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SHORT RESEARCH AND DISCUSSION ARTICLE

^{210}Po and ^{210}Pb bioaccumulation and possible related dose assessment in parasol mushroom (*Macrolepiota procera*)

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Abstract Presented are results of a study on accumulation and distribution of ^{210}Po and ^{210}Pb in the fruitbodies of parasol mushroom (*Macrolepiota procera*) and risk to human consumer due to exposure from highly radiotoxic decay particles emitted by both radionuclides. Mushrooms were collected from 16 forested places in central and northern regions of Poland. Activity concentrations of ^{210}Po and ^{210}Pb were determined after radiochemical separation of nuclides and subsequent measurement using validated method and alpha spectrometer. Results showed on spatially heterogeneous distribution of the ^{210}Po and ^{210}Po activity concentrations in *M. procera* and two interpolation maps were prepared. Activity concentrations of nuclides in dried caps of *M. procera* were in the range from 3.38 ± 0.41 to 16.70 ± 0.33 Bq·^{210}Po ·kg^{-1} and from 5.11 ± 0.21 to 13.42 ± 0.30 Bq·^{210}Pb ·kg^{-1}. Consumption of *M. procera* foraged in central and northern Poland should not contribute significantly to the annual effective radiation doses from ^{210}Po and ^{210}Pb due to amount of both nuclides accumulated by fungus in caps.

Keywords Polonium ^{210}Po · Radiolead ^{210}Pb · Accumulation · Mushrooms · Foraging · Effective radiation dose

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Introduction

It is proved that many edible and also inedible fungi, if compared to other vegetable foods, can accumulate certain metallic elements in the fruitbodies (mushrooms) at elevated or great concentrations even if it grew in soil unpolluted with heavy metals or soil not enriched geochemically in toxic elements. What is more, certain fungi can take up and accumulate in flesh some trace elements and radioisotopes in a species-specific mode (Falandysz and Borovička 2013). Factors considered as influencing bioavailability, bioaccumulation, and biological importance of metallic elements accumulated in plants are pH, Eh, water regime, clay content, organic matter content, cation exchange capacity, nutrient balance, concentration of other trace elements, higher ambient temperature, and climatic conditions due to water flow phenomenon (Kabata-Pendias 2011), while soil factors other than pollution or geochemical anomaly that can influence process of metallic elements accumulation by fungi are considered as little known or generally without effect (Baptista et al. 2009; Falandysz and Borovička 2013). Naturally or artificially elevated content of some metallic elements in forest topsoil can result in heightened accumulation in mushrooms (Kojta et al. 2012). Concentrations of Hg, Cd, and Pb can be relatively high both in certain terrestrial saprotrophs but also in ectomycorrhizal fungi that usually accumulate higher amounts of alkali metals, Rb, and Cs (Borovička and Řanda 2007; Falandysz et al. 2015, 2017a, b, c, d; Řanda and Kučera 2004).

There are numerous reports describing mushrooms as efficient accumulators and bioindicators of environmental diffusion of radionuclides even though the capacity to sequester radioactive elements and the rates of their uptake vary significantly between species. As stated by the authors Falandysz and Borovička (2013), there is no reason for mushrooms to discriminate between stable and radioactive isotopes of elements when absorbed and accumulated in the mushroom flesh, while their accessibility and availability at sites where the mycelium lives can be different. Frequently studied is ^{137}Cs that polluted the surface of the Earth due to global radioactive fallout after using and testing of nuclear weapons and from nuclear power plant accidents but also ^{40}K that is of natural terrestrial origin (Mietelski et al. 2010; Steinhauser et al. 2014; Falandysz et al. 2016; Zalewska et al. 2016; Falandysz et al. 2017d). Other radionuclides studied in mushrooms included ^{210}Po, ^{210}Pb, ^{234,238}U, ^{228,230,232}Th, ^{238}Pu, $^{239+240}$Pu (Mietelski et al. 2002; Vaaramaa et al. 2009; Guillén and Baeza 2014; Strumińska-Parulska et al. 2016).

Both ^{210}Po and ^{210}Pb are daughters of uranium ^{238}U and radioecologically interesting natural elements to investigate due to their high radiotoxic characteristics (Strumińska-Parulska 2015). Both are natural radionuclides and their half-lives are 138.38 days for ^{210}Po and 22.3 years for ^{210}Pb (Boryło et al. 2013; Persson and Holm 2011). These natural radionuclides are found in varying concentrations in soil, sand, sediment, and natural water and constitute an important component of the natural background radiation. They are introduced into the biosphere through various routes of terrestrial and marine pathways and continuously deposited from the atmosphere in association with aerosols. ^{222}Rn constantly emanates from the ground, decaying through short-lived radon daughters to ^{210}Pb, ^{210}Bi, and ^{210}Po which attach to airborne particles and return to the earth as dry fallout or are washed out in rain (Struminska-Parulska et al. 2010; Persson and Holm 2011). ^{210}Po and ^{210}Pb are known to significantly contribute to the radiation dose of the population (Persson and Holm 2011). Anthropogenic sources of these radionuclides are burning of fossil fuels, tetraethyl lead in petrol, dust storms, refineries, superphosphate fertilizers, the sintering of ores in steelworks, and the burning of coal in coal-powered power stations (Boryło et al. 2012) but also so called technologically enhanced naturally occurring radioactive materials (TENORM) including enriched in ^{210}Po and ^{210}Pb phosphogypsum that stored in stacks can increase these radionuclides concentrations in nearby soils, biota, and water (Boryło et al. 2013; Olszewski et al. 2015, 2016). ^{210}Po is highly toxic and its presence in soils may be traced to the decay of radionuclides of the ^{238}U chain in the soil (Aslani et al. 2005).

Macrolepiota procera (Scop. Fr.) Singer (1948), formerly also called *Lepiota procera* and known under the common name parasol mushroom, field parasol, or shaggy parasol, is widely collected in temperate regions and sub-tropical regions such as India, Thailand, China, Pakistan, and across Europe. It has an edible and delicious pileus, highly valued by locals, cooked fresh—sautéed, roasted, fried in butter or grilled, roasted with eggs or stuffed, and broiled (Falandysz et al. 2017a). Parasol mushroom prefers lighted and warm places, especially in calcareous and sandy soils that are well drained in forests, meadows, and gardens (Rizal et al. 2015). Parasol mushrooms are common in Poland, can be purchased commercially, but because of the relatively fragile structure of its cap, it is rarely offered at the rural markets, while easily available from roadside sellers in the countryside (Gucia et al. 2012a, b). Due to this, many elements alongside with their bioconcentration factors have been analyzed in parasol mushrooms (Falandysz et al. 2007, 2008; Jarzyńska et al. 2011; Kułdo et al. 2014; Stefanović et al. 2016a, b).

The aims of the study were to determine ^{210}Po and ^{210}Pb content in fruiting bodies (caps and whole mushrooms) of parasol mushroom (*Macrolepiota procera*); calculate the values of ^{210}Po/^{210}Pb activity ratios; recognize radionuclide distribution; estimate possible annual effective radiation; and evaluate the level of their radiotoxicity.

Materials and methods

Fruiting bodies of wild edible parasol mushroom (*Macrolepiota procera*) were collected from 16 forested places across central and northern Poland: near Bydgoszcz, Łuby, Osiek, Gdańsk, island of the Jeziorak lake, Kartuzy, Kukawy, Warta Landscape Park, Vistula Spit, Olsztyn, Augustów, Lębork, Sarnówek, Toruń, Poniatowa, and Włocławek (Figs. 1 and 2). The exact procedure for samples preparation for ^{210}Po and ^{210}Pb analysis was described by Strumińska-Parulska et al. (2016). The radiochemical analysis of ^{210}Po included polonium autodeposition on silver discs and its measurement using an alpha spectrometer equipped with semiconductor silicon detectors and a 450-mm^2 active surface barrier (Alpha Analyst S470, Canberra-Packard, USA) according to the method described by Skwarzec (1997). Activity concentration of ^{210}Pb in analyzed samples was calculated indirectly via its daughter ^{210}Po activity measurement. After 6 months, polonium was again autodeposited on silver discs and activities of ingrown ^{210}Po were measured in alpha spectrometer (Alpha Analyst S470, Canberra-Packard). ^{210}Pb activities at the time of mushrooms collection were calculated using the simplified equation for the daughter activity as a function of time (Skwarzec 1997). The ^{210}Po and ^{210}Pb yield in the analyzed mushroom and soil samples ranged from 95 to 100%. The results of ^{210}Po and ^{210}Pb activity concentrations were given with standard deviation (SD) calculated for 95% confidence intervals. The accuracy and precision of the radiochemical method were positively evaluated using IAEA

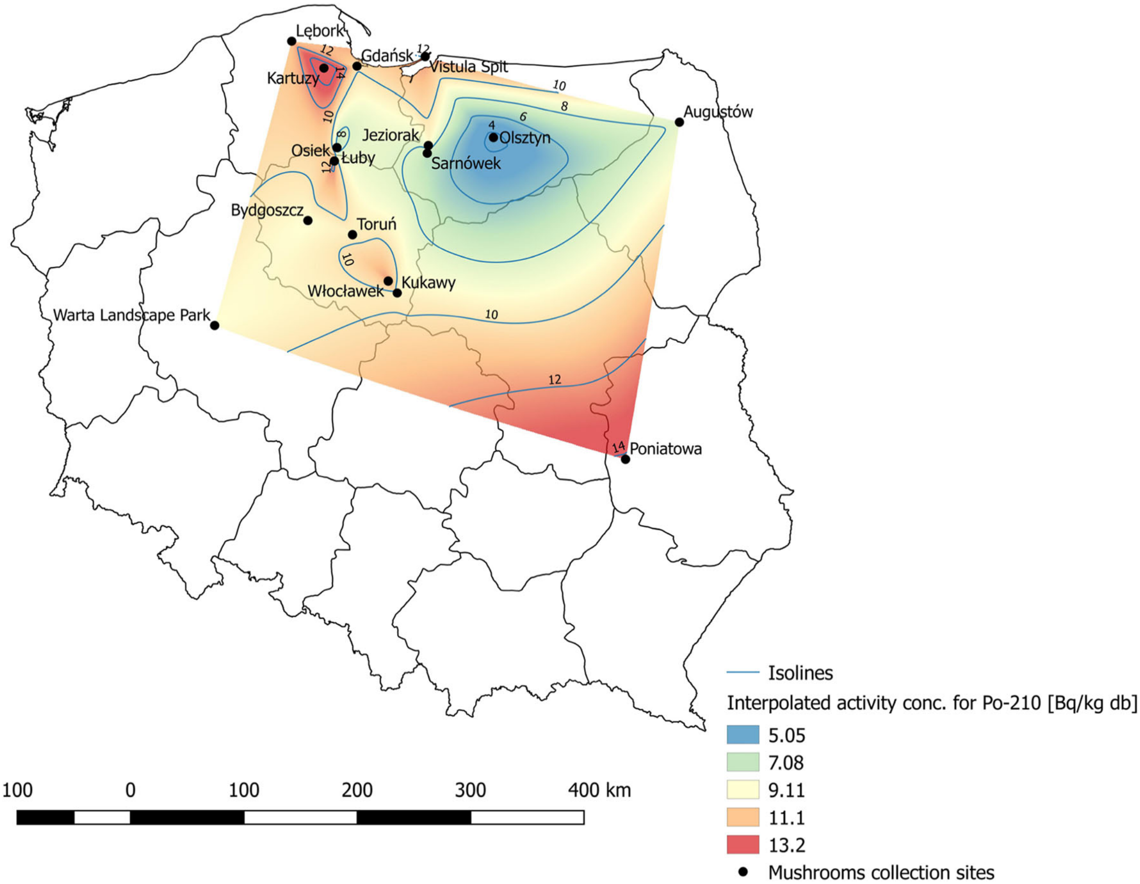

Fig. 1 Interpolation for ^{210}Po activity concentrations in parasol mushroom (*Macrolepiota procera*)

reference materials (IAEA-414; IAEA-384), and both were estimated at less than 5%.

Results and discussion

^{210}Po and ^{210}Pb activity concentrations in parasol mushroom

The results of ^{210}Po and ^{210}Pb activity concentrations in parasol mushroom (*M. procera*) samples were presented in Table 1. The highest ^{210}Po activity concentrations were measured in caps of *M. procera* collected in Kartuzy (16.70 ± 0.33 Bq·kg^{-1} dry biomass), while the lowest in caps collected in Olsztyn (3.38 ± 0.41 Bq·kg^{-1} db). Similar observations were noticed for ^{210}Pb—the highest ^{210}Pb activity concentration was determined in parasol mushroom caps collected in Kartuzy (13.42 ± 0.30 Bq·kg^{-1} db), while the lowest in samples from Łuby (5.11 ± 0.21 Bq·kg^{-1} db). Using mass units, ^{210}Po concentration in caps was from $2.05 \cdot 10^{-11}$ to $1.01 \cdot 10^{-10}$ mg·kg^{-1}, while ^{210}Pb ranged from $1.82 \cdot 10^{-9}$ to $4.79 \cdot 10^{-9}$ mg·kg^{-1}. Similar results were received in Finland where the highest activity concentration of ^{210}Pb was detected in red-banded *Cortinarius* (*Cortinarius armillatus* Fr) (16.2 Bq·kg^{-1} db) and the lowest in foxy bolete (*Leccinum vulpinum* Watling) (1.38 Bq·kg^{-1} db) (Vaaramaa et al. 2009). In general, the pattern of the ^{210}Pb and ^{210}Po distribution within the mushrooms seems to be species dependent (Guillén and Baeza 2014), e.g., in some species of the *Boletaceae* family, the cap content was higher than that of the stipe (Vaaramaa et al. 2009). Similarly, the average concentrations of heavy metals (Ni, Cr, Pb, Cd, and Hg) in the anatomical parts of the fruiting body (cap and stipe) were considerably different (Falandysz et al. 2008; Baptista et al. 2009; Širić et al. 2017; Falandysz et al. 2017a).

In order to present the distribution of ^{210}Po and ^{210}Pb activity concentrations in Poland, based on analyzed *M. procera* samples, we prepared interpolation maps (Figs. 1 and 2) using natural neighbor interpolation method (Sibson 1981) and QGIS software (QGIS Development Team). In our previous study on red-capped scaber (*Leccinum aurantiacum*), we noticed that differences in activity concentrations (in Bq·kg^{-1} db) between caps and whole mushrooms were vague and statistically insignificant (Mann-Whitney *U* test $p > 0.05$) (Strumińska-Parulska et al. 2016). In this case, for the purpose of map preparation, we decided to use both *M. procera* caps and whole mushrooms ^{210}Po and ^{210}Pb activity

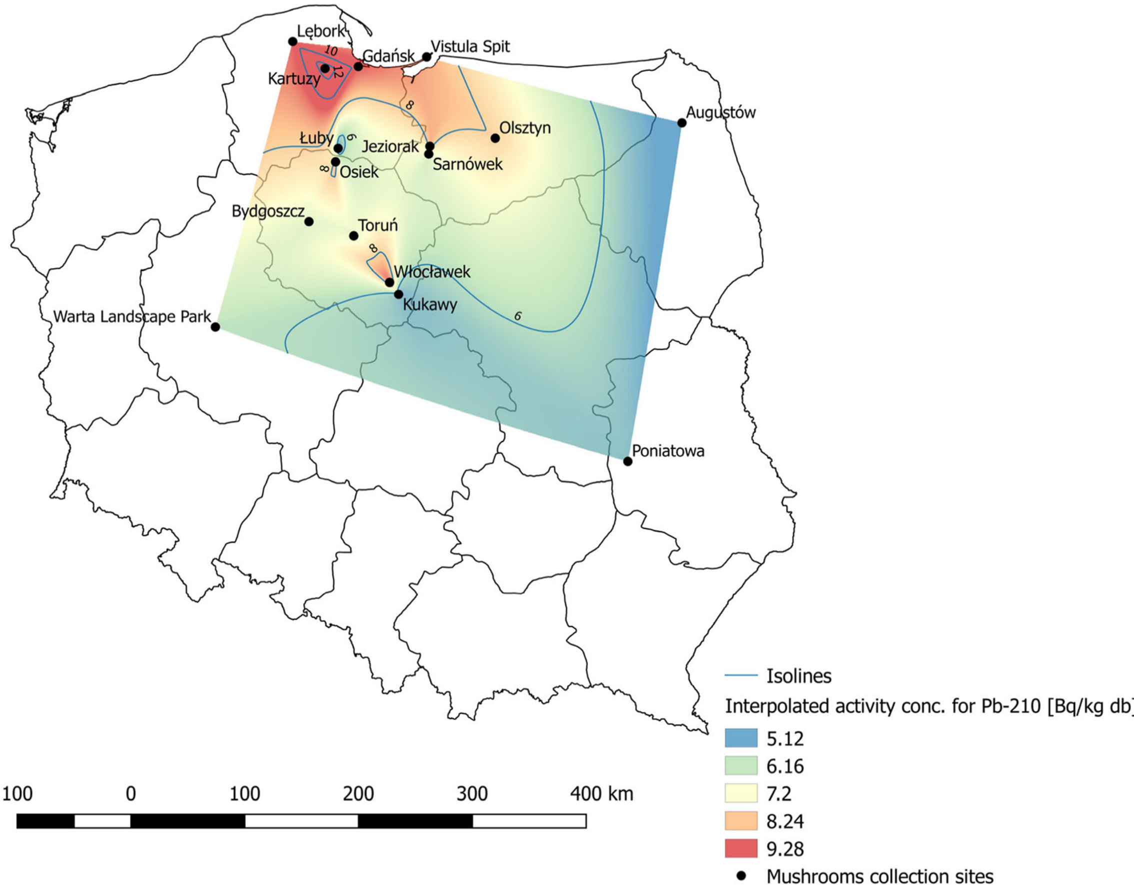

Fig. 2 Interpolation for ^{210}Pb activity concentrations in parasol mushroom (*Macrolepiota procera*)

Table 1 ^{210}Po and ^{210}Pb activity concentrations, the values of ^{210}Po/^{210}Pb activity ratio, and annual effective radiation doses for analyzed parasol mushrooms (*Macrolepiota procera*)

Sampling site	Activity concentration (Bg·kg^{-1} dry biomass)		^{210}Po/^{210}Pb activity ratio	Annual effective radiation dose (μSv·year^{-1})	
	^{210}Po	^{210}Pb		^{210}Po	^{210}Pb
Caps					
Gdańsk	10.13 ± 0.50	9.31 ± 0.26	1.09 ± 0.06	6.08 ± 0.30	3.21 ± 0.09
Jeziorak	9.43 ± 0.25	8.46 ± 0.24	1.11 ± 0.04	5.66 ± 0.15	2.92 ± 0.08
Kartuzy	16.70 ± 0.33	13.42 ± 0.30	1.24 ± 0.04	10.02 ± 0.20	4.63 ± 0.10
Warta Landscape Park	9.09 ± 0.74	6.10 ± 0.16	1.49 ± 0.13	5.45 ± 0.45	2.11 ± 0.06
Vistula Spit	12.07 ± 0.74	8.49 ± 0.21	1.42 ± 0.09	7.24 ± 0.44	2.93 ± 0.07
Olsztyn	3.38 ± 0.41	7.94 ± 0.17	0.43 ± 0.05	2.03 ± 0.25	2.74 ± 0.06
Augustów	8.18 ± 0.42	4.63 ± 0.11	1.77 ± 0.10	4.91 ± 0.25	1.60 ± 0.04
Lębork	11.10 ± 0.30	9.21 ± 0.27	1.21 ± 0.05	6.66 ± 0.18	3.18 ± 0.09
Sarnówek	6.78 ± 0.20	6.51 ± 0.23	1.04 ± 0.05	4.07 ± 0.12	2.24 ± 0.08
Toruń	9.57 ± 0.62	6.82 ± 0.12	1.40 ± 0.09	5.74 ± 0.37	2.35 ± 0.04
Włocławek	11.90 ± 0.32	9.14 ± 0.28	1.30 ± 0.05	7.14 ± 0.19	3.15 ± 0.10
Łuby	6.84 ± 0.17	5.11 ± 0.21	1.34 ± 0.06	4.11 ± 0.10	1.76 ± 0.07
Whole mushrooms					
Bydgoszcz	9.02 ± 0.26	6.72 ± 0.26	1.34 ± 0.07	5.41 ± 0.16	2.32 ± 0.09
Osiek	12.52 ± 0.33	8.36 ± 0.25	1.50 ± 0.06	7.51 ± 0.20	2.88 ± 0.09
Kukawy	9.53 ± 0.49	5.36 ± 0.19	1.78 ± 0.11	5.72 ± 0.30	1.85 ± 0.07
Poniatowa	14.13 ± 0.62	5.52 ± 0.19	2.56 ± 0.14	8.48 ± 0.37	1.91 ± 0.07

concentrations. Interpolation map for ^{210}Po (Fig. 1) clearly shows that *M. procera* with the highest activity concentrations is located in northern Poland, close to Gdańsk agglomeration and Kartuzy, while the lowest near Olsztyn agglomeration in Warmia region. According to ^{210}Pb interpolation map (Fig. 2), this dispersion is more heterogeneous. Southern parts of sampled region have lower ^{210}Pb activity concentrations in *M. procera*, while northern parts are characterized with higher values. These differences might be connected with numerous factors. This region of Poland is mostly characterized by the same types of soils: brown earth on the north and podzols and lessive soil on the south. Differences in these soil properties could have major impact on ^{210}Po and ^{210}Pb mobility and bioavailability. It must be noted that almost all mushrooms were collected in forests that are usually semi-natural ecosystems, much different from agricultural cultivated lands. In forests, mushrooms and microbes biological activities effect on long-term radionuclides retention in organic layers of forest soil (Strumińska-Parulska et al. 2016). The levels of ^{210}Pb and ^{210}Po activity concentrations in top soil layers can be correlated with the amount of atmospheric precipitation (Persson and Holm 2011). In this case, top layers of the soil that are mostly dependent on ^{210}Po and ^{210}Pb activity concentrations in aerial deposition represent the major reservoir of radionuclides for mushrooms.

Values of ^{210}Po/^{210}Pb activity ratios in parasol mushroom

All mushrooms, except one, contained more ^{210}Po than ^{210}Pb. The values of ^{210}Po/^{210}Pb activity ratios ranged from 0.43 ± 0.05 for Olsztyn (cap) to 2.56 ± 0.14 for Poniatowa (whole mushroom). These results are comparable with previously obtained by Strumińska-Parulska et al. (2016). Observed differences could be a result of disparities in dry atmospheric fallout and its impact on topsoil including air dust but also mineral particles from dusty soils that can be moved by wind and cover the surface of plants and ground. These variations may be also connected with different ^{210}Po and ^{210}Pb bioavailability in soils.

Value of ^{210}Po/^{210}Pb activity ratio lower than 1 could suggest that wet and dry deposition was significant source of ^{210}Po and ^{210}Pb radionuclides in *M. procera* collected near Olsztyn. The value of ^{210}Po/^{210}Pb activity ratio in air deposition was calculated at 0.03–0.05 (Vaaramaa et al. 2010). Up to 80% of natural ^{210}Po and ^{210}Pb radioactivity in wild plants are connected to wet and dry deposition of ^{222}Rn decay products (Persson and Holm 2011). The residence time of tropospheric aerosols is varying with latitude and is shorter in low latitudes due to high amounts of precipitation. This leads to disequilibrium between ^{222}Rn, ^{210}Pb, ^{210}Bi, and ^{210}Po that affect mushrooms and vascular plants (Baskaran 2011). There are multiple factors affecting ^{210}Pb in air including the seasons, atmospheric pressure variations that affect the sources of air masses, local radon emanation rates, height of the atmospheric boundary layer, temperature inversions, diurnal and seasonal variations of meteorological parameters, frequency and amount of precipitation as well as soil moisture content and presence of snow cover which affect the ^{222}Rn emanation (Baskaran 2011). We also noticed that tree coverage over mushrooms might decrease the influence of wet and dry deposition on soils which results in indirect increment of the value of ^{210}Po/^{210}Pb activity in mushrooms closer to 1 (unpublished data).

A value higher than 1 could suggest, particularly in soil, that ^{210}Po was more mobile and thus more bioavailable for mushrooms when compared to ^{210}Pb. This aspect is also relevant for vascular plants (Olszewski et al. 2016). What is more, most mushrooms accumulate ^{210}Po preferentially over ^{210}Pb, and typical values of ^{210}Po/^{210}Pb activity ratios are in the range of 0.87 to 320 (Guillén and Baeza 2014; Gwynn et al. 2013; Vaaramaa et al. 2009). In general, ^{210}Po in soils may be of two origins: supported, from ^{238}U decay chain present in the soil; and unsupported, from precipitation of ^{222}Rn decay products (Persson and Holm 2011). There are numerous factors governing the mobility and bioavailability of metals from the geological bedrock to mushrooms. The influence of these factors on trace element uptake in macrofungi has been poorly investigated, and in case of soil pH and organic matter content, no correlation was found (Falandysz and Borovička 2013). In majority of soils, ^{210}Po is in equilibrium with its parent ^{210}Pb which implies that this radioisotope is the main source of ^{210}Po irreversibly adsorbed on clay and organic colloids (Persson and Holm 2011).

Annual effective radiation doses

In order to identify the potential radiotoxicity, on the basis of previously calculated ^{210}Po and ^{210}Pb content in dried fruiting bodies of parasol mushrooms (*M. procera*), the annual effective radiation doses were calculated (Tab. 1). The effective dose conversion coefficients from ^{210}Po and ^{210}Pb ingestion for adult members of the public recommended by ICRP are 1.2 and 0.69 μSv·Bq^{-1}, respectively (ICRP 2012). The average mushroom consumption in Poland was calculated at 0.5 kg dry biomass, but in some cases, consumption exceeds 1–1.5 kg dry biomass (Strumińska-Parulska et al. 2016). In case of analyzed *M. procera* samples, the consumption of 0.5 kg dry biomass caps and whole mushrooms could in the case of ^{210}Po lead to annual effective radiation dose from 2.03 ± 0.25 to 10.02 ± 0.20 μSv, while in the case of ^{210}Pb decay from 1.60 ± 0.04 to 4.63 ± 0.10 μSv. The total annual effective dose from natural radiation in Poland, including ^{222}Rn, was estimated at 2.1–2.6 mSv, while the average annual effective dose from ^{210}Po and ^{210}Pb intake with different types of food and water was estimated at 54 μSv for both radionuclides (Pietrzak-Flis et al. 1997). Thus, the calculated annual effective doses from analyzed mushrooms seem to be low when compared to other Polish food products (Skwarzec et al. 2001; Skwarzec et al. 2003; Skwarzec et al. 2004; Strumińska-Parulska 2015; Strumińska-Parulska et al. 2016; Strumińska-Parulska and Olszewski 2017).

Conclusions

The studies showed that edible wild parasol mushroom (*M. procera*) accumulated ^{210}Po and ^{210}Pb at different levels. Created interpolation maps showed different distribution of ^{210}Po and ^{210}Pb. Activity concentrations of these two radionuclides were dependent on many factors, i.e., soil properties and wet and dry air deposition. Values of ^{210}Po/^{210}Pb activity ratio for most of the analyzed samples were higher than 1, suggesting that ^{210}Po was preferentially accumulated by *M. procera*. Consumption of analyzed mushrooms (0.5 kg of dry mushrooms per year) in the case of ^{210}Po could lead the annual effective dose at 2.03–10.02 μSv, while in the case of ^{210}Pb at 1.60–4.63 μSv per year. This indicated that analyzed parasol mushrooms were safe from a radiological point of view.

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