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Artificial environmental radionuclides in Europe and methods of lowering their foodstuff contamination – a review

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Abstract

This review discusses the consequences of the food chain contamination with radionuclides, especially focusing on the radiocaesium impact after the Chernobyl nuclear accident. In particular, the ^{137}Cs isotope still represents a risk. Until present it is still detectable in the meat of game animals, especially in wild boar, but also in elk and reindeer. Although the occurrence of highly contaminated foods in most of Europe is currently limited, along the German-Czech border (the Šumava Region) the activity concentration of the ^{137}Cs isotope in the meat of wild boar exceeds the acceptable limit several times. Additionally, the article describes simple processing technologies (cooking, pickling etc.) that lead to reduction of radionuclides in contaminated food.

Chernobyl, ^{137}Cs, food safety, radiocaesium reduction, wild boar

Within the terrestrial environment of Europe, contamination by anthropogenic radionuclides comes from two different sources. The first source was the global atmospheric fallout, which appeared after the start of intensive atmospheric nuclear weapon testing in the 1950s, and has been observed for a long time after their completion in 1963.

The second source of terrestrial contamination was the fallout after the nuclear reactor accident at Chernobyl in 1986. In terms of time, it was a one-time contamination, where the intensity of radioactive fallout was determined by the local meteorological situation and movement of contaminated air masses. The Fukushima nuclear accident which contaminated mainly Japan and a significant part of Asia, only had a limited impact on Europe.

Radionuclide contamination

Relevant radionuclides

From the radio-ecological perspective, only the long-lived components of nuclear weapons testing are relevant (^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239}Pu). The most important long-lived contaminant was the caesium ^{137}Cs isotope (Högberg 2013). The level of contamination in a certain area was dependent on the latitude and long-term weather situation, particularly precipitation (Csupka et al. 1978). Currently, the ^{137}Cs from this source, is already largely immobilized in the clay fraction of the soil, with limited access for plant roots (Nimis 1996).

The Chernobyl nuclear accident on April 26, 1986, caused a release of radioactive caesium at the amount up to 3.8×10^{16} Bq. The ratio of ^{137}Cs to ^{134}Cs long-lived radionuclides released was approximately 2 : 1 (UNSCEAR 1988). The radioactive cloud passed over

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the continent three times. On some territories with intensive rainfalls, increased soil contamination by large amounts of radionuclides was observed. Besides the Ukraine, Belarus and Russia, the most affected parts of Europe also included Norway, where the surface activity values reached up to 500 kBq·m^{-2} (Pedersen et al. 1998).

Soil contamination

Due to the elapsed time and the nature of the Chernobyl accident, currently the only artificial radionuclide present in the soil is ^{137}Cs, with a physical half-life of 30.17 years (Krolak et al. 2010). The ^{137}Cs quickly integrated into the biological cycle, similarly to potassium. Generally, radionuclides are characterized by their mobility in soil (Gadd 1996). Analysis has shown that the diffusion coefficient of radionuclides in the soil is affected by the soil moisture, presence of chemical homologs determining the capacity of the exchange process in the soil, soil acidity, soil humus content, and temperature (Koprda 1986).

High mobility of radiocaesium gradually decreased after its deposition in the soil. Differences in mobility were observed especially in the non-cultivated meadow and forest soils, and were associated with the soil depth. In mineral soils with increasing depth, the amount of ^{137}Cs accessible by plants may rise, however it depends on many factors and circumstances (Schimmack and Bunzl 1996). Tomioka et al. (1992) reported several conditions which play an important role in the maintenance of radiocaesium stability in the surface layers of forest soils especially soil bacteria.

Soil contamination by different radionuclides significantly affects the level of terrestrial radiation in a given area. Together with cosmic rays they represent the main sources of external exposure. Gamma spectrometric analysis of soil samples is an integral part of continuous long-term monitoring and evaluation of radiation.

As one of few components of the environment, ^{137}Cs is well detectable in soils until present (Palagyi et al. 2013). Modified sequential extraction analysis published by Tessier (Tessier et al. 1979; Pipiška et al. 2004; Galamboš et al. 2012) was used to study the distribution, determination of bioavailability and uptake of radionuclides by plants.

Radiocaesium is most strongly retained in clay minerals. For this radionuclide, its residual fraction is the most important (Bowen 1979; Ciuffo et al. 2003). The water-soluble and removable fractions of radiocaesium defining the intensity of vertical migration in the soil samples represent only 5%. In the annual sediments of the Morava River and the Brno Dam, ^{137}Cs was found to a depth of 42 mm of the upper sediment, and to a depth of 2.9 mm in the lower sediment (Sedláček et al. 2013).

Plant contamination

Some plants have a high ability of ^{137}Cs uptake from the environment and thus are considered to be bioindicators (Vaaramaa et al. 2009; Rulík et al. 2014). Mosses are able to capture up to 93% of airborne radionuclides (Heinrich et al. 1989). Bilberry (*Vaccinium myrtillus*) belongs among boreal forest plants that are spread mainly by the vegetative way of reproduction, creating dense intertwined root systems, where one plant often occupies an area of few square metres (Nimis 1996). Even 20 years after the Chernobyl accident, ^{137}Cs may under certain conditions and in some areas increasingly penetrate bilberry fruits (Nimis 1996; Commission Recommendation 2003) which are very popular and heavily harvested forest fruits in many European countries.

In 1999–2002 the highest radioactivity contamination of ^{137}Cs measured in bilberries from the French Alps was 5–140 Bq·kg^{-1} fresh weight (Pourcelot et al. 2003).

In the context of reports from other countries on cases of exceeding ^{137}Cs concentrations in bilberries and bilberry products in the Rapid Alert System and the European Commission recommendations (Commission Recommendation 2003) at the end of the summer of

2005, The State Veterinary and Food Administration of the Slovak Republic carried out a targeted control of all commodities containing berries commercially distributed and sold in Slovakia. From among all of the controlled frozen bilberry samples, the highest activity (535.7 $Bq \cdot kg^{-1}$) was found in fruits originating from Romania. Sample contamination was close to the hygienic limit of 600 $Bq \cdot kg^{-1}$ valid for the total caesium content (^{134}Cs + ^{137}Cs) in foodstuffs within EU countries (Council Regulation 2000). The lowest activity was found in bilberries from Belgium (7.58 $Bq \cdot kg^{-1}$). From samples of bilberry compotes, the highest activity (33.3 $Bq \cdot kg^{-1}$) was measured in a sample coming from the Czech Republic (Miššík et al. 2006).

Mushroom contamination

Fungi, as one of the most important constituents of forest ecosystem are capable of accumulating a significant amount of radionuclides including ^{137}Cs (Heinrich 1991; Škrkal et al. 2013; Guillen and Baeza 2014). It is due to their heterotrophic metabolism, significantly different from green plants, and dependence on the supply of final organic compounds (Yoshida and Muramatsu 1994).

Some species of fungi, including the edible ceps (*Boletaceae* family) that grow in the deciduous forests of Central Europe, are not only bioindicators of environmental contamination by radiocaesium (and heavy metals), but their consumption represents a potential hygienic and health risks (Linkov et al. 2000; Kaláč 2001; Bystrzejewska-Piotrowska et al. 2003; Dvořák et al. 2006).

Radiocaesium distribution in different mushroom body parts was uneven but higher activity was determined in the mushroom caps (Heinrich 1993; Mukhopadhyay et al. 2007). The ability to accumulate radionuclides from the environment differs among different species of ceps (Kaláč 2001). This corresponds with the finding that increased accumulation of ^{137}Cs in the bay bolete (*Xerocomus badius,* syn. *Boletus badius*) is mainly due to the presence of the pigment norbadion A, present in the mushroom cap (Aumann et al. 1989). It is assumed that, as in higher plants, radiocaesium activity in fungi is associated with the growth phase, and that the total radiocaesium activity decreases with gradual growth. In terms of health, the most risky are therefore the smallest fruiting bodies, which are most commonly used for canning.

Mushroom samples collected in coniferous forests are characterized by a high content of radionuclides compared to those collected in deciduous forests (Čipáková 2004).

Findings on the radiocaesium mass activity from different areas of the Czech and Slovak Republics in 2000–2004 were published by Dvořák et al. (2006a). The highest ^{137}Cs activity of 6,263 $Bq \cdot kg^{-1}$ dry weight (measured by gamma spectrometry method) was found in *Xerocomus badius* from the Old Ransko area (Czech-Moravian Highlands). The highest ^{137}Cs concentration of 966 $Bq \cdot kg^{-1}$ dry weight in Slovakia was measured in *Suillus luteus*, in the area of Senica. For comparison, ^{137}Cs activity in the sample of mixed dried ceps (*Boletaceae* family) coming from the 30 km Chernobyl border zone, was 6,000 $Bq \cdot kg^{-1}$ dry weight. Results for dried mushrooms show that there is currently no ^{137}Cs activity dependency related to the distance from the place of the radioactive accident or the location altitude. These results also indicate significantly lower values of ^{137}Cs activity in the Slovak Republic compared to the Czech Republic, despite the fact that Slovakia is closer to Ukraine. Explanations should be sought in the airborne radioactive cloud movement through various parts of Europe after the Chernobyl disaster.

In the Czech Republic mainly the Czech-Moravian Highlands region was monitored, as it was impacted by radioactive clouds passing over it for three times. Slovakia was affected considerably less.

In fresh mushrooms collected in the French Alps in 1999–2002, ^{137}Cs activity ranged from 273–1,165 $Bq \cdot kg^{-1}$ (Pourcelot et al. 2003).

Animal contamination

Due to the fact that Scandinavia was heavily contaminated by the Chernobyl radiocaesium fallout, considerable attention within the food chain was paid to elk (Palo et al. 2003) and reindeer meat (Skuterud et al. 2004). Similar attention was paid also to marine organisms (seafood), such as crab from the north of Ireland (Copplestone et al. 2004). In Central Europe, the animal product most contaminated from the Chernobyl accident was game meat (Sprem et al. 2013). Relatively higher activity of ^{137}Cs in wild animals is based on the mosaic pattern of the contamination area after the Chernobyl accident, in the way the wild animals searches and acquires food (especially wild boar), and in a significantly greater mobility and persistence of radiocaesium in the forest ecosystems, compared to intensively used agricultural land (Vaaramaa et al. 2009).

The Recommendation of the European Commission (2003) highlights the limit for radiocaesium activity in game meat, and calls on the Member States to act upon it in order to protect the consumers. Member states should implement steps to ensure that the limits set by the Directive no. 737/90 / EEC (2000) for marketing game meat, wild berries, mushrooms, and predatory lake fish, are respected. At the same time it recommends warning the inhabitants of affected regions of the health risks resulting from contaminated food consumption. Member States are asked to prepare a feedback on the implementation of this Directive for the European Commission and other EU Member States.

After a gradual decline of the ^{137}Cs activity in game muscles in the 1990s, unexpected increase of activity occurred after the floods in North-Eastern Moravia in 1997. In the meat of wild boar, the activity exceeded the hygienic limit set for the EU countries (600 $Bq \cdot kg^{-1}$), especially in the age category up to 1 year. Since 2000, the ^{137}Cs activity has been reduced back to the level before the floods (Obzina 2002). Seasonal fluctuation of ^{137}Cs activity in the wild boar meat samples was observed in the forests on Southern Rhineland. In the examined muscle tissue the median activity was 129 $Bq \cdot kg^{-1}$, and the maximum reached was 5,573 $Bq \cdot kg^{-1}$ (Hohmann and Huckschlag 2005). From 1998 to 2008, 656 samples from the wild boar were analysed in the district of Ravensburg (Southern Germany). The activity was variable from less than 5 up to 8,266 $Bq \cdot kg^{-1}$, depending on the season, weather conditions and the associated changes in dietary habits and food availability (Semizhon et al. 2009). High radioactivity concentrations (up to 10,699 $Bq \cdot kg^{-1}$) were reported in the wild boar muscles from the Šumava Region of the Czech Republic (Latini 2011). In 2012, the highest value measured was 14,252 $Bq \cdot kg^{-1}$ (Kouba et al. 2013). Wild boar muscle contamination is mainly due to the consumption of the underground fruiting bodies of the mushroom genus of *Elaphomyces* sp. (*E. granulatus* - deer balls, hart's truffles) (Hohmann and Huckschlag 2005; Dvořák et al. 2010). The highest ^{137}Cs specific activity of 4,743 $Bq \cdot kg^{-1}$ was detected in the mushroom fruiting bodies in the area of Šabrava, while the other components of the food chain of wild boar did not exceed a few tens of $Bq \cdot kg^{-1}$ (Dvořák et al. 2010).

Possibilities of reduction of food contamination

Reduction of radiocaesium transfer to animals

A wide range of different and effective measures were implemented to reduce the contamination of livestock by radiocaesium. Administration of potassium in the diet caused an increase in radiocaesium excretion (Mraz et al. 1958; Mraz 1959; Johnson et al. 1968), while the administration of stable caesium was not efficient (Oughton et al. 1989; Rundo 1964). Different complexes or chelating additives are used successfully, with the ability to absorb and/or to bind radionuclides forming chemical compounds not absorbed by the digestive tract but easily excreted from the body (Giese 1988; Giese 1989; Pöschl and Baláš 1999; Pöschl and Řezáč 2004). The same positive effect on

^{137}Cs contamination reduction was observed by adding 5% bentonite into a feeding ration (Andersson et al. 1990). In ruminants grazing on mountain pastures in Norway, salt lick with 2.5% AFCF (Giese salt) was used with a positive effect (Hove et al. 1990). Capsules containing 15–20% AFCF reduced the radiocaesium concentration in meat by 50–80% (Hove 1993).

Publications on the reduction of radionuclides directly in the foodstuff are very rare, and were mainly focused on mushrooms and meat. However, a marked reduction to acceptable concentrations of radiocaesium in foodstuffs was achieved by leaching, macerating, marinating, soaking and/or boiling.

Caesium has a number of physical and chemical properties that are similar to those of potassium. *In vivo*, the two elements behave similarly, and they do not influence each other. Due to good water solubility, caesium is very well and relatively evenly absorbed, and distributed in the body. After the Chernobyl accident, from the total amount of radiocaesium present in different types of foodstuffs meat accounted for 50%, foods of plant origin approximately 30%, and milk 14% (Wagner 1988).

Reduction of radiocaesium contamination in mushrooms

For ^{137}Cs activity reduction in fungi, some preservation methods can be used. The basic form of fungi storage include drying, preservation in vinaigrette, or freezing, with drying representing the oldest preservation method. During drying, 80–90% of the mushroom water content is evaporated. Due to the relatively long half-life of radiocaesium, ^{137}Cs remains in the dry matter as a source of contamination for a long time.

Almost complete ^{137}Cs decontamination was achieved by dipping frozen or dried mushrooms in brine (Neukom and Gisler 1991). Similar results in mushrooms were achieved by using boiling in water (over 80% decontamination) and salted boiling water (Steger et al. 1987; Klan et al. 1988).

Canning of mushrooms in vinaigrette is one of the most popular preservation methods, as mushrooms treated in this way have multiple uses. Significant ^{137}Cs content reduction (59–73%) was achieved in fungi by soaking them in vinegar brine (2% solution of acetic acid) (Dvořák et al. 2006b). One load in the vinegar brine showed a very positive effect on ^{137}Cs activity reduction in both dried and fresh mushrooms. Nevertheless by repeating the process, mushroom consistency deteriorated, especially in mushrooms that were thawed or fresh. Dried mushrooms maintained their consistency for the whole period of the experiment.

As the decrease in ^{137}Cs activity achieved by 2% acetic acid solution treatment has an exponential character, in areas significantly affected by post-Chernobyl radiation, the vinaigrette preservation technology for mushrooms can be recommend. However, it needs to be pointed out that the vinaigrette itself must not be consumed (Dvořák et al. 2006b).

Reduction of radiocaesium contamination in meat

Elimination or at least reduction of radioactivity in the red deer (*Cervus elaphus*) meat can be achieved by marinating, soaking in brine or water (Jandl et al. 1989). This procedure is one of the oldest method of preserving meat, using long-term brine treatment, or using salty mixtures composed of table salt with the addition of 3% sodium nitrate or potassium. After two days, a 50% reduction of ^{137}Cs is achieved in comparison to the sample starting value. The second and third loading for additional 2+2 days decreased radiocaesium activity to 20% and 10%, respectively. It has been shown that increasing temperatures speeds up the radiocaesium release process in reindeer meat. Meat cooking was only slightly less effective in radiocaesium reduction than meat treatment in salt solutions or plain water. A 1% salty water solution was used for heat treatment (meat cooking) (Lofti et al. 1989).

Washing meat samples under running water for 17 h showed a decrease of ^{137}Cs by up to 10% of the starting level (Petäjä et al. 1992).

After heat pressure treatment the mean ^{137}Cs activity in the meat was reduced by 50% (Dvořák and Kunová 2006).

To reduce meat contamination by brining, both pure 8% sodium chloride and a mixture of 8% sodium chloride and potassium nitrate were used. The brine was exchanged each 24 h and the process was repeated × 4 for pure 8% sodium chloride and × 3 for the mixture of 8% sodium chloride and potassium nitrate. Pure 8% sodium chloride brine reduced ^{137}Cs concentration by 72%; the mixture of 8% sodium chloride and potassium nitrate by 77% (Dvořák et al. 2008).

Impact of using salt treatment on radiocaesium reduction was also studied in fish (Petäjä et al. 1992). The radiocaesium activity in fish meat was reduced by 50% after 4 h either by using 5% saline and water, or by watering for 8 h (Petäjä et al. 1992).

Conclusion

Despite the fact that shortly the 30th anniversary of the Chernobyl accident will be commemorated, we still face its dire consequences to this day. Excessive values of ^{137}Cs measured in the past years in the meat of the wild boar population in Germany and the Czech Republic represent a potential radioactive risk for humans. For this reason, radioactivity tracking as well as studying new methods of food decontamination is still highly actual.

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