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^{210}Po and ^{210}Pb in forest mushrooms of genus *Leccinum* and topsoil from northern Poland and its contribution to the radiation dose

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HIGHLIGHTS

- The study on ^{210}Po and ^{210}Pb in soil and mushrooms of genus *Leccinum* is presented.
- ^{210}Po and ^{210}Pb are important radiotoxic natural isotopes due to potential internal expose.
- The analyzed mushrooms are not hyperaccumulators in case of ^{210}Po and ^{210}Pb.
- The higher the bioconcentration factor (BCF) the lower the translocation factor (TF).
- Consumption of analyzed mushrooms is safe from radiological point of view.

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ABSTRACT

Wild growing mushrooms are traditional food items for man and also an important source of nutrients for small and big wildlife. Nevertheless, they can be species - specifically vulnerable for contamination with heavy metals and radionuclides. We studied a less known phenomenon of accumulation of highly toxic, the alpha-radiation emitter such as ^{210}Po and the beta emitter ^{210}Pb by three *Leccinum* mushrooms: orange oak bolete *L. aurantiacum* (Bull.) Gray (previous name *Leccinum aurantiacum* var. *quercinum* Pilát), foxy bolete *L. vulpinum* Watling and slate bolete *L. duriusculum* (Schulzer ex Kalchbr.) Singer. Fungal and soil materials were collected from areas of a different geochemical composition in the northern regions of Poland. In parallel evaluated was the risk to human consumer due to possible intake of ^{210}Po and ^{210}Pb with a mushroom meal. Results showed a heterogeneous distribution of ^{210}Po and ^{210}Pb activity concentrations within caps and stipes of fruiting bodies. Overall activity concentration for whole dried fungi material ranged from 0.59 ± 0.38 to 3.2 ± 0.2 Bq ^{210}Po kg^{-1} and from 0.45 ± 0.04 to 3.1 ± 0.2 Bq ^{210}Pb kg^{-1}. Evaluation showed that *Leccinum* mushrooms consumed by locals in typical quantity of 0.5 kg (dry biomass) can contribute into annual effective radiation dose at 0.90–3.81 μSv from ^{210}Po decay and 0.31–2.14 μSv from ^{210}Pb decay, which is a small portion of the annual effective radiation dose of ^{210}Po and ^{210}Pb for human inhabiting the northern regions of Poland.

1. Introduction

^{210}Po and ^{210}Pb are both of natural origin radionuclides and daughters of uranium ^{238}U that are interesting to investigate due to their radiotoxic features. Their half-lives are 138.38 days for ^{210}Po and 22.3 years for ^{210}Pb (Persson and Holm, 2011). ^{210}Po is the most stable natural alpha emitting isotope among Po isotopes and highly radiotoxic (dose conversion factor 1.2 μSv Bq^{-1}) with specific activity 166 TBq·g^{-1} (Holm, 2009; ICRP, 2012). ^{210}Pb is the beta-emitter, less radiotoxic to humans due to its longer half-life and decay type (dose conversion factor 0.69 μSv Bq^{-1}), but is very important contaminant as it decays (via ^{210}Bi) to ^{210}Po. Nevertheless, its chemical toxicity is the same as of stable lead isotopes (Heiserman, 1997; ICRP, 2012).

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Both ^{210}Po and ^{210}Pb can be observed in fluctuating concentrations in soil, sediment and water medias and constitute a significant part of the Earth's natural background radiation (Persson and Holm, 2011). Both ^{210}Po and ^{210}Pb occur in the atmosphere also, as an effect of continuous ^{222}Rn emission from the lithosphere – radon decays to its short-lived daughters, ^{210}Pb, ^{210}Bi, ^{210}Po, and they are continuously deposited in association with aerosols as dry fallout or washed out in rain (Persson and Holm, 2011; Strumińska-Parulska et al., 2010). Anthropogenic sources of these radionuclides are related to combustion of fossil fuels or use of phosphate fertilizers (Olszewski et al., 2015, 2016). At a regional scale, because of anthropogenic pollution (Technologically Enhanced Naturally Occurring Radioactive Material) or geology (100 μg of Po per 1 ton of uranium ore), exposure can be elevated (Heiserman, 1997; El Mrabet et al., 2005; Giri et al., 2013; Gwynn et al., 2013; Arunchalam et al., 2014; Persson, 2014).

The ubiquity of ^{210}Po and ^{210}Pb in the environment results in their food-chain transfer and occurrence in foodstuffs. Both nuclides are common trace-compounds in foods and this usually results in low dietary intake but their physical properties and radiotoxicity lead to significant contribution to the radiation dose of the population. Main route of ^{210}Po exposure for human is food (Hill, 1966), while ^{210}Pb is mainly inhaled (Holtzman, 1964). Among of all natural radionuclides ingested by human ^{210}Po is the most important and gives the highest radiation dose (Pietrzak-Flis et al., 1997; Persson and Holm, 2011; Strumińska-Parulska, 2015). ^{210}Po and ^{210}Pb content of food products highly depend on the geological, climatic and agricultural conditions of their geographic origin (Persson and Holm, 2011; Strumińska-Parulska and Olszewski, 2018).

Wild mushrooms are a highly biodiverse group of organisms – as a source of food for forest animals and human on one side and as creatures “involved in soil mineral weathering, organic substrate decomposition and element cycling” on the other (Falandysz and Treu, 2017). There are many data published about nuclides migration but mushrooms have not been considered in any Wildlife Transfer Database yet (Copplestone et al., 2013; Howard et al., 2013; Wood et al., 2013; Guillen et al., 2017). And there is still not much known about the uptake mechanisms of radionuclides (i.e. ^{137}Cs and ^{210}Po) by fungal mycelia and their bioconcentration in fruiting bodies (Gwynn et al., 2013). Accumulation and hyper-accumulation of toxic elements by mushrooms from soil might have consequences for health of human and animals, including accumulation in game animals which eat mushrooms. Contaminated food ingestion is the main route for internal exposure with anthropogenic radionuclides and i.e. wild boars (*Sus scrofa*) are well-known accumulators of radiocesium in their soft tissues and organs (Steinhauser et al., 2017). Wild boars feed on cesium-rich food, including mushrooms, and the impact of nuclear accidents on the ^{137}Cs activities could be found in their organisms (Steinhauser and Saey, 2016). However, wild boars (which forage for food below the soil surface) contained much more ^{137}Cs when compared to red or roe deer (Škrkal et al., 2015). The results on ^{210}Po determination in livers, kidneys and muscles of red deer, roe deer and fallow deer form northern Poland showed the differences in accumulation of ^{210}Po in tissues of various species were not significant as well as independent of age, gender and hunting place. Also the food preferences between all three species had no significant influence on excess in accumulation of ^{210}Po or other nuclides (Skwarzec and Prucnal, 2007; Skwarzec et al., 2010; Škrkal et al., 2015).

Circumstances considered as influencing bioavailability and bioconcentration of metallic elements by mushroom are or can be such as: pH and Eh of soil, co-occurrence of other inorganic ions and ligands at substantial quantity, clay content, organic matter content, cation exchange capacity, water regime, and climatic conditions due to water flow phenomenon (Gadd, 2007; Baptista et al., 2009; Falandysz and Borovička, 2013). Although the primary source of many inorganic compounds for fungi seem to be the substratum (e.g. decaying litter, organic or mineral layer of soil), the movement of water and migration of water soluble compounds through the soil horizon can also count (Falandysz et al., 2015a, 2015b). Still, elevated content of some metallic elements or radionuclides in forest topsoil usually results in higher accumulation in mushrooms (Kojta et al., 2012; Falandysz et al., 2018). Thus far most often studied was ^{137}Cs that contaminated Earth surface due to global radioactive fallout after use and testing of nuclear weapons and from nuclear power plant accidents but also natural terrestrial origin ^{40}K (Mietelski et al., 2010; Steinhauser et al., 2014; Falandysz et al., 2015a, 2017a; 2018; Zalewska et al., 2016; Chatterjee et al., 2017; Cocchi et al., 2017). Other radionuclides were studied at a lesser extent, i.e. ^{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, 2017).

Leccinum (Gray) is fungi genus that belongs to the family Boletaceae, has widespread distribution, generally found in the woodlands of Europe, Asia, and North America, and covers about 75 species (Škubla, 2007; Kirk et al., 2008; Falandysz et al., 2012, 2015b). *Leccinum* species form ectomycorrhizal associations with trees and most of these mushrooms consociate with trees of a single genus (Škubla, 2007). Ectomycorrhizal fungi may dissolve a variety of toxic metal-bearing minerals including phosphates (Gadd, 2007). All three analyzed mushroom species are edible and often collected. There exists some debate about the classification of *Leccinum quercinum* (Pilát) Green & Watling and *Leccinum aurantiacum* Bull.) Gray as separate species. Nevertheless, analyzed orange oak bolete *L. quercinum* as well as red aspen bolete *L. rufum* (Schaeff.) Kreisel are currently classified as *L. aurantiacum* (Index Fungorum, 2018). The mushroom is mycorrhizal most commonly with poplars and aspen and with oak trees. Its cap is usually greyish brown to orange brown, grows up to 25 cm in diameter, hymenium is cream-colored. The scales on the white long stem become dark brown with age (Garnweidner, 1994; Škubla, 2007). Foxy bolete (*L. vulpinum* Watling 1961) is also rare species, grows in association with pines and bearberries and resembles spruce bolete (*L. piceinum*) and orange birch bolete (*L. versipelle*). The cap can be from orange to purple-brown, grows 3–12 cm in diameter, hymenium is white or cream-colored. The stem is white and becomes darker with age (Wojewoda and Gumińska, 1988). Slate bolete (*L. duriusculum* Singer, 1947) grows under different aspen species, mainly Eurasian aspen (*Populus tremula*), and looks similar to birch bolete (*L. scabrum*) which grows under birches. The cap is 4–15 cm colored from gray-violet to red-brown with light gray tubes and the stem is white (Wojewoda and Gumińska, 1988; Škubla, 2007).

The aim of the study was to determine ^{210}Po and ^{210}Pb content in forest mushrooms of the genus *Leccinum*: *L. aurantiacum* (previously *L. quercinum*), *L. vulpinum* and *L. duriusculum*, and their soil substrate and fill the gaps in existing knowledge. On the basis of the obtained results this study allowed to check if ^{210}Po and ^{210}Pb are accumulated and uniformly distributed in analyzed forest mushrooms and verify if top soils affected their accumulation. Additionally, the possible annual effective radiation doses from analyzed ^{210}Po and ^{210}Pb to the public based on dietary statistics were calculated as well as the risk from analyzed radionuclides consumption with mushrooms was assessed.

2. Materials and methods

The *Leccinum* genus mushrooms species such as orange oak bolete (*L. aurantiacum* (Bull.) Gray), foxy bolete (*L. vulpinum* Watling) and slate bolete (*L. duriusculum* (Schulzer ex Kalchbr.)