

DETERMINATION OF RADIONUCLIDE ^{137}Cs AND ^{134}Cs ACTIVITY IN HONEY AND POLLEN SAMPLES HARVESTED IN VARIOUS AREAS OF ROMANIA

DETERMINAREA ACTIVITĂȚII RADIONUCLIZILOR ^{137}Cs ȘI ^{134}Cs ÎN PROBE DE MIERE ȘI POLEN RECOLTATE DIN DIFERITE ZONE ALE ROMÂNIEI

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ABSTRACT | REZUMAT

Honey is a concentrated product in which, certain environmental contaminants such as $^{137}\text{Caesium}$ (Cs) and ^{134}Cs radionuclides can appear naturally, caused by reducing the water content of flower nectar. Cs is an environmental contaminant, a fission product with a long lifespan (radioactive half-life of 30 years) dispersed globally as a result of atmospheric nuclear weapons testing in the mid-20th century. The hive products are considered of animal origin, and the determination of the radioactive contamination degree is subject to current legislation. In order to determine the degree of radioactive contamination of animal-origin foodstuffs, namely the determination of ^{137}Cs and ^{134}Cs radionuclides in samples collected from hives (honey and pollen), were analysed by Gamma spectrophotometry with an HPGe detector at a reference laboratory in Romania (IISPV Bucharest). Results obtained in the radionuclides analysis of ^{137}Cs and ^{134}Cs in honey and pollen from 10 counties in Romania were all below the limit of the specific detectable activity. Specific detectable activity analysis results show that the combined radioactivity of ^{137}Cs and ^{134}Cs had values between $<4.22 - <9.47 \text{ Bq/kg}$ in honey, and between $<6.54 - <8.40 \text{ Bq/kg}$ in pollen, compared to the level allowed by legislation which is up to 600 Bq/kg . Radioactivity does not pose a danger to humans from the consumption of honey, but the cumulative bee consumption of ^{137}Cs and ^{134}Cs from honey and pollen can shorten their life and lead to depopulation, especially during over-wintering. Based on the produced honey, bees have been proven to be a useful indicator of air pollutants and modern pollution hotspots.

Keywords: radionuclide, ^{137}Cs , ^{134}Cs , honey, pollen

Mierea este un produs concentrat care prin reducerea conținutului în apă al nectarului provenit din flori, poate concentra în mod natural și pe unii contaminanți din mediu precum radionuclizii de $^{137}\text{Cesiu}$ (Cs) și ^{134}Cs . Cs este un contaminant al mediului, un produs de fisiune cu durată lungă de viață (timp de înjumătățire radioactivă de 30 de ani) dispersat la nivel global ca rezultat al testării armelor nucleare atmosferice de la mijlocul secolului XX. Produsele stupului sunt considerate produse de origine animală, iar pentru determinarea gradului de contaminare radioactivă sunt supuse legislației actuale. În vederea determinării gradului de contaminare radioactivă a produselor alimentare de origine animală, respectiv determinarea radionuclizilor de ^{137}Cs și ^{134}Cs din probe provenite din stupi (*miere și polen*), au fost efectuate analize prin spectrofotometrie gamma cu detector HPGe la un laborator de referință din România (IISPV București). Rezultatele obținute la analiza pentru radionuclizi de ^{137}Cs și ^{134}Cs în miere și polen din 10 județe din România s-au situat toate sub limita activității specifice detectabile. Analiza rezultatelor activității specifice detectabile arată că radioactivitate cumulată de ^{137}Cs și ^{134}Cs a prezentat valori cuprinse între $< 4,22 - < 9,47 \text{ Bq/kg}$ la miere, și între $< 6,54 - < 8,40 \text{ Bq/kg}$ la polen, față de nivelul permis de legislația în vigoare de până la 600 Bq/kg . Radioactivitatea nu prezintă un pericol pentru oameni în urma consumului de miere, dar consumul cumulat de ^{137}Cs și ^{134}Cs prin miere și polen de către albine poate duce la scurtarea vieții albinelor și la depopulări în special în sezonul inactiv. Albinele, prin mierea produsă, sunt un indicator util al contaminanților din atmosferă și de identificare a „punctelor fierbinți” de poluare modernă.

Cuvinte cheie: radionuclizi, ^{137}Cs , ^{134}Cs , miere, polen

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$^{137}\text{Caesium}$ (Cs) is a long-life fission product (radioactive half-life of 30 years) dispersed globally as a result of atmospheric nuclear weapons testing since the mid-20th century, when five countries tested more than 500 nuclear weapons, which taken together, emitted much more ionizing radiation into the atmosphere than any other event in human history. Most of these weapons were detonated in a few locations in the northern hemisphere: the Marshall Islands in the Pacific

Ocean (USA) and Novaya Zemlya (former USSR), which hosted over 75% of the energy efficiency of all tests. Many of the aerial detonations were so powerful that dozens of radioactive fission products were injected into the stratosphere (^{137}Cs , ^{90}Sr , ^{131}I) and distributed globally, with a residual time of about 1 year before their deposition by precipitation. In 1963, the Nuclear-Test-Ban Treaty effectively restricted atmospheric testing, with the exception of a few dozen smaller detonations, which took place in China and Africa in the 1960s and 1970s. The presence of nuclear tests' radioactive pollution is ubiquitous worldwide and can be detected on every continent even in deep ocean waters (6). While most of the radiation produced by the detonation of a nuclear weapon decreases in the first few days, one of the most longevity and abundant fission products is ^{137}Cs , which has a radioactive half-life of 30.2 years (5). Recent research suggests that even small amounts of ^{137}Cs can kill organisms and disrupt essential ecosystem services. Cs is not necessary for the growth and functioning of plants, but because Cs has an ionic charge and a radius similar to potassium (mimics potassium), which is an essential element for plants; ^{137}Cs is absorbed from the soil into vegetation through potassium-specific membrane transporters. This pathway of ^{137}Cs from soil to plant and then into the human diet was predicted decades ago, leading to a radionuclide surveillance program supported by the US government, which conducted extensive milk testing in the late 19th century (1950–1980). A US study showed the first measurements of ^{137}Cs in honey from the eastern US, in which it was observed that the concentrated effects on soil-plant-pollinator can increase by a few magnitude orders ^{137}Cs in honey from specific geographical regions with a low potassium soil content. Potassium in soil decreases plant uptake of ^{137}Cs and ultimately limits this radionuclide by several mechanisms. Experiments on a number of plant species indicate that Cs is not required for vegetation and is therefore never absorbed preferentially over K, but rather that Cs is taken up because its ionic properties are almost similar to K and allow its mobility through the transport channels of K. However, increasing the availability of potassium simply reduces the absorption of Cs, due to the mass action relationships associated with the plant-soil cation exchange; thus, K-based fertilizers are a proven method of reducing the absorption of ^{137}Cs by food crops from heavily contaminated soils (12). Ammonium ion (NH_4^+) also competes with Cs in these reactions, further explaining why honey from such agricultural systems tends to be very low in ^{137}Cs . Soil mineralogy likely plays an important role in seizing ^{137}C plants. K-rich soils tend to have a family of clay minerals with a strong ability to absorb or include Cs, thus reducing its bioavailability (4). Vegetation

that is thousands of miles away from nuclear test sites continues to attract ^{137}Cs , as it mimics potassium and, as a result, bees increase this radionuclide in honey. In the eastern United States, although there have been no tests for atmospheric nuclear weapons, most honey contains detectable ^{137}Cs at over 0.03 Bq / kg, while in the south-eastern United States, the activities of ^{137}Cs radionuclide may be > 500 times larger. Using honey measurements, US researchers presented regional models in the biogeochemical cyclization of ^{137}Cs and concluded that plants and animals receive a disproportionately high exposure to ^{137}Cs ionizing radiation in low-potassium soils. In several cases, the presence of ^{137}Cs has, even more, doubled the ionizing radiation from gamma and X-rays in honey, which indicates that despite its radioactive half-life, the environmental legacy of regional pollution by ^{137}Cs can persist for more than six decades (4).

Studies conducted in Japan on the blue butterflies *Zizeeria maha* have shown an increase in deformation and mortality when these insects consume radioactive vegetation ranging from 2.5–48 Bq / kg (7). Subsequent studies on a different species of butterfly, *Pieris rapae*, also indicate that ingestion of low levels of radiocesium, even at levels lower than the natural activity of ^{40}K , has had negative biological effects. Other recent studies indicate that the negative effects of ionizing radiation on insects begin only above a threshold of 40 $\mu\text{Gy h}^{-1}$ (4).

Several long-standing international agreements on nuclear non-proliferation and weapons control have been dismantled in recent years, and certain leaders have begun to place greater emphasis on the role of nuclear weapons in their military strategies. These developments may lead to the renewal of arms testing activities by the nine nuclear-capable nations and may encourage other nations to start their own testing programs. In addition, given the relatively low efficiency and footprint of nuclear greenhouse gases for electricity generation, it is likely that in the coming years there will be an increased dependence on nuclear processes as a source of fuel for domestic and military needs. Thus, there is a likelihood of future releases of fission products into the environment (4).

Given that pollinating insects provide vital services to the world's ecosystem, which are essential for maintaining global food security, more research is needed to help us better understand how ionizing pollution threatens their health and survival.

As honey is a concentrated product, reducing the water content of the flower nectar can naturally create five times the amount of environmental contaminants (3). In the years following the 1986 event at the Chernobyl nuclear power plant, several researchers documented the presence of ^{137}Cs in honey and pollen from the European nations affected by the event,

stating that, with only a few exceptions, the load of ^{137}Cs in honey is generally safe for human consumption and that honey is a useful indicator of airborne contaminants and for the "hot spots" identification of the modern pollution (4). The purpose of the present research is to determine the degree of radioactive contamination of bee products (honey and pollen) produced in Romania. These researches are sporadically found in the literature and refer only to samples from bee colonies in Romania (2).

MATERIALS AND METHODS

According to the legislation in force, the products of the hive are considered products of animal origin. In order to determine the degree of radioactive contamination, samples of honey and pollen were taken through a specific procedure of sampling, transport, and storage of samples, according to the legislation in force, from private apiaries located in different Romanian Counties (PH, SJ, AG, IS, CT, TL, BR, AR, VN, and IL), in the beekeeping season 2020-2021. The location of the counties of origin of honey and pollen samples on the Romanian map is represented schematically in Fig. 1.



Fig. 1. The counties of origin of the investigated honey and pollen samples

A total of 17 samples were collected from 10 apiaries, including 8 samples of honey (500 g / sample), 2 samples of honeycomb with honey (500 g), and 7 samples of pollen (200 g / sample). A double sample of both honey and pollen was collected from 7 of the studied apiaries. The predominant type of honey was polyfloral, with samples of rapeseed and honeydew honey, and pollen was predominantly rapeseed. Radioactive contamination control was performed at the IISPV Accredited Chemistry and Radioactivity Laboratory (11), subordinated to the competent authority, respectively ANSVSA (9). In order to determine the degree of radioactive contamination of animal-origin

food, respectively the determination of ^{137}Cs and ^{134}Cs radionuclides from hives samples (honey and pollen), analyzes were performed by gamma spectrophotometry with an HPGe detector.

RESULTS AND DISCUSSIONS

The laboratory analyses results received from the Bucharest Institute of Veterinary Hygiene and Public Health for determining the specific activity of radionuclides by gamma spectrophotometry with HPGe detector in honey and pollen samples are presented in Tables 1 and 2. The measurement results met the requirements specified in Ord. common no. 1805 / 29.12.2006 to MSP no. 286 / 08.12.2008 to ANSVSA, no. 314 / 06.2006 of CNCAN and Regulation (Euratom) no. 52 / 15.01.2016 of the EU Council to which RD4 (Non-Binary Decision Rule) applies, which takes into account the measurement uncertainty ($W = IM$). The specific risk of the result, represented by the probability of false acceptance is $<2.5\%$.

The cumulative radioactivity results of ^{137}Cs and ^{134}Cs radionuclides in honey and pollen samples are shown in Table 3. The analysis results for ^{137}Cs and ^{134}Cs radionuclides in honey and pollen from the 10 counties in Romania were all below the limit for the specific detectable activity. The analysis results obtained for the cumulative radioactivity of ^{137}Cs and ^{134}Cs show values in the range of $<4.22 - <9.47 \text{ Bq / kg}$ for honey, and between $<6.54 - <8.40 \text{ Bq / kg}$ for pollen, values that represent a percentage between 0.7-1.57% (for honey) and 1.09-1.40% (for pollen) compared to the permitted level of 600 Bq / kg for food of animal origin. This level of cumulative radioactivity of ^{137}Cs , ^{134}Cs is close to that found by Croatian honey researchers (1).

The cumulative radioactivity of ^{137}Cs and ^{134}Cs in bee honey samples was significantly lower (0.7-1.57%) than the maximum permitted values for food of animal origin, with higher values obtained in the polyfloral honeycomb sample from Sălaj county ($<9.47 \text{ Bq / kg}$), followed by rapeseed honey ($<5.24 \text{ Bq / kg}$) from Prahova County (samples shaded with green colour in table 3). The lowest cumulative radioactivity activity of ^{137}Cs and ^{134}Cs was detected in the sunflower honey sample ($<4.22 \text{ Bq / kg}$) from Tulcea county (shaded in yellow in Table 3). Regarding the pollen, the cumulative radioactivity of ^{137}Cs and ^{134}Cs was between $<7.08 \text{ Bq / kg}$ (Iasi county) and $<8.40 \text{ Bq / kg}$ (Arges county), which represents a percentage between 1.09-1.40% of the permitted level of 600 Bq / kg for food of animal origin. For pollen, as for honey, the values obtained are smaller than the minimum detectable specific activity. The found activities of ^{137}Cs and ^{134}Cs radionuclides in honey and pollen did not reach the permitted level of radionuclides for animal products. The activities of ^{137}Cs and ^{134}Cs radionuclides measured

were comparable to the values of honey samples collected in Europe (Czech Rep., Poland, Croatia) and those studied in Romania (2). The results obtained by us in Romania (<2.07 - <5.42 Bq /kg) indicate lower ^{137}Cs radionuclide values than those found in the Czech Rep. (max. 39.16 Bq /kg) (Pöschl 2011), Poland (max.

46.3 Bq/kg) and Croatia (34.3 Bq/kg) (13). The measurement results showed that the samples were compliant, as no cumulative radioactivity of ^{137}Cs and ^{134}Cs greater than 600 Bq/kg was detected, which is the permitted level of radioactivity in products of animal origin, in accordance with the legislation in force (9, 10).

Table 1

The results of the radionuclides' specific activity by gamma spectrophotometry with HPGe detector in honey samples determination

Sample no.	Analyzed sample type (honey)	Samples origin area	Test / analysis method / performance parameters	
			RESULTS $\pm$ IM (UM)	
			^{137}Cs (Bq / kg) * Specific Detectable Minimum Activity (AMD)	^{134}Cs (Bq / kg) * Specific Detectable Minimum Activity (AMD)
1.	Rapeseed	PH	<3.24 (AMD = 3.24) Bq / kg	<2.00 (AMD = 2.00) Bq / kg
2.	Honeydew	CT	<2.59 (AMD = 2.59) Bq / kg	<2.26 (AMD = 2.26) Bq / kg
3.	Polyfloral	AG	<2.64 (AMD = 2.64) Bq / kg	<1.96 (AMD = 1.96) Bq / kg
4.	Polyfloral	IS	<2.29 (AMD = 2.29) Bq / kg	<2.19 (AMD = 2.19) Bq / kg
5.	Polyfloral	TL	<2.07 (AMD = 2.07) Bq / kg	<2.15 (AMD = 2.15) Bq / kg
6.	Honeycomb with polyfloral honey	SJ	<5.42 (AMD = 5.42) Bq / kg	<4.05 (AMD = 4.05) Bq / kg
7.	Polyfloral	BR	<2.68 (AMD = 2.68) Bq / kg	<2.15 (AMD = 2.15) Bq / kg
8.	Polyfloral	AR	<2.34 (AMD = 2.34) Bq / kg	<2.33 (AMD = 2.33) Bq / kg
9.	Polyfloral	VN	<2.54 (AMD = 2.54) Bq / kg	<2.22 (AMD = 2.22) Bq / kg
10.	Honeycomb	IL	<2.49 (AMD = 2.49) Bq / kg	<2.36 (AMD = 2.36) Bq / kg

* Becquerel (Bq): the special name of the activity unit. A Becquerel is equivalent to one disintegration per second

Table 2

Results of the radionuclides specific activity determination by gamma spectrophotometry with HPGe detector in pollen samples

Sample no.	Analyzed sample type (honey)	Samples origin area	Test / analysis method / performance parameters	
			RESULTS $\pm$ IM (UM)	
			^{137}Cs (Bq / kg) * Specific Detectable Minimum Activity (AMD)	^{134}Cs (Bq / kg) * Specific Detectable Minimum Activity (AMD)
1.	Pollen	PH	<3.95 (AMD = 3.95) Bq / kg	<3.33 (AMD = 3.33) Bq / Kg
2.		CT	<3.36 (AMD = 3.36) Bq / kg	<3.18 (AMD = 3.18) Bq / Kg
3.		AG	<4.45 (AMD = 4.45) Bq / kg	<3.95 (AMD = 3.95) Bq / Kg
4.		IS	<4.10 (AMD = 4.10) Bq / kg	<2.98 (AMD = 2.98) Bq / Kg
5.		TL	<4.35 (AMD = 4.35) Bq / kg	<2.98 (AMD = 2.98) Bq / Kg
6.		SJ	<3.71 (AMD = 3.71) Bq / kg	<3.59 (AMD = 3.59) Bq / Kg
7.		BR	<4.38 (AMD = 4.38) Bq / kg	<3.79 (AMD = 3.79) Bq / Kg

* Becquerel (Bq): the special name of the activity unit. A Becquerel is equivalent to one disintegration per second

Table 3

Cumulative radioactivity of ^{137}Cs and ^{134}Cs in honey and pollen samples

Sample no.	Analyzed sample type	Samples origin area	Specific activity of radionuclides		Cumulative radioactivity ^{137}Cs and ^{134}Cs (Bq / kg)
			^{137}Cs (Bq / kg) *	^{134}Cs (Bq / kg) *	
1.	Rapeseed honey	PH	<3.24	<2.00	<5.24
2.	Honeydew honey	CT	<2.59	<2.26	<4.85
3.	Polyfloral honey	AG	<2.64	<1.96	<4.60
4.	Polyfloral honey	IS	<2.29	<2.19	<4.48
5.	Sunflower honey	TL	<2.07	<2.15	<4.22
6.	Honeycomb with polyfloral honey	SJ	<5.42	<4.05	<9.47
7.	Polyfloral honey	BR	<2.68	<2.15	<4.83
8.	Polyfloral honey	AR	<2.34	<2.33	<4.67
9.	Polyfloral honey	VR	<2.54	<2.22	<4.76
10.	Honeycomb	IL	<2.49	<2.36	<4.85
11.	Pollen	PH	<3.95	<3.33	<7.28
12.	Pollen	CT	<3.36	<3.18	<6.54
13.	Pollen	AG	<4.45	<3.95	<8.40
14.	Pollen	IS	<4.10	<2.98	<7.08
15.	Pollen	TL	<4.35	<2.98	<7.33
16.	Pollen	SJ	<3.71	<3.59	<7.30
17.	Pollen	BR	<4.38	<3.79	<8.17

Radionuclide ^{137}Cs and ^{134}Cs values identified in the honey and pollen samples in Romania are not a source of concern for consumers or the environment. International studies claim that although honey does not endanger human consumption, it is not as safe for the bees that produce it (a key indicator of what is happening in the environment) and it has become increasingly clear that insects suffer significant negative consequences due to radiation doses that were considered safe in the past. Other studies show that the level of Cs pollution can be fatal to bees, thus taking into account the continuation of research on bees from different areas, correlated with depopulation whose cause is not established.

The limits of the current research consist of the limited capacity to ensure the representativeness of the results for the entire area of Romania. Future research can be done on a representative sample based on characteristics such as distance from major cities and the apary keeping system.

Our research has confirmed the importance of monitoring the cumulative radioactivity of ^{137}Cs and ^{134}Cs in honey and bee pollen, as honey is a useful indicator of airborne contaminants and the identification of "hot spots" of modern pollution, but also regarding the fact

that plants and animals receive a disproportionately high exposure to ionizing radiation from ^{137}Cs in soils with the low potassium content.

CONCLUSIONS

The results obtained in honey and pollen ^{137}Cs and ^{134}Cs radionuclides analysis from 10 counties in Romania were all below the limit for the specific detectable activity. The analyses results of the specific detectable activity show that the cumulative radioactivity of ^{137}Cs and ^{134}Cs showed values between <4.22 - <9.47 Bq / kg for honey and between <6.54 - <8.40 Bq / kg for pollen, compared to the level allowed by the legislation in force up to 600 Bq / kg. The cumulative radioactivity of ^{137}Cs and ^{134}Cs in the honey samples showed higher values in the honeycomb sample with polyfloral honey from Sălaj county (<9.47 Bq / kg), followed by rapeseed honey (<5.24 Bq / kg) from Prahova County. The lowest cumulative radioactivity activity of ^{137}Cs and ^{134}Cs was detected in the sample of sunflower honey (<4.22 Bq / kg) from Tulcea County. The cumulative radioactivity of ^{137}Cs and ^{134}Cs for pollen samples was between <7.08 Bq / kg (Iasi County) and <8.40 Bq / kg (Argeş County). The values of ^{137}Cs and ^{134}Cs radionu-

clides identified in the samples of honey and pollen in our country are not a source of concern for consumers or the environment, but although honey does not pose a danger to human consumption, it is not as safe for bees which produce it, the insects suffering significant negative consequences due to radiation doses that were considered safe in the past. Given that pollinating insects provide vital services to the world's ecosystem, which are essential for maintaining global food security, more research is needed to help us better understand how ionizing pollution threatens their health and survival.

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