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Caesium radioactivity in mushrooms in Northwest Croatia

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SUMMARY. – After the nuclear accident at Chernobyl, some mushrooms have become the greatest source of radiocaesium among other foodstuffs. ^{137}*Cs concentrations in different species are extremely variable despite relatively little variation in potassium. The excess* ^{137}*Cs from the pre-Chernobyl fallout was found to affect* ^{134}*Cs/*^{137}*Cs concentration ratios in mushrooms. No correlation between the naturally occurring* ^{40}*K and fallout radiocaesium in mushrooms was established. As human consumption of mushrooms in the diet is small, the resulting committed effective dose equivalent would be a few percentage of the dose from natural background radiation.*

INTRODUCTION

Mushrooms tend to accumulate some radionuclides, particulary caesium isotopes. Selective resorption of radiocaesium from the soil is due to the strong preference of mushrooms for the chemically very similar alkali metal potassium which is their principal inorganic constituent. Radioactivity measurements of mushrooms therefore provide a relevant measure of the extent of radioactive contamination of the ecosystem. Pedological characteristics affect the concentration of particular radionuclides in mushrooms. The ions of some elements which are more tightly bound to soil particles are almost immobile. For instance, ^{137}Cs and ^{134}Cs are more tightly bound to soil particles than ^{90}Sr. As a consequence, more than 20 years after the major period of nuclear weapon tests performed in the atmosphere, radiocaesium in uncultivated (undisturbed) soils is still confined to the upper 20 centimeters. By function minimization of experimental data *(1)* it can be shown that the half-value depth of radiocaesium penetration is less than five centimeters which was observed elsewhere *(5)*.

Following the nuclear accident at Chernobyl (1986, April 26) the released radionuclides were dispersed with air masses over Europe. In some parts of Europe, rainfall coincident with the passage of the radioactive cloud caused high wet deposition. In the Zagreb area, deposition in the top 5-cm layer of the soil in May of 1986 was 1.9 kBqm^{-2} ^{131}I, 1.2 kBqm^{-2} ^{103}Ru, 2 kBqm^{-2} ^{137}Cs and 1.0 kBqm^{-2} ^{134}Cs *(2)*. These radionuclides were subsequently found in elevated concentrations in soils, grass and many kinds of plants *(1)*.

MATERIAL AND METHODS

The sampling sites were a microlocation on Mt. Medvednica, North of Zagreb and various locations in Northwest Croatia. Intensive sampling took place in the autumn of 1989 and 1990. Unfortunately, some mushroom species, with the exception of *Rozites caperata* were not available every year.

At each site at least 100 g of mushrooms were collected. Before counting, the soil was carefully removed. Soil cores were collected under the mushroom stalks.

A gamma-ray spectrometry system based on a low-level ORTEC Ge(Li) detector (FWHM 1.82 keV at 1.33 MeV), coupled to a computerized data acquisition system (4096-channel pulse height analyzer and personal computer), was used to determine ^{40}K, ^{134}Cs, ^{137}Cs, ^{226}Ra and ^{235}U levels in the specimens from their gama-ray spectra.

As naturally occurring ^{226}Ra was detected in some species by gammaspectrometry, quantitative results were obtained after radiochemical separation, by alpha spectrometry. ^{235}U activity in *Coprinus comatus* was not only measured by gamma ray spectrometry, but also on the basis of radium data obtained by alpha spectrometry (no radium was found). The alpha spectrometry system used was a semiconductor Si alpha detector (EG & G ORTEC) coupled to a multi-channel analyzer system.

Efficiency calibration was carried out using sources provided by the International Atomic Energy Agency (IAEA) and World Health Organization (WHO). In addition, intercalibrations on samples provided by the IAEA and WHO have been part of the Department's activity for many years.

Samples were measured in cylindrical plastic containers of appropriate volume which were placed directly on the detector.

Counting time depended on sample activity, but was not less than 10 000 seconds.

RESULTS AND DISCUSSION

Mushrooms, as well as lichens and mosses, have been recognized as ideal indicators of radionuclides in the ecosystem. The results of the investigations of radioactive contamination of mushrooms performed in 1989 and 1990 are summarized in Table 1. By that time, ^{131}I, (physical half-life $t_{1/2}$ = 8 days), ^{103}Ru ($t_{1/2}$ = 371.6 days), ^{144}Ce ($t_{1/2}$ = 284.9 days) and some other anthropogenic nuclides released in the atmosphere at the time of the Chernobyl nuclear accident were no longer present in the environment, because of their relatively short physical half-live compared with the elapsed time between the accident and the time of sampling. The caesium radionuclide ^{137}Cs ($t_{1/2}$ = 30.14 years) as the main long-lived component of the radioactive fallout and ^{134}Cs ($t_{1/2}$ = 2.06 years) were found in substantial concentrations in some species.

TABLE 1

^{137}Cs, ^{134}Cs and ^{40}K activities in various species of mushrooms collected in 1989 and 1990 on Mt. Medvedica

Year	Species	Activity as Bqkg^{-1} fresh weight (% error) at date of sampling ^{137}Cs	^{134}Cs	^{40}K
1989	Armillariella mellea	30.5 (13 %)	5.6 (20 %)	472 (13 %)
	Armillariella tabescens	LLD	LLD	400 (13 %)
	Boletus edulis	LLD	LLD	LLD
	Cantharellus cibarius	70.0 (7 %)	17.1 (12 %)	160 (19 %)
	Clitocybe nebularis	76.2 (8 %)	16.1 (16 %)	LLD
	Coprinus comatus	22.4 (13 %)	LLD	201 (19 %)
	Craterellus cornucopioides	82.5 (7 %)	9.4 (17 %)	264 (16 %)
	Hygrophorus russula	1364.0 (2 %)	177.9 (4 %)	255 (16 %)
	Laccaria amethystina	112.0 (16 %)	LLD	LLD
	Lactarius vellereus	39.1 (9 %)	LLD	283 (15 %)
	Lepista nuda	69.2 (8 %)	15.4 (15 %)	278 (16 %)
	Polyporus pes-caprae	LLD	LLD	LLD
	Rozites caperata	1880.0 (2 %)	210.7 (7 %)	LLD
	Russula cyanoxantha	LLD	LLD	LLD
1990	Amantia muscaria	LLD	LLD	LLD
	Agaricus silvicola	26.9 (12 %)	LLD	LLD
	Cantharellus cibarius	50.8 (6 %)	LLD	254 (11 %)
	Hygrophorus russula	371.8 (3 %)	48.3 (8 %)	213 (14 %)
	Laccaria amethystina	77.9 (11 %)	31.8 (16 %)	LLD
	Lactarius vellereus	133.5 (3 %)	10.3 (11 %)	186 (11 %)
	Macrolepiota procera	20.0 (12 %)	LLD	LLD
	Rozites caperata	777.8 (4 %)	83.6 (11 %)	LLD
	Verpa bohemica	7.6 (16 %)	LLD	231 (13 %)

LLD – below lower limit of detection

Great differences in the extent of radiocaesium accumulation by mushrooms depended on the species and the local soil properties. The classification of mushrooms into saprophytes and symbionts reflects the different soil nutrition horizons. Those relate to each species type since different species have different mycelium depth. Because of the great extension of mycelium into the soil, some mushroom species represent contamination of large surfaces and can be used as bioindicators of radiocaesium in pedological horizons. Yearly variations of the contamination were also caused by caesium migration into the soil.

Since potassium and caesium have similar chemical properties, it was interesting to compare their concentrations in mushrooms. No correlation was found between the activities of ^{40}K and radiocaesium, which was also observed elsewhere *(6)*. The observed ^{40}K concentrations ranged from 160 Bqkg^{-1} in *Cantharellus cibarius* to 472 Bqkg^{-1} in *Armilariella mellea*. In other *Armilariella* species, *Armilariella tabescens*, ^{40}K, was present in the amount of 400 Bqkg^{-1}, which was higher than in the other analyzed mushrooms.

The concentration of radiocaesium isotopes in *Boletus edulis, Russula cianoxantha* and some other species was quite low, reflecting a mean mycelium depth at soil horizons below 5 cm. The affinity of these mushrooms for caesium was very low and even mushrooms found on soils with relatively high concentrations of caesium were not active.

The ^{134}Cs/^{137}Cs activity ratio ranged between, 0.08 in *Lactarius vellereus* to 0.41 in *Laccaria amethistina*. The high caesium concentrations and the high ^{134}Cs/^{137}Cs activity ratio in *Laccaria amethistina*, also reported elsewhere *(3)*, reflect a shallow mean depth of the mycelium in the soil, the deeper layers of which contained only pre-Chernobyl ^{137}Cs, and no ^{134}Cs at all.

Table 2 shows ^{137}Cs, ^{134}Cs and ^{40}K activities in the commonest species of mushrooms collected in 1991 at various locations in Northwest Croatia. ^{134}Cs activities were found only in *Rozites caperata*. As before, concentrations of caesium radioisotopes in *Boletus edulis*, commonest edible mushroom, were found to be below the lower limit of detection at all locations.

TABLE 2

^{137}Cs, ^{134}Cs and ^{40}K activities in various species of mushrooms collected in 1991 at locations in Nortwest Croatia

Location	Species	Activity as Bqkg^{-1} fresh weight (% error) at date of sampling ^{137}Cs	^{134}Cs	^{40}K
Ivanec	Amanita muscaria	LLD	LLD	LLD
Čakovec	Boletus edulis	LLD	LLD	LLD
Ivanec	Boletus edulis	LLD	LLD	LLD
Samobor	Boletus edulis	LLD	LLD	LLD
Samobor	Cantharellus cibarius	LLD	LLD	175 (16 %)
Petrinja	Cantharellus cibarius	17.3 (12 %)	LLD	190 (17 %)
Čakovec	Craterellus cornucopioides	36.8 (11 %)	LLD	331 (15 %)
Jastrebarsko	Coprinus comatus	22.4 (13 %)	LLD	201 (19 %)
Ivanec	Lycoperdon perlatum persoon	LLD	LLD	LLD
Klanjec	Macrolepiota procera	10.0 (11 %)	LLD	LLD
Kašina	Rozites caperata	253.2 (3 %)	23.3 (6 %)	LLD
Medvednica	Rozites caperata	543.6 (1 %)	44.7 (2 %)	LLD
Klanjec	Xerocomus badius	12.0 (14 %)	LLD	275 (13 %)

LLD – below lower limit of detection

With the half-life of 2.06 years, in 1986 ^{134}Cs was no longer present in the environment as the consequence of atmospheric nuclear weapon tests. The amount of caesium released after the reactor explosion at Chernobyl was 3.7×10^{16} Bq of ^{137}Cs (13% of total reactor inventory) and 1.9×10^{16} Bq of ^{134}Cs (10% of total reactor inventory) *(7)*. This was more than 3% of radiocaesium that had been released to the environment by all nuclear weapon tests conducted in the atmosphere.

As the half-life of ^{137}Cs is much longer (30.14 years), the ^{134}Cs/^{137}Cs activity ratio decreased from the initial value of 0.5 in May 1986.

Using radioactive decay law and fractions of caesium isotopes released to the atmosphere, it can be shown that the time dependent ^{134}Cs/^{137}Cs activity ratio R(t) decreases according to the equation:

$$R(t) = \frac{1.9\times10^{16}}{3.7\times10^{16}} \times e^{\ln(2)\times t\times\left(\frac{1}{t_1}-\frac{1}{t_2}\right)}$$

where t is the elapsed time after the Chernobyl accident, t_1 and t_2 are physical half-lives for ^{137}Cs and ^{134}Cs.

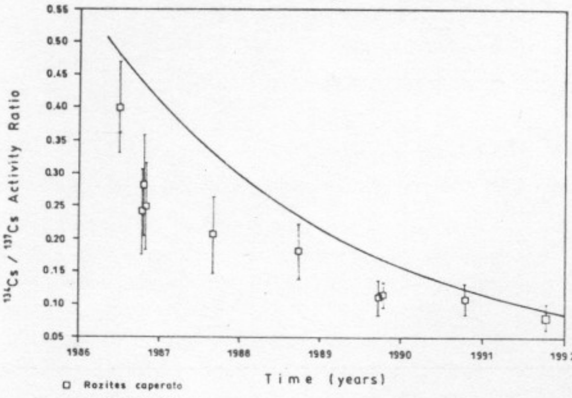

FIGURE 1. *Observed and theoretical activity ratios of ^{134}Cs and ^{137}Cs in Rozites Caperata*. Bars represent standard errors.

Figure 1 shows the observed and theoretical activity ratio of ^{134}Cs and ^{137}Cs in *Rozites caperata*.

The excess ^{137}Cs in soil, from the pre-Chernobyl fallout, affected the ^{134}Cs/^{137}Cs concentration ratios. As ^{134}Cs penetrated to deeper layers of soil, the observed ^{134}Cs/^{137}Cs concentration ratios approached the values theoretically predicted.

The accumulation ratio in *Rozites caperata* found by dividing the radiocaesium specific activity in mushroom by that in adjacent soil showed a significant enrichment of radiocaesium in the mushroom. The species was found to be a strong caesium accumulator with a factor of 4.3 for ^{137}Cs and 4.2 for ^{134}Cs.

In some species natural radionuclides were detected such as ^{226}Ra and ^{235}U. ^{226}Ra was found in *Craterellus cornucopioides* (292 Bqkg^{-1}), *Amanita muscaria* (223 Bqkg^{-1}) and *Lepista nuda* (222 Bqkg^{-1}). Traces of ^{235}U were present in *Coprinus comatus*. In that species uranium was also found at few other sampling sites (Jastrebarsko, Kašina, Zagreb).

Using environmental radioactivity data for Croatia (1), daily intake of caesium by food is estimated to be under 10 Bq. A single meal of mushrooms may increase the normally incorporated content to a considerable degree. Dose conversion factors are $1.3\text{X}10^{-8}$ SvBq^{-1} for ^{137}Cs and $1.9\text{X}10^{-8}$ SvBq^{-1} for ^{134}Cs *(4)*. According to Table 1, if an adult person had eaten 1 kg of *Rozites caperata* in October 1990, he would have received a dose:

$$1\ \text{kg}\times(780\ \text{Bqkg}^{-1}\times1.3\times10^{-8}\text{SvBq}^{-1}+85\ \text{BqKg}^{-1}\times1.9\times10^{-8}\ \text{SvBq}^{-1})\approx12\mu\text{Sv}$$

This value is still two orders of magnitude lower than the allowed dose (1 mSvy^{-1}), i.e. the committed effective dose equivalent due to consumption of mushrooms is very low compared to dose from natural background radiation. The actual incorporation of radiocaesium will depend upon the way of mushroom preparation. By boiling in a salt water they lose about 50% of initial caesium content.

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RADIOCEZIJ U GLJIVAMA SJEVEROZAPADNE HRVATSKE

SAŽETAK. – Jedna je od posljedica nuklearne nesreće u Černobilju i znatna kontaminacija nekih vrsta gljiva radiocezijem. Iako su varijacije koncentracija kalija u ispitivanim gljivama male, koncentracije su ^{137}Cs vrlo različite. Na koncentracijski omjer $^{134}Cs/^{137}Cs$ u gljivama djeluje i ^{137}Cs iz pred-černobiljskih radioaktivnih padalina. U gljivama nije nađena korelacija između prirodnog ^{40}K i radiocezija. Kako je udio gljiva u ljudskoj ishrani malen, rezultirajući godišnji efektivni dozni ekvivalent iznosi nekoliko postotaka od doze primljene uslijed osnovnog zračenja.