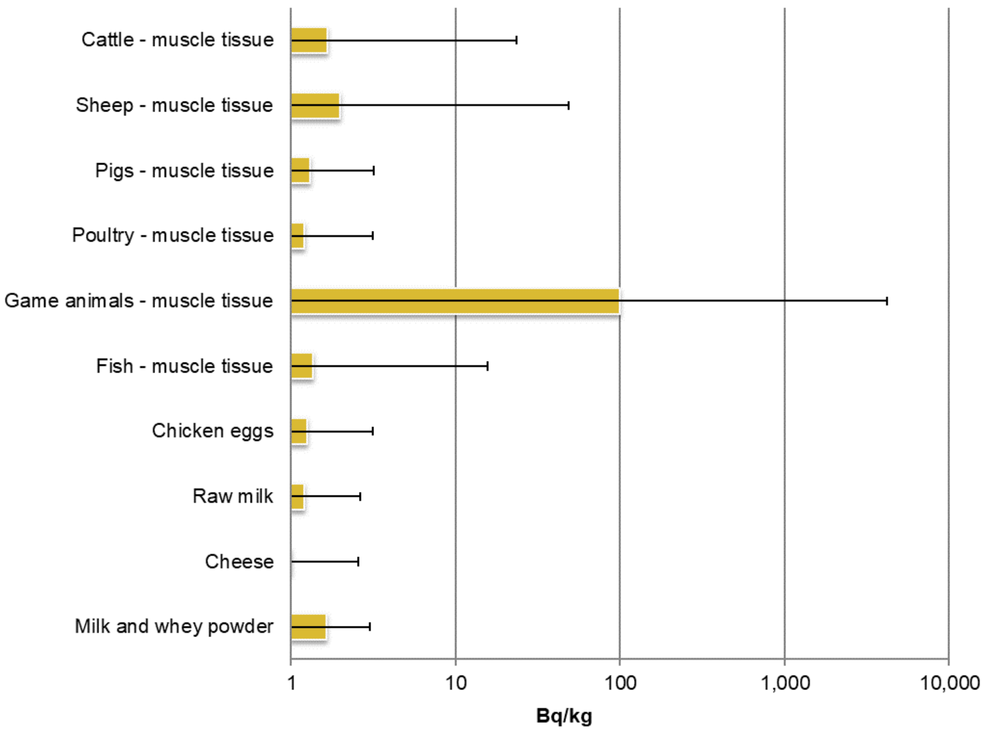

Fig. 3. Summary presentation of the caesium-137 activity in all types of tested samples
brown highlighting – average of all samples of a matrix; whiskers – maximum values. The scale is logarithmic for better representation of all matrices in one graph accommodating the over tenfold greater caesium-137 activity in game animal muscle tissue than in other matrices

Measurement of ^{137}Cs activity in game animal muscle tissue. Results for 114 wild-game muscle samples were obtained. Among the animals tested in this group were wild boar, roe deer and European deer. Muscle samples from wild boar predominantly showed significant caesium-137 activities, with the highest being 4,136.8 ± 238 Bq/kg w.w. Importantly, in this sample type exclusively, there were more results above the MDC (67 samples) than results below it (47 samples). The average concentration of measured activities in wild game muscle samples was 113.7 ± 27.5 Bq/kg w.w.

Measurement of ^{137}Cs activity in fish muscle tissue. Another numerous product group tested (195 samples) was fish. These were freshwater fish (*e.g.* carp, trout, bream and pike) from farmed fisheries operated in ponds as well as from lakes and rivers. Values above the MDC were recorded in 4 samples, of which the highest measured activity was 15.6 ± 1.55 Bq/kg w.w. The average of the measured activities was 7.62 ± 0.48 Bq/kg w.w.

Measurement of ^{137}Cs activity in chicken eggs. Another tested matrix, quite important in terms of consumption in Poland, was chicken eggs. Homogenised whole eggs (yolk and albumen) were used for the analysis. All 193 samples tested had values below the MDC.

Measurement of ^{137}Cs activity in milk and dairy products. The final product group tested was raw milk and dairy products (cheese, whey and milk powder), which also account for a significant share of all food consumption in the country. A total of 235 samples was tested. Of the 203 raw milk samples tested, only 3 samples yielded a value above the MDC. The average concentration was 1.21 ± 0.12 Bq/kg. The remaining samples did not exceed the indicated MDC level. As for cheese, 26 samples of this matrix were tested. In 25 samples, the activities obtained were below the MDC and only 1 sample was above it (2.57 ± 0.44 Bq/kg w.w.). In dairy products such as milk powder and whey, of the 6 samples tested, 5 had values slightly above the MDC value. The average concentration was 1.93 ± 0.42 Bq/kg.

Percentage of the results obtained above and under the MDC. The percentages of results for each product group in which the measured activity exceeded or was below the MDC are shown in Table 2. The data presented in the table allow a preliminary assessment of the tested matrices in which there is a higher probability of ^{137}Cs penetration. Foods for which the vast majority of results (93–100%) did not exceed the MDC value included pork, poultry, fish, chicken eggs, milk and dairy products. On the other hand, three groups of products can be distinguished for which the number of results exceeding the MDC value was noteworthy. These were primarily the meat of game animals (principally wild boar), in 59% of the samples of which concentrations above the MDC were found. The next two food products in which a higher number of results above the MDC were noted were mutton (25% of samples) and beef (14% of samples).

Effective equivalent dose. The consumer exposure to ionising radiation from caesium-137 resulting from the consumption of one kg of a given product was calculated from the results. These calculations of the effective equivalent dose were used to determine the exposure (Table 3) as first described by Šprem *et al.* (23).

The average activity from all tested samples of a given matrix was used to determine the effective dose. This made it possible to determine any possibility of exposure, both from contact with samples in which no activity was found, and from contact with samples with significant activity. After the effective equivalent doses for each matrix had been calculated, only game muscle had a value which was higher than the overall trend, this value being 0.83 μSv/kg. In the other matrices, the calculated value was around 0.02 μSv/kg.

Table 2. Number of samples and % of results below and above MDC values for each matrix

Matrix	<MDC		>MDC	
	n	%	n	%
Cattle – muscle tissue	177	86	28	14
Sheep – muscle tissue	59	75	20	25
Pigs – muscle tissue	182	99	1	1
Poultry – muscle tissue	212	100	-	-
Game animals – muscle tissue	47	41	67	59
Fish – muscle tissue	191	93	4	7
Chicken eggs	193	100	-	-
Raw milk	200	98	3	2
Cheese	25	96	1	4
Milk and whey powder	1	17	5	83

MDC – minimum detectable concentration

Table 3. Effective equivalent dose determined for each matrix

Matrix	Average concentration of ^{137}Cs (Bq/kg w.w.)	H_E per kg of product (μSv)
Cattle – muscle tissue	1.66	0.02
Sheep – muscle tissue	1.98	0.03
Pig – muscle tissue	1.31	0.02
Poultry – muscle tissue	1.21	0.02
Game animals – muscle tissue	99.7	0.83
Fish – muscle tissue	1.36	0.02
Chicken eggs	1.25	0.02
Raw milk	1.21	0.02
Cheese	0.42	0.01
Milk and whey powder	1.65	0.02

Discussion

When ionising radiation is present in food products, there is potential for exposure to radiation through the consumption of this contaminated food. Indeed, food taken into the body is one of the main routes of possibly harmful exposure to ionising radiation. Depending on the type and amount of radiation, damage to cellular DNA structures and internal organs can occur, and is certain to occur at high radiation doses (30).

Data on amounts and types of food consumed is important for determining consumer exposure to harmful ionising radiation from caesium-137. The investigations provided information on the levels of caesium-137 in the studied food groups of animal origin, which were basic constituents of the Polish diet. Similarly to Grabowski *et al.* (8), we found low levels of ^{137}Cs in foods in Poland. The calculated effective doses indicate very low consumer exposure resulting from the consumption of each matrix tested.

Statistics from recent years show meat continuing to be an important part of the Polish food basket (14, 24). The most commonly consumed types of meat in the country include pork, poultry and beef. Meat is a matrix susceptible to absorption of radioactive ^{137}Cs. This is confirmed by the results of muscle samples of animals living in a highly contaminated environment. In the present study, measurable caesium-137 activity above MDC values was found mainly in muscle tissue from game animals, sheep and cattle. As a result of the migration of caesium-137 from the soil to vegetation and fungi, there is a risk of radioactive contamination of animals concentrated in pastures (sheep and cattle) and a risk of significant radioactive contamination of wildlife consuming contaminated vegetation and fungi in forest ecosystems (game animals). This is partly due to the diet of wild animals and the way they eat. Forest mushrooms and berries, which constitute a significant part of the diet of game animals, are characterised by a particularly high concentration of caesium-137 activity. Some species of fruit plants and fungi found especially in forest environments are prone to uptake of radionuclides from the soil (12).

In addition, the lack of major intervention in forest ecosystems favours the accumulation of ^{137}Cs in the soil The surface layers of soil have accumulated this radionuclide as a result of the radioactive fallout (26). On the other hand, long-term soil reclamation in urban areas may have reduced the presence of ^{137}Cs in human-inhabited areas. The results confirm the transfer of caesium-137 from soil to vegetation, which occurs to a greater extent in forest ecological systems than from land used for livestock grazing. Because game species derive nutrition from forests, of all the matrices studied, wild game muscle tissue represents a unique group in terms of its potential for uptake of radioactive caesium-137 from the environment. Similarly to Zalewski and Szymczyk-Kobrzyńska (29), Rachubik (18, 19) and Vilic *et al.* (27), we found the phenomenon of samples with high activities in these animals' samples, sometimes exceeding the current permissible limits under the regulations. As shown in Table 2, 59% of the tested samples of this matrix showed activities above the MDC. The highest caesium-137 concentrations obtained were recorded for wild boar. The results of our study pertaining to game were generally consistent with those of Oloś and Dołhańchuk-Śródka (15,16), while activities above the permissible limit of 1,250 Bq/kg were found among wild game samples only in our study. The boar sample tested in which a value of 4,136.8 ± 238 Bq/kg was recorded is evidence of there being sites in Poland that are quite heavily contaminated with caesium-137, as are also described in the literature (10, 20, 29). Livestock that do not graze on meadows or pastures have negligible environmental uptake of caesium-137. In the case of muscle samples from pigs, the great majority of the results obtained did not exceed MDC values.

Another important group of food products for Polish consumers, and one which can derive from animals which graze, is milk and its products, mainly cheese. These foodstuffs also were generally only contaminated to an extent below the detectable concentration. Significant consumption of poultry and chicken eggs is also a stable aspect of Polish dining habits. Fortunately, no instances of samples exceeding the MDC were noted in these matrices.

In contrast, lower consumption is noticeable in the case of fish. Values above the MDC were reported for some fish samples. Fish (*e.g.* carp, trout, bream, pike) from breeding fisheries operated in ponds as well as fish from lakes and rivers were tested. According to the literature, contamination with radioactive caesium is more serious and persistent in freshwater fish, especially those living in rivers and ponds, than in marine fish. This is due to the continuous uptake of radiocaesium by the food for these fish. How contaminated freshwater fish become with radionuclides depends on the habits and size of the fish, as well as the biotic properties in the water and the surrounding environment (28).

The effective equivalent dose calculated for each matrix tested made it possible to determine the degree of exposure resulting from the ingestion of the amount of caesium-137 expected in 1 kg of the product tested. Based on the calculations, there is no danger of exposure to ionising radiation from the intake of significant amounts of the food materials investigated. Despite the muscle tissue of game animals standing out from the other food matrices in the concentration of radioactivity, the obtained value of 0.83 μSv/kg in this matrix also poses no danger. The average annual consumption of products from each study group does not expose the person eating food of that group to radiation exceeding the background, which is 2.4 mSv/year.

The tested food of animal origin intended for consumption in Poland is fully safe and poses no health risks in respect of radionuclide activity. However, it should be noted that there is a possibility of wild boar meat being consumed which could be contaminated with caesium-137 and could emit ionising radiation. For the safety of consumers who might frequently make the choice to eat wild boar, moderate consumption of such meat is recommended.

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