Environmental Science and Pollution Research (2021) 28:52925–52935
https://doi.org/10.1007/s11356-021-14423-2

RESEARCH ARTICLE

Natural radioactivity and total K content in wild-growing or cultivated edible mushrooms and soils from Galicia (NW, Spain)

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Received: 8 February 2021 / Accepted: 10 May 2021 / Published online: 21 May 2021
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Abstract
The radioactive isotope, ^{40}K, of naturally occurring potassium (0.012%) is present in the Earth's crust in a low percentage of all potassium, leading to its presence in almost all foodstuffs. The impact of ^{40}K activity concentrations was assessed in wild and cultivated edible mushrooms and in growing substrates. Samples were analysed by gamma spectroscopy. In the wild mushroom species, the average activity concentration of ^{40}K was 1291 Bq kg^{-1} dry weight (dw), approximately 140 Bq kg^{-1} fresh weight (fw), with a range of average values per species from 748 in *Lactarius deliciosus* to 1848 Bq kg^{-1} dw in *Tricholoma portentosum*. The cultivated species presented an average value of 1086 Bq kg^{-1} dw; and the soils, compost of cultivation and wood of substrate are 876, 510 and 59.4 Bq kg^{-1} dw, respectively. The total K content reached a maximum of 59,935 mg kg^{-1} dw in *T. portentosum*. The transfer factors (TF > 1) suggested that mushrooms preferentially bioconcentrated ^{40}K. *Cantharellus cibarius*, *Craterellus tubaeformis*, *Hydnum repandum* and *T. portentosum* by most TF could be considered as bioindicators of ^{40}K. Taking into account that the annual radiation dose of ^{40}K due to the average consumption of mushrooms analysed (0.15 μSv/year) is very low, it can be concluded that the consumption of these mushrooms does not represent a toxicological risk for human health. Finally, according to the total K content, from the nutritional point of view, these mushrooms could be considered as a potential source of potassium for the human diet.

Keywords Potassium · Radionuclide · Soils · Mushrooms · Transfer factors · Gamma spectroscopy · Health risk

Introduction

In recent years, information about mushrooms has increased, about their culinary properties and beneficial effects on human health, but also about the potential risks associated with their intake. In this sense, the study of the content of heavy metals, metalloids and in a smaller proportion of radionuclides has been especially considered in fungi (macroscopic fungi), all of them possible contaminants that are transferred from the soil (Falandysz and Borovička, 2013; Govorushko et al. 2019; Kalač 2012, 2019). These studies have been carried out with both wild and cultivated fungal species, and this accumulation has been shown to be species-dependent (Alonso et al. 2013; Caridi and Belmusto, 2017; Falandysz et al. 2018; Kalač 2019; Melgar et al. 2016).

Radioactivity can be found in many foods, including mushrooms (Baeza et al., 2004a, b; de Castro et al. 2012; Falandysz et al. 2017; Falandysz et al. 2018; Kioupi et al. 2016; Lee et al. 2018). In soils, the source of heavy metals can be geological and anthropogenic, and above all, the concentrations of natural radionuclides in soils can also vary due to human activities (Ribeiro et al., 2018; Ivanić et al., 2019). Radioactive potassium of natural origin, ^{40}K, is found in almost all foods, since a small percentage of all potassium is found naturally in the Earth's crust. Natural uranium and thorium can be found in fish, grains, and leafy vegetables, due to their decomposition in water. On the other hand, through photosynthesis and the absorption of water in plants, cosmogenic radionuclides can enter the food chain; therefore, living beings are continuously exposed to these natural radionuclides, which lead to an inevitable dose of more or less stable radiation (Brandhoff et al. 2016). Furthermore, many of these natural radioactive elements (uranium, thorium, potassium, etc.) can also be treated as pollutants, through technologically enhanced natural

Responsible Editor: Georg Steinhauser

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radioactive materials (TENORM) that generate industrial waste or by-products enriched with radioactive elements found in the environment (Escareño-Juárez et al., 2019; Strumińska-Parulska and Falandysz, 2020).

In addition, anthropogenic radioisotopes can be released to the environment by discharges into air or water, during normal practice or incidents. The activity of airborne can contaminate the plants by the dry deposition of aerosols into the water that is used for irrigation (Brandhoff et al. 2016).

The ^{40}K radioisotope (~ 0.012% natural potassium) is usually determined together with the radiocaesium, but these two elements present a chemical analogy between them, a correlation between ^{137}Cs and ^{40}K which has not been found, suggesting different absorption mechanisms (Baeza et al. 2004a; Falandysz et al. 2020; Mietelski et al. 2010). These radioisotopes can be transferred to food, such as fungi, and in edible species, numerous studies have been carried out that analyse the main elements and the accumulation of toxic and radioactive elements (García et al. 2015; Melgar et al. 2014; Melgar et al. 2016; Zocher et al. 2018).

Potassium in fungi is an essential nutrient; its range of variation is limited, and the levels reported, more frequently, for ^{40}K range between 1000 and 2000 Bq kg^{-1} dw, depending on the species (Guillén and Baeza 2014; Kalač 2001).

No difference has been reported regarding nutritional mechanism; mycorrhizal and saprotrophic mushrooms have similar contents of ^{40}K, but among the fungi, there is no homogeneity in the distribution, and the cap + gills have a higher content than the stipe (Baeza et al. 2006; Guillén and Baeza 2014), although, Kalač (2001) found that potassium concentrations decrease in the following order: cap > stipe > gills or tubes in spores-forming part (hymenium) > spores.

Literature data shows that the activity concentrations of ^{40}K in mushrooms are between 0.8 and 1.5 kBq kg^{-1} dw for both wild-growing and cultivated edible species. In *Imleria badia* (before called *Xerocomus badius*), *Lycoperdon perlatum* and *Amanita rubescens*, the transfer factors (TF) for ^{40}K ranged between 1.5 and 22.7, with values higher than 10 (Eckl et al. 1986). On the other hand, in cultivated mushrooms (lignicolous), *Flammulina velutipes*, *Lentinula edodes* and *Ganoderma lucidum*, values of 7.2, 1.8 and 1.6 were observed, respectively (Kalač 2001; Wang et al. 1998).

Numerous investigations were carried out to study how radionuclides behave and are transported in soil, plants and fungi (de Castro et al. 2012; Falandysz et al. 2016, 2017; García et al. 2015; Lee et al. 2018; Malinowska et al. 2006; Mietelski et al. 2010; Yamada, 2013). Although the TF in fungi are higher for both ^{137}Cs and ^{40}K than for vegetables, not all fungi accumulate caesium at the same levels as potassium. For example, TF of caesium was lower than that of potassium in *L. edodes* (Yamada 2013).

Potassium is the major monovalent element that bioconcentrates in mushrooms and is an essential component of cell protoplasm (Ayaz et al. 2011; López-Vázquez and Prieto-García 2016; López Vázquez et al. 2016; Nnorom et al. 2020; Wang et al. 2015). Some authors have evaluated, in various species of fungi, the relationship between ^{40}K and total (stable) K bioconcentrated with the BCF (bioconcentration factor), due to its impact on the human diet because it is an electrolyte and vital ion to body liquids and an almost constant component of lean body tissues (López Vázquez et al. 2016). Since different amounts of K are often associated at the age of maturity of mushrooms, studies have been carried out on this, even evaluating the influence of cooking these mushrooms in their development stages towards maturity and the potassium content (Falandysz and Borovička 2013; Falandysz et al. 2020; Falandysz et al., 2021a.

Fungi bioconcentrate heavy metals; therefore, they could be considered good bioindicators of environmental contamination depending on the species, among other factors (de Castro et al. 2012; Guillén and Baeza 2014; Świsłowski et al. 2020). Most radionuclides, whether anthropogenic or naturally occurring (as potassium (^{40}K), uranium (^{234}U/^{238}U), thorium (^{230}Th/^{232}Th) and radium-lead-polonium (^{226}Ra, ^{210}Pb and ^{210}Po)), can be bioconcentrated by mushrooms (Guillén and Baeza 2014). Ivanić et al. (2019) determined the activity concentrations of natural (^{238}U, ^{235}U and ^{232}Th decay chains, ^{40}K and ^{7}Be) and anthropogenic (^{137}Cs, ^{134}Cs) concluding that they accumulate in mushrooms. In areas heavily contaminated by radioactive fallout can negatively impact consumer health (Grodzinskaya et al. 2003; Grodzinskaya et al. 2011). Thus, Guillén and Baeza (2014) reported an exhaustive review of what is known about the radionuclide content in mushrooms worldwide. The objectives were to identify which radionuclides could constitute a hazard to the consumer health and the conditioning factors. They include the ^{40}K content in a very wide range (70–3520 Bq/kg dw) and geographically as the most widespread in many European countries and in some countries in Asia and Central America. Moreover, Guillén et al. (2017) estimated the total dose rate radioactivity of mushrooms using the ERICA Tool, assuming different fruiting body geometries, a single ellipsoid and more complex geometries considering the different components of the fruit body (cap, gills, steam) and the mycelium and their differing radionuclide contents (especially from ^{226}Ra and ^{210}Po). The study was carried out in the Mediterranean ecosystem (Spain), and the species considered were *Agaricus bisporus* and *Macrolepiota procera*.

Since uranium and thorium are not essential elements for the development of fungi, their content by fungi is lower than that of potassium. No influence of nutritional mechanisms has been reported; however, it is known that thorium and uranium isotopes are preferentially detected in the cap and gills, and given that the ratios ^{234}U/^{238}U and ^{230}Th/^{232}Th of isotopes in mushrooms samples are close to unity, it could be indicative of a single absorption pathway for all uranium and thorium isotopes. However, some human activities (NORM) can increase the content of these radionuclides in the environment, such as the uranium mining industry, and in this case, mushrooms could also be used as bioindicators for these radionuclides found as pollutants. The isotope ^{210}Pb occurs as a descendant of the decay of ^{226}Ra which could end up as a direct deposition onto the fruiting bodies; however, comparisons with stable lead uptake showed that mushrooms mainly take ^{210}Pb up directly from the soil (Guillén and Baeza 2014).

The article presents the determined levels of a natural radioisotope potassium (^{40}K), including total potassium (K), and other radionuclides (^{7}Be, ^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{210}Pb, ^{228}Ac, ^{224} Ra, ^{212}Pb, ^{208}Tl, ^{235}U) in the several species of wild and cultivated edible mushrooms, in their growth substrates and in soils of Galicia (NW Spain), and bioconcentration values (TF/BCF), in order to assess its impact on consumer health.

Materials and methods

Sampling: mushrooms and soils

The study was carried out in Galicia (NW Spain), placing the location of sampling in various areas of its 4 provinces (A Coruña, Lugo, Ourense and Pontevedra), as shown in Fig. 1. The mushrooms collected belong to the class *Basidiomycetes* and represent 9 species, 48 samples of edible wild mushrooms and 5 samples of edible cultivated mushrooms (Table 1). Mushroom names in this study follow the nomenclature of the Index Fungorum (2020). Taking into account their mode of nutrition, which can condition the biological accumulation of radionuclides, all the species were mycorrhizal, except the cultivated species that are saprotrophic. Mushroom species were selected based on availability in the study areas and also according to their commercialization and culinary quality.

After sampling, the fungal samples were cleaned to remove all impurities and substrate debris, dried at 110 °C and pulverized. Then, they were ashed at 430 °C (Alonso et al., 2013; García et al., 2015). Finally, they were homogenized up to fine powder. This powder samples were analysed by the LAR (Laboratory of Radiation Analysis, University of Santiago de Compostela). The mushroom analysis was carried out jointly without distinguishing between anatomical parts (A_M).

A total of 18 soil samples (0–10-cm depth) were collected from the same sampling sites as the mushrooms and were examined under the same conditions as fungi (A_S).

At each sampling site, soil samples (4–8) consisting of samples and their replicates were collected by a mechanical procedure using a helical tube, to obtain 1 kg of sample.

The sieving, drying and calcining processes of the samples were adjusted to the protocols (weight, temperature and time) described in other authors (Baeza et al., 2003; Herranz et al., 2003; García et al., 2015). At the end of these treatments, sample was transferred to perfectly identified 500-ml Duchess Shutter flasks. They were selected as measurement geometries: Petri model (Petri dishes) of 90 mm diameter for biological samples (4 g) and Duchess model (Duchess vessels) of 500 ml for soil samples (150 g) (Baeza et al., 2004b; García et al., 2015).

Analysis of mushrooms and soils

The activity level of natural radionuclides ^{40}K, ^{7}Be, ^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{210}Pb, ^{228}Ac, ^{224} Ra, ^{212}Pb, ^{208}Tl and ^{235}U for both fungi and the soil samples were measured by an ORTEC gamma spectroscopy system, using a Hyper-Pure Germanium (HPGe) detector. The resolution was 2.2 KeV at 1.33 MeV ^{60}Co, and a data analysis software GammaVision V. 5.31 was used. Calibration of the detector in energy and efficiency was performed for the two applied geometries. The matrixes, with a density similar to the samples, were activated with a certified standard solution (P925/LMR/RN/336, CIEMAT), according to the integration times described for soils and biological samples by Alonso et al. (2013) and García et al. (2015).

The amount of mass available for analysis, the geometry used and the need to reach the lowest possible detection limits depending on the available time detector determined the difference in integration time.

All samples, both mushrooms and soil samples, were analysed in triplicate, and the final results were the averages of the three analyses. Concentrations of total (stable) K were calculated from ^{40}K activity concentration (Escareño, 2012).

The radionuclide TF expressed as Bq kg^{-1} dw was applied to quantify the translocation and uptake of stable elements and their radioactive isotopes from soil to fungal fruiting bodies (Baeza et al., 2005; Karadeniz and Yaprak, 2011).

Statistical study of the results

IBM SPSS Statistics software, version 25, was used to carry out the statistical analysis of the data. The Kolmogorov-Smirnov test was used to check whether the data fit a normal distribution and to evaluate the homogeneity of the variations the Levene's test was used. The influence of certain factors in the level of activity of ^{40}K, ^{235}U, ^{234}Th, ^{224}Ra, ^{226}Ra and ^{210}Pb in fruiting bodies of mushrooms, such as the type of collection (wild or cultivated) and fungal species, was assessed by analysis univariate variation (ANOVA). To apply ANOVA considering these criteria, a logarithmic transformation of the data was previously performed.

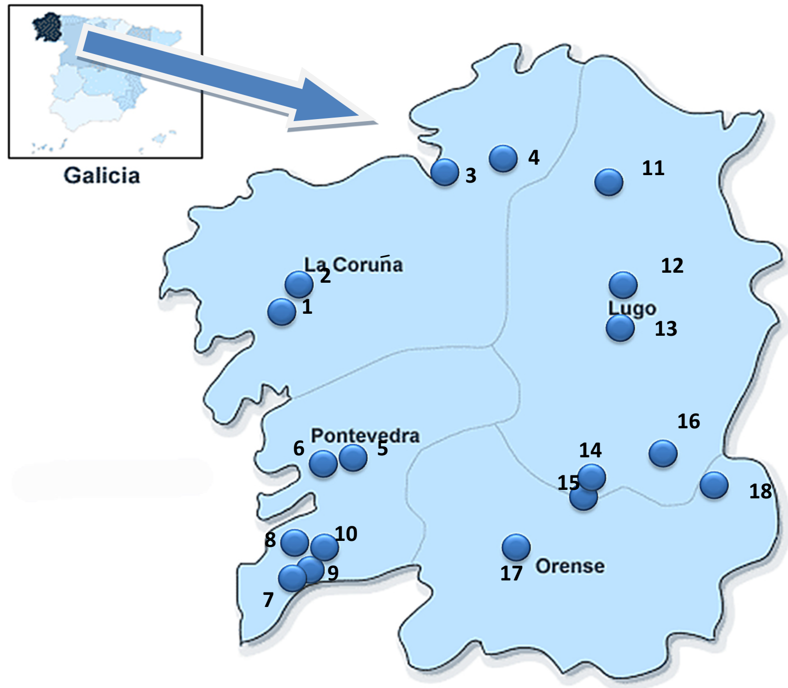

Fig. 1 Location of the sampling areas in the Provinces of Galicia, NW of Spain (more than one species of some were collected from some sites)

Sampling zones:

1. Liñaio, Negreira (La Coruña)
2. Zas, Negreira (La Coruña)
3. Miño (La Coruña)
4. As Pontes (La Coruña)
5. Bora, Pontevedra (Pontevedra)
6. Lourizán, Pontevedra (Pontevedra)
7. A Laxe, Pontevedra (Pontevedra)
8. A Picoña, Pontevedra (Pontevedra)
9. Cabreira, Pontevedra (Pontevedra)
10. Cristiñade, Pontevedra (Pontevedra)
11. Romariz, Abadín (Lugo)
12. Meilán, Lugo (Lugo)
13. Lamablanca-Coeses, Lugo (Lugo)
14. Vilaoscura, Sober (Lugo)
15. Anllo, Sober (Lugo)
16. Vilar de Lor, Quiroga (Lugo)
17. San Xurxo, Taboadela (Orense)
18. Roblido, A Rúa (Orense)

Results and discussion

General

The results of analyses, given as Bq kg^{-1} on a dry weight basis (Bq kg^{-1} dw), are presented in Table 1 for mushroom and soil samples. The total K content was expressed as mg kg^{-1} dw. In order to transform Bq kg^{-1} fw into Bq kg^{-1} dw, a dry/wet ratio of 0.1 was assumed.

Radionuclide activity concentrations in mushrooms and total K content

The mean concentration in the wild mushroom samples (all mycorrhizal species) for ^{40}K was 1291 Bq kg^{-1} dw (approximately 140 Bq kg^{-1} fw), with a range of average values by species from 748 in *L. deliciosus* to 1848 Bq kg^{-1} dw in *T. portentosum*. Cultivated species presented an average level of activity of 1086 Bq kg^{-1} dw; soils, 877 Bq kg^{-1} dw; compost culture, 510 Bq kg^{-1} dw; and wood substrate, 59.4 Bq kg^{-1} dw. These activity concentrations were higher than those presented in other foods (100–600) Bq kg^{-1} dw and similar to those of potassium-rich foods like spinach, potatoes, nuts or some seafood (Aloraini et al. 2018; Garcêz et al., 2018; González and Bonzi 2012; Kalač 2012; Quintero et al. 2007).

In this study, according to the statistically significant differences ($p<0.001$), 2 very different groups were clearly observed (Fig. 2): the species *Boletus* section *edules* and *L. deliciosus* with concentrations below 1000 Bq kg^{-1} dw and TF below 2 and the other species with values between 1100 and 1900 Bq kg^{-1} dw and TF between 3 and 4. However, the distribution of the concentrations obtained is quite symmetrical, and the deviations of the results in each

Table 1 Activity concentrations of ^{40}K (Bq kg^{-1} dw) and concentration of total K (mg kg^{-1} dw) in the analysed species of mushrooms collected in Galicia

Species	Nutritional group	n	^{40}K (Bq kg^{-1} dw)	^{40}K range	TF/BCF	K (mg kg^{-1} dw)
Boletus edulis Bull.	M	6	801 ± 129	698–980	1.62	25,978 ± 4184
Boletus pinophilus Pilat & Dermek	M	5	1003 ± 102	860–1086	1.81	32,530 ± 3308
Boletus reticulatus Schaeff.	M	4	963 ± 52	789–1089	1.14	31,232 ± 1688
Cantharellus cibarius Fr.	M	9	1715 ± 207	1321–1992	3.96	55,622 ± 6714
Cantharellus subpruinosus Eyssart & Buyck	M	2	1544 ± 47	1511–1577	2.59	50,076 ± 1524
Craterellus tubaeformis (Fr.) Quél.	M	6	1408 ± 202	1095–1661	3.42	45,665 ± 6551
Hydnum repandum L.	M	7	1756 ± 230	1255–1890	3.26	56,950 ± 7460
Lactarius deliciosus (L.) Gray	M	4	748 ± 42	690–788	1.06	24,260 ± 1362
Tricholoma portentosum (Fr.) Quél.	M	5	1848 ± 146	1670–1995	3.57	59,935 ± 4735
Pleurotus ostreatus (Jacq.) P. Kumm	S	1	1309		2.57	42,454
Lentinula edodes (Berk.) Pegler	S	1	749		12.61	24,292
Agaricus bisporus (J.E. Lange) Imbach	S	1	1366			44,303
Agaricus brasiliensis Peck	S	1	1761			57,114
Trametes versicolor (L.) Lloyd	S	1	246			7978
Compost cultivation of *Pleurotus*/compost *Pleurotus*		1	510			16,540
Wood cultivation of *Lentinula*/ wood *Lentinula*		1	59.4			1927
Soils		18	877 ± 47	174–1474		28,443

The number of samples (n), the mean concentrations, the standard deviations, the range and the transfer factors are indicated

M mycorrhizal, *S* saprotrophic

species from the average value are much lower than those observed for the ^{137}Cs (García et al. 2015).

The cultivated species presented an average value of 1086 Bq kg^{-1} dw, but a clear difference was also observed between species cultivated in compost (*Pleurotus ostreatus*, *A. bisporus* and *Agaricus brasiliensis*) with significantly higher average levels similar to those of the species wild than those cultivated in wood (*L. edodes* and *Trametes versicolor*). This may be due to the low concentration of potassium in the wood in which they grow and feed, with respect to the compost or soils in which other species grow.

In general, the levels obtained in this study are located in ranges of activity similar to those usually indicated in other countries, which are generally averaged between 800 and 2000 (Table 2). The fairly homogeneous and symmetrical distribution of ^{40}K and the similarity data obtained by all authors, as well as the fact that the potassium is an abundant element and a nutrient in which the isotopic mixture (isotopes of ^{39}K, ^{40}K and ^{41}K) is fairly constant, representing ^{40}K the 0.012% of the mixture, suggest that the incorporation of K is self-regulated (bio-adjustable) by the fungus itself, due to the vital role of potassium in hydration (the moisture content of fresh *B. edulis* is around 90%) (Falandysz et al., 2021b) and is carried out together with stable potassium (Baeza et al. 2004b; Falandysz and Borovička 2013; Kalač 2012).

In wild mushroom samples, the total K content, estimated from the corresponding ^{40}K data (Table 1), was in the range of 24260 ± 1362 (*L. deliciosus*) to 59935 ± 4735 mg kg^{-1} dw (*T. portentosum*). *H. repandum* species (56950 ± 7460 mg kg^{-1} dw) and *Cantharellus* genus (mean 52849 ± 4119 mg kg^{-1} dw) stand out for their content. Moreover, being known that the Boletaceae family is rich in K, in this study with 3 species, the total K content in the whole fruiting bodies was in the range of 25978 ± 4184 (*B. edulis*) to 32530 ± 3308 mg kg^{-1} dw (*B. pinophilus*); these data obtained, in our work, agree with the results shown for Boletaceae family that grows in Southwest China and in European forests (Falandysz et al. 2011; Falandysz et al. 2020; Zhang et al. 2010).

Comparing our results of the total K content, referenced to the ^{40}K, with data of other studies whose results were obtained by the ICP-OES/AAS analytical method (Ayaz et al. 2011; López-Vázquez and Prieto-García 2016) and the review carried out for 5 years (2011–2015) by López Vázquez et al. (2016), a parallel trend is observed in the levels of potassium, being among the coincident genera, the most accumulating: *Tricholoma*, *Hydnum*, *Cantharellus* and *Agaricus*.

These data contrast with the much lower potassium levels observed in mushrooms grown in compost and in wood

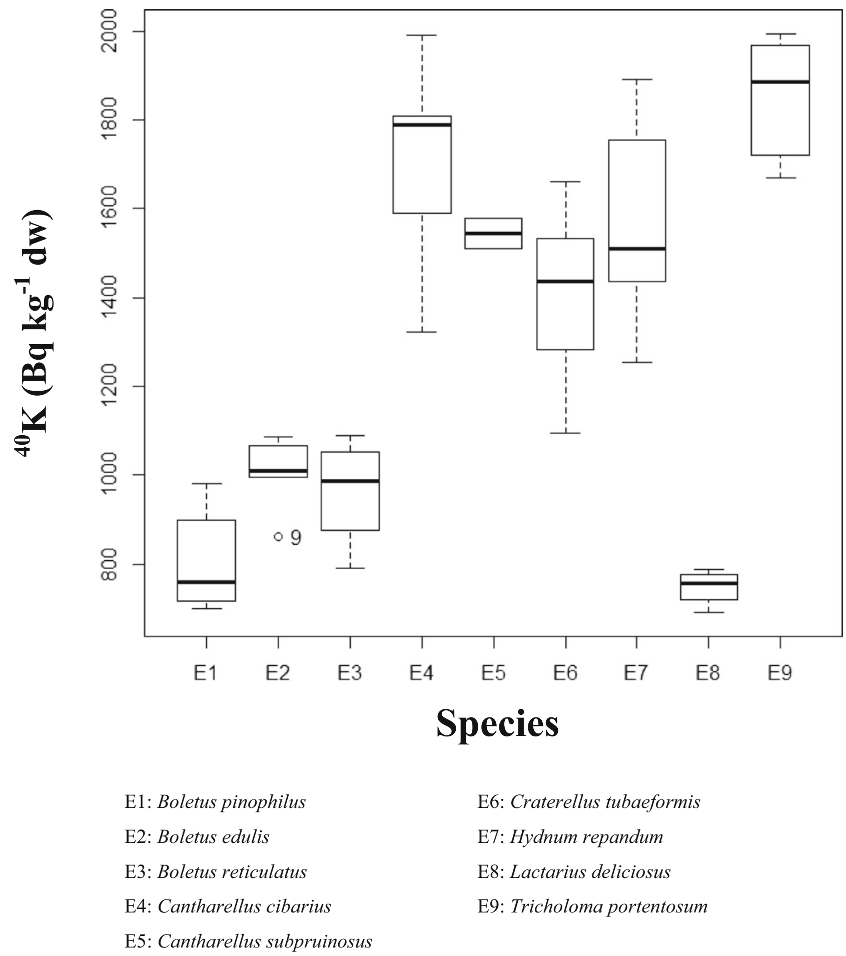

Fig. 2 Activity concentrations of ^{40}K, expressed in Bq kg^{-1} dw, in the mushroom species

(Table 1), but they are even higher than those observed in Nigerian forests by Nnorom et al. (2020).

Radionuclide activity concentrations in soils and bioconcentration in mushrooms (TF/BCF)

The average potassium value in soils was 877 Bq kg^{-1} dw, with a range from 174 (zone 13 in Lugo) to 1474 Bq kg^{-1} dw (area 3 in A Coruña). The mean levels in soils were as follows: A Coruña 843 ± 37 Bq kg^{-1} dw, Lugo 791 ± 37 Bq kg^{-1} dw, Ourense 988 ± 64 Bq kg^{-1} dw and Pontevedra 886 ± 51 Bq kg^{-1} dw (Table 3). These levels were slightly higher than those found (473–621 Bq kg^{-1}) in Croatia by Ivanić et al. (2019) and also in Brazil (12−1042 Bq kg^{-1}) by Ribeiro et al. (2018).

A global average of 412 Bq kg^{-1} dw was established (UNSCEAR 2010), with a usual average range between 140 and 850 and higher values in the environment of 1200 Bq kg^{-1} dw for granitic and marble soils.

In our study, levels above 1000 Bq kg^{-1} dw (Fig. 3) refer to granitic soils, coinciding with those indicated in Extremadura (Spain) by Baeza et al. (1994) on surface soils, although

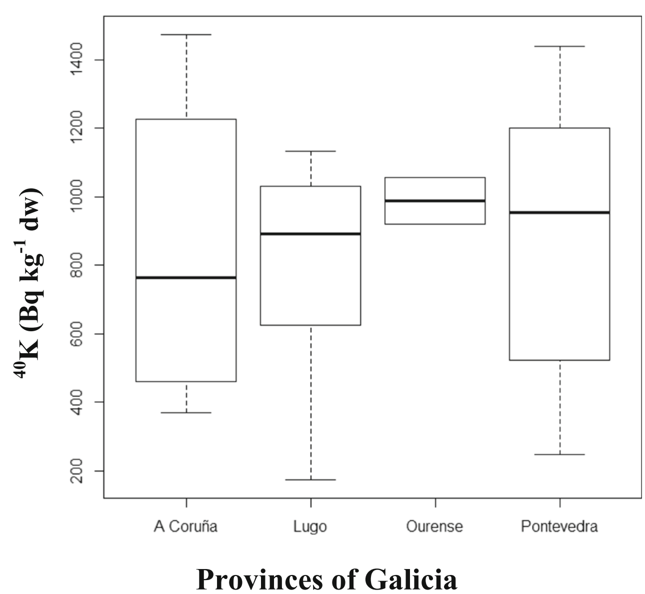

Fig. 3 Activity concentrations of ^{40}K, expressed in Bq kg^{-1} dw, in different soils sampling of mushroom

Table 2 Activity concentrations of ^{40}K in mushrooms reported by other authors

References	Country	Species	Activity concentrations (Bq kg^{-1} dw)
Baeza et al. (2005)	Extremadura (Spain)	*L. deliciosus*	852 ± 25
Barua et al. (2019)	Bangladesh	*P. ostreatus*	440 ± 61.6
Caridi and Belmusto (2017)	Italy	*B. edulis*	978 ± 113
		L. deliciosus	1487 ± 118
Castro et al. (2012)	Sao Paulo (Brazil)	*A. bisporus*	753 ± 3
		L. edodes	753 ± 3
		P. ostreatus	776 ± 4
Falandysz et al. (2016)	Poland, China	*Cantharellus*	1500 ± 50
Falandysz et al. (2017)	China	*Boletus* sp.	1300 ± 200
Falandysz et al. (2021)	Poland	*B. edulis*	696 ± 130
Kioupi et al. (2016)	Greece	*Boletus* sp.	1685
Malinowska et al. (2010)	Poland	*X. badius*	1030 ± 101
		B. edulis	723 ± 77
Mietelski et al. (2010)	Poland	*B. edulis*	1389 ± 228
		P. ostreatus	1130 ± 323
Szántó et al. (2007)	Belgium	*B. edulis*	1060
		C. cibarius	1380
Tuo et al. (2017)	China	*B. edulis*	758
		L. edodes	629
		C. cibarius	1306
Turkekul et al. (2018)	Turkey	*B. edulis*	570
		L. deliciosus	505
Wang et al. (1998)	Taiwan	*L. edodes*	540 ± 117

Ribeiro et al. (2018) found correlation between the radionuclide activities and soil characteristics (pH, organic matter content) and not with the bedrock composition.

No statistically significant correlations have been observed between the ^{40}K levels in soils and those corresponding to the samples of fungi grown in them, since, as already indicated, potassium is an essential nutrient and its absorption is self-regulated by the fungus in function to its physiological needs.

In relation to transfer factors, all wild species of the present study showed mean TF higher than 1. Transfer factors suggest that fungi, preferentially, uptake and bioconcentrate ^{40}K, that is, they increase in the fungi the levels corresponding to their soils of growth, although in a magnitude, in general, quite discreet (between 1 and 4 according to species), suggesting that potassium is essential for fungi (Baeza et al. 2005).

Table 3 Activity concentrations of potassium (Bq kg^{-1} dw) in soils of the 4 provinces from Galicia

Provinces of Galicia	n	Mean ± SD	Range
A Coruña	4	843 ± 37	371–1474
Lugo	6	791 ± 37	174–1132
Pontevedra	6	886 ± 51	248–1439
Ourense	2	988 ± 64	920–1055
Total	18	877 ± 47	174–1474

Number of samples (n), mean concentrations, standard deviations, and range are indicated

The results for ^{40}K were within the range of variation reported in previous studies (Baeza et al. 2005; Falandysz et al. 2017; Karadeniz and Yaprak 2011; Tuo et al. 2017), although below the average ratios were reviewed (between 20 and 40) by Kalač (2012).

Regarding the cultivated species, the highest transfer factor was observed in the species *L. edodes* (12.6), whose levels of ^{40}K were not very high (749 Bq kg^{-1}dw), being lower than in the wild species, but the very low concentration of potassium in the wood (59.4 Bq kg^{-1}dw), with respect to other substrates, explains this high transfer factor. Rakić et al. (2014) noted that, generally, species with fleshy basidiomata, lignicolous as well as mycorrhizal showed higher transfer for ^{40}K and ^{137}Cs. As the species *C. cibarius*, *C. tubaeformis*, *H. repandu*m and *T. portentosum* showed the highest activity concentration and the highest TFs (> 3), they could be considered as bioindicators for ^{40}K in these habitats.

Considering the bioconcentration factor (BCF) of the total K content in mushrooms (Table 1), our values (BCF > 1) between 1.06 (*L. deliciosus*) and 3.96 (*C. cibarius*) were very similar to those determined in a study carried out in China (Wang et al. 2015) by ICPE on the anatomical parts of 10 *Boletus* species.

All relevant gamma emission isotopes (^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{210}Pb, ^{228}Ac, ^{224} Ra, ^{212}Pb, ^{208}Tl, ^{235}U) belonging to the three natural decay chains (^{238}U, ^{232}Th, ^{235}U) were present in all samples (Table 4), except for one sample (S4), corresponding to A Coruña, in which ^{238}U if exists presents an activity below the limit of detection (0.4 Bq kg^{-1} dw). Regarding the gamma-emitting elements analysed belonging to the ^{238}U chain, the presence of secular equilibrium between ^{226}Ra and $^{214}Pb/^{214}Bi$ has been confirmed. This equilibrium was evident from the first measurement performed on the samples, except in the case of sample S12. For samples S3, S4, S7, S10, S11, S16 and S18, the equilibrium extends to all sections of the chain. While the establishment of the first equilibrium is expected if we consider a long storage period of the sample without losses of ^{222}Rn, the establishment of the second implies, at least, unfavourable conditions for the mobilization of radon and radius from the soil. The absence of ^{235}U in sample S4 implies a significant isotopic imbalance in the terrain in which the sample was taken. Values higher than the minimum detection for ^{7}Be have not been obtained, although the short half-life of this element (53.12 days) makes its detection unlikely if the samples have been stored for more than 6 months. The activity concentrations in mushrooms were below the minimum detectable activity (MDA). This fact shows that there is no transfer to the mushrooms, which implies that they are not bioconcentrated and transfer factors cannot be calculated.

Some authors have detected these radionuclides in mushrooms; thus, Szymańska et al. (2020) described low levels of uranium (^{234}U, ^{238}U) and thorium (^{230}Th, ^{232}Th) in mushrooms of the genus *Leccinum* and *Leccinellum* from Yunnan (China). In other study, in Yunnan (China), Strumińska-Parulska et al. (2020) detected that a 100 g daily portion could provide a radiation of Pb of 0.02–0.06 μSv.

Table 4 Activity concentrations of radionuclides (Bq kg^{-1} dw) in soils of the 4 provinces from Galicia

Serie	Isotope	A Coruña	Lugo	Ourense	Pontevedra
Serie ^{238}U	^{234}Th	53 ± 10	48 ± 10	82 ± 12	129 ± 19
	^{226}Ra	65 ± 14	44 ± 13	104 ± 19	138 ± 22
	^{214}Pb	66 ± 4	47 ± 4	82 ± 5	126 ± 7
	^{214}Bi	66 ± 5	46 ± 4	105 ± 6	112 ± 7
	^{210}Pb	101 ± 22	78 ± 21	103 ± 25	136 ± 29
Serie ^{232}Th	^{228}Ac	22 ± 5	62 ± 5	29 ± 6	113 ± 8
	^{224}Ra	51 ± 15	88 ± 24	87 ± 24	164 ± 34
	^{212}Pb	33 ± 2	45 ± 3	57 ± 3	104 ± 5
	^{208}Tl	14 ± 2	19 ± 2	19 ± 2	40 ± 3
Serie ^{235}U	^{235}U	3.7 ± 1	2,1 ± 0.8	6 ± 1	6.3 ± 1.3

Mean concentrations and standard deviations are indicated

Repercussions in food

Real Decreto 30/2009 (2009) on the health/sanitary conditions for the marketing of mushrooms for food establishes that commercially available mushrooms must be free of pesticide residues, chemical contaminants and radioactivity, above the legally established limits.

However, unlike artificial radionuclides, such as ^{137}Cs, there are no national or international regulations that establish limits for the presence of ^{40}K in fungi or other foods. This is because, although ^{40}K is normally the main source of internal radiation because of its presence in food and beverages, the diet contribution to the total radiation dose is discrete, about 7.7% in Spain (CSN 2010), and in addition by its natural origin, the levels of ^{40}K are usually quite stable.

Considering for ^{40}K the similarity with ^{137}Cs and the new EU Regulation (Commission Implementing Regulation (EU) 2020/1158), the levels observed in mushrooms, in this study, were much lower than the maximum levels allowed for the importation of mushrooms from third countries (Albania, Byelorussia, Bosnia and Herzegovina, Kosovo, North Macedonia, Moldavia, Montenegro, Russia, Serbia, Switzerland, Turkey, Ukraine, the UK (except Northern Ireland)) into the EU, set at 600 Bq kg^{-1} fw (about 6000 Bq kg^{-1} dw) for mushrooms. If to this limit for ^{137}Cs applies the correction according to the conversion factor or coefficient for each radionuclide, which for the ^{40}K is 6.2×10^{-9} Sv Bq^{-1}, the limit for ^{40}K would be 12,581 Bq kg^{-1} dw (Guillén and Baeza 2014). No samples of fungi studied previously, including those of the present study (mean values, 748–1848 Bq kg^{-1} dw), reached similar levels, and these levels were usually about 10 times lower.

Health risk can also be calculated on the basis of the radiation dose according to food consumption (fungi in this study) through the following formula (Kalač 2012): $E = Y \times Z \times Dc$, where E is the annual effective radiation dose, Y annual intake of mushrooms (kg of dry matter per person), Z activity concentration (Bq kg^{-1} dw) and Dc dose coefficient (conversion factor) defined as the dose received by an adult per unit intake of radioactivity (for ^{40}K is 6.2×10^{-9} Sv Bq^{-1}). Agencia Española de Seguridad Alimentaria y Nutrición (AESAN, 2011) established the average data of consumption in Spain in 2 kg of fresh mushrooms/year/person. According to this study results and considering a mixed consumption of wild mushrooms with a mean value of 1291 Bq kg^{-1} dw and cultivated 1086 Bq kg^{-1} dw (mean for calculation of 1189 Bq kg^{-1} dw, i.e. approximately 118.9 Bq kg^{-1} fresh weight) would produce an annual effective dose of 0.15 μSv/year, a smaller contribution than that obtained for ^{137}Cs (0.32 μSv/year) for the same samples (García et al. 2015).

With data of this study, what risk does it really represent? If it takes as a reference the equivalence above indicated with respect to the legal limit for the ^{137}Cs, which would suppose a theoretical limit for the ^{40}K of approximately 12,500 Bq Kg^{-1} dw? It can see that the values found are much lower. In Spain, according to the CSN (2010), the average radiation dose received by the population is estimated at 3700 μSv/year, of which 2400 are by natural sources, and of these, 290 μSv are derived from the diet (200–800 μSv), of which 170 are due to ^{40}K. Therefore, 0.15 μSv/year indicates that the consumption of fungi supposes less than 0.09% of the annual radioactive dose due to the ^{40}K normally provided by the food and beverages, a very small amount and low contribution to consider them a food risk.

Another factor to be in account is that mushrooms are not usually eaten raw. They are generally cooked and consumed immediately or preserved. Some studies focused on radiocaesium reported that it, and by extension other radionuclides, could significantly reduce its content in mushrooms after undergoing cooking procedures (Guillén and Baeza 2014).

However, recent studies showed that, during cooking procedures (blanching, frying, braising and similar), a shrinking (loss of mass) of mushrooms and only a partial leak of the ^{137}Cs and ^{40}K, total K, makes that levels of $^{137}Cs/^{40}K$, total K ratio in fried or braised mushrooms (wet weight), higher than in fresh (raw) mushrooms used for cooking (wet weight), which supposes an enrichment of ^{137}Cs. This may be due to the difference in the distribution of caesium and potassium in cell structures and their binding sites. In the other hand, the breakdown of the cell wall because of high temperature cell shrinkage during stir-drying can favour the release of ^{40}K-total K but can have a lower effect on ^{137}Cs (Falandysz et al. 2020). In another study using household processes (Saba and Falandysz 2021), it was concluded that blanching of fungal materials always decreased activities resulting from ^{137}Cs and ^{40}K, but also the total of K content of the product, relative to the substrate, when the data were expressed in dry weight (biomass). In addition to the mushroom cooking procedures, it has been shown that there is a dependence on the stages of maturity of the mushrooms, observing that the meals made from button stage braised *B. edulis* presented higher ^{137}Cs activity concentrations than those made from more mature fruit bodies (Falandysz et al., 2021a, b).

The edible mushrooms in this study, especially the wild ones, constitute an important component of the diet in Galicia. Based on the estimates calculated for the total concentration of K in all fruiting bodies and the recommendations for daily intake of adults (3500 or 4700 mg of K according to AESAN (2019) or NIH (2020), respectively), these fungi could provide a significant amount of K to the diet.

Conclusions

The activity levels of ^{40}K in the species analysed, wild and cultivated edible mushrooms, in Galicia were within the usual ranges for this radionuclide, but the concentrations in soils were slightly above the usual averages, although within normal ranges considering the granitic character of many of the analysed soils.

The species *C. cibarius*, *C. tubaeformis*, *H. repandum* and *T. portentosum* by greater TF could be considered as bioindicators of ^{40}K.

The effective annual radiation dose of ^{40}K for the normal consumption of mushrooms analysed is very small, even lower than the corresponding to the ^{137}Cs, and, therefore, is not considered a health risk.

Finally, from the nutritional point of view and according to the total K content, these mushrooms could be considered as a potential source of potassium for the human diet.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11356-021-14423-2.

Availability of data and materials We include, in supplementary material, 2 excel files with the data of the materials worked: mushrooms and soils (Natural-Radionuclides-Mushrooms.xlsx and Natural-Radionuclides-Soils.xlsx) and a Technical-analytical report.

Author contribution All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by María Julia Melgar and María Ángeles García. The first draft of the manuscript was written by María Ángeles García, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Funding This work was supported by CETAL (Centro Tecnológico Agroalimentario de Lugo). The study was conducted in collaboration with “Federación Galega de Micoloxía”.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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