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To cite this article: Velibor Andric and Maja Gajic-Kvascev 2021 IOP Conf. Ser.: Earth Environ. Sci. 854 012003

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The radioactivity parameters in the food chain – legislation, control and critical points

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Abstract. Radioactivity, whether natural or artificial, today constitutes a significant segment in the process of protecting human and animal health. Natural radioactivity is an integral part of our ecological system, so it has long been present in the food we eat. However, intensive industrial processes in some areas have disturbed the natural ecological balance and thus introduced into the environment natural radionuclides in quantities that can affect the quality of human life and the state of the environment. On the other hand, artificial radionuclides reach the environment only in the case of accident situations in nuclear facilities. In the nuclear era so far, two accidents, Chernobyl in 1986 and Fukushima in 2011, affected the environment globally with the significant impact. Consequently, a system of monitoring radioactivity in the environment was introduced, which includes foodstuffs as well as animal feed. The paper describes the sources of radioactive contamination and the most critical raw materials and products in the human and animal food chain, as well as a critical review of current legislation.

1. Introduction
Natural radioactivity is an integral part of our ecological system, so it has long been present in the food we eat. The human body is thus in a balance between natural and cosmic radiation because there are natural processes of repairing cells damaged by that radioactive radiation. By consuming food during their life, a person also increases the total radioactivity of their body, because part of the ingested radionuclides is retained in the body and is included in metabolic processes. This is the reason we need to monitor the annual consumption and, consequently, intake of natural radionuclides in the population, to keep it at the safe level.

However, intensive industrial processes in some areas have disturbed the natural ecological balance and thus introduced into the environment natural radionuclides in quantities that can affect the quality of human life and the state of the environment. Such materials are called NORM (naturally occurring radioactive material), while a special category consists of TENORM products (technologically-enhanced NORM), in which the original content of natural radionuclides is increased by the technological process itself, making them even more dangerous to the environment [1].

The rapid alert system for food and feed (RASFF) monitors the results of control of foodstuffs and products, as well as animal feed, from the point of view of all quality parameters, including radionuclides.
2. The source of radioactivity

The concentration of natural radionuclides depends on the location and is highest in places where mineral raw materials are exploited, which includes radionuclides from the uranium and thorium series, together with all their radioactive descendants, the most important among them being radon gas (\(^{222}\text{Rn}\)). This radionuclide is not characteristic of industrial processes, so we will not consider it further.

Besides the ore and coal mining industries, the technology of production of mineral phosphate and potassium fertilizers for plant nutrition and mineral additives for animal feed also significantly contribute to the introduction of natural radionuclides into the environment. The reason is that most phosphate ores are characterized by the presence of radioactive elements from the uranium and thorium series, especially \(^{226}\text{Ra}\) and \(^{238}\text{U}\), so the total specific activity of these ores can reach several thousand Bq/kg. Therefore, crude phosphate as a semi-processed product and mineral fertilizers with potassium and phosphorus also contain significant amounts of natural radionuclides. These radionuclides are regularly introduced into the ecosystem through plant nutrition processes, but also through technology of wastewater and waste treatment, because unwanted contamination and exposures can originate from radionuclides that enter the environment due to inadequate waste treatment, especially via phosphogypsum, which is a by-product of phosphate production. Mineral additives for animal feed, based on phosphates, can significantly increase the amount of natural radionuclides in diet and consequently in the meat and other products.

In that way, a part of these radionuclides through the animal nutrition chain, but also deposition, can end up in foodstuffs. It is estimated that the total contribution of this industry, together with the oil and gas exploration industry, is almost 95% of the total input of natural radionuclides into the environment.

In addition to the mentioned natural radionuclides, one of the most important sources of natural radiation is potassium, i.e., its radioactive isotope \(^{40}\text{K}\). Unlike natural radionuclides from the uranium and thorium series, which, due to their chemical structure (heavy metals), are not included in metabolic processes, \(^{40}\text{K}\) acts like natural potassium, fully participates in human and animal metabolism and finally is deposited in bones. It is found in soils in quantities that correspond to specific activities in the range of 600-900 Bq/kg, and consequently, it is also found in various products for human and animal nutrition. During life, a person constantly ingests new amounts of \(^{40}\text{K}\) through food, with a contribution of approximately 65 Bq/kg body weight, so this radionuclide together with \(^{14}\text{C}\) makes up almost the total radioactivity of the human body, which for a person of 80 kg is about 5200 Bq \(^{40}\text{K}\) and 3400 Bq \(^{14}\text{C}\).

On the other hand, artificial radionuclides reach the environment only in the case of accident situations in nuclear facilities. The highest global radioactive contamination that has affected the environment was registered after the 1986 Chernobyl and 2011 Fukushima disasters. The accident at the Chernobyl nuclear power plant reactor was recorded as the accident that led to the greatest radioactive pollution of the environment, especially in Eastern and Northern Europe. The consequences of that accident are still measurable, and some of the artificial radionuclides that were emitted at that time are still detected in samples from the environment, including foodstuffs. Of course, due to the natural process of radioactive element decay, their concentration is now much lower than in the years immediately after the accident. Due to its longer half-life of about 30 years, \(^{137}\text{Cs}\) is the primary radionuclide that can still be detected in the environment. Significant amounts of radioactive \(^{137}\text{Cs}\) originating from this accident are today mostly detected in forest fruits, mushrooms and wild game meat since this forest ecosystem is mostly not mechanically processed and treated, which are procedures that reduce the specific activity of radionuclides in the surface layer of the soil. Given the much smaller share of these foods compared to other agro-industrial products, the contribution of this radioactivity to the total dose received by the population is relatively small.

Forest wild mushrooms can have a \(^{137}\text{Cs}\) specific radioactivity of several hundred Bq/kg, and exceptionally, up to 1000 Bq/kg depending on mushroom species, the altitude at which they were collected and the characteristics of the environment itself. In addition to mushrooms, berries can also have increased radioactivity due to the presence of \(^{137}\text{Cs}\).

Wild game, especially boar and deer, also can produce meats that contain increased amounts of radioactive Cs as a result of their diet and the accumulation of radionuclides in the animal’s body.
Therefore, some European countries have defined a limit of 3000 Bq/kg for $^{137}$Cs in wild meat from certain areas (Norway) [2]. Based on the data published in Germany for 2017-2019, it was concluded that the maximum values of specific activity of radioactive $^{137}$Cs in wild boar meat were up to 1600 Bq/kg, which excluded further sale of that meat and its restriction to personal use only [3].

3. The current legislation

European legislation defines the maximum values of permitted contamination by appropriate regulations, especially foodstuffs and feed originating from third countries, are particularly important items to be controlled for radionuclide content and their specific activity (Commission Implementing Regulation (EU) 2020/1158 of 5 August 2020 on the conditions governing imports of food and feed originating in third countries following the accident at the Chernobyl nuclear power station).

In relation to the previous edition of this rulebook from 2008, the radionuclide $^{134}$Cs was excluded from the control system, considering that 10 periods of its half-life (approximately 2 years) had already passed since the accident, and it no longer needed to be controlled.

The prescribed limits of permitted maximum specific activity of $^{137}$Cs are 370 Bq/kg in milk, dairy products and food for infants and young children and 600 Bq/kg for all other products. The stated maximum values do not apply to private consumption, only to products for further sale. It is estimated that the contribution of food and water radioactivity to the total dose received by the population is in the order of 10%, but would be greater in any population that has specific eating habits. In numbers, it would look like this: ingestion of food containing 80 kBq of $^{137}$Cs corresponds to exposure with a dose of 1 mSv, which is approximately one third of the total dose received by the population during one year from all sources. Most countries in the region that are not EU members have accepted the EU legislation in the accession process, so the same maximum allowed values of $^{137}$Cs contamination are used on their markets.

The Russian authorities have defined the issue of radioactivity in food much more precisely, as stated in Annex No. 4 to the Technical regulation of the Customs Union On Food Safety (TR TS 021/2011), where the maximum values for radionuclides $^{137}$Cs and $^{90}$Sr are defined (Table 1).

Table 1. Maximum allowable levels of radionuclides cesium-137 and strontium-90

| No. | Groups of Food Products                                                                 | Specific Activity of $^{137}$Cs, Bq/kg(l) | Specific Activity of $^{90}$Sr, Bq/kg(l) |
|-----|---------------------------------------------------------------------------------------|------------------------------------------|-----------------------------------------|
| 1   | Meat, meat products and by-products                                                     | 200                                      | -                                       |
| 2   | Venison, game                                                                         | 300                                      | -                                       |
| 3   | Fish and fish products                                                                 | 130                                      | 100                                     |
| 4   | Dried fish and stockfish                                                               | 260                                      | -                                       |
| 5   | Milk and products of milk processing (except for condensed, concentrated, canned, dry, | 100                                      | 25                                      |
|     | cheese products, butter, and butter paste from cow milk; cream-and-vegetable spread   |                                          |                                          |
|     | and-vegetable melted mixture, concentrates of dairy proteins, lactulose, lactose,     |                                          |                                          |
|     | casein, caseinates, hydrolysates of dairy proteins                                    |                                          |                                          |
| 6   | Concentrates of dairy proteins, lactulose, lactose, casein, caseinates, hydrolysates   | 300                                      | 80                                      |
|     | of dairy proteins                                                                     |                                          |                                          |
| 7   | Products of milk processing, dry, freeze-dried                                        | 500                                      | 200                                     |
| 8   | Cheese and cheese products                                                             | 50                                       | 100                                     |
| 9   | Products of milk processing, concentrated, condensed; dairy, composite dairy, milk-   | 300                                      | 100                                     |
|     | containing canned food products                                                        |                                          |                                          |
10 Butter, butter paste from cow milk, milk fat 200 (for milk fat 100) 60 (for milk fat 80)
11 Cream-and-vegetable spread, cream-and-vegetable melted mixture 100 80
12 Dry nutritional media on a milk basis 160 80
13 Vegetables, root crops including potatoes 80 (600(2)) 40 (200(2))
14 Bread and bake goods 40 20
15 Flour, grits, meals, cereals, alimentary products, 60 -
16 Wild berries and preserved wild berry products 160(800(2)) -
17 Fresh mushrooms 500 -
18 Dried mushrooms 2500 -
19 Specialized baby foods ready to serve(1) 40 25
20 Vegetable oils 40 80
21 Vegetable oils (fats) interesterified refined deodorized; oils (fats) hydrogenated refined deodorized; margarines; special purpose fats, including cooking, confectionary, and baking fats; milk fat replacers; cocoa butter equivalents, cocoa butter improvers of SOS-type, cocoa butter substitutes of POP-type, non-tempering cocoa butter substitutes, vegetable-and-fat spread, vegetable-and-fat melted mixtures, sauces on the basis of vegetable oils, mayonnaises,
22 Vegetable-and-cream spreads, vegetable-and-cream melted mixtures 100 80

(1) – in the case of freeze-dried products, the specific activity is determined for a reconstituted product.
(2) – permissible level for a dry product.

Legislation in the Republic of Serbia regarding radioactivity is within the competence of the Directorate for Radiation and Nuclear Safety and Security of Serbia and is formulated in two documents: Rulebook on control of radioactivity of goods during import, export and transit, 86/19 and 90/19 and the Ordinance on the limits of radionuclide content in drinking water, foodstuffs, animal feed, medicines, items of general use, construction materials and other goods placed on the market (Official Gazette 36/18).

Goods for which gamma spectrometric analysis is mandatory during import, export and transit are listed in Annex 1 of the Ordinance on the control of radioactivity of goods during import, export and transit (Official Gazette 44/11, 86/19 and 90/19).

Goods under the jurisdiction of phytosanitary inspection and for which gamma spectrometric examination is performed are:
1. Herbs
2. Mushrooms, in any form and products of which they are a part
3. Blueberries, in any form and products of which they are a part
4. Cranberry, in any form and products of which they are a part
5. Forest fruits, in any form and products of which they are a part
6. Mineral phosphate fertilizers (finished product)

Goods under the responsibility of veterinary inspection and for which gamma spectrometric analysis is performed in accordance with the risk analysis are:
1. Meat and products
2. Milk and milk products
3. Edible products of animal origin
4. Fish caught in the sea and their products
5. Other goods under the jurisdiction of the veterinary inspection

The limits of maximum permitted values are prescribed by the Ordinance on the limits of radionuclide content in drinking water, foodstuffs, animal feed, medicines, general use items, construction materials and other goods placed on the market and are based on the values of derived radionuclide concentration in water and food which induces an annual dose of 0.1 mSv. These sizes depend on the annual quantities of these products that are consumed, so based on WHO recommendations, an average value of 730 l of annual drinking water consumption per capita was taken.

\[
IK_v = \frac{GD}{e_g V_v}
\]

GD - annual dose, maximum value 0.1 mSv.
Ikv - derived concentration of radionuclides in water.
e_g - received effective dose at unit intake.
V_v - annual amount of consumption (for water 730l).

Table 2. Derived specific activities of some radionuclides in drinking water

| Radionuclide | IK_v, Bq/l |
|--------------|------------|
| \(^{238}\)U | 3          |
| \(^{226}\)Ra | 0.5        |
| \(^{210}\)Pb | 0.2        |
| \(^{14}\)C | 240        |
| \(^{90}\)Sr | 4.9        |
| \(^{60}\)Co | 40         |
| \(^{134}\)Cs | 7.2        |
| \(^{137}\)Cs | 11         |

The same Rulebook defines the specific activity of \(^{137}\)Cs in individual food categories, which are shown in Table 3.

Table 3. Derived \(^{137}\)Cs radionuclide specific activities in particular product categories

| Product categories                                                                 | IK_v, Bq/kg, Bq/l |
|------------------------------------------------------------------------------------|------------------|
| Milk and milk products, infant formulas, vegetables, fruits, cereals, meat and meat products, eggs, other foods such as lard, oil, sugar, sweets, alcoholic and non-alcoholic beverages | 15               |
| Powdered milk, wild berries (blueberries, cranberries, blackberries, strawberries, raspberries, currants, gooseberries), game, fish, seafood, mushrooms (fresh and mushroom products), herbs, teas and coffee | 150              |
| Dried mushrooms, flavourings, spices and other foods used less than 2 kg per year  | 600              |

These two tables shows that the defined value of IK_v for \(^{137}\)Cs is determined arbitrarily, because it would imply that all these categories of food are consumed in amounts per year of 535 kg or l for the first category and 53.5 kg or l for the second category, which is certainly overestimated value.

4. The critical points
Previously, the system of radioactivity control of goods in Serbia included a large number of products, and control, and sampling was performed at the border, so it had a preventive effect in some way. The
import control system with the new editions of the Rulebook excludes certain categories of foodstuffs that have been regularly controlled so far, so that imported cereals are no longer controlled, even though a large part of these products comes from Eastern Europe. Also, the established control system is more focused on imported products, while domestic products are generally more often controlled during exports or monitoring.

As critical points in the current national legislation, which defines radioactivity as a parameter of food and feed safety as well as of other goods placed on the market, we can list the following:

- inconsistency of ordinances describing the control of goods at import and placing of goods on the market.
- certain product categories are excluded from the control process, although they can significantly increase the radiation load of the population (cereals and plant products).
- the categorisation of animal feed is not defined, but a historical approach is used and boundaries for humans are applied, although the diet and quantities are not the same.
- non-compliance of product and raw material categories, e.g., for forest fruits and products (blueberries).
- lack of information for end users about the content of radionuclides in certain types of products and procedures for their safe use (mineral fertilizers, some mineral premixes for animal nutrition).

Compared with the allowed values of specific activity for $^{137}\text{Cs}$ in national and European regulations and in regulations valid in Russia and some former Soviet republics, we notice deviations and differences that can cause misinterpretations and problems in the trade of such goods in import and export. A significant difference is the introduction of control of the content of the radionuclide $^{90}\text{Sr}$, which is analytically much more complicated than analysis of $^{137}\text{Cs}$.

5. Conclusion

In conclusion, we point out the fact that Serbia’s national legislation is stricter than the European one in terms of radioactive contamination, but that it will be harmonized with it in time. The current situation can lead to certain problems when importing goods that are safe on other markets but cannot be marketed in Serbia because they do not meet the national legislation. Even in the EU system with higher permitted contamination limits than are allowed in Serbia, during 2019, the RASFF recorded 13 cases of food and animal feed with high radionuclide content [4]. Mechanisms for controlling radioactivity in food and animal feed need to be developed and improved, since there are several older generation nuclear power plants in the vicinity of Serbia that are still operational. The security systems of nuclear facilities are certainly one of the most advanced creations of the human mind, but cases like Chernobyl or Fukushima must suggest the need to constantly improve the process of protecting the population from the harmful effects of radioactivity.

References

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