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ACTIVITY CONCENTRATION OF Cs-137 AND K-40 IN WILD EDIBLE *Craterellus cornucopioides* MUSHROOM GATHERED 31 YEARS AFTER THE CHERNOBYL POWER PLANT ACCIDENT IN BATAK MOUNTAIN, BULGARIA

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ABSTRACT

The aim of this study was to evaluate Cs-137 and K-40 activity concentrations and effective dose in selected mushroom species *Craterellus cornucopioides* from Mount Batak from 2015 to 2017. *Craterellus cornucopioides* is in the Rapid Alert System for Food and Feed database for excess radioactivity and strongly accumulates Cs-137 and K-40. It was found that the activity concentrations of Cs-137 in selected mushroom species *Craterellus cornucopioides* in 2015 was 106.13 (Bq kg^{-1} DM), which is 29 years after the Chernobyl accident. In 2016 (30 years after the Chernobyl accident) there was a sharp decline in the activity concentrations of Cs-137 from 106.13 (Bq kg^{-1} DM) to 53.00 (Bq kg^{-1} DM). In 2017 (31 years after the Chernobyl accident) the activity concentrations of Cs-137 was 11.67 (Bq kg^{-1} DM), and according to literature data the Cs-137 was disintegrated after 30.5 years. It was calculated that the effective dose for Cs-137 was in the range of 3.40e-06 per year, and for the K-40 was in the range of 5.70e-05, which was below 0.1% of the natural radioactive background. It was found that cesium half-life is longer than 30.17 years at high altitudes in soils with a low clay content and an acidic reaction.

Keywords: Cs-137 and K-40, effective dose, wild edible mushroom species *Craterellus cornucopioides*, Bulgaria.

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AIMS AND BACKGROUND

The accident in the Chernobyl nuclear power plant (ChNPP) caused the largest uncontrolled radioactive release into the environment dispersed on over the entire northern hemisphere. It was estimated that about 85 PBq of Cs-137 was released from the accident[1]. The radionuclides with long half-life like Cs-137 deposited after the accident will remain in the environment, mainly in the soil, for decades and will be relevant for environmental monitoring. Bulgaria, South Bulgaria in particular, was among the European countries with relatively high contamination as a result of the Chernobyl accident. According to UNSCEAR (Ref. 2), the deposition of Cs-137 on the southern part of Bulgaria is 12 kBq m^{-2}, which is significantly higher than the deposition for countries like France (max 3.2 kBq m^{-2}), Belgium (max 0.84 kBq m^{-2}), Poland (max 5.2 kBq m^{-2}), Hungary (4.8 Bq m^{-2}) and Greece (8 kBq m^{-2}). In the same report Southern Bulgaria is indicated as a territory with Cs-137 deposition density higher than 5 kBq m^{-2}, while for a large part of the European territory it is between 1 and 5 kBq m^{-2}. The activity concentration of man-made radionuclides in the Bulgarian soils increased considerably. Up until now, 30 years later, Cs-137 is still detected in all soil samples and represents a potential danger for the contamination of the plant production through root feeding[3–6]. Interest in the accumulation of radioactive Cs was first aroused much earlier than the Chernobyl disaster. Caesium-137 (and probably Caesium134) was released after the disaster at the graphite reactor at Windscale (now Sellafield) in 1957 and research was done on the presence of this radionuclide in food products. During the 1960s and 1970s extensive research was done on the uptake and accumulation of Cs-137 (Cs-134 was hardly present) that was present in the world-wide fallout from the above ground tests with nuclear weapons. The accumulation of radioactive Cs was first found in lichens and mosses in 1961 and later (in 1963) in fungal fruitbodies[7]. The difference is that lichens and mosses are accumulators of aerial fallout, but that fungi take up the Cs from the substrate on which they grow.

Mushrooms are often considered as excellent bioindicators for evaluation of environmental pollution, since they are known to accumulate metals and other elements[8–13]. Cs-137 in wild mushroom species can be detected consistently, due to atmospheric radioactive fallout in aerosol particle and precipitation form, initially as a result of the explosion of nuclear devices in the atmosphere, and subsequently the Chernobyl nuclear accident in 1986 (Refs 14–19). Cs-137 values in mushrooms can be used to trace and evaluate fallout of radioactive from past and future nuclear accidents. Furthermore, mushrooms are also consumed by man and directly eaten by animals. The European mushrooms in 1986–2015 may have given humans a greater amount of Cs-137 compared to any other kind of food[20]. Regular consumption of some types of mushroom species or animals that eat them may pose a human health concern[8]. The Rapid Alert System for Food and Feed (RASFF) database contains 24 overdose radioactivity signals in wild mushrooms originating in Bulgaria for the period 1998 to 2011 (Ref. 21). Therefore, it is important to have information on radioactivity concentration of mushrooms originated from Bulgaria.

The aim of this study was to evaluate Cs-137 and K-40 activity concentrations and effective doses in *Craterellus cornucopioides* mushroom gathered in Batak Mountain from 2015 to 2017. The time interval is chosen purposely as 2016 marks 30.17 years elapsing from the Chernobyl nuclear disaster, equal to the half-life of Cs-137. Thus, the difference between laboratory and real conditions will be evaluated. The *Craterellus cornucopioides* mushroom was chosen as it is present in the RASFF database for excessive radioactive contamination and substantial accumulation of Cs-137 and K-40.

EXPERIMENTAL

Mushroom samples. The Batak mountain is located in western Rhodopes, Bulgaria. Its western border is defined by the Chepinska river, the southern border – by Dospatska river and Dospat dam, the eastern border – by Vacha river and the northern border – by the Thracian Plane (GPS41°46′02.6″N 24°08′48.4″E) (Fig. 1). The regions is industry-free and is characterised with forests, land and low buildings.

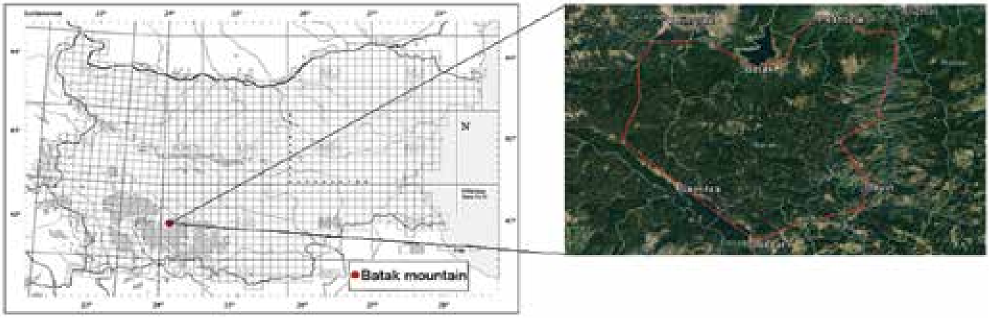

Fig. 1. Location of sampling sites

Mushroom samples from the species *Craterellus cornucopioides* were collected in 2015 and 2017 from the Batak Mountain by the authors themselves. The Batak mountain territory was conditionally divided into 15 regions. From each region, we collected 0.5 kg fresh mushroom. After sampling, the fungal specimens, were immediately transferred to the laboratory, they were cleaned from soil and other impurities first by hand and then washed with tap water and finally rinsed with bidistilled water in order to be perfectly free from residuals. After that, they were dried. The dried samples were ground, then homogenised and stored in polyethylene bottles until analysis. Then, they were ashed at 360°C (> 90% recovery of radionuclides according to ERL treatment protocols, IAEATRS No 118 of 1970 and IAEATRS No 295 of 1989) until constant weight which was measured and the percentage ratio (ashing factor) was estimated. Finally, they were homogenised up to fine powder and enclosed in white cups for immediate gamma measurement.

Reagents. All chemicals were at least of analytical-reagent grade. Water was de-ionised in a Milli-Q system (Millipore, Bedford, MA, USA) to a resistivity of 18.2 MΩ cm. All plastic and glassware were cleaned by soaking in diluted HNO_3 (1/9, v/v) and were rinsed with distilled water prior to use.

Gamma spectrometry. The Laboratory of Radioactivity and Radioisotopes Research at IC FT is accredited by EA 'BAS' for the determination of radioactive elements in soils, plants, foods, food products and waters.

The development and validation of the radioactivity and radionuclides in low-background determination of natural and technogenic gamma-emitters in agricultural objects allows for the simultaneous determination of a large number of radionuclides with energies ranging from 50 to 2000 keV by means of spectrum, produced by semi-conductive Ge detector with high resolution. The used gamma-spectrometric system, produced by Canberra USA, includes the following modules:

– coaxial detector of extremely pure Ge with diameter of the drill 76 mm and relative efficiency 20% and resolution at 661, 4 keV of Cs-137 – 1.3 keV.

– multi-channel analyser (MCA) 'Canberra 85' with low background protective camera of old steel. The working volume is suitable for installing containers, type 'Marinelli' with capacity 0.5 and 1 l.

In this way, the coupled low background system strongly reduces the natural background and allows direct spectrometry of samples.

Potassium-40 activity concentrations were calculated by its single peak at 1460.8 keV and Cs-137 activity concentrations through its only peak at 661.7 keV, in cases where more than two peaks were used, the calculation of the mean value was performed.

Radioactivity units and legislation. One Bq (Becquerel) has been the unit for the activity of a radioactive source in which one atom decays per second on average. Activity concentration, that is activity per dry matter unit, is used in this review. The usual statutory limit for foods has been 600 Bq per kg of fresh weight, i.e. 6 kBq per kg of dry matter for mushrooms. However, in response to the Chernobyl disaster, the European Communities published Council Regulation defining values for the maximum permitted levels of foodstuff radioactive contamination[22]. The regulation was established with a view to responding to accidents of a similar magnitude to the Chernobyl disaster. Under this regulation, the maximum permitted level of Cs-137 for foodstuffs such as mushrooms, was 1.25 kBq kg^{-1} fresh weight (i.e. 12.5 kBq kg^{-1} DM for mushrooms). A similar limit of 1.0 kBq kg^{-1} fresh weight (i.e. 10 kBq kg^{-1} DM for mushrooms) was recommended by the International Atomic Energy Agency[23].

Effective dose. A possible risk of radioactivity for human health is expressed by the effective dose (E) given in mSv (millisievert) per year. The acceptable yearly burden for an adult of the public, recommended by the International Commission for Radiation Protection, has been 5 mSv. A contribution to the yearly effective dose to an adult from mushroom consumption may be calculated as follows[24]:

$$E = Y \times Z \times d_{\mathrm{k}},$$

where Y is the annual intake of mushrooms (kg DM per person): Z – the activity concentration (kBq kg^{-1} DM); d_{k} – the dose coefficient (conversion factor) defined as the dose received by an adult per unit intake of radioactivity. Their values are 1.3 × 10^{-8} and 6.2 × 10^{-9} Sv Bq^{-1} for Cs-137 and K-40, respectively. During an explosive fission reaction many radionuclides are produced, among them Cs-137 with long half-lives 30.17.

Statistical analysis. All analyses were carried out in triplicate and the data were reported as means ± standard deviation (SD). All statistical computing, test and graphics were performed within the statistical software R version 3.5.1 (2018-07-02). The results were analysed for normality through Shapiro-Wilks test (chosen alpha level was 0.05) and density plot. The null-hypothesis of Shapiro-Wilks test is that the population is normally distributed. If p-value > 0.05, then the null hypothesis that the data came from a normally distributed population can not be rejected[25].

RESULTS AND DISCUSSION

The activity concentrations for Cs-137 and Cs-137 in the samples were checked for normality using the Shapiro–Wilks test and density plot (Table 1 and Fig. 2). For the radionuclides K-40 and Cs-137, the hypothesis that they are normally distributed cannot be rejected based on their p-values (Table 1).

Table 1. Descriptive value for Cs-137 and K-40 activity concentration (Bq kg^{-1} DM) in *Craterellus cornucopioides*

Activity concentration	Cs-137			K-40		
Year	2015	2016	2017	2015	2016	2017
Mean	106.13	53.00	11.67	1780.33	1762.80	2060.27
Std. dev.	24.18	12.87	5.45	55.72	69.14	84.85
Median	107.00	54.00	11.00	1795.00	1760.00	2060.00
Minimum	70.00	30.00	4.00	1670.00	1655.00	1950.00
Maximum	150.00	76.00	20.00	1850.00	1884.00	2250.00
Range	80.00	46.00	16.00	180.00	229.00	300.00
Shapiro–Wilks Test	0.971	0.985	0.939	0.905	0.971	0.916
p-value	0.873	0.994	0.372	0.114	0.884	0.166
Effective dose	7.20e-06	3.40e-06	7.92e-07	5.76e-05	5.70e-05	6.67e-05

The present study showed that the concentration of Cs-137 in the selected mushroom species *Craterellus cornucopioides* in 2015, e.g. 29 years after Chernobyl, was 106.13 (Bq kg^{-1} DM). In 2016 (30 years after the Chernobyl disaster), a sharp decline in Cs-137 activity concentrations was noted – from 106.13 (Bq kg^{-1} DM) to 53.00 (Bq kg^{-1} DM). In 2017 (31 years after the Chernobyl disaster) Cs-137 activity concentration was 11.67 (Bq kg^{-1} DM), while literature data show a half-life of radiocesium of 30.17. Time evolution of Cs-137 and K-40 specific activity in *Craterellus cornucopioides* mushrooms in the years 2015–2017 (Fig. 3).

The effective dose per capita in Bulgaria is calculated for average consumption of 50 kg fresh weight or 5 kg dry weight mushrooms. The effective annual dose of Cs-137 was 3.40e-06, while for K-40 was 5.70e-05, e.g. < 0.1% of the natural background radiation. This fact suggests that the consumption of mushrooms, even at the background of high levels of radioactive contamination, has a little contribution to the effective dose for Bulgarian population.

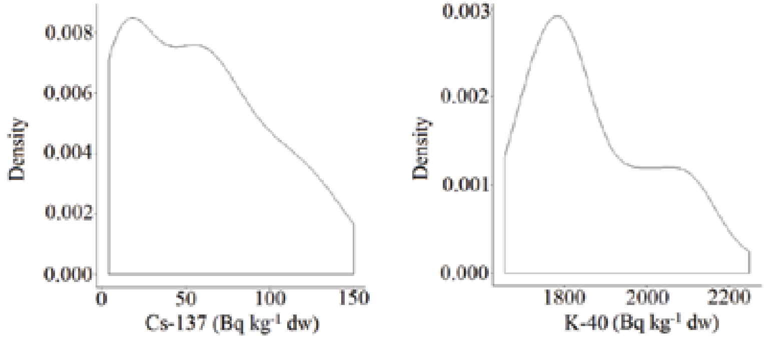

Fig. 2. Density plot of the distribution of Cs-137 and K-40

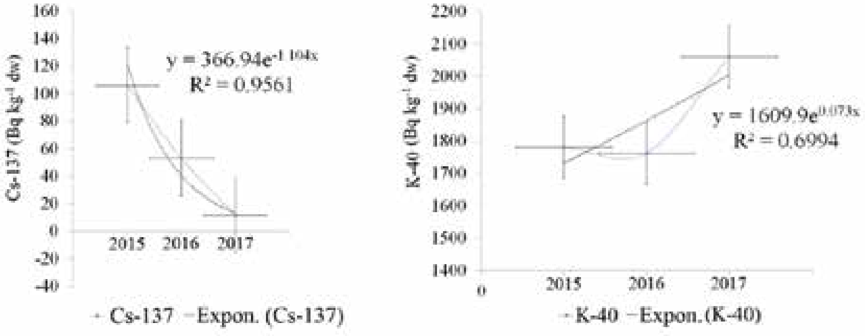

Fig. 3. Time evolution of Cs-137 and K-40 specific activity in *Craterellus cornucopioides* mushrooms in the years 2015–2017

CONCLUSIONS

The mushrooms analysed by us during the three years do not exceed the EU approved standards for Cs-137.

It was calculated that the effective dose for Cs-137 was in the range of 3.40e-06 per year, and for the K-40 was in the range of 5.70e-05, which was below 0.1% of the natural radioactive background.

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