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EXAMINATION OF THE DYNAMICS OF ANALYZES PERFORMED FOR THE PRESENCE OF RADIONUCLEIDE RESIDUES IN FOOD IN THE PERIOD 2019-2023 IN THE REPUBLIC OF NORTH MACEDONIA

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Abstract

The main focus of this research is to determine the extent of analyzes performed for the presence of radionuclides in food, their dynamics, which radionuclides are present in food, and how much their radioactivity was in the period 2019-2023. The research covers the period from January 01, 2019. until December 31, 2023. The data we used to conduct this research were obtained from the Public Health Institute of R. N. Macedonia. The tests were carried out with accredited methods and by the Regulation on the maximum allowed amounts of radionuclides in food, water, air, land, products, and raw materials of animal and plant origin and objects for general use. Relative numbers and trends are used as statistical methods of work. According to the results, in the period 2019-2023, radionuclides were analyzed in 6065 samples (2711 samples from imports, and 3354 samples from domestic production). The largest amount of radioactivity analyzes were performed in the group of mushrooms (51%) and the group of grain and flour (44%). The scope of analyzes performed for the presence of radionuclide residues in food in R.N. Macedonia shows a downward trend. In the analyzed products, no radioactivity above the limit values was detected (except in 2020 when only in 1 product from the mushroom group - dry morel) detected a value for Cs-137 = 662.8 Bq/kg, and the maximum allowed value is 600 Bq/kg). It can be concluded from the study that food is safe in terms of the presence of radionuclide residues, but work should be done to increase the volume of analyzes performed for the presence of radionuclides in food, as well as to increase the coverage of different groups of products (fruit, vegetables, coffee, tea, etc.) in which such analyzes would be performed.

Keywords: radioactivity, radionuclides, food, analyses, trend, dynamics.

1. Introduction

Radioactivity occurs when the nuclei of atoms split into the nuclei of other elements, emitting beams of particles or electromagnetic waves (radiation), a process known as radioactive decay. A radioactive element is one whose nuclei gradually split in this way. Such nuclei split because they are unstable. This usually happens because they have either very large mass numbers, or an imbalance of protons and neutrons.

Radioisotope or radioactive isotope is a general term for a radioactive substance because they are all isotopes. Several radioisotopes are naturally occurring, such as carbon-14 and uranium-238, and others are created in several different ways [1].

There are several types of radiation. Radiation from radioactive substances can be: alpha-, beta- and gamma rays (see Table 1) [2].

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Table 1: Types of radioactive radiation and their characteristics

Kinds	Alpha particles (α)	Beta particles (β)	Gamma rays (γ)
	Each particle is composed of 2 protons and 2 neutrons	Every particle is an electron	Electromagnetic waves similar to X-rays
Charging	+	-	There is no charge
Mass	Much bigger compared to beta	Very small	...
Speed	Over 1/10th the speed of light	Over 9/10 of the speed of light	Speed of light
Ionization effect / breakthrough effect	Strong, not very penetrating: prevented by a thick sheet of paper or leather	Weak, penetrable: prevented by an aluminum plate of about 5 mm	Very weak, very penetrating: never fully absorbed, but a 25 mm lead plate cuts it in half
The effect of charges	They bend in magnetic and electric field	Many deflect in magnetic and electric fields	They do not deviate in magnetic and electric fields

The unit of radioactive irradiance is the becquerel (Bq).

1 Bq = 1 s-1

This means that one atom of the radioactive substance decays every second. The natural radioactivity of the human organism is greater than 4000 Bq [3]. Another commonly used unit of measurement for radioactive exposure is the curie (Ci). One curie is equal to 3.7 x 1010 becquerels. [1]

Half-life (t1/2) is the time required for half of a radioactive substance to decay. The unit of half-life is second (s). For different nuclides, the half-life is different and serves for their identification (see Table 2). [3]

Table 2: Half-life of certain radionuclides

A nuclide	Half -life
Tellurium - 130	1,4 x 10^{21} year
Uranus- 238	4,5 x 10^{9} year
Radium- 226	1,6 x 10^{3} year
Polonium - 211	0,52 s
Polonium - 212	3 x 10^{-7} s

Radioactive decay is a process in which the nuclei of a radioactive element undergo a series of disintegrations (decay series) and become stable nuclei of another element. [1] The disintegration of radioactive elements occurs according to the Law of Displacement of Soddy-Fajans (F. Soddy – K. Fajans).

According to the Soddy-Fajans Displacement Act (F. Soddy – K. Fajans):

- when an element emits α-rays, the newly created element is located two places to the left in the periodic table in relation to the outgoing element;

$$\frac{234}{92}U \rightarrow \frac{230}{90}Th + \frac{4}{2}He$$

- if the radioactive element emits β-rays, the newly created element is located one place to the right, in relation to the starting element, in the periodic system;

$$\frac{234}{92}U \rightarrow \frac{230}{90}Th + \frac{4}{2}He$$

- emitting γ-rays does not change either the atomic or mass number of the element. During γ-decay, the atom moves from an excited state to a lower energy state by emitting electromagnetic waves (γ-quanta).

Radioactivity has a wide range of applications:

- in medicine;
- the chemical industry;
- the so-called radiography to detect hidden defects in metals and other materials;
- activators of lighting systems (luminophores): e.g. lighting systems for tunnels, mines, lighthouses, clock lighting systems, etc.; [3]
- Food industry: treating food, such as fruit, with gamma rays to keep it fresh, etc. [1]

2. Goals

The main objectives of this research are:

- to determine the volume of analyzes performed for the presence of radionuclides in food in the period 2019-2023;
- to determine the development tendency of the volume of performed analyzes of radioactivity in food products;
- to determine the volume of radioactivity analyzes performed by groups of food products in the period 2019-2023;
- to determine which radionuclides are present in the food that was examined in the period 2019-2023;
- what is the value of the detected radioactivity in the food that was examined in the period 2019-2023.

3. Material and methods

3.1. Research material: The main focus of this research is to determine the extent of analyzes performed for the presence of radionuclides in food in the period 2019-2023, as well as to determine which radionuclides are present in food in the specified period and how much their radioactivity is. The research was conducted by: The Faculty of Food Technology and Nutrition at the University of Tetovo and VT Diet Club - Bitola. The research covers the period from 01.01.2019. until 31.12.2023 The data we used to conduct this research was obtained from the Institute of Public Health of the Republic of North Macedonia. The research is of a retrospective type. 1.1. Research methodology

In order to conduct this research, it was necessary to meet certain criteria:

- to determine the volume of performed radioactivity analyzes by groups of food products in the period 2019-2023;
- the tests should be carried out in accordance with the Rulebook on maximum permitted amounts of radionuclides in food, water, air, land, products and raw materials of animal and plant origin and objects for general use (Official Gazette of the Republic of Moldova No. 163/2009);
- the tests should be performed with accredited methods.

3.2. Statistical method of data processing: Relative numbers and trend are used as statistical methods of work. The data are presented tabularly and graphically.

4. Results

Table 3 shows the distribution of performed radioactivity analysis in the period 2019-2023

Table 3: Distribution of performed radioactivity analyzes in the period 2019-2023.

Year	Import	Trade and domestic production	Total
2019	1421	670	2091
2020	774	608	1382
2021	340	627	967
2022	113	709	822
2023	63	740	803
In total	2711	3354	6065

In the period 2019-2023, radionuclides were analyzed in 6065 samples. 2711 samples were examined from imports, and 3354 samples were from domestic production.
Graph 1 shows the distribution of radioactivity analyzes performed by groups of food products in the period 2019-2023.

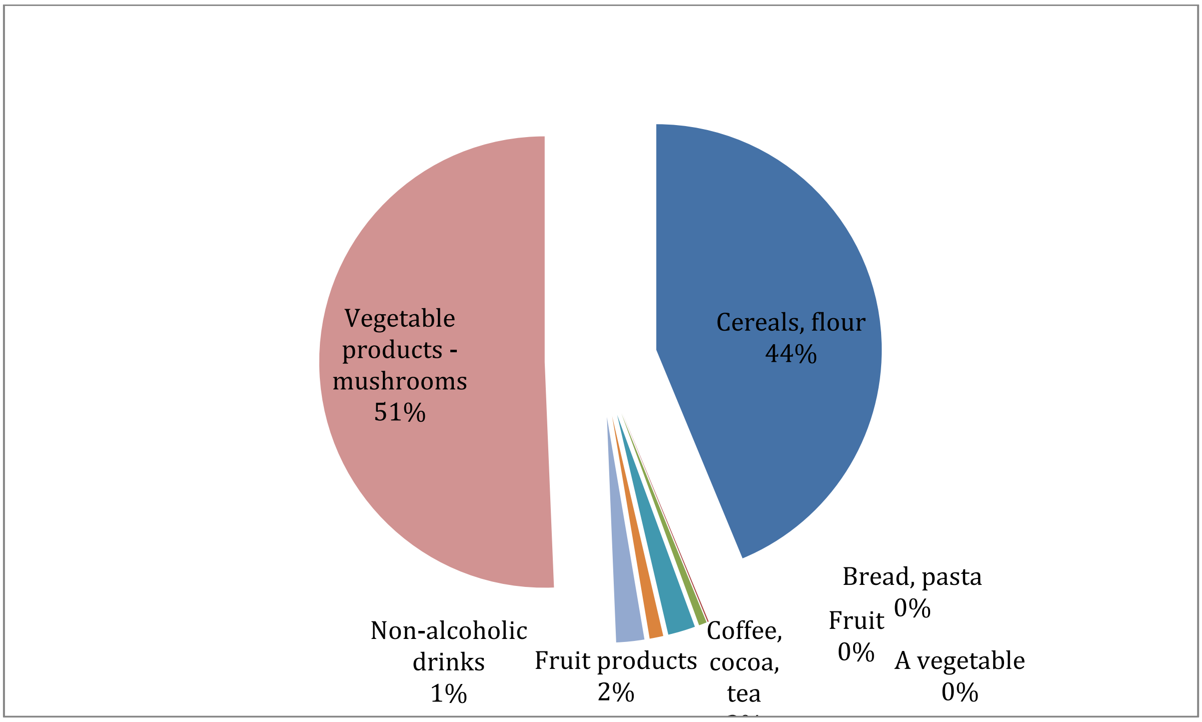

Chart 1: Distribution of performed radioactivity analyzes by groups of food products in the period 2019-2023.

According to graph 1, the largest volume of radioactivity analyzes were performed in the group of mushrooms (51%) and the group of grain and flour (44%) in the period 2019-2023.

Chart 2 shows the variations in the volume of radioactivity analyzes performed during the period 2019-2023.

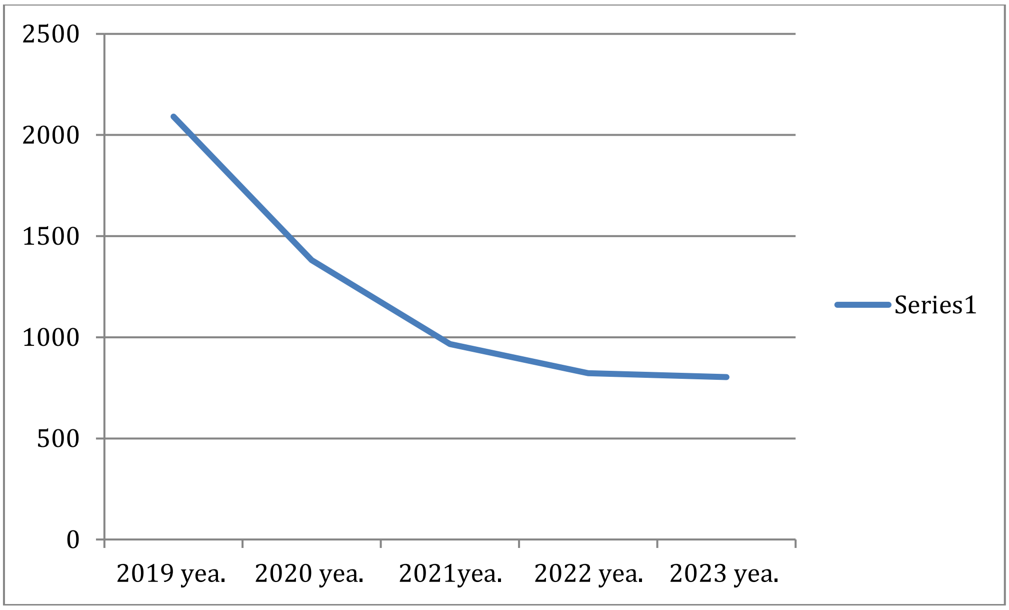

Chart 2: Variations in the volume of radioactivity analyzes performed in the period 2019-2023.

From graph 2, it is not possible to see the development tendency of the volume of performed radioactivity analyzes in the examined period. To determine the development tendency, a trend is created.

There are several types of trend: linear, parabolic, logarithmic, etc. The line trend is mostly used. It is made according to the formula:

$$y = a + bx$$

In the formula, y represents the trend value, a and b are trend coefficients, and x represents time. The value of the trend coefficients is determined by the formulas:

$$a = \frac{\sum y}{N}$$

$b = \frac{\sum xy}{\sum x^2}$In Table 4, the time values (X) are determined in such a way that the middle year (2021) in the analyzed period is marked with 0 (X = 0). The values of (X) above zero are indicated as: -1, -2, and below zero 1, 2. Y shows the individual frequencies of radioactivity analyzes performed in the analyzed period. XxY shows the individual products, and when calculating the sum, attention should be paid to the negative sign.

Table 4: Volume of performed radioactivity analyzes in the period 2019-2023.

Year	X	Y	X x Y	X x X
2019	-2	2091	-4182	4
2020	-1	1382	-1382	1
2021	0	967	0	0

2022	1	822	822	1
2023	2	803	1606	4
Σ	/	6065	-3136	10

By replacing the corresponding values, the values of the trend coefficients, as well as the trend itself, are determined:

$$a = \frac{\sum y}{N} = \frac{6065}{5} = 1213$$

$$b = \frac{\sum xy}{\sum x^2} = \frac{-3136}{10} = -313{,}6$$

$$y = a + bx = 1213 - 313{,}6x$$

In further elaboration, the trend values for each year are determined (see table 5):

Table 5: Determination of trend values

Year	X	y = 1213 - 313,6x
2019	-2	1840.2
2020	-1	1526.6
2021	0	1213
2022	1	899.4
2023	2	585.8
2024	3	272.2
2025	4	-41.4
2026	5	-355

The obtained individual trend values in the examined period are applied to a coordinate system, and the graphic display shows that the volume of radioactivity analyzes performed in the Republic of North Macedonia shows a decreasing tendency (see graph 3).
For the years 2024, 2025 and 2026, a forecast was made for the trend values (in the trend formula only the values for time X are changed), and it can be seen that even in the years of the forecast, the volume of radioactivity analyzes performed shows a decreasing tendency. [4]

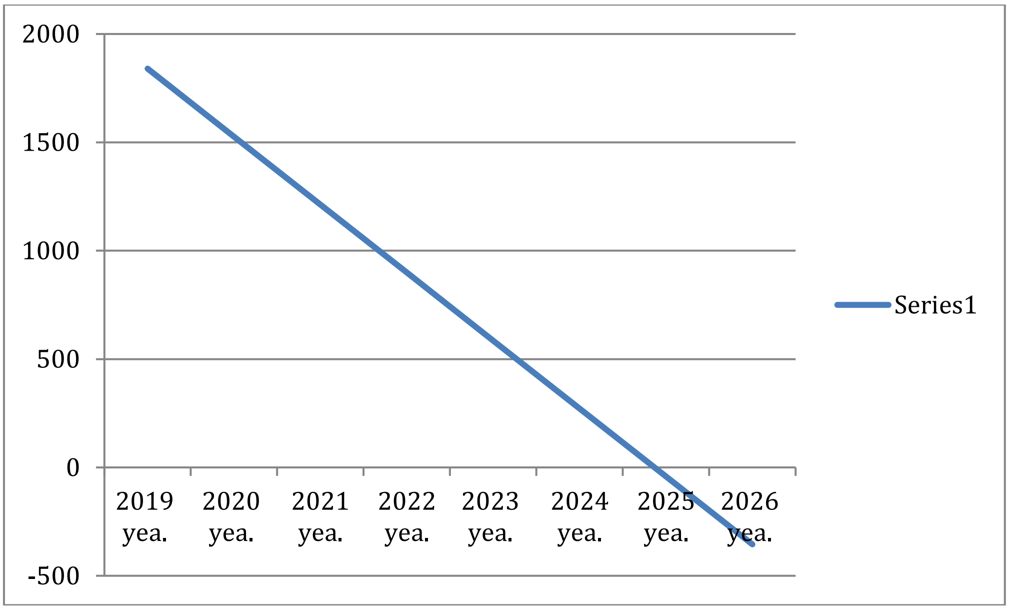

Chart 3: Volume of performed radioactivity analyzes in the period 2019-2023. – trend

Table 6: Distribution of analyzed products in which radioactivity above the limit values was detected in the period 2019-2023.

Year	**Number of defective samples**
2019	0
2020	1
2021	0
2022	0
2023	0

From table 6, it can be concluded that radioactivity above the limit values was detected in only 1 product in 2020. According to the information we received from the Institute of Public Health, it is dry snot with a detected value for Cs-137 = 662.8 Bq/kg, with a maximum allowed value of 600. For all other analyzed products in the period 2019-2023. no radioactivity above the limit values was detected.

4. Discussion

The average radiation exposure in FR Germany in 1975 was estimated to be 172 mrad, of which 21 mrad was attributable to internal radiation from natural radionuclides embedded in the body (about 90% from 40K, the rest from 14C) and less than 1 mrad from nuclides from atmospheric fallout from nuclear explosion tests (50% of 137Cs, a radionuclide with a half-life of 30 years but rapidly excreted from the body; approximately 50% of 90Sr, the most dangerous radioisotope, capable of inducing leukemia and bone cancer; and traces of 14C and tritium).

137Cs and 90Sr are companion elements of potassium and calcium. Radionuclide contamination of food in FR Germany peaked in 1964/65, when the daily food intake per person was 240 pCi of 137Cs and 30 pCi of 90Sr. Until the Chernobyl reactor accident in April 1986, the intake was less than 10% of previous values as a result of the moratorium on atmospheric testing of atomic weapons. Radionuclide residues in food were not hazardous to health.
In 1986, the Chernobyl accident caused an additional dietary intake of radionuclides estimated at (children under 1 year of age / adults) 1779/4598 Bq/year of 131I, 986/1758 Bq/year of 134Cs, and 1849/3399 Bq / year of 137Cs. The resulting additional effective equivalent dose for people in FR Germany is estimated at 0.06-0.22 mSv. For comparison, natural radiation exposure is about 2 mSy per year, of which 0.38 mSv / year is caused by radionuclides in food. As a precautionary measure, maximum values of 500 Bq / l and 250 Bq / kg are provided for milk and vegetables. For comparison, the activity of natural radionuclides (mainly 40K) in food is: milk 40-50 Bq / kg, milk powder 400-500 Bq / kg, fruit juice concentrates 600-800 Bq / kg and soluble coffee (powder) > 1000 Bq / kg.
The level of tritium in the biosphere is expected to increase further due to increased activity of nuclear power plants worldwide. [5]

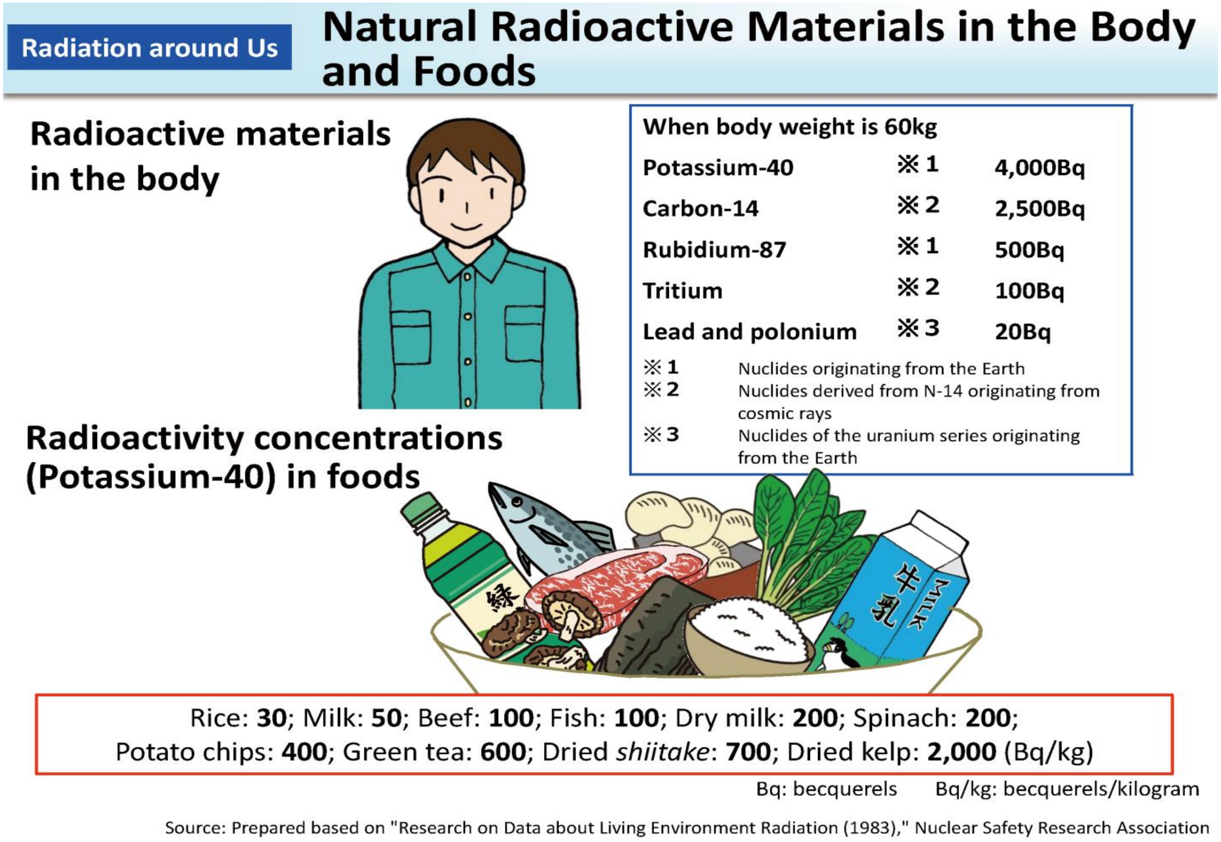

Figure 1: Natural radioactive materials present in the human body and food

Pursuant to the Rulebook on maximum permitted amounts of radionuclides in food, water, air, land, products and raw materials of animal and plant origin and objects for general use (Official Journal of the Republic of Macedonia No. 163/2009) which is valid in our country, the import of products as well as forest fruits originating from countries affected by the Chernobyl accident shall not be carried out if the total specific activity of Cs-134 and Cs-137 in them exceeds:

- 370 Bq/kg in milk and milk products, as well as for products intended for infant nutrition in the first four to six months, if this is explicitly declared on the product packaging;
- 600 Bq/kg for other products and forest fruits. Also, in the Rulebook on maximum permitted amounts of radionuclides in food, water, air, land, products and raw materials

of animal and plant origin and objects for general use, the maximum permitted amounts of radionuclides in food and animal feed intended for the market after nuclear accident or radiation emergency (see table 7). [6]

Table 7: Maximum allowed amounts of radionuclides in food (Bq/kg)

A radionuclide	Baby food	Milk and milk products	Food excluding non-essential food	Non-essential food	Liquid food and drinking water
Strontium - 90	75	125	750	7500	125
Iodine -131	150	500	2000	20000	500
Alpha emitters, plutonium isotopes especially Pu-239 and Am-241	1	20	80	800	20
Other radionuclides with a half-life of 10 days, especially Cs-134 and Cs-137 (without K-40, H-3, C-14)	400	1000	1250	12500	1000

In Japan following the TEPCO Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, the current limits for radioactive contamination of food are: 100 Bq/kg for general food, 50 Bq/kg for milk and infant food, and 10 Bq/kg for water for drinking. These limits were established based on an effective dose of 1 mSv per year, according to international standards to mitigate the exposure of the general public to radiation. Measures have also been taken, including the withdrawal or restriction of food in cases where these restrictions have been breached. As a result of these efforts, the actual effective doses of radioactive cesium (134Cs + 137Cs) in food approximately one year after the FDNPP accident were below 0.01 mSv per year. [7]

In the period from January 2016 to February 2017, 900 food samples were collected in South Korea and the radioactive contamination of food with 134Cs, 137Cs, 239Pu, 240Pu and 90Sr was investigated. The results of the study show that the activity concentrations of 137Cs in fish range from minimum detectable activity (MDA) to 340 mBq/kg fresh weight. The concentration factor (CF) determined for 137Cs as a measure of its bioavailability is calculated to be about 74 and is found to be very similar to that (100) recommended by the International Atomic Energy Agency. With an MDA of <0.221 mBq/kg, the results reveal that the values of 239Pu and 240Pu in fish are below the MDA. Activity concentrations of 137Cs and 90Sr are lower than MDA in both shellfish and seaweed, while activity concentrations of 239Pu and 240Pu in shellfish range from 2.07 to 2.18 mBq/kg, and in seaweed samples range from 2.07 to 3.07 m Bq/kg. The ratio of 240Pu/239Pu atoms in shellfish caught off the Korean coast varied from 0.209 to 0.237, with a mean value of 0.227. The higher ratio of 240Pu/239Pu atoms found in the shells is thought to be caused by plutonium transported from the Pacific fields, rather than from other sources, such as the Fukushima nuclear plant accident. Activity concentrations of 137Cs in mushrooms have been shown to vary from 1 to 21.4 Bq/kg. 134Cs were detected in three mushroom samples collected from Jeju Island and about 3-3.6% of 137Cs present in wild mushrooms collected from Jeju Island originated as a result of the Fukushima nuclear power plant accident. The conclusion of the study states that the annual effective doses of 137Cs received through the consumption of mushrooms and fish are 2.0 x 10-4 mSv per year and 3.9 x 10-5 mSv per year and those values are negligible compared to the annual limit of effective dose of 1 mSv per year. [8]

On the other side of the Pacific, in Buena, a thorium-rich region of Brazil, research indicates that the concentration values of radionuclides in food and drinking water are 100 times higher than international reference values. Studies show that concentration values for 210Pb and radium isotopes in Buena drinking water are among the highest values reported in the literature. 228Ra is the most important radionuclide ingested through food and water in Buena residents. [9]

It is also significant to mention an older scientific study from the USA conducted in the period 1987-1992. According to the results of this study, a continuation of the general decrease in the intake of 90Sr, which was ascertained already in 1961, was detected. Also, within this study approximately 2600 samples of imported food were analyzed for contamination related to the Chernobyl nuclear accident. Concentrations of radionuclide activity were below detection limits for the vast majority of imported foods tested, but in 23 samples they were above levels of concern. According to the results of this study, since 1986, the proportion of imported food samples tested with measurable amounts of contamination has declined, as have the average concentrations of radionuclide activity, but contamination is still occasionally found. The conclusion states that continuous monitoring of domestic and imported food is needed. [10]

From the overall review of the scientific literature that has been made in this section, it can be seen that studies on the presence of radionuclides in food are of great interest worldwide because exposure to radionuclides, especially in food, can endanger the health of consumers.

5. Conclusion

In all the analyzed products in the period 2019-2023, no radioactivity above the limit values was detected (except in 2020 when only 1 product from the group of mushrooms - dry morel) a value for Cs-137 = 662.8 Bq/kg was detected, and the maximum the permitted value is 600 Bq/kg). However, it is worrying that the volume of radioactivity analyzes performed in the Republic of North Macedonia shows a decreasing tendency. Also, the largest volume of radioactivity analyzes were performed in the group of mushrooms (51%) and the group of grain and flour (44%) in the period 2019-2023. We believe that in the future we should work on increasing the volume of analyzes performed for the presence of radionuclides in food, as well as increasing the coverage of other groups of products (fruit, fruit products, vegetables, coffee, cocoa, tea, soft drinks and Fig.) in which such analyzes would be performed.

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