https://doi.org/10.17221/140/2019-CJFS

Activity of natural radionuclide ^{210}Pb in Czech foodstuffs and its annual intake

Miluše Bartusková[1], Irena Malátová[2], Věra Bečková[2], Jiří Hůlka[2]*

[1]*Branch Ostrava, National Radiation Protection Institute, Ostrava, Czech Republic*

[2]*National Radiation Protection Institute, Prague, Czech Republic*

**Corresponding author: miluse.bartuskova@sujb.cz*

Citation: Bartusková M., Malátová I., Bečková V., Hůlka J. (2019): Activity of natural radionuclide ^{210}Pb in different foodstuffs in the Czech Republic and its annual intake. Czech J. Food Sci., 37: 463–468.

Abstract: Activities of ^{210}Pb in 18 sorts of foodstuffs were determined by radiochemical analysis. The lowest activity was found in carrots (0.01 Bq/kg) and cabbage (0.03 Bq/kg), the highest in banana (1.54 Bq/kg). The activity in potatoes, milk, wheat rolls, dairy products and beef was below the limit of detection. Annual intake of ^{210}Pb by inhabitants of the Czech Republic was calculated using food basket and it was 117.7 Bq/year; without banana 101.2 Bq/year. It is higher than the average value given by UNSCEAR Report for the world as well as for Europe. The reason of this finding is discussed.

Keywords: food; radioactivity; lead-210; ingestion; dose

The environment including living organisms contains varying concentrations of natural radionuclides, which enter human body mainly through inhalation and ingestion. Knowledge of their content in food is thus important in order to estimate doses to human organism. Since the first use of atomic bombs, following massive atmospheric test of nuclear weapons and development of peaceful uses of nuclear energy, occurrence of radioactive substances in food has been surveyed worldwide. Mostly man-made radionuclides were in focus at first, but since the ban of atmospheric tests, natural radioactivity of foodstuffs and intake of natural radionuclides by ingestion and inhalation has been intensively studied worldwide (UNSCEAR 2000). Isotope of lead of mass number 210 – ^{210}Pb is one of the most important contributors to natural radiation background. It is a member of the uranium decay chain; its half-life is 22.3 years (Chu *et al.* 1999). Main source of ^{210}Pb in the atmosphere is emanation ^{222}Rn from soil and its decay through short-living products. Worldwide ^{210}Pb content in air of (0.74–1.85) × 10^{15} Bq (Beneš 1988; Rulík *et al.* 1993) and activity concentration of 28–2250 Bq/m^3 are estimated (UNSCEAR 2000). Activity concentration strongly depends on distance of a location from the seashore, on altitude and on geographical longitude (Preiss *et al.* 1996). Further sources of ^{210}Pb, though less important than the former, are for example fossil fuels burning, phosphate fertilisers production and use (Salmon *et al.* 1996). From the atmosphere ^{210}Pb is washed out by rain or, comes to Earth surface as dry fallout, penetrates to plants and animals and through them to the human food chain.

Content of ^{210}Pb in food is important due to doses for population, considering its decay products ^{210}Bi (β) and ^{210}Po (α). Lead accumulates in bone, whereas polonium is distributed mainly to soft tissues.

In order to calculate activity of ^{210}Pb ingested with food it is necessary to know composition of food basket for the average consumer and levels of activity concentrations in particular food components.

MATERIAL AND METHODS

Food consumption. Consumption of food has been monitored since 1948, the data are nowadays provided by Czech statistical institute (CSI). These data analysed National Radiation Protection Institute in 2003 (Bartusková 2005), later analyses were published in

https://doi.org/10.17221/140/2019-CJFS

(Bartusková *et al.* 2017) and (Škrkal *et al.* 2017). Another data sets about consumption of food in the Czech Republic (CR) are available from the Czech National Institute of Public Health (Ruprich *et al.* 2006; Dofková *et al.* 2009), on European Food Safety Authority (EFSA).

Progression of food categories consumption in period 1948–2017, in terms of weight per average person and year, is shown in Figure 1, using (CSI 2017) data. In 2003 eighteen food components whose consumption proportion was bigger than 0.5% of total annual consumed amount had been chosen. Their list is in Table 1. From year 2003, when ^{210}Pb activities were determined, changes in consumption are not so apparent as before 2000 (Figure 1.)

Determination of ^{210}Pb activity concentrations. ^{210}Pb activities were determined by radiochemical analysis, described in (Bartusková *et al.* 2006). Food samples were bought in 3 shops in 2 big cities in the northern Moravia, north-eastern part of the CR. Samples were dryashed for 48 h at temperature 350°C, then wetashed with nitric and hydrochloric acid. The extraction chromatographic material SrResin® was used for the Pb separation in hydrochloric medium (Vajda *et al.* 1997) and Pb was eluted with 6 M HCl. Eluted lead was precipitated with oxalic acid. After 22 days the activity of the ingrown ^{210}Bi was measured using gas-flow proportional detector (POB 302E; EMPOS, Czech Republic). Measuring times ranged from 90 000 to 150 000 s. Activity concentrations of ^{210}Pb in Bq/kg of samples, *a*, were calculated (Equation 1):

$$a = \frac{\dfrac{\dfrac{N_s}{t_S} - \dfrac{N_b}{t_b}}{n \times m \times Y}}{1 - e^{-\ln 2 \frac{t}{T_{1/2}}}} \qquad (1)$$

where: N_S – counts from sample; N_b – counts from background; t – measuring time of sample; t_b – measuring time of background; h – counting efficiency; m – sample weight; Y – chemical yield of separation; t – time between separation and measurements; $T_{1/2}$ – half-life of ^{210}Bi.

Uncertainty is given as combined standard deviation *U*. Minimum detectable activity of a determination, *MDA*, was calculated using equations (Equations 2 and 3) (Currie 1968):

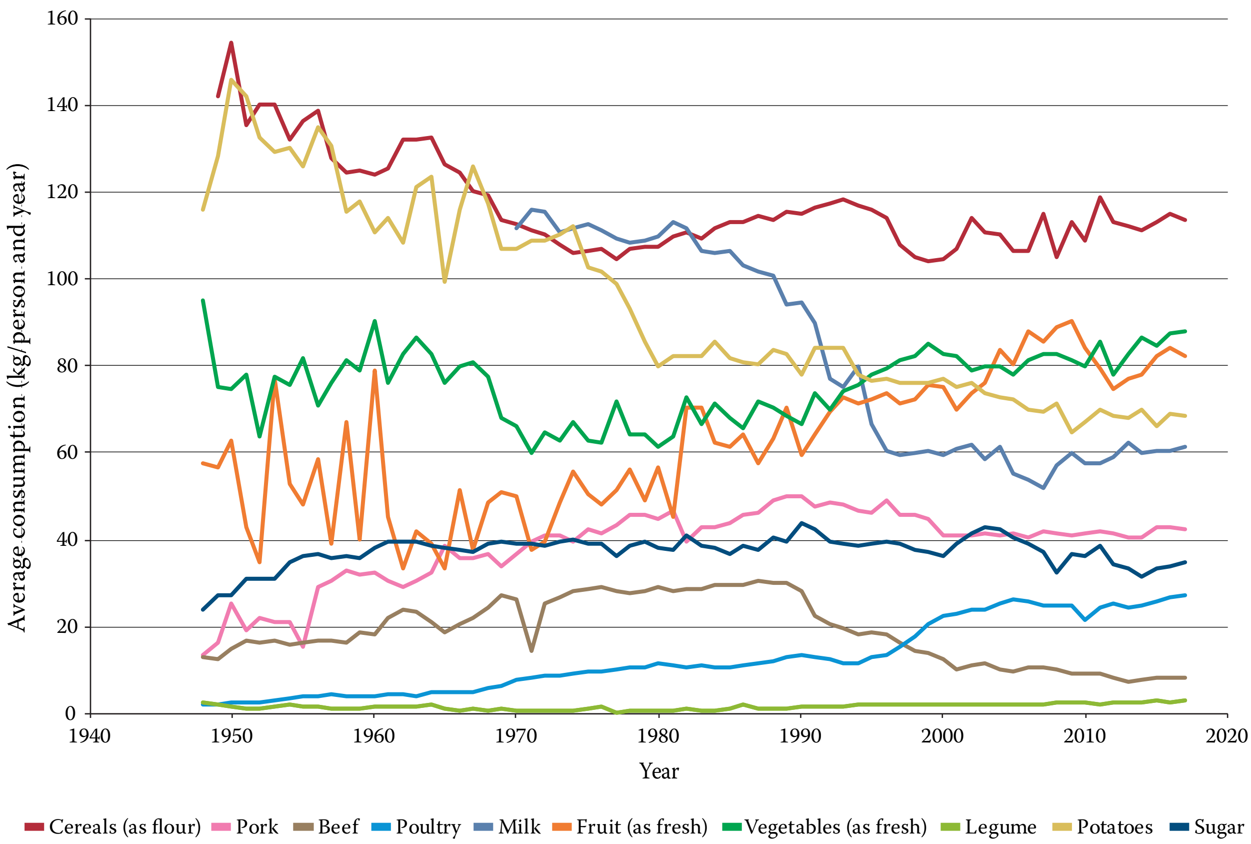

Figure 1. Average per capita annual consumption for main food categories in period 1947–2017 (data from CSI 2017)

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$$MDA = \frac{u^2}{t_S \times \eta \times m \times Y} + 2MSA \quad (2)$$

$$MSA = u \times \frac{\sqrt{N_b\left(1+\frac{t_b}{t_s}\right)}}{\eta \times t_b \times m \times Y} \quad (3)$$

where: *MSA* – minimum significant activity; *u* – quantile of normal distribution.

Annual ^{210}Pb ingestion intakes *I* were calculated as a product of determined ^{210}Pb activities *a* and annual consumption *S* of a single food (Equation 4):

$$I = a \times S \quad (4)$$

Annual effective dose for single food groups *E(t)* was calculated using equation (Equation 5) with effective dose coefficient for ingestion intake for adults *e* value 6.9×10^{-7} Sv/Bq (ICRP 2012) (Equation 5):

$$E(\tau) = I_0 \times e \quad (5)$$

RESULTS AND DISCUSSION

Activity concentrations are given in the 1st column of Table 1. ^{210}Pb activities varied from 0.010 ± 0.009 Bq/kg for carrots to 1.54 ± 0.40 Bq/kg for bananas. For five food groups the determined activities were lower than MDA, therefore values of ¼ of MDA were used in next calculations. Intakes and annual effective doses for each year in period 2003–2017 and the arithmetical average for the whole period were calculated (Equations 4 and 5). Minimum annual intake from ingestion of chosen food categories was 111.2 Bq/year in 2013, highest 125.5 Bq/year in 2004. Average value for period 2003–2017 was 117.7 ± 3.7 Bq/year. Average daily intake was calculated as 0.32 Bq/day.

Minimum calculated ingestion annual effective dose $E_{(t)}$ from ^{210}Pb was 7.7×10^{-5} Sv/year in year 2013, highest 8.7×10^{-5} Bq/year in year 2004. Average for period 2003–2017 was 8.1×10^{-5} Bq/year

In Table 2 data on ^{210}Pb activity concentrations for Czech Republic (Bartuskova *et al.* 2006), Great Britain (Smith-Briggs *et al.* 1986), France (Renaud *et al.* 2015), Europe (UNSCEAR 2000) and for banana in Vietnam (Tran *et al.* 2018) are presented. Czech annual consumption of several food groups were compared with reference values (whole world average) from the UNSCEAR (2000) publication – given in Table 3, as well as calculated ratio our consumption/reference values. This comparison is only rough due to very different

Table 1: ^{210}Pb activities, annual consumptions and proportions of chosen foods

Food	^{210}Pb activity concentration (Bq/kg)		Consumption (kg/person and year)		Proportion of consumption (%)	
	determined	¼ MDA	2003	2017	2003	2017
Bread	0.60 ± 0.19	–	54.3	39.2	7.6	5.3%
Sugar	0.22 ± 0.09	–	43.0	34.9	6.0	4.7
Pork	0.21 ± 0.03	–	41.5	42.3	5.8	5.8
Apple	0.09 ± 0.03	–	23.8	22.3	3.3	3.0
Egg	0.19 ± 0.06	–	12.8	12.7	1.8	1.7
Tomato	0.14 ± 0.03	–	12.6	11.2	1.8	1.5
Orange	0.53 ± 0.12	–	12.5	12.3	1.7	1.7
Chicken	0.19 ± 0.06	–	23.8	27.3	3.3	3.7
Cabbage	0.03 ± 0.01	–	11.0	8.0	1.5	1.1
Banana	1.54 ± 0.04	–	10.4	11.5	1.5	1.6
Carrot	0.010 ± 0.009	–	6.1	7.1	0.9	1.0
Pasta	0.34 ± 0.09	–	5.6	7.8	0.8	1.1
Rice	0.62 ± 0.21	–	5.0	6.5	0.7	0.9
Potatoes	< 0.36	0.09	73.6	68.5	10.3	9.3
Milk	< 0.22	0.06	58.4	61.3	8.2	8.3
Wheat rolls/pastries	< 1.5	0.38	43.8	50.4	6.1	6.9
Dairy products	< 0.30	0.08	29.4	34.4	4.1	4.7
Beef	< 1.6	0.40	11.5	8.4	1.6	1.1
Sum			479.1	466.0	67.0	63.4
Annual consumption			715.2	735.5		

< means lower than MDA

https://doi.org/10.17221/140/2019-CJFS

Table 2. ^{210}Pb activity concentrations comparison

Food group	^{210}Pb activity concentration (Bq/kg)				
	CR[a]	GB[b]	France[c]	Europe[d]	Vietnam[e]
Milk products	< 0.22	< 0.1	0.12	0.005–0.280	–
Meat	0.19; < 1.6	< 0.14	0.34	0.015–3.700	–
Cereals	0.6; < 1.5	0.066; 0.093 (bread only)	0.41	0.040–4.000	–
Leafy vegetables	0.03	–	0.34	0.004–4.100	–
Root vegetables and fruits	0.01–1.54	0.049; 0.068	0.12	0.018–4.900	–
Banana	1.54 ± 0.04	–	–	–	0.36 ± 0.17

[a] Bartuskova *et al.* (2006); [b] Smith-Brigg *et al.* (1986); [c] Renaud *et al.* (2015); [d] UNSCEAR (2000); [e] Tran *et al.* (2018)

food basket composition on individual continents (GNPC 2018). Czech consumption of milk and milk products, the same as of meat, is higher than word reference values and on the other hand, it is lower for root vegetables and fruits, cereals and for leafy vegetables (only 0.2 of reference value). The sum of all compared groups (in kg) in the Czech Republic is 1.1 times higher than that of reference values. Changes in food basket composition between years influenced the intakes and annual effective doses calculation. Even though Czech food basket has not changed dramatically after 2003, some trends in consumption (decrease of potato and sugar consumption) are mirrored in our results.

Differences in amount of consumed food are mentioned by Mertens *et al* (2019). They compiled data obtained from national dietary surveys made in 4 European countries: Denmark, France, Italy and Czech Republic. Quite big differences, both between individual countries and between individual respondents in one country were found. Mean fruit intake ranged from 118–199 g/day, for vegetables from 95 to 239 g/day, for fish from 12 to 45 g/day, for dairy from 129–302 g/day, with higher intakes in Italy for fruits, vegetables and fish, and in Denmark for dairy foods (Mertens *et al*. 2019).

Pietrzak-Flis *et al.* (1997) found the largest contribution to ^{210}Pb intake in central Poland for flour and meat (approx. 50%). For the Czech Republic it was more, about 60%. Founded rate for banana was about 16%, because the result for ^{210}Pb activity in banana is abnormally high even though the proportion of the consumption is not very high (banana are not domestic fruit in CR).

Calculated intakes of ^{210}Pb from experimentally determined activity concentrations were compared with ^{210}Pb intakes for adults, published in UNSCEAR (2002). Though worldwide reference value is 30 Bq/year, published values for Europe ranged in interval (18–84) Bq/year. Czech calculated average annual value for period 2003–2017 is 117.7 Bq/year, intake without banana is 101.2 Bq/year. Our ^{210}Pb intakes are approximately 1.4 times higher than the highest published intake in Europe. The reason probably lies in food consumption in CR which is higher than the average European consumption.

Several published sources present intakes as daily intake. In GB daily ^{210}Pb intake 0.082 Bq/day was found by Smith-Briggs *et al.* (1986), Persson and Holm (2011) presented in their review average median of daily dietary intakes of ^{210}Pb for the adult

Table 3. Annual food groups consumption comparison for Czech consumers in 2017 (CSI 2017) and annual reference values for adult people (world average) from UNSCEAR (2000)

Food groups	Annual consumption (kg/year)		Ratio of consumption in CR and reference value
	CR	reference values (UNSCEAR)	
Milk products	246.5*	105	2.4
Meat	80.3	50	1.6
Cereals	105.2	140	0.8
Leafy vegetables	10.8	60	0.2
Root vegetables and fruits	159.6	170	0.9
Sum	602.4	525	1.1

*in terms of milk (CSI 2017)

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world population as 0.11 Bq/day. In another review source Salmon *et al.* (1998) gives daily ^{210}Pb intake for Poland 0.12 Bq/day, for Germany 0.17 Bq/day and for Russia 0.23 Bq/day. According to the present study, daily ^{210}Pb intake in the CR is 0.32 Bq/day, without banana 0.28 Bq/day, what is most similar to the Russian intake.

Annual effective doses, caused by ^{210}Pb consumed with food were compared with the value for Europe, published in UNSCEAR (2000). Published values ranged between 1.2×10^{-5} and 5.8×10^{-5} Sv/year, with reference value for average adult world inhabitant 2.1×10^{-5} Sv/year. Persson and Holm (2011) published in their review 2.8×10^{-5} Sv/year, Pietrzak-Flis *et al.* (1997) determined 4.5×10^{-5} Sv/year for central Poland.

Persson and Holm (2011) published annual effective dose from ^{210}Pb in only vegetarian food, which was estimated as 1.0×10^{-5} Sv/year. Other data were published in Poland (Strumińska-Parulska and Olszewski 2018) for vegetables and cereals, both from traditional and ecological agriculture. For ecological agriculture they calculated 1.3×10^{-6} Sv/year, for traditional agriculture then 1.9×10^{-6} Sv/year. In this case, however, the annual effective doses were lower than for classical food basket.

Czech calculated average value is 8.1×10^{-5} Sv/year, without banana 7.0×10^{-5} Sv/year. Probable reason for the high values is higher food consumption.

CONCLUSIONS

Generally, activities of ^{210}Pb found in several foodstuffs consumed in the Czech Republic are comparable with activities published in other countries. However, both annual and daily ^{210}Pb intakes and annual effective doses are higher. Therefore, comparison of activity concentrations is more reasonable than comparing annual intakes, where the choice of food basket is a major factor. Another probable source of higher estimated values is a relatively high occurrence of foods in which values were under relatively high MDA. To decrease MDA and broaden the sort of foodstuff has to be subject of further research.

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Received: 2019–05–23
Accepted after corrections: 2019–10–14