^{137}Cs and ^{40}K in *Cortinarius caperatus* mushrooms (1996–2016) in Poland - Bioconcentration and estimated intake: ^{137}Cs in *Cortinarius* spp. from the Northern Hemisphere from 1974 to 2016

Jerzy Falandysz, Tamara Zalewska, Alwyn Fernandes

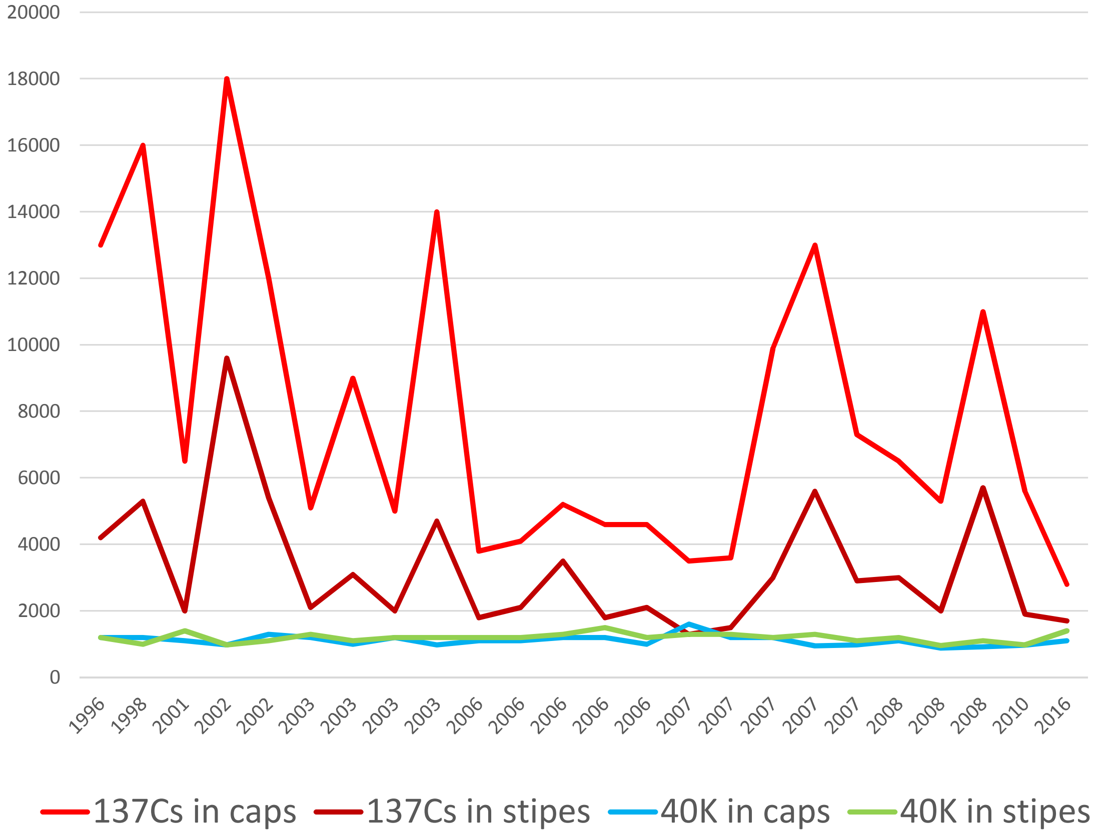
PII: S0269-7491(19)33913-2
DOI: https://doi.org/10.1016/j.envpol.2019.113208
Reference: ENPO 113208

To appear in: *Environmental Pollution*

Received Date: 17 July 2019
Revised Date: 6 September 2019
Accepted Date: 6 September 2019

Please cite this article as: Falandysz, J., Zalewska, T., Fernandes, A., ^{137}Cs and ^{40}K in *Cortinarius caperatus* mushrooms (1996–2016) in Poland - Bioconcentration and estimated intake: ^{137}Cs in *Cortinarius* spp. from the Northern Hemisphere from 1974 to 2016, *Environmental Pollution* (2019), doi: https://doi.org/10.1016/j.envpol.2019.113208.

C. caperatus - Hailuogou Glacier Forest Park, Minya Konka (Gongga Shan), Eastern Tibetan Plateau. *Photo JF*



^{137}Cs and ^{40}K in ***Cortinarius caperatus***
from the northern regions of Poland
(Bq kg^{-1} dry weight)

^{137}Cs and ^{40}K in *Cortinarius caperatus* mushrooms (1996 – 2016) in Poland - bioconcentration and estimated intake: ^{137}Cs in *Cortinarius* spp. from the Northern Hemisphere from 1974 – 2016

Jerzy Falandysz[1,2,3], Tamara Zalewska[4] and Alwyn Fernandes[5]

[1]University of Gdańsk, Environmental Chemistry & Ecotoxicology, 80-308 Gdańsk, Poland
[2]Environmental and Computational Chemistry Group, School of Pharmaceutical Sciences, Zaragocilla Campus, University of Cartagena, 130015 Cartagena, Colombia+
[3]Institute of Medicinal Plants, Yunnan Academy of Agricultural Sciences, Kunming 650200, China
[4]Institute of Meteorology and Water Management – Maritime Branch, National Research Institute, 42 Waszyngtona Av., 81-342 Gdynia, Poland
[5]School of Environmental Sciences, University of East Anglia, Norwich NR4 7TJ, UK

Correspondence: jerzy.falandysz@gmail.com (J. Falandysz); +visiting professor

Highlights

- Gypsy mushroom (*C. caperatus*) is an efficient fungal accumulator of radiocaesium
- Decades after Chernobyl accident *C. caperatus* could exceed radiocaesium safety limits
- Activity concentrations of *C. caperatus* fluctuate over time
- Recent examples of *C. caperatus* from hot-spots can show elevated ^{137}Cs levels
- Dietary intake of some Polish C. caperatus can provide relatively high radioactive dose

Abstract

Cortinarius caperatus grows in the northern regions of Europe, North America and Asia and is widely collected by mushroom foragers across Europe. This study shows that in the last three decades since the Chernobyl nuclear accident, *C. caperatus* collected across much of Northern Poland exhibited high activity concentrations of radiocaesium (^{137}Cs) - a long-lived radionuclide. The mushroom appears to efficiently bioconcentrate ^{137}Cs from contaminated soil substrata followed by sequestration into its morphological parts such as the cap and stipe which are used as food. The gradual leaching of ^{137}Cs into the lower strata of surface soils in exposed areas are likely to facilitate higher bioavailability to the mycelia of this species which penetrate to relatively greater depths and may account for the continuing high activity levels noticed in Polish samples (e.g. activity within caps in some locations was still at 11000 Bq kg^{-1} dw in 2008 relative to a peak of 18000 in 2002). The associated dietary intake levels of ^{137}Cs have often exceeded the tolerance limits set by the European Union (370 and 600 Bq kg^{-1} ww for children and adults respectively) during the years 1996 to 2010. Human dietary exposure to ^{137}Cs is influenced by the method of food preparation and may be mitigated by blanching followed by disposal of the water, rather than direct consumption after stir-frying or stewing. It may be prudent to provide precautionary advice and monitor activity levels, as this mushroom continues to be foraged by casual as well as experienced mushroom hunters.

Key words: exposure, food, fungi, forest, radiocaesium, radiotoxicity, soil

Introduction

Surface vegetation and fungi, within the humified layer of soil are part of the forest ecosystem that is susceptible to pollution with caesium (^{134}/^{137}Cs – half-life 2.1/30 yrs respectively) particularly from radioactive fallout after nuclear events (Römmelt et al., 1990; Suchara, 2017). Mushrooms, the fruiting bodies of fungi, exhibit a remarkable aptitude and propensity to bioconcentrate a variety of metallic elements and metalloids, both of a beneficial (Cu, K, Se, Zn) or harmful nature (As, Ag, Cd, Hg, Pb, radiocaesium) (Cocchi et al., 2017; Frankowska et al., 2010; Giannaccini et al., 2012; Ingrao et al., 1992; Jarzyńska et al., 2012a, 2012b; Komorowicz et al., 2019; Melgar et al., 1998; Vukojević et al., 2019). This tendency arises from genetic features of the species which include a wealth of transport genes and binding ligands which act as drivers of metallic element accumulation on the one hand combined with site-specific features related to soil geochemistry, biology and pollution on the other. Mushrooms that grow in soil contaminated due to mining or processing of metal ores, metallurgy, metal waste management, chemical waste disposal, nuclear explosions and nuclear accidents are often considerably contaminated with specific metal or metalloid elements, e.g. arsenic (As), cadmium (Cd), lead (Pb), mercury (Hg), nickel (Ni), silver (Ag) or radionuclides (^{134}Cs, ^{137}Cs) (Barcan et al., 1988; Borovička et al., 2014; Falandysz, 2016, 2017; Kojta et al., 2012; Komárek et al., 2007; Larsen et al., 1998; Mleczek et al., 2016; Petkovšek and Pokorny, 2013; Steinhauser et al., 2014). Similarly, those that grow in soils with a naturally high polymetallic background from regions with geochemical anomalies can display high levels of Hg and As, e.g. affected mushrooms in the mercuriferous belt in the Yunnan province of China (Falandysz and Rizal, 2016; Falandysz et al., 2014, 2015a, 2015b; Kojta et al., 2015), and those susceptible to cinnabar deposition from sites and mining activity in Europe in the Middle Spiš in Slovakia, Monte Amiata in Italy or the Idrija area in Slovenia (Árvay et al., 2014; Bargagli and Baldi 1984; Vogel-Mikuš et al., 2016).

Among monovalent alkali elements, stable caesium (^{133}Cs), lithium (Li) and sodium (Na) occur as minor constituents in mushrooms compared to the high proportions of potassium (K) with lesser amounts of rubidium (Rb) (Falandysz and Borovička, 2013). The edible Gypsy mushroom, *Cortinarius caperatus* (Pers.) Fr., shows a high propensity to absorb radioactive caesium ($^{134/137}$Cs) from soil (Haselwandter et al., 1988). The contents of K, Rb, Na and Cs (^{133}Cs) in *C. caperatus* (Pers.) Fr., collected from Precambrian shales in the Middle Bohemia region (background area without metallic ores) were respectively 45000 ± 1400 mg kg^{-1} dry biomass (db), 243 ± 8 mg kg^{-1} db, 720 ± 25 mg kg^{-1} db and 8.4 mg kg^{-1} db (Řanda and Kučera, 2004). K, Rb and Na in *C. caperatus* (n = 3) sampled in a Norwegian mountain area in 1988 occurred at concentrations of 55000, 173 and 102 mg kg^{-1} db, respectively, with total Cs at 3.6 ± 1.6 mg kg^{-1} db (including ^{134}/^{137}Cs at activity concentration of 282000 ± 162000 Bq kg^{-1} db) (Bakken and Olsen, 1990b).

The content of ^{133}Cs in the fruiting bodies of fungi is positively correlated to the value of the bioconcentration factor/transfer factor (BCF/TF) for radiocaesium in mushrooms from the soil substrata (Olsen, 1994). It is therefore unsurprising that the level of radiocaesium activity correlates well with ^{133}Cs at equilibrium state in mushrooms (Ismail, 1994; Karadeniz and Yaprak, 2007; Rühm et al., 1999; Yoshida et al., 2004). Nevertheless, due to the high biodiversity within mushrooms there is an insufficient quantity of good quality data on this topic.

The activity concentration of ^{137}Cs is often also positively correlated with Rb content (Vinichuk et al., 2011) but not with K (^{40}K), Na and Li or P (Bakken and Olsen, 1990b; Bem et al., 1990; Falandysz et al., 2019a; Ismail et al. 1995; Karadeniz and Yaprak, 2010; Strandberg, 1994; Vinichuk et al., 2011). Nyholm and Tyler (2000) noted that: “low K status (pool of exchangeable K) in the soil, usually aggravated by high soil acidity which causes K leaching losses, is compensated by increased uptake of Rb by plants and fungi". Nevertheless, any dependence on uptake or a substantial relationship between ^{133}Cs and $^{134/137}$Cs on the one hand and Rb and K on the other has not been studied so far in mushrooms.

C. caperatus is a prized edible species with a fruiting body of appreciable size. The stipe height is typically from 6 to 12 (15 cm) with a thickness ranging from 1 to 2 (3 cm) over most of the length, widening towards the base, and a cap of up to 12 cm in diameter. Its occurrence is widespread in the northern regions of Europe and it also occurs locally in other regions of Europe, North America and Asia (section 3.3.). Its flesh has a mild smell (Laessoe et al., 1996) and nutty flavor when cooked.

The culinary treatment and preservation of mushrooms including *C. caperatus* vary, depending on the strain, abundance, region, local culinary tradition or the specific needs of collectors or growers. *C. caperatus* can be prepared for consumption as fried, stewed or pickled (caps with a short part of the stipe prepared from young or button stage fruiting bodies are especially preferred). Blanching (parboiling) with high excess of water and blanching & pickling can significantly decrease the content of radiocaesium in mushrooms prepared for consumption due to high predilection and leakage of the element into the water phase, and hence reduce exposure. However, blanching, in common with stir-frying, frying or stewing, causes, among other effects, a partial dehydration and shrinkage of the prepared mushrooms leading to an apparent increase in the elemental content of the cooked produce when expressed on a whole (wet) weight basis (relative to the uncooked mushroom weight). This can result in an apparent increase in the intake of a compound or radionuclide (Falandysz et al., 2019b, 2019c, 2019d, 2019f), relative to calculations/estimations that are based on dry weight results for both uncooked and cooked mushrooms and can lead to misinterpretation. Stewing or frying at higher temperatures lead to denaturation, hydrolysis and dehydration, and can cause partial leaching, but not loss of the radiocaesium (and other metallic elements) content which shows an apparent increase when results are expressed on a whole (wet) weight basis for substrate (fresh mushrooms) and product (stewed or fried mushrooms). During frying there can be partial leakage of minerals into the oil residue (this is sometimes recovered and used as a sauce and consumed, or withdrawn), but at the same time due to the high temperature of the oil (around 160 °C during stir-frying in deep oil), partial dehydration also occurs, resulting in enhancement of the metallic and metalloid element content, including radiocaesium (Falandysz et al., 2019b-e). The process of stewing (often with other vegetables, spices and a handful of butter or vegetable oil), in a covered pot, results in almost all minerals remaining with the dish. Hence, due to the high bioaccumulation potential for radiocaesium and regardless of the kind of culinary processing, *C. caperatus* collected from areas experiencing radioactive fallout can be a particularly risky source of exposure to consumers. In this study the activity concentration and bioconcentration of ^{137}Cs and ^{40}K in *C. caperatus* mushrooms collected from soil substrata (0 – 10 cm layer) sampled from twenty two locations in the northern part of Poland in 1996 – 2016 was evaluated. Existing data on the radiocaesium contamination in mushrooms (sixty one species) of the genus *Cortinarius* from locations in Europe, Japan and China during 1974 – 2016 have also been collated and discussed.

2. Materials and methods

2.1. Mushrooms and soil substrate

Samples of *C. caperatus* matured fruiting bodies (sporocarps) and generally, the underlying soil substrata (0 - 10 cm upper layer) of humified and mineral soil (ca. 100 g) were collected from 22 locations in 20 geographically discrete, distributed forested areas in the northern part of Poland in 1996 – 2016 (Fig. 1). Between 8 and 70 individual fruiting bodies were collected per sampling location. The fresh fruiting bodies were routinely cleaned to remove any visible plant vegetation and soil debris at the site using a plastic knife, and the bottom part of the stipe was cut-off. The cleaned samples were air-dried for a few days. With the exception of two sets from sites for which the whole fruiting bodies were examined, each individual fruiting body was then separated into cap (with skin) and stipe, and dried at 85 °C to constant mass. The dried fungal materials were pulverized in a porcelain mortar and kept in screw sealed plastic (low density polyethylene) bags under dry conditions.

In parallel with the mushrooms from 17 locations, samples of corresponding topsoil layer (0–10 cm) of humified and mineral forest soil were collected from directly beneath the fruiting bodies, and were pooled for each site (ca. 100 g of dried soil within a pool). After the removal of any small stones, sticks and leaves, the pooled samples were air dried at room temperature (constant 18 - 22 °C) for several weeks under clean (preventing from dust) and dry conditions. When constant air-dried weight was reached, the soil samples were graded through a plastic sieve of 2 mm pore size, into sealed polyethylene bags and kept under dry and clean conditions in the laboratory sample store.

2.2. Analysis

^{137}Cs and ^{40}K contents were determined using a gamma spectrometer with a coaxial HPGe detector with a relative efficiency of 18 % and a resolution of 1.9 keV at 1.332 MeV. All measurements of the fungal materials were preceded by a background measurement (time 80,000 s or 250,000 s), and background counts were subtracted (the GENIE 2000 program). The equipment was calibrated using a multi-isotope standard and the method was fully validated. The reference solution 'Standard solution of gamma emitting isotopes, code BW/Z-62/27/07' produced at the IBJ-Świerk near Otwock in Poland was used for preparing reference samples for equipment calibration. The same geometry of cylindrical dishes with 40 mm diameter (as applied for environmental samples) was used for reference samples during equipment calibration. Data obtained were recalculated for dehydrated materials (dried at 105 °C) and results were decay corrected back to the time of mushrooms and soil sample collection (Falandysz et al., 2015a, 2017; Zalewska and Saniewski, 2011). Concentrations of stable K were calculated from the ^{40}K data (Table 1).

3. Results and discussion

3.1. ^{137}Cs and ^{40}K in mushrooms

As in other locations in northern Europe, *C. caperatus* that grows in the northern regions of Poland is found in coniferous (spruce and pine) and beech woods on poor and acidic sandy soils, both humid and dry. Seasonally, in Poland, it can proliferate in forests in the late summer and autumn. Caps in this study showed higher concentrations of ^{137}Cs than stipes, the median value of the quotient between level of the activity concentration in caps and stipes (index $Q_{C/S}$) for all locations was 2.4 (mean: 2.4 ± 0.5), and minimum and maximum values were in the range 1.5 to 3.3 (Table 1).

As reported in Table 1, the determined activity concentrations of ^{137}C in Bq per kilogram of dry weight (dw) in pooled samples and according to the period and place of collection were in the range of 1500 ± 16 to 9600 ± 71 Bq kg^{-1} dw in stipes to 2800 ± 52 to 18000 ± 140 Bq kg^{-1} dw in caps and fluctuated to some degree depending on region (Fig. 2). Mushrooms from a few sites in the Warmia region and from one site from the Mazovia

(Mazowsze) region all showed relatively high ^{137}Cs activity (Table 1). Nevertheless, a high degree of contamination was also noted in *C. caperatus* at a site in the Pomerania region. The activity concentration of ^{137}Cs in the pooled sample of the whole fruiting bodies collected in 2010 was 3700 ± 30 Bq kg^{-1} dw.

C. caperatus is considered to have a high potential to accumulate ^{137}Cs (Bakken and Olsen, 1990a; Strandberg, 2004). Indeed, the activity concentrations of ^{137}Cs in caps of this species collected over 22 to 24 years after the Chernobyl accident and examined in this study were as high as 11000 ± 82 Bq kg^{-1} dw in the Orzechowo place (Warmia region) and 5600 ± 58 Bq kg^{-1} dw in the Śliwice place (central area of the Tuchola Pinewoods) (Table 1). The substantial variability in the activity concentrations of ^{137}Cs within the caps and stipes of *C. caperatus* observed in fruiting bodies seem to reflect location-specific differences in the degree of soil pollution (Figs. 2 and 3). For example, activity in caps at four locations in 2003 was in the range 5000 ± 49 to 14000 ± 110 Bq kg^{-1} dw, and in 2007 at five other locations was in the range 3500 ± 38 to 13000 ± 100 Bq kg^{-1} dw (Table 1).

Edible mushrooms with greater gastronomic value such as *Imleria badia* (Fr.) Fr. (previous name *Xerocomus badius*), and others such as *Boletus edulis* Bull., *Boletus pinophilus* Pilát & Dermek, *Boletus reticulatus* Schaeff., *Leccinum scabrum* (Bull.) Gray or *Cantharellus cibarius* Fr. are also considered as good accumulators of ^{137}Cs. During the last four decades these mushrooms that are foraged in the same regions as those in this study and elsewhere in Poland have been found to be clearly less contaminated with radiocaesium than the *C. caperatus* in this study (Bem et al., 1990; Falandysz et al., 2015a, 2016; Malinowska et al., 2006). An exception could be mushrooms foraged from the hot spot area in the southwestern region of Poland (cumulative deposition of ^{137}Cs was 64 ± 2 kBq m^{2} - as calculated in autumn 2006) and described by Mietelski et al. (2010; see also Fig. 3). Considerable contamination with radiocaesium could also be expected in mushrooms foraged from the region of Śnieżnik in the Sudety Mountains in the south-west of Poland (Fig. 3). *B. edulis* collected in 1998 from the Kłocka Dale in Sudety Mountains showed concentration level of ^{137}Cs at 5722 ± 5 Bq kg^{-1} db (Falandysz et al., 2015a), but there is no data available for other species from this location.

The degree of maturity of the fruiting body is a possible variable that could influence the content of accumulated ^{137}Cs as well as certain other metallic and metalloid elements in mushrooms, as seen for example, in *Amanita muscaria* (L.) Lam.) (Falandysz et al., 2019a, 2019g), but there are no similar observations that are specific to *C. caperatus* from this study Clearly, the characteristics of accumulation of ^{137}Cs and ^{40}K by *C. caperatus* vary, regardless of location and year of collection, or morphological parts such as caps and stipes (Table 1, Fig. 2).

Unlike ^{137}Cs, ^{40}K activity concentrations (and hence also total K content) were more uniform across the range of sampling locations and over time, i.e. the median and mean values in stipes was 1200 Bq kg^{-1} dw and 1200 ± 140 Bq kg^{-1} dw respectively (range 960 ± 130 to 1500 ± 150 Bq kg^{-1} dw) (see also Fig. 2). The corresponding values for caps were 1100 Bq kg^{-1} dw and 1100 ± 160 Bq kg^{-1} dw respectively (range 880 ± 140 to 1600 ± 130 Bq kg^{-1} dw) (Table 1). Relative to ^{137}Cs however, the morphological distribution was different in the fruiting bodies. The stipes were usually richer in potassium compared to the caps. The median value of the quotient ($Q_{C/S}$) for ^{40}K was 0.92 (mean: 0.95 ± 1.2 and range 0.73 to 1.2) (Table 1).

Potassium generally occurs to a high level and is an important functional metal in the flesh of mushrooms. It is essential as an intracellular cation for osmotic regulation of water content and as a co-factor in enzymes. The reported potassium content in a large set of fruiting bodies of the mycorrhizal (mutualistic) mushrooms such as King Bolete (*Boletus edulis*) and Fly Agaric (*Amanita muscaria*) was in the range 20000 to 38000 mg kg^{-1} db (median values) and 37000 to 45000 mg kg^{-1} db (Drewnowska et al., 2013; Falandysz et al., 2007b; Lipka and Falandysz, 2017). Saprotroph mushrooms are also rich in potassium, e.g. in caps of Parasol Mushroom (*Macrolepiota procera*) potassium occurs in the range 33000 to 46000 mg kg^{-1} db (Gucia et al., 2012; Jarzyńska et al., 2011; Kojta et al., 2011 and 2016; Kułdo et al., 2014) with higher concentrations in the fruitbodies of *Coprinus micaceus*, i.e. from 99000 to 135000 mg kg^{-1} db (Tyler, 1980), 79500 (74500 to 87000) mg kg^{-1} db (Seeger, 1978) and 69000 ± 2400 mg kg^{-1} db – all values rounded (Vetter, 1994). On the other hand, mycorrhizal species whose mycelia extract nutrition from wood, e.g. *Auricularia auricula-judae* (Bull.) Quél have considerably lower potassium levels than saprotrophs (4300 mg K kg^{-1} db) (Vetter, 1994).

The value of $Q_{C/S}$ for ^{40}K in fruiting bodies of many species of mushrooms is usually above 1.0 (Falandysz et al., 2017 and 2018). In a recent study of *A. muscaria*, the caps to stipe quotients ($Q_{C/S}$) of ^{40}K decreased with increasing development of the mushroom, i.e. from 1.5 (1.4 to 1.6) in the button stage and young individuals (n = 97) down to 1.0 (0.62 to 1.2) in the older and mature specimens (n = 144) (Falandysz et al., 2019a). However, based on the results for one species in one location, it would be difficult to generalize on the influence of the development stage of fruiting bodies on the $Q_{C/S}$ values for other species, as there is no other reported data on this parameter.

3.2. ^{137}Cs and ^{40}K in the soil beneath fruiting bodies and bio-concentration

^{137}Cs activity concentrations in the forest topsoil (mixed layers including organic and mineral layer) samples showed values in the range from 10 ± 1 (Lębork site, Pomerania, 2007) to 70 ± 3 Bq kg^{-1} dm (Strzebielino, Pomerania, 2006). Soils sampled from other locations during 1996 to 2008 contained ^{137}Cs at level 33±1 to 41±2 Bq kg^{-1} dm (Table 1). This level of ^{137}Cs activity is similar to that reported earlier (Malinowska et al., 2006) for soils sampled below the fruiting bodies of *I. badia*, that were collected from sites in the northern and north-eastern regions of Poland in 1996-1998. ^{137}Cs activity concentrations were in the range of 34 ± 3 to 100 ± 4 (total 11 to 260) Bq kg^{-1} dm, with areas in the north-eastern region showing higher pollution.

Bioconcentration factors (BCFs) and aggregated transfer factors (TFs) are generally calculated to understand the potential of some species including mushrooms, to accumulate chemical elements contained in the soils or substrates in which they grow. BCFs and TFs are ratios of radionuclide specific activity in mushrooms to the activity concentration in the underlying soil (0-10 cm layer) or the surface activity of the soil (m^2 kg^{-1}) respectively. It is evident that the BCF estimate is highly specific because it relates to the soil collected directly beneath the sampled fruiting body of the mushroom while the distribution of radiocaesium in the surrounding surface soils can be more heterogeneous.

The variability of ^{137}Cs pollution of Polish soil resulting from the Chernobyl accident is evident from geospatial imagery (Fig. 3) which presents a slightly mosaic-like distribution highlighting areas of higher concentration. This detailed picture differs form more generalized images (Betti et al., 2016). Fig. 3 can be particularly useful in identifying possible “hot spot” forested areas, where mushrooms can show site-specific levels of high pollution, both for one-off sampling or for trends from longer term sampling.

An earlier study (Borio et al., 1991) showed no reliable correlation between the radiocaesium activity concentration of mushrooms and the underlying soil. However, mushrooms collected from forested areas (Falandysz and Borovička, 2013; Falandysz et al., 2008; Mietelski et al., 2010) that corresponded to “hot spot” visualized in Fig. 3, with soils showing substantially elevated ^{137}Cs levels (due to the Chernobyl fallout), appear to be more contaminated with ^{137}Cs. Moreover the elevation in ^{137}Cs levels for these areas seems to be regardless of the species of mushroom under study.

The relationship between substrate areas of mycelial proliferation and growth (either within layers or within the soil horizon), the severity of the radioactive fallout, speed of ^{137}Cs infiltration to deeper layers and the nuclide activity concentration accumulated by mushrooms can be effectively illustrated using the example of the amethyst deceiver mushroom (*Laccaria amethystina* Cooke), that feeds on decaying litter. Immediately after the Chernobyl accident, high levels of ^{137}Cs and ^{134}Cs were found in *L. amethystina,* which is known to accumulate activity through surface mycelia that absorb the easily available radiocaesium from fresh fall-out (Stijve and Poretti, 1990). This is because forest topsoil rich in organic matter (humus) can adsorb and retain a large portion of radiocaesium from airborne deposition (Lehto et al., 2013).

According to a number of studies, the infiltration of airborne ^{137}Cs from the surface to wider soil horizons (or deeper layers) is a slow process (Mietelski et al., 2010; Niesiobędzka, 2000). The soil layer with the highest density of mycelia and the extent of depth and space to which the hyphae penetrate, largely depends on the type of mushroom. Therefore, bulk (0-10 cm layer) sampling of soils can only give a general idea of ^{137}Cs contamination and its availability from soils. To better assess the efficiency of ^{137}Cs uptake and sequestration by *C. caperatus,* a more detailed study would be required to identify the specific soil layers (e.g.: 0-1, 1-2, 2-3 cm etc.) that correlate with the highest density of *C. caperatus* mycelia and the ^{137}Cs content.

For ectomycorrhizal fungi, the penetration zone of hyphae is likely to be species-dependent and if the soil profile is favorable they can follow the roots to deeper levels. For example, the saprobic *Agaricus bernardii* hyphae lives at least down to a depth of 30 cm (Borovička et al., 2010). Ingrao et al. (1992) noted, that one of difficulties in estimating the bio-concentration potential of mushrooms to accumulate metallic and metalloid elements and their suitability or not, as indicative species in environmental (soil) pollution monitoring, is that hyphae can live down in soil to depths of 50 cm. On other occasions, mushrooms can be suitable in prospecting of the metal/metalloid resources and geochemical anomalies (Borovička et al., 2010; Falandysz et al., 2015a).

Relative to K, both ^{137}Cs and ^{133}Cs (stable) only occur at trace concentrations in *C. caperatus*, like other metallic and metalloid elements in most species of macromycetes (Falandysz and Borovička, 2013). The relatively high levels of ^{137}Cs that accumulated in many mushroom species shortly after the Chernobyl accident have been interpreted as possibly being due to its "better availability/accessibility" when compared to stable ^{133}Cs, and a possible role (direct or indirect) of K in terms of absorption pathway. The latter influence (of K availability) can in turn dependent on multiple factors such as the chemical state in which K exists in the associated soil, its bio-availability in soil and other competing alkali ions during homeostasis.

As the mycelia of *C. caperatus* penetrate to deep levels in the soil, the samples of soils collected from 0-10 cm layer can give only a general idea of ^{137}Cs pollution about the soil horizons and its availability from forest soils. ^{137}Cs has been found to have penetrated into deeper soil layers, and radiocesium levels in this particular mushroom have long been on the rise because it is increasingly available to the mycelium at lower depths. Ismail et al. (1995) observed that activity concentration of ^{137}Cs in *C. caperatus* increased by around 20 percent each year between 1991 and 1993 (Table S1). The same was observed by Daillant et al. (2013) in *C. caperatus* sampled in 1992, 1993 and 1995, while levels dropped in 1998 and 2011 (Table S1).

3.3. Risk of ^{137}Cs intake from *C. caperatus*

The maximum and minimum values of ^{137}Cs activity concentration for caps of *C. caperatus* in this study in the years 1996 – 2016 were 2800 and 18000 Bq mg kg^{-1} db (280 and 1800 Bq mg kg^{-1} on a fresh biomass basis - assuming a moisture content of 90%). In view of the food tolerance limits for radiocaesium that are 370 and 600 Bq kg^{-1} whole (fresh) weight within the European Union for children and adults respectively, the *R. caperatus* collected in this study (in practice the caps with a piece of the uppermost part of stipes are used as food) (Table 1) often exceeded these limits in the years 1996 to 2010. Thus, while this species may be avoided by knowledgeable and informed mushroom hunters (mushroomers), the precaution may not extend to casual or opportunistic foragers. Due to their fresher appearance, it is possible that mushroomers preferentially collect young fruiting bodies, which can be more contaminated with ^{137}Cs than more mature examples as has been observed recently for *Amanita muscaria* (Falandysz et al., 2019a).

The mode of preparation of a mushroom dish can significantly influence the content of the metallic elements including ^{137}Cs (Drewnowska et al., 2017; Falandysz et al., 2019b-f; Shutov et al., 1996). Blanching for 10 min or longer in boiling water (often slightly salty) leaches radioactivity into the water and can remove from around 20 to 90% - (based on dry biomass data) of the radiocaesium from the mushrooms, although there is no original data that is specific to *C. caperatus*, (or knowledge of such estimations when based on whole (wet) weight basis). On the other hand, stir-frying or stewing is much less efficient at causing leaching and removal of metallic and metalloid elements or radiocaesium from mushroom meals than blanching (Falandysz et al., 2019b-f; Shutov et al., 1996).

Conclusions

The mushroom *Cortinarius caperatus* has been seen to efficiently bioconcentrate ^{137}Cs that is contained in the soil substratum in which its mycelia live. The mushroom sequesters this element in substantial amounts in its morphological parts such as the cap and stipe which can be foraged and used as food.

The mechanistic pathways that lead to a higher pollutant loading over time, in this species is not fully confirmed, but it is thought that the gradual leaching of ^{137}Cs from the Chernobyl fallout, into lower strata of surface soil and decay combined with the relatively greater depths to which the mycelia of this species penetrate, may account for the higher levels of activity noticed in the samples collected in Poland.

Intakes of radiocaesium arising from the consumption of *C. caperatus* collected in this study (and based on data collated from literature) have often exceeded the tolerance limits set by the European Union for radiocaesium (370 and 600 Bq kg^{-1} ww for children and adults respectively) during the years 1996 to 2010. Human dietary exposure to ^{137}Cs is influenced by the method of food preparation and may be mitigated by blanching mushrooms for 10 min or longer in boiling water (followed by disposal of the water), rather than direct consumption after stir-frying or stewing.

As this mushroom continues to be foraged by casual as well as experienced mushroom hunters, it would be prudent to monitor levels of ^{137}Cs in this species and mushrooms within this region, in general, and provide precautionary advice depending on the findings.

Acknowledgements

Technical assistance by Katarzyna Bargłowska, Ada Borkowska, Ilona Bochentin, Karolina Czapiewska, Anna Dryżałowska, Dorota Górska, Aleksandra Jabłońska, Grażyna (Jarzyńska) Krasińska), Anna K. Kojta, Arleta Naczk, Daniela Orzłowska, Damroka

Reglińska, Daniel Siwicki, Aleksandra Stefańska, Dorota Stenka, Monika Szlosowska, Malwina Szczypińska and Joanna Szubstarska is acknowledged.

Disclaimer

The authors assert no conflict of interest.

Supplementary information

Table s1.

References

Amundsen I, Gulden G, Strand P (1996) Accumulation and long term behaviour of radiocaesium in Norwegian fungi. Sci Total Environ 184:163-171

Árvay J, Tomáša J, Hauptvogl M, Kopernická M, Kováčik A, Bajčan D, Massányi P (2014) Contamination of wild-grown edible mushrooms by heavy metals in a former mercury-mining area. J Environ Sci Health Part B 49:815-827

Bakken LR, Olsen RA (1990a) Accumulation of radiocaesium in fruit bodies of fungi, pp. 664-668. *In* Desmet G, Nassimbeni P, Belli M (eds.); 707 p; ISBN 1-85166-539-0; Worldcat; 1990; p. 664-668; Elsevier Applied Science; London (UK); Workshop on the transfer of radionuclides in natural and semi-natural environments; Udine (Italy); 11-15 Sep 1989

Bakken LR, Olsen RA (1990b) Accumulation of radiocaesium in fungi. Can J Microbiol 36:704-710

Barcan VS, Kovnatsky EF, Smetannikova MS (1988) Absorption of heavy metals in wild berries and edible mushrooms in an area affected by smelter emissions. Water Air Soil Poll 103:173-195

Bargagli R, Baldi F (1984) Mercury and methylmercury in higher fungi and their relation with the substrata in a cinnabar mining area. Chemosphere 13:1059–1071

Bem H, Lasota W, Kuśmierek E, Witusik M (1990) Accumulation of ^{137}Cs by mushrooms from Rogoźno area of Poland over the period 1984-1988. J Radioanal Nucl Chem Lett 145:39-46

Betti L, Palego L, Lucacchini A, Giannaccini G (2017)^{137}Caesium in samples of wild-grown *Boletus edulis* Bull. from Lucca province (Tuscany, Italy) and other Italian and European geographical areas. Food Addit Contam A 34:49-55

Borio R, Chiocchini S, Cicioni R, Degli Esposti P, Rongoni A, Sabatini P, Scampoli P, Antonini A, Salvadc P (1990) Uptake of radiocesium by mushrooms. Sci Total Environ 106: 183-190

Borovička J, Mihaljevič M, Gryndler M, Kubarová J, Žigarová A, Hršelova H, Řanda Z (2014) Lead isotopic signatures of saprotrophic macrofungi of various origins: Tracing for lead sources and possible applications in geomycology. Appl Geochem 43:114-120

Borovička J, Kotrba P, Gryndler M, Mihaljevič M, Řanda Z, Rohovec J, Cajthaml T, Stijve T, Dunn CE. Bioaccumulation of silver in ectomycorrhizal and saprobic macrofungi from pristine and polluted areas. Sci Total Environ 2010:408, 2733-2744.

Butkus D, Dimavičienė D (2009) ^{137}Cs pernašos sistemoje „dirvožemis – grybai – žmogus“ tyrimas. J Environ Eng Landscape Manag 17:44-50

Byrne AR (1988) Radioactivity in fungi in Slovenia, Yugoslavia, following the Chernobyl accident. J Environ Radioact 6:177-183

Cocchi L, Kluza K, Zalewska T, Apanel A, Falandysz J (2017) Radioactive caesium (^{134}Cs and ^{137}Cs) in mushrooms of the genus *Boletus* from the Reggio Emilia in Italy and Pomerania in Poland. Isotopes Environ Health Stud 53:620–627

Consiglio G, Gattavecchia E, Tonelli D, Cocchi L (1990) La radioattivitá nei finghi: attualitá del problema ed opportunitá di un approfondimento. Rivista di nucologia : bollettino dellAssociazione micologica Bresadola 33:227-231

Čadová M, Havránková R, Havránek J, Zölzer F (2017) Radioactivity in mushrooms from selected locations in the Bohemian Forest, Czech Republic. Radiat Environ Biophys 56:167-175

Čipáková A (2005) Migration of radiocaesium in individual parts of the environment. Nukleonika 50:S19−S23

Daillant O, Boilley D, Josset M, Hettwig B, Fischer HW (2013) Evolution of radiocaesium contamination in mushrooms and influence of treatment after collection. J Radioanal Nucl Chem 297:437–441

Dietl G, Breitig D (1988) Radioactive cesium in mushrooms from Schwaebisch Gmuend area, F.R.G. Northeastern Baden-Wuertemberg (*In German*). Z Mykol 54:109-112

Drewnowska M, Lipka K, Jarzyńska G, Danisiewicz-Czupryńska D, Falandysz J (2013) Investigation on metallic elements of Fly Agaric, *Amanita muscaria*, fungus and the forest soils from the Mazurian Lakes District of Poland. Fresenius Environ Bull 22:455-460

Eckl P, Hofmann W, Türk R (1986) Uptake of natural and man-made radionuclides by lichens and mushrooms. Radiat Environ Biophys 25:43-54

Elstner EF, Fink R, Höll W, Lengfelder E, Ziegler H (1987) Natural and Chernobyl-caused radioactivity in mushrooms, mosses and soil-samples of defined biotops in SW Bavaria. Oecologia 73:553-558

Elstner EF, Fink R, Höll W, Lengfelder E, Ziegler H (1989) Radioactivity in mushrooms, mosses and soil samples of defined biotops in SW Bavaria- two years after "Chernobyl". Oecologia 80:173-177

Falandysz J (2014) Distribution of mercury in Gypsy *Cortinarius caperatus* mushrooms from several populations: an efficient accumulator species and estimated intake of element. Ecotoxicol Environ Saf 110:68-72

Falandysz J (2016) Mercury bio-extraction by fungus *Coprinus comatus*: a possible bioindicator and mycoremediator of polluted soils. Environ Sci Pollut Res 23:7444-7451

Falandysz J (2017) Mercury accumulation of three Lactarius mushroom species. Food Chem 214:96-101

Falandysz J, Borovička J (2013) Macro and trace mineral constituents and radionuclides in mushrooms: health benefits and risks. Appl Microbiol Biotechnol 97:477-501

Falandysz J, Rizal LM (2016) Arsenic and its compounds in mushrooms: a review. J. Environ. Sci. Health Part C 34:217-232

Falandysz J, Kunito T, Kubota R, Lipka K, Mazur A, Falandysz JJ, Tanabe S (2007b) Selected elements in Fly Agaric *Amanita muscaria*. J Environ Sci Health A 42:1615-1623

Falandysz J, Kunito T, Kubota R, Bielawski L, Frankowska A, Falandysz J, Tanabe S (2008) Multivariate characterization of elements accumulated in King Bolete *Boletus edulis* mushroom at lowland and high mountain regions. J Environ Sci Health Part A 43:1692-1699

Falandysz J, Zalewska T, Krasińska G, Apanel A, Wang Y, Pankavec S (2015a) Evaluation of the radioactive contamination in Fungi genus *Boletus* in the region of Europe and Yunnan Province in China. Appl Microbiol Biotechnol 99:8217-8224

Falandysz J, Zhang J, Wang Y, Krasińska G, Kojta A, Saba M, Shen T, Li T, Liu H (2015b) Evaluation of the mercury contamination in mushrooms of genus *Leccinum* from two different regions of the world: accumulation, distribution and probable dietary intake. Sci Total Environ 537:470-478

Falandysz J, Zalewska T, Apanel A, Drewnowska N, Kluza K (2016) Evaluation of the activity concentrations of ^{137}Cs and ^{40}K in some Chanterelle mushrooms from Poland and China. Environ Sci Pollut Res 23:20039-20048

Falandysz J, Zhang J, Zalewska T (2017) Radioactive artificial ^{137}Cs and natural ^{40}K activity in 21 edible mushrooms of the genus Boletus species from SW China. Environ Sci Pollut Res 24:8189–8199

Falandysz J, Saniewski M, Zheng J, Zalewska T, Liu H, Kluza K (2018) Artificial ^{137}Cs and natural ^{40}K in mushrooms from the subalpine region of the Minya Konka summit and Yunnan Province in China. Environ Sci Pollut Res 25:615–627

Falandysz J, Saniewski M, Zalewska T, Zhang J (2019a) Radiocaesium pollution of fly agaric *Amanita muscaria* in fruiting bodies decreases with developmental stage. Isotop Environ Health Stud 55:317–324

Falandysz J, Zhang J, Mędyk M, Zhang X (2019b) Mercury in stir-fried and raw mushrooms from the *Boletaceae* family from the geochemically anomalous region in the Midu county, China. Food Control. 102:17–21

Falandysz J, Dryżałowska A, Zhang J, Wang Y (2019c) Mercury in raw mushrooms and in stir-fried in deep oil mushroom meals. J Food Comp Anal 82:103239

Falandysz J, Saniewski M, Zalewska T (2019d) Effect of a cooking (drying, blanching, pickling, macerating and deep freezing – blanching/pickling) on content of ^{40}K (total K) and ^{137}Cs in *Boletus edulis*, *Leccinum scabrum* and *Leccinum versipelle* mushrooms. submitted

Falandysz J, Wang Y, Zalewska T (2019e) ^{137}Cs, ^{40}K and K in raw mushrooms and stir-fried in deep oil mushroom meals from Yunnan in China. submitted

Falandysz J, Zhang J, Saniewski M, Zhang X (2019f) Pollution with ^{137}Cs and content of ^{40}K/K in raw bolete mushrooms grow in Midu area of SW China and in stir-fried mushroom meals from the *Boletaceae* family in of Yunnan, China. submitted

Falandysz J, Hanć A, Barałkiewicz D, Zhang J, Treu R (2019g) Metallic and metalloid elements in various developmental stages of *Amanita muscaria*. submitted

Fraiture A, Guillitte O, Lambinon J (1990) Interest of fungi as bioindicators of the radiocontamination in forest ecosystems. *In* Desmet G, Nassimbeni P, Belli M (eds.); 707 p; ISBN 1-85166-539-0; Worldcat; 1990; p. 477-484; Elsevier Applied Science; London (UK); Workshop on the transfer of radionuclides in natural and semi-natural environments; Udine (Italy); 11-15 Sep 1989

Frankowska A, Ziółkowska J, Bielawski L, Falandysz J (2010) Profile and bioconcentration of minerals by King Bolete (*Boletes edulis*) from the Płocka Dale in Poland. Food Addit Contam B 3:1-6

Ftanić Z, Senčar J, Bauman A (1992) Caesium radioactivity in mushrooms in Northwest Croatia. Period Biol 94:115-120

Fujii A, Koura T, Yoshimoto T, Kawabata T, Nakamura Y, Hamamichi K, Kakimoto H, Yamada Y, Yoshioka M, Yamamoto M, Hayakawa K (2014) Concentrations of ^{137}Cs and ^{40}K in wild mushrooms collected in a forest on Noto Peninsula, Japan. J Radioanal Nucl Chem 300:707-717

Gaso MI, Segovia N, Cervantes ML, Herrera T, Perez-Silva E (2000) internal radiation dose from ^{137}Cs due to the consumption of mushrooms from a Mexican temperate mixed forest. Radiation Protection Dosimetry 87:213–216

Giannaccini G, Betti L. Palego L, Mascia G, Schmid L, Lanza M, Mela A, Fabbrini L, Biondi L, Lucacchini A (2012) The trace element content of top-soil and wild edible mushroom samples collected in Tuscany, Italy. Environ Monit Assess 184:7579-7595

Grodzinskaya AA, Berreck M, Haselwander K, Wasser, SP (2003) Radiocesium contamination of wild-growing medicinal mushrooms in Ukraine. Int J Med Mush 5:61-86

Grodzinskaya AA, Syrchin SA, Kuchma ND, Wasser SP (2013) Macromycetes accumulative activity in radionuclide contamination conditions of the Ukraine territory. Part 6. - P.217-260, 368-373. *In* Mycobiota of Ukrainian Polesie: Consequences of the Chernobyl disaster. Kiev: Naukova dumka (In Russian)

Gucia M, Jarzyńska G, Kojta AK, Falandysz J (2012) Temporal variability in twenty chemical elements content of Parasol Mushroom (*Macrolepiota procera*) collected from two sites over a few years. J Environ Sci Heal B 47:81-88

Gulden G, Amundsen I (1994) Radioaktivitet i Norske sopper perioden 1988-1993. Våre Nyttevekster 2:36-47

Guillitte O, Fraiture A, Lambinon J (1990) Soil-fungi radiocesium transfers in forest ecosystems. *In* Desmet G, Nassimbeni P, Belli M (eds.); 707 p; ISBN 1-85166-539-0; Worldcat; 1990; p. 468-476; Elsevier Applied Science; London (UK); Workshop on the transfer of radionuclides in natural and semi-natural environments; Udine (Italy); 11-15 Sep 1989

Haselwandter K, Berreck M, Brunner P (1988) Fungi as bioindicators of radiocaesium contamination: pre- and post-Chernobyl activities. Trans Br Mycol Soc 90:171-174

Heinrich G (1993) Distribution of radiocesium in the different parts of mushrooms. J Environ Radioact 18:229-245

Heinrich G, Müller HI, Oswadl K, Gries A (1989) Natural and artificial radionuclides in selected Styrian soils and plants before and after the reactor accident in Chernobyl. Biochem Physiol Pflanzen 185:55-67

Hove K, Pedersen Ø, Garmo TH (1990) Fungi: A major source of radiocesium contamination of grazing ruminants in Norway. Healfh Physics 59:189-192

Ingrao G, Belloni P, Santaroni GP (1992) Mushrooms as biological monitors of trace elements in the environment. J Radioanal Nucl Chem 161:113-120

Isajenko K, Piotrowska B, Fujak M, Kardaś M (2012) Atlas Rdiologiczny Polski 2011 (Radiation Atlas of Poland 2011) Główny Inspektorat Ochrony Środowiska, Centralene laboratorium Ochrony Radiologicznej, Warszawa 2012. http://www.gios.gov.pl/images/dokumenty/pms/monitoring_promieniowania_jonizujscego/Atlas_Radiologiczny_Polski_2011.pdf

Ismail SS (1994) Distribution of Na, K, Rb, Cs, and ^{137}Cs in some Austrian higher fungi. Biol Trace Elem Res 43–45:707-714

Ismail SS, Dolezel P, Xarg V (1995) ^{137}Cs, trace and toxic elements distribution in Austrian Mushrooms. J Radioanal Nucl Chem Letters 200:315-336

Jarzyńska G, Gucia M, Kojta AK, Rezulak K, Falandysz J (2011) Profile of trace elements in Parasol Mushroom (*Macrolepiota procera*) from Tucholskie Forest. J Environ Sci Health Part B 46:741-751

Jarzyńska G, Falandysz J (2012a) Metallic elements profile of Slate Bolete (*Leccinum duriusculum*) mushroom and associated soil horizon. J Geochem Expl 121:69-75

Jarzyńska G, Chojnacka A, Dryżałowska A, Nnorom IC, Falandysz J (2012b) Concentrations and bioconcentration factors of minerals by Yellow-cracking Bolete (Xerocomus subtomentosus) mushroom collected in Noteć Forest, Poland. J Food Sci 77:H202-H206

Kabai E, Hiersche L, Poppitz Spuhler A, Baginski K (2016) Radioiaktive Kontamination von Speisepilzen. Aktuelle Messwerte (Standt: 2015) Fachbereich Strahlenschutz und Umwelt. Bundesamt für Strahlenschutz.

Kammerer L, Hiersche L, Wirth E (1994) Uptake of radiocaesium by different species of mushrooms. J Environ Radioact 23:135-150

Karadeniz Ö, Yaprak G (2007) Dynamic equilibrium of radiocesium with stable cesium within the soil-mushroom system in Turkish pine forest. Environ Poll 148:316-324

Karadeniz Ö, Yaprak G (2010) ^{137}Cs, ^{40}K, alkali–alkaline earth element and heavy metal concentrations in wild mushrooms from Turkey. J Radioanal Nucl Chem 285:611–619

Kirchner G, Daillant O (1998) Accumulation of ^{210}Pb, ^{226}Ra and radioactive cesium by fungi. Sci Total Environ 222:63-70

Kojta AK, Gucia M, Jarzyńska G, Lewandowska M, Zakrzewska A, Falandysz J, Zhang D (2011) Phosphorous and metallic elements in Parasol Mushroom (*Macrolepiota procera*) and soil from the Augustowska Forest and Ełk regions in north-eastern Poland. Fresenius Environ Bull 20:3044-3052

Kojta A, Jarzyńska G, Falandysz J (2012) Mineral composition and heavy metal accumulation capacity of Bay Bolete (*Xerocomus badius*) fruiting bodies collected near a former gold and copper mining area. J Geochem Expl 121:76-82

Kojta AK, Gucia M, Krasińska G, Saba M, Nnorom IC, Falandysz J (2016) Mineral constituents of edible field parasol (*Macrolepiota procera*) mushrooms and the underlying substrate from upland regions of Poland: Bioconcentration potential, intake benefits, and toxicological risk. Polish J Environ Stud 26:2445-2460

Komárek M, Chrastný V, Štichová J (2007) Metal/metalloid contamination and isotopic composition of lead in edible mushrooms and forest soils originating from a smelting area. Environ Intern 13:677-684

Komorowicz I, Hanć A, Lorenc W, Barałkiewicz D, Falandysz J, Wang Y (2019) Arsenic speciation in mushrooms using dimensional chromatography coupled to ICP-MS detector. Chemosphere 233:223-233

Kostiainen E (2007) ^{137}Cs in Finnish wild berries, mushrooms and game meat in 2000-2005. Boreal Environ Res 12: 23-28

Kułdo E, Jarzyńska G, Gucia M, Falandysz J (2014) Mineral constituents of edible parasol mushroom *Macrolepiota procera* (Scop. ex Fr.) Sing and soils beneath its fruiting bodies collected from a rural forest area. Chem Pap 68:484-492

Kuwahara C, Fukumoto A, Ohsone A, Furuya N, Shibata H, Sugiyama H, Kato F (2005) Accumulation of radiocesium in wild mushrooms collected from a Japanese forest and cesium uptake by microorganisms isolated from the mushroom-growing soils. Sci Total Environ 345:165-173

Larsen EH, Hansen M, Gössler W (1998) Speciation and health risk considerations of arsenic in the edible mushroom *Laccaria amethystina* collected from contaminated and uncontaminated locations. Appl Organom Chem 12:285-291

Laessoe, T., Del Conte, A., Gary Lincoff, G. (1996). The mushroom book How to identify, gather and cook wild mushrooms and other fungi. A DK Publishing Inc., New York, USA. ISBN 0-7894-1073-7.

Lehto J, Vaaramaa K, Leskinen A (2013) ^{137}Cs, ^{239,240}Pu and ^{241}Am in boreal forest soil and their transfer into wild mushrooms and berries. J Environ Radioact 116:124-132

Lipka K, Falandysz J (2017) Accumulation of metallic elements by *Amanita muscaria* from rural lowland and industrial upland regions. J Environ Sci Health B 52:184-190

Malinowska E, Szefer P, Bojanowski R (2006) Radionuclides content in *Xerocomus badius* and other commercial mushrooms from several regions of Poland. Food Chem 97:19–24

Marović G, Franić Z, Senčar J, Bituh T, Vugrinec O (2008) Mosses and some mushroom species as bioindicators of radiocaesium contamination and risk assessment. Coll Antropol 32:Suppl 2:109–114

Mascanzoni D (1990) Uptake of 90Sr and 137Cs by mushrooms following the Chernobyl accident. *In* Desmet G, Nassimbeni P, Belli M (eds.); 707 p; ISBN 1-85166-539-0; Worldcat; 1990; p. 459-466; Elsevier Applied Science; London (UK); Workshop on the transfer of radionuclides in natural and semi-natural environments; Udine (Italy); 11-15 Sep 1989

Mascanzoni D (1992) Determination of ^{90}Sr and ^{137}Cs in mushrooms following the Chernobyl fallout. J Radioanal Nucl Chem Articles 161:483-488

Mascanzoni D (2001) Long-term ^{137}Cs contamination of mushrooms following the Chernobyl fallout. J Radioanal Nucl Chem 249:245-249

Matishov GG, Kasatkina NE, Usyagina IS, Farion DA (2014) Artificial radionuclides in edible wild mushrooms and berries of the Murmansk region. Doklady Akademii Nauk 455:589–594

Melgar MJ, Alonso J, Perez-Lopez M, García MA (1998) Influence of some factors in toxicity and accumulation of cadmium from edible wild macrofungi in NW Spain. J Environ Sci Health Part B 33:439-455

Mietelski JW, Dubchak S, Błażej S, Anielska T, Turnau K (2010) ^{137}Cs and ^{40}K in fruiting bodies of different fungal species collected in a single forest in southern Poland. J Environ Radioact 101:706-711

Mleczek M, Niedzielski P, Kalač P, Budka M, Siwulski M, Gąsecka M, Rzymski P, Magdziak Z, Sobieralski K (2016) Multielemental analysis of 20 mushroom species growing near a heavily trafficked road in Poland. Environ Sci Pollut Res 23:16280–16295

Molzahn D, Reinen D, Behr H, Kocksholt P, Patzelt P (1989) The radioactive contamination of mushrooms by cesium – results from the Hessian rural district of Marburg-Biedenkopf. (*In German*) Z Mykol 55:135-148

Nakai W, Okada N, Ohashi S, Tanaka A (2015) Evaluation of ^{137}Cs accumulation by mushrooms and trees based on the aggregated transfer factor. J Radioanal Nucl Chem 303:2379-2389

Nakashima K, Orita M, Fukuda N, Taira Y, Hayashida N, Matsuda N, Takamura N (2015) Radiocesium concentrations in wild mushrooms collected in Kawauchi Village after the accident at the Fukushima Daiichi Nuclear Power Plant. PeerJ 3:e1427; DOI 10.7717/peerj.1427

Niesiobędzka (2000) Mobile forms of radionuclide ^{137}Cs in sandy soils in northeastern Poland. Polish J Environ Stud 9:133-136

Nikolova I, Johanson KJ, Dahlberg A (1997) Radiocaesium in fruitbodies and mycorrhizae in ectomycorrhizal fungi. J Environ Radioact 37:115-125

Nyholm NEI, Tyler G (2000) Rubidium content of plants, fungi and animals closely reflects potassium and acidity conditions of forest soils. Forest Ecol Management 134:89±96

Olsen RA (1994) The transfer of radiocaesium from soil to plants and fungi in seminatural ecosystems., pp. 265-286. *In* Nordic radioecology the transfer of radionuclides through Nordic ecosystems to man. Dahlgaard H (ed.). Elsevier, Amsterdam

Orita M, Nakashima K, Taira Y, Fukuda T, Fukushima Y, Kudo T, Endo Y, Yamashita S, Takamura N (2017) Radiocesium concentrations in wild mushrooms after the accident at the Fukushima Daiichi Nuclear Power Station: Follow-up study in Kawauchi village. Sci Rep 7:6744 | DOI:10.1038/s41598-017-05963-0

Petkovšek SA, Pokorny B (2013) Lead and cadmium in mushrooms from the vicinity of two large emission sources in Slovenia. Sci Total Environ 443:944-954

Řanda Z, Benada J, Horyna J, Kláan J (1990) Mushrooms – significant source of internal contaminationby radiocaesium. *In* Desmet G, Nassimbeni P, Belli M (eds.); 707 p; ISBN 1-85166-539-0; Worldcat; 1990; p. 169-178; Elsevier Applied Science; London (UK); Workshop on the transfer of radionuclides in natural and semi-natural environments; Udine (Italy); 11-15 Sep 1989

Řanda Z, Kučera, J (2004) Trace elements in higher fungi (mushrooms) determined by activation analysis. J Radioanal Nucl Chem 259: 99-107

Römmelt R, Hiersche L, Schaller G, Wirth E (1990) Influence of soil fungi (basidiomycetes) on the migration of Cs 134+137 and Sr 90 in coniferous forest soils. *In* Desmet G, Nassimbeni P, Belli M (eds.); 707 p; ISBN 1-85166-539-0; Worldcat; 1990; p. 152-160; Elsevier Applied Science; London (UK); Workshop on the transfer of radionuclides in natural and semi-natural environments; Udine (Italy); 11-15 Sep 1989

Rühm W, Yoshida S, Muramatsu Y, Steiner M, Wirth E (1999) Distribution patterns for stable ^{133}Cs and their implications with respect to the long-term fate of radioactive ^{134}Cs and ^{137}Cs in a semi-natural ecosystem. J Environ Radioact 45:253-270

Seeger (1978) Kaliumgehalt höherer Pilze. Z Lebensm Unters Forsch 167:23 31

Shutov NV, GYa Bruk, Basalaeva LN, Vasilevitskiy VA, Ivanova NP, Kaplun IS (1996) The role of mushrooms and berries in the formation of internal exposure doses to the population of Russia after the Chernobyl accident. Radiation Prot Dosim 67: 55-64

Strandberg M (1994) Radiocesium in a Danish pine forest ecosystem. Sci. Total Environ 157:125-132

Strandberg M (2004) Long-term trends in the uptake of radiocesium in *Rozites caperatus*. Sci Total Environ 327:315-321

Steinhauser G, Brandl A, Johnson TE (2014) Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts. Sci Total Environ 470-471:800-817

Stijve T, Poretti M (1990) Radiocesium levels in wild-growing mushrooms from various locations. Mushroom the Journal, Summer 1990, 5-9

Suchara I (2017) The Distribution of ^{137}Cs in selected compartments of coniferous forests in the Czech Republic. *In*: Gupta D, Walther C (eds) Impact of cesium on plants and the environment. Springer, Cham.

Sugiyama H, Terada H, Shibata H, Morita Y, Kato F (2000) Radiocesium concentrations in wild mushrooms and characteristics of cesium accumulation by the edible mushroom (*Pleurotus ostreatus*). J Health Sci 46:370-375

Tyler G (1980) Metals in sporophores of *Basidiomycetes*. Trans Br mycol Soc 74:41-49

Vetter J (1994) Kalium-Gehalt von eßbaren Wildpilzen. Z Lebensm Unters Forsch 198:33-35

Vinichuk M, Rosén K, Johanson KJ, Dahlberg A (2011) Correlations between potassium, rubidium and cesium (^{133}Cs and ^{137}Cs) in sporocarps of *Suillus variegatus* in a Swedish boreal forest. J Environ Radioact 102:386-392

Vinichuk M, Rosén K, Dahlberg A (2013) ^{137}Cs in fungal sporocarps in relation to vegetation in a bog, pine swamp and forest along a transect. Chemosphere 90:713-720

Vogel-Mikuš K, Debeljak M, Kavčič A, et al. (2016) Localization and bioavailability of mercury and selenium in edible mushrooms *Boletus edulis* and *Scutiger pes caprae*. Abstract Book of the 18th International Conference on Heavy Metals in the Environment, 12 to 15 September 2016, Ghent, Belgium:611-612

Vukojević V, Đurđić S, Mutić J (2019) Accumulation of U, Th, Pb, V, Rb, and Ag in wild mushrooms *Macrolepiota procera* (Scop.) Singer from Goč, Serbia. Environ Sci Pollut Res 26:13147–13158

Yoshida T, Muramatsu Y (1998) Concentrations of alkali and alkaline earth elements in mushrooms and plants collected in a Japanese pine forest, and their relationship with 137Cs. J Environ Radioact 41:183-205

Yoshida T, Muramatsu Y, Dvornik AM, Zhuchenko TA, Linkov I (2004) Equilibrium of radiocesium with stable cesium within the biological cycle of contaminated forest ecosystems. J Environ Radioactivity 75:301-313

Zalewska T, Cocchi L, Falandysz J (2016) Radiocaesium in *Cortinarius* spp. mushrooms in the regions of the Reggio Emilia in Italy and Pomerania in Poland. Environ Sci Pollut Res 23:23169-23174

Zalewska T, Saniewski M (2011) Bioaccumulation of gamma emitting radionuclides in red algae from the Baltic Sea under laboratory conditions. Oceanologia 53:631-650

FIGURE LEGENDS

Figure 1. Location of the *C. caperatus* and surface soil sampling places in Poland (for name and ID of the places see also Table 1).

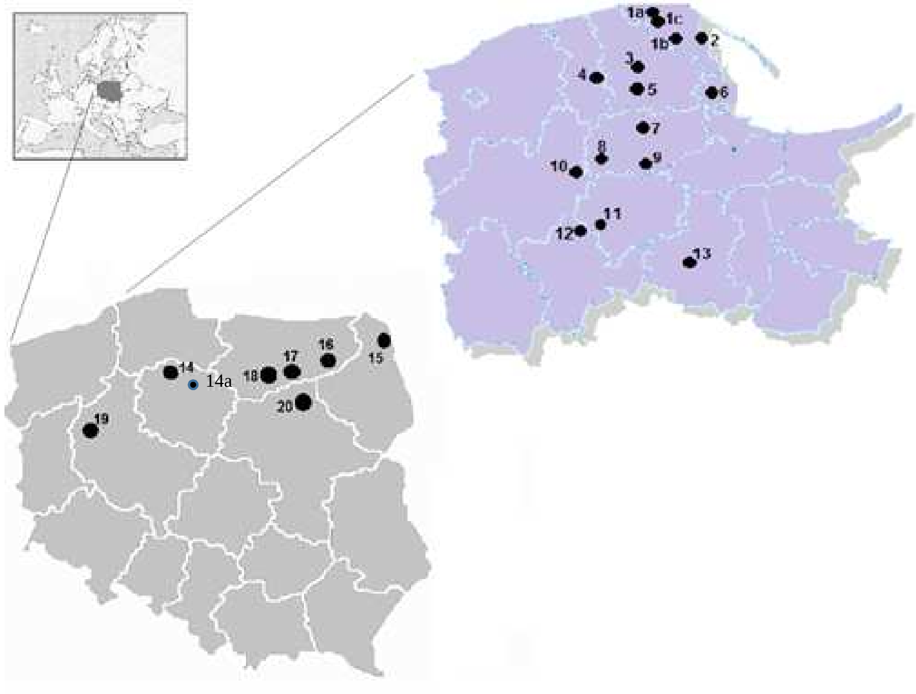
Figure 2. Spatial visualisation of the distribution of ^{137}Cs and ^{40}K activity concentrations in surface layer of soils in Poland (Isajenko et al., 2012).

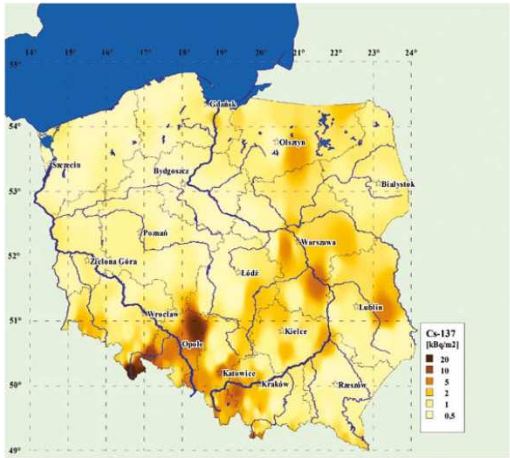
Figure 3. ^{137}Cs and ^{40}K in caps and stipes of *C. caperatus* from the northern regions of Poland (Bq kg^{-1}dry weight).

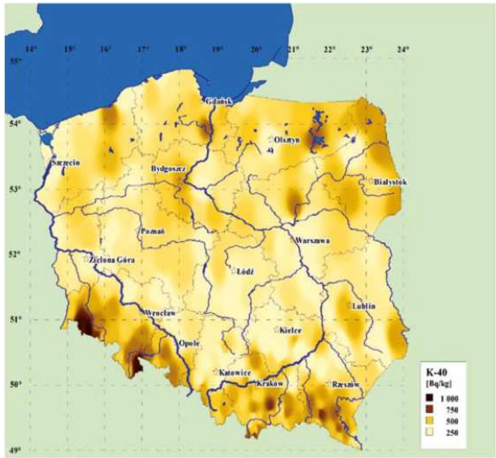
Table 1. ^{137}Cs and ^{40}K activity of *C. caperatus* – composite samples and beneath soils

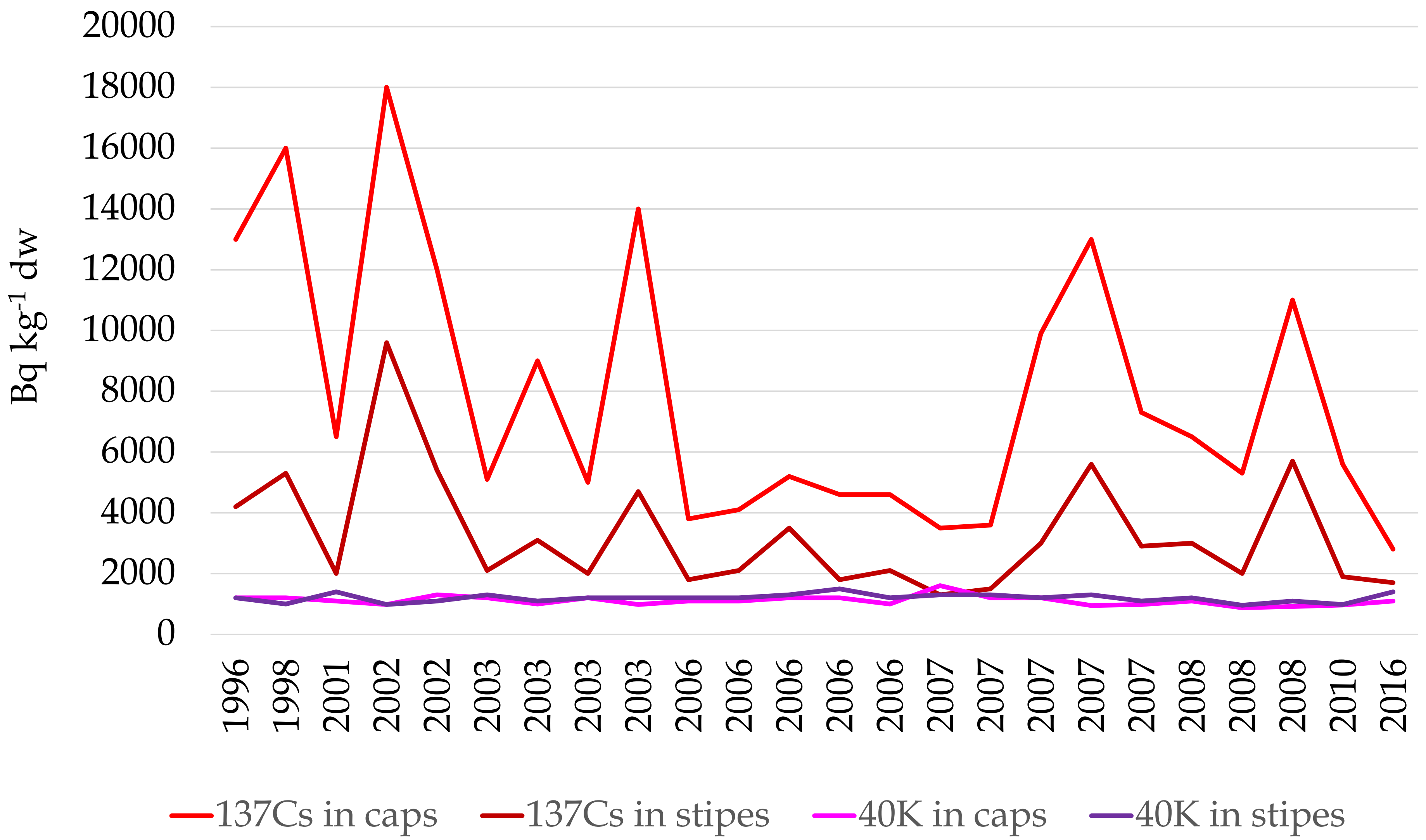
	^{137}Cs (Bq kg^{-1} db) Fruiting body			^{137}Cs (Bq kg^{-1} dm)		^{40}K (Bq kg^{-1} db) Fruiting body			^{40}K (Bq kg^{-1} dm)	
Region, site, year and site ID[#] (see Fig. 1)	Cap	Stipe	$Q_{C/S}$	Soil	$BCF_{C\,//}\,BCF_S$	Cap	Stipe	$Q_{C/S}$	Soil	$BCF_{C\,//}\,BCF_S$
Pomerania; Trójmiejski Landscape Park, 1996 (n=20)[*] [6][#]	13000 ± 110[¶]	4200 ± 45	3.1	33 ± 1	390 // 130	1200 ± 110	1200 ± 140	1.0	150 ± 41	8.0 // 8.0
Pomerania, Wdzydze Landscape Park, 1998 (n=15) [5]	16000 ± 180	5300 ± 65	3.0	WD[*]	WD	1200 ± 130	1000 ± 120	1.2	WD	WD
Pomerania, Darżlubska Wilderness, 2001 (n=15) [1a]	6500 ± 60	2000 ± 19	3.3	19 ± 2	340 // 100	1100 ± 140	1400 ± 100	0.79	110 ± 74	10 // 13
Pomerania, Darżlubska Wilderness, 2003 (n=53) [1b]	5100 ± 43	2100 ± 19	2.4	24 ± 1	210 // 87	1200 ± 100	1300 ± 91	0.92	120 ± 36	10 // 9.2
Masuria, Napiwodzko-Ramucka Wild., 2002 (n=15) [17]	18000 ±140	9600 ± 71	1.9	37 ± 1	490 // 260	980 ± 120	980 ± 97	1.0	180 ± 34	5.4 // 5.4
Przymuszewo Forest Inspectorate, 2002 (n=16) [10]	12000 ± 94	5400 ± 43	2.2	WD	WD	1300 ± 130	1100 ± 91	1.2	WD	WD
Pomerania, Dziemiany, 2003 (n=14) [4]	9000 ± 69	3100 ± 27	2.9	17 ± 1	530 // 180	1000 ± 110	1100 ± 98	0.91	97 ± 34	10 // 11
Pomerania, Kępice; 2003 (n=8) [12]	5000 ± 49	2000 ± 19	2.6	16 ± 2	310 // 120	1200 ± 140	1200 ± 100	1.0	100 ± 69	12 // 12
Masuria, Piska Wilderness, 2003 (n=52) [18]	14000 ± 110	4700 ± 40	3.0	WD	WD	980 ± 130	1200 ± 100	0.82	WD	WD
Pomerania, Ostrowo, 2006 (n=53) [2]	3800 ± 36	1800 ± 17	2.1	23 ± 1	165 // 78	1100 ± 130	1200 ± 100	0.92	170 ± 41	6.5 // 7.0
Pomerania, Seaside Landscape Park, 2006 (n=43) [2]	4100 ± 33	2100 ± 18	1.9	16 ± 2	260 // 130	1100 ± 100	1200 ± 90	0.92	< 37	61 // 67
Pomerania, Seaside Landscape Park, 2007 (n=16) [2]	3500 ± 38	1300 ± 17	2.6	46 ± 3	76 // 28	1600 ± 130	1300 ± 130	1.2	110 ± 86	14 // 12
Pomerania, Sulęczyno, 2006 (n=70) [3]	5200 ± 50	3500 ± 25	1.5	43 ± 1	120 // 81	1200 ± 140	1300 ± 100	0.92	83 ± 39	14 // 16
Pomerania, Strzebielino, 2006 (n=16) [7]	4600 ± 42	1800 ± 23	2.6	70 ± 3	66 // 26	1200 ± 130	1500 ± 150	0.80	120 ± 73	10 // 12
Pomerania, Kobylnica region, 2006 (n=61) [8]	4600 ± 39	2100 ± 18	2.2	25 ± 1	180 // 84	1000 ± 110	1200 ± 89	0.83	< 57	36 // 43
Pomerania, outskirts of the town of Lębork, 2007 (n=31) [3]	3600 ± 36	1500 ± 16	2.4	10 ± 1	360 // 150	1200 ± 140	1300 ± 120	0.92	< 40	60 // 65
Pomerania, Gołubie, 2008 (n=15) [11]	6500 ± 47	3000 ± 22	2.2	18 ± 1	360 // 120	1100 ± 100	1200 ± 80	0.92	150 ± 36	7.3 // 8.0
Augustowska Primeval Forest, Suwałki, 2007 (n=17) [15]	9900 ± 99	3000 ± 32	3.3	24 ± 1	410 // 120	1200 ± 120	1200 ± 120	1.0	230 ± 37	5.2 // 5.2
Mazovia, Olszewo-Borki, 2007 (n=19) [20]	13000 ± 100	5600 ± 59	2.3	WD	WD	950 ± 120	1300 ± 150	0.73	WD	WD
Notecka Wilderness, Lubusz region, 2008 (n=32) [19]	5300 ± 50	2000 ± 17	2.7	WD	WD	880 ± 140	960 ± 130	0.92	WD	WD
Warmia, Orzechowo, 2008 (n=52) [16]	11000 ± 82	5700 ± 44	1.9	41 ± 2	270 // 140	920 ± 120	1100 ± 100	0.92	180 ± 82	5.1 // 6.1
Pomerania, Commune Parchowo, 2010 (n=15) [9]	3700 ± 30		NA	WD	WD	1100 ± 93		NA	WD	WD
Pomerania, Tuchola Pinewoods, Lubichowo, 2007 (n=53) [13]	7300 ± 60	2900 ± 24	2.5	20 ± 1	360 // 140	980 ± 120	1100 ± 93	0.89	140 ± 36	7.0 // 7.9
Pomerania, Tuchola Pinewoods, Śliwice, 2010 (n=16) [14]	5600 ± 58	1900 ± 24	3.0	WD	WD	970 ± 150	980 ± 140	0.99	WD	WD
Pomerania, Tuchola Pinewoods, SE, 2016 (n=15) [14a]	2800 ± 52	1700 ± 84	1.6	WD	WD	1100 ± 21	1400 ± 96	0.79	WD	WD
Mean	NA	NA	2.4	NA	280 // 110	1100	1200	0.95	110	17 // 18
SD	NA	NA	0.5	NA	140 // 51	160	140	0.12	57	17 // 19
Median	NA	NA	2.4	24	260 // 130	1100	1200	0.92	110	10 // 12
Minimal	2800	1500	1.5	10	66 // 26	880	960	0.73	< 40	5.1 // 5.2
Maximal	18000	9600	3.3	70	530 // 260	1600	1500	1.2	230	61 // 67

Notes: $Q_{C/S}$ (The quotient of the activity concentration in cap and stipe); BCF (bioconcentration factor – cap or stipe); [*]Number of fruiting bodies in composite sample – in parentheses (from 15 to 152 specimens per site, respectively); [#]Sampling site – in brackets (see Fig. 1); [¶]Activity concentration ± measurement uncertainty; WD (without data); NA (not applicable)


1a
1c
1b
2
3
4
5
6
7
8
9
10
11
12
13
14
14a
15
16
17
18
19
20


14°
15°
16°
17°
18°
19°
20°
21°
22°
23°
24°
55°
54°
53°
52°
51°
50°
49°
Gdańsk
Olsztyn
Szczecin
Bydgoszcz
Białystok
Poznań
Warszawa
Zielona Góra
Łódź
Wrocław
Lublin
Opole
Kielce
Katowice
Kraków
Rzeszów
Cs-137
[kBq/m2]
20
10
5
2
1
0.5


14°
15°
16°
17°
18°
19°
20°
21°
22°
23°
24°
55°
54°
53°
52°
51°
50°
49°
Gdańsk
Olsztyn
Szczecin
Bydgoszcz
Białystok
Poznań
Warszawa
Zielona Góra
Łódź
Wrocław
Lublin
Opole
Kielce
Katowice
Kraków
Rzeszów
K-40
[Bq/kg]
1 000
750
500
250


Bq kg-1 dw
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0
1996
1998
2001
2002
2002
2003
2003
2003
2003
2006
2006
2006
2006
2006
2007
2007
2007
2007
2007
2008
2008
2008
2010
2016
137Cs in caps
137Cs in stipes
40K in caps
40K in stipes

Highlights

- Gypsy mushroom (*C. caperatus*) is an efficient fungal accumulator of radiocaesium
- Decades after Chernobyl accident *C. caperatus* could exceed radiocaesium safety limits
- Activity concentrations of *C. caperatus* fluctuate over time
- Recent examples of *C. caperatus* from hot-spots can show elevated ^{137}Cs levels
- Dietary intake of some Polish C. caperatus can provide relatively high radioactive dose