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Mosses and Some Mushroom Species as Bioindicators of Radiocaesium Contamination and Risk Assessment

Gordana Marović[1], Zdenko Franić[1], Jasminka Senčar[1], Tomislav Bituh[1] and Ozren Vugrinec[2]

[1] Radiation Protection Unit, Institute for Medical Research and Occupational Health, Zagreb, Croatia,
[2] School of Medicine, University of Zagreb, Zagreb, Croatia

A B S T R A C T

Mosses, lichens, mushrooms are able to efficiently accumulate different radioactive elements from their environment to a much higher degree than other vegetation. They are sensitive bioindicators of radioactive contamination for various ecosystems, particularly in the event of a nuclear accident and uncontrolled emission oh fission products. Results of systematic, long-term measurements of ^{137}Cs activities in mosses and in some edible mushroom species in North Croatia for the post-Chernobyl period (1986–2007) are summarized. The study was conducted in the Radiation Protection Unit of the Institute for Medical Research and Occupational Health in Zagreb, as a part of an extensive monitoring program of the Croatian environment. In the overall observed period the highest activity concentration of ^{137}Cs deposited by fallout has been recorded in 1986, which is the year of Chernobyl accident, causing peak ^{137}Cs activity concentration in moss of 8800 Bq/kg in May 1986. In the same period mean ^{137}Cs activity concentration in grass was 390 Bq/kg. The highest value of ^{137}Cs activity concentration in Cortinarius caperatus mushrooms of 1351 Bq/kg has been recorded in 1989. Fitting the measured ^{137}Cs activity concentrations to the theoretical curve the ecological half-life of ^{137}Cs in moss was found to be around 978 days, in grass around 126 days in the period 1986–1990, in Cortinarius caperatus mushroom around 5865 days (16.1 years). Regarding the risk assessment to Croatian population, due to consumption of mushrooms, the collective effective dose for Croatian population, estimated to be about 35 mSv per year, was found to be quite low. Therefore, it can be concluded that mushroom consumption was not a critical pathway for the transfer of radiocaesium from fallout to humans after the Chernobyl accident.

Key words: *radiocaesium, ecological half-life, risk, moss, grass, mushroom*

Introduction

Mosses, lichens, mushrooms and some perennial plants are able to efficiently accumulate different pollutants from their environment to a much higher degree than other vegetation (vascular plants). These plants also tend to accumulate various radioactive elements, particularly caesium isotopes. This preference for caesium is due to its chemical similarity to potassium, which is their principal inorganic constituent. Consequently, they are sensitive bioindicators of radioactive contamination for various ecosystems[1,2].

In addition, contaminated edible mushroom might pose certain radiological risk if consumed too extensively[3]. One of such mushrooms is *Cortinarius caperatus* in which quite high radiocaesium activity concentrations have been found ever since the Chernobyl accident. Commonly known as »The Gipsy Mushroom«, *Cortinarius caperatus* is edible mushroom of the genus *Cortinarius*, regularly found in Europe and North America. It should be noted that this mushroom has been previously known as *Rozites caperata* before recent genetic study placed it within gens *Cortinarius*.

In the event of a nuclear accident and uncontrolled emission oh fission products, these plant species can be used as bioindicators for monitoring geographical and seasonal distribution of radioactive contamination.

The study of long-term behaviour of ^{137}Cs activity concentrations in mosses and grass and in some edible mushroom species in North Croatia for the post-Cher-

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nobyl period (1986–2007) has been carried out as a part of an extended monitoring program of radioactive contamination of human environment in Croatia[4–7].

Among the other investigated edible mushrooms special attention has been paid to *Boletus edulis* and *Cantharellus cibarius* as those mushrooms are not only widespread, but also a highly esteemed delicacy.

Material and Methods

The samples of moss and grass were collected from the several locations in Croatia. The primary sampling site for mushroom collection is micro-location on Mt. Medvednica north of Zagreb, the capital of Croatia. From this site, 1–2 kg of each mushroom species had been obtained, if it was possible. Mushrooms were cut into small pieces in order to obtain the composite sample and then dried in an oven. The ^{137}Cs and ^{134}Cs activity concentrations are reported as wet weight. Grass and moss samples were dried before the analysis in an oven and usually ashed after that.

All the samples were gamma-spectrometrically analyzed using HPGe and/or Ge(Li) detector (resolution 1.78 keV on 1.33 MeV ^{60}Co, relative efficiency 16.8%; resolution 1.56 keV on 1.33 MeV ^{60}Co, relative efficiency 18.7%) coupled to data acquisition system. The samples were measured in Marinelli beakers of 1 L in volume. Counting time was 80,000 s or longer (depend on sample activities).

Quality assurance and intercalibration measurements were performed through participation in an International Atomic Energy Agency (IAEA), World Health Organization (WHO) and Joint Research Centre (JRC) international intercalibration programs, which also include the regular performance of blank and background as well as quality control measurements.

Results and Discussion

The results of ^{137}Cs activity concentrations in mosses are shown in Figure 1 and ^{137}Cs activity concentrations in grass are shown in Figure 2.

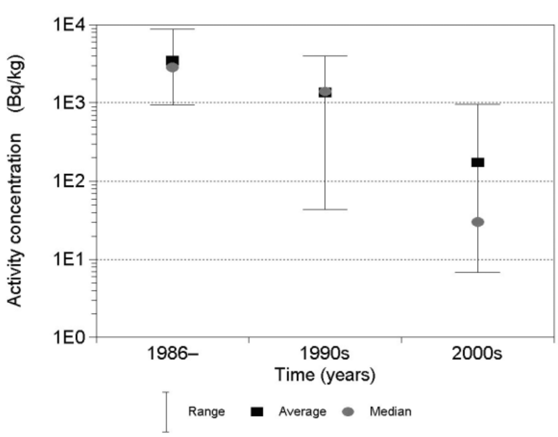

Fig. 1. ^{137}Cs activity concentrations in moss.

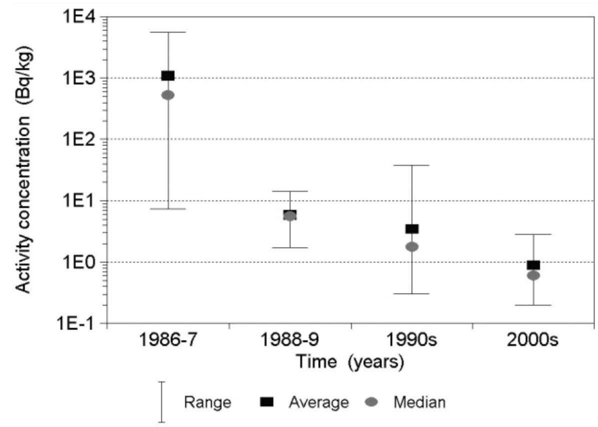

Fig. 2. ^{137}Cs activity concentrations in grass.

In the overall observed period the highest activity concentration of ^{137}Cs deposited by fallout (6410 Bq/m^2) has been recorded in 1986, which is the year of Chernobyl accident, causing peak ^{137}Cs activity concentration in moss (maximum value being 8800 Bq/kg on May 24 and mean value 3150 Bq/kg). For comparison, in the same year mean concentration in grass was 919 Bq/kg, maximum 5500 Bq/kg on June 6.

Radiocaesium activity concentrations in mosses are in good correlation with fallout activities. However, such correlation has not been found for the *Cortinarius caperatus* mushrooms as they extremely efficiently accumulate radiocaesium from the soil, regardless of deposition of fresh fallout.

Ten years after Chernobyl accident, in 1996, mean ^{137}Cs activity concentration in moss was 1345 Bq/kg with maximum value of 3940 Bq/kg, mean ^{137}Cs activity concentration in grass was 3 Bq/kg with maximum value of 38 Bq/kg. In 2006, mean ^{137}Cs activity concentration in mosses was 172 Bq/kg with maximum value of 955 Bq/kg, mean ^{137}Cs activity concentration in grass was 1 Bq/kg with maximum value of 3 Bq/kg.

The highest value of ^{137}Cs activity concentration in *Cortinarius caperatus* mushrooms (1351 Bq/kg) has been recorded in 1989 and gradually decreasing to the value of 189 Bq/kg till 2006. It should be noted, however, that the samples for year 1986 were not available.

Ecological half-life of 137Cs

To study the effective ecological half-life of ^{137}Cs in moss, grass and mushrooms, observed values of ^{137}Cs activity concentrations in analyzed samples have been graphed as a function of time. The distribution in moss and grass showed peak ^{137}Cs activity in the year of the Chernobyl accident. For mushrooms, the samples for the year 1986 were not available.

A first order kinetic equation was used to parameterize time changes in the ^{137}Cs activity concentrations for the period followed by peak activity concentration:

$$A_m(t) = A_m(0)\, e_{-kt} \qquad (1)$$

where:

$A_m(t)$	is time-dependant activity concentration of ^{137}Cs in sample (Bq/kg wet weight),
$A_m(0)$	peak activity concentration of ^{137}Cs in sample (Bq/kg wet weight) and
$ln(2)/k=T_{1/2,eff}$	effective (observed) ecological half-life of ^{137}Cs in moss/grass (years).

As shown in Figure 3, by fitting the measured ^{137}Cs activity concentrations in moss to the theoretical curve (1) the ecological half-life of ^{137}Cs in moss was found to be around 978 days. As shown in Figure 4, by fitting the measured ^{137}Cs activity concentrations to the theoretical curve (1) the bimodal behavior of the ecological half-life of ^{137}Cs in grass has been found. The ecological half-life of ^{137}Cs in moss is around eight times higher than ^{137}Cs ecological half-life observed in grass in the first period after the Chernobyl accident. For the period 1986–1990 the ecological half-life of ^{137}Cs was found to be around 126 days. However, the ecological half-life of ^{137}Cs in grass after 1991 was significantly higher (around 2503 days).

Regarding the mushrooms, the distribution showed peak ^{137}Cs activity (1351 ± 529 Bq/kg wet weight) in 1989, i.e. three years after the Chernobyl accident. This peak has been followed by relatively constant decrease to 179 ± 27 Bq/kg wet weight in 2006.

Due to the unavailability of samples it was not possible to estimate the ecological half-life of ^{137}Cs. However, it can be argued that bimodal behavior is also exhibited in mushroom samples since ^{137}Cs activity concentrations in years 1987 and 1988 (602 and 373 Bq/kg respectively) are much smaller compared to ^{137}Cs activity concentration in year 1989 (1351 Bq/kg). The increase of ^{137}Cs activity concentration in mushroom in year 1989 can be explained by penetration of Chernobyl radiocaesium to deeper soil layers where it was more efficiently absorbed by mycelium.

By fitting the measured ^{137}Cs activity concentrations to the theoretical curve (1) the ecological half-life of ^{137}Cs in *Cortinarius caperatus* mushroom was found to be around 5865 days (16.1 years), which is a little bit more than two times higher than ^{137}Cs ecological half-life observed in grass. The observed and modeled ^{137}Cs activity concentrations in *Cortinarius caperatus* mushroom are shown in Figure 5.

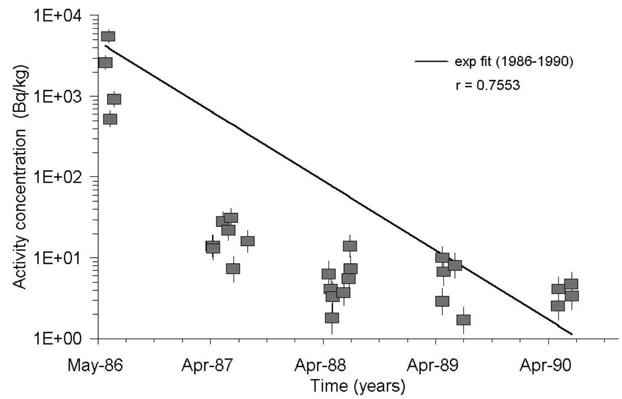

Fig. 4. Observed and modeled ^{137}Cs activity concentrations in grass.

In case of a nuclear accident and uncontrolled emission oh fission products, these species of plant can be used as bioindicators for monitoring geographical and seasonal distribution of radioactive contamination.

134Cs/137Cs activity ratio

In May 1986, the presence of ^{134}Cs radionuclide (half-life of 2.06 years) was detected for the first time in the environment in Croatia. It could be immediately attributed to the Chernobyl nuclear accident, because being a »shielded radionuclide« it is not produced in the nuclear weapon explosions. This means that the ^{134}Xe nuclide that would produce ^{134}Cs by β decay following production in the fission process is, by itself, stable. Therefore, the mass 134 fission product decay chain stops with ^{134}Xe

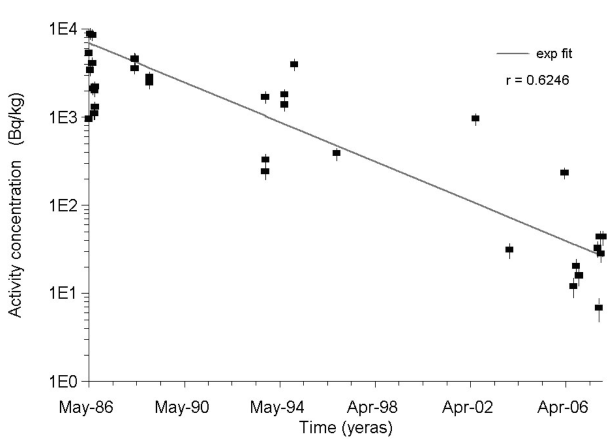

Fig. 3. Observed and modeled ^{137}Cs activity concentrations in mosses.

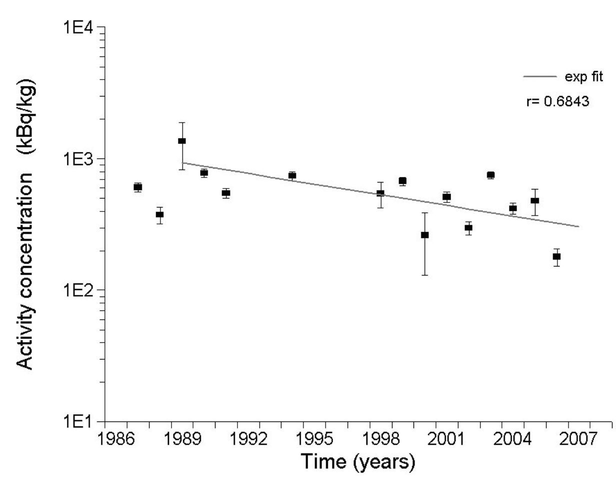

Fig. 5. Observed and modeled ^{137}Cs activity concentrations in Cortinarius caperatus.

and ^{134}Cs is not formed. ^{134}Cs is found, however, in reactor fission product inventories due to long irradiation times for nuclear fuel, which has a typical residence time in the reactor core of 3 years. This permits the build-up of the stable end-product nuclide ^{133}Cs in the core and the corresponding neutron capture by the ^{133}Cs results in the ingrowth of significant quantities of radioactive ^{134}Cs. However, this does not occur in the nuclear weapons blast which is terminated in milliseconds.

The estimated 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[8]. Thus, in the Chernobyl plume the initial value for the $^{134}Cs/^{137}Cs$ activity ratio in May 1986 was 0.51.

The environmental pathways and consequent impact of some chemical elements sometimes depend on chemical or isotope form, (tritium being the most striking example). However, ^{134}Cs and ^{137}Cs have undergone no selective removal in transit between the accident site in Chernobyl and Croatia, as their activity ratio has not been altered along the pathway of radioactive plume.

As the half-life of ^{137}Cs (30.14 years) compared to the half-life of ^{134}Cs is about 15 times longer, the $^{134}Cs/^{137}Cs$ activity ratio, R(t), decreased due to differential radioactive decay, from the initial value of 0.51 according to relation:

$$R(t) = \frac{1.9 \times 10^{16}}{3.7 \times 10^{16}} \times e^{\ln(2) \times t \times (\frac{1}{T_1} - \frac{1}{T_2})} \qquad (2)$$

where:

t is time elapsed after the Chernobyl accident and

T_1 and T_2 are respective physical half-lives for ^{137}Cs and ^{134}Cs.

The similar ratio, decreasing according to equation (2) has been found in most of the other environmental samples[4–7, 9,10].

However, in the case of grass, moss and mushrooms, the situation is more complex. Figure 6 shows $^{134}Cs/^{137}Cs$ activity ratios in grass and moss. It is clearly visible that decrease rate of $^{134}Cs/^{137}Cs$ activity ratio for moss and grass is different, which can be explained by different mechanisms of caesium absorption.

The similar behavior has also been observed in mushrooms[3]. Namely, in mushrooms the excess of ^{137}Cs from the pre-Chernobyl fallout affects $^{134}Cs/^{137}Cs$ concentration ratios. As ^{134}Cs penetrated to deeper layers of soil, the measured $^{134}Cs/^{137}Cs$ concentration ratios approximated the theoretically predicted values.

It could be argued that the results of long-term investigations of radiocaesium activity concentrations in grass, moss and mushrooms show that this biota to be efficiently used as a bioindicator of geographical and seasonal distribution of environmental contamination with anthropogenic, i.e., fission radionuclides.

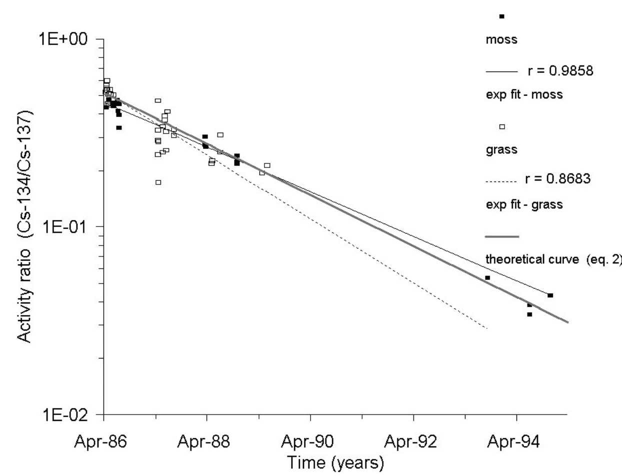

Fig. 6. Observed and theoretically predicted ^{134}Cs:^{137}Cs activity ratios in moss and grass.

Dosimetry

Due to relatively high contribution of ingestion dose to total dose received by general population as a consequence of nuclear fallout, originating from nuclear accidents or bomb tests[11], a reliable knowledge of ingestion dose is of particular importance. Therefore, it is interesting to estimate the upper limit for the effective dose for an adult member of Croatian population due to ^{137}Cs ingestion by consumption of two most common species of edible mushrooms, i.e., *Boletus edulis* and *Cantharellus cibarius*. In 2007, the maximal ^{137}Cs activity concentration in *Cantharellus cibarius* was found to be 20 Bq/kg and 15 Bq/kg in *Boletus edulis*.

Effective dose incurred due to consumption of particular foodstuff over certain time period, depends on the activity of a respective radionuclides present in this foodstuff and on the quantity of food that is consumed. The dose can be expressed as:

$$E = \sum_m C_m \sum_n D_n^{cf} A_{mn} \qquad (3)$$

where:

E is the effective dose in Sv,

C_m total annual *per caput* consumption of given foodstuff m (kg),

D_n^{cf} dose conversion factor for radionuclide n, i.e. effective dose per unit input, which converts the ingested activity to effective dose ($SvBq^{-1}$) and

A_n mean annual specific activity of radionuclide n in foodstuff m ($Bqkg^{-1}$).

For ^{137}Cs dose conversion factor D_n^{cf}, i.e. effective dose per unit intake via ingestion for the member of public older than 17 is 1.3×10^{-8} Sv/Bq [12].

Because the large quantities of those two mushrooms are picked and consumed by individuals, it is very difficult to gain good estimate of the current consumption of *Cantharellus cibarius* and *Boletus edulis* in Croatia. We estimate that in Croatia about 75 tons of each species is annually consumed. Applying equation (2) this yields an upper limit of collective effective dose for Croatian population (4.5 million inhabitants) to be about 35 mSv per year.

Conclusion

Grass, as a widespread plant species, moss and some mushroom species have been recognized as efficient bioindicators of environmental contamination by radionuclides of anthropogenic origin, particularly by radiocaesium isotopes. In addition in many countries, including Croatia, many mushroom species are being regarded as a non-negligible component of overall diet.

For the immediate post-Chernobyl period relatively strong decrease of radiocaesium activity concentrations has been observed in grass followed by much smaller decrease rate. It could be argued that similar bimodal behavior is exhibited in *Cortinarius caperatus* mushroom. However, for mushrooms, due to unavailability of samples, it was not possible to estimate value for the ecological half-life of ^{137}Cs. In grass samples such bimodal behavior has not been clearly observed.

Since radiocaesium activity concentrations in beef are in good correlation with radiocaesium activity concentrations in fallout, this enables development of simple mathematical models as useful tools for quick prediction of meat contamination in the case of nuclear accidents.

Generally, a few years after the Chernobyl nuclear accident the activity concentrations of ^{137}Cs in all analyzed samples were quite low, while ^{134}Cs activity concentrations were below the detection limit of the instruments after the year 1991.

Regarding the risk assessment to Croatian population, due to consumption of mushrooms, the collective effective dose for Croatian population, estimated to be about 35 mSv per year, was found to be quite low.

Therefore, it can be concluded that mushroom consumption was not a critical pathway for the transfer of radiocaesium from fallout to humans after the Chernobyl accident. It should be noted that using simple procedures the efficient decontamination of radiocaesium from mushrooms could be performed. This is of special importance for implementing the protective measures in those regions in which mushrooms are either traditionally esteemed delicacy or important foodstuff, like in some East European countries. The same procedure is planned to be studied in some other mushroom species in Croatia that are still notably contaminated by ^{137}Cs, like *Hydnum repandum* (up to 130 Bq/kg in 2007).

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G. Marović

Institute for Medical Research and Occupational Health, Ksaverska cesta 2, 10000 Zagreb, Croatia
e-mail: marovic@imi.hr

MAHOVINE I NEKE VRSTE GLJIVA KAO BIOINDIKATORI KONTAMINACIJE RADIOCEZIJEM I PROCJENA RIZIKA

S A Ž E T A K

Mahovine su osjetljivi biološki indikacijski organizmi u primjeni biomonitoringa. Pomoću njih se mogu prikupiti vjerodostojne informacije o prostornoj i vremenskoj razdiobi i trendovima onečišćenja zraka i okoliša radioaktivnim tvarima, posebno u uvjetima nuklearne nesreće i nekontrolirane emisije fisijskih produkata. U radu su prikazani rezultati dvadesetogodišnjih istraživanja koncentracija aktivnosti cezija u mahovini, travi i nekim vrstama gljiva. Istraživanja su provedena u Jedinici za zaštitu od zračenja Instituta za medicinska istraživanja i medicinu rada u Zagrebu kao dio proširenog programa praćenja stanja radioaktivnosti uzoraka životne sredine na području Republike Hrvatske. U promatranom razdoblju najviša izmjerena vrijednost koncentracije aktivnosti ^{137}Cs u mahovini bila je 8800 Bq/kg u svibnju 1986. godine. Za usporedbu u istom je razdoblju prosječna koncentracija aktivnosti trave bila devet puta manja 390 Bq/kg, uz maksimalnu vrijednost od 5500 Bq/kg izmjerenu u lipnju 1986. godine. Najviša vrijednost koncentracije aktivnosti ^{137}Cs u gljivi (*Cortinarius caperatus*) od 1351 Bq/kg bila u 1989. godini. Iz izmjerenih vrijednosti koncentracija aktivnosti ^{137}Cs izračunato je i procijenjeno srednje vrijeme boravka (tj. ekološkog vremena boravka) ^{137}Cs u mahovini, travi i gljivama. Procijenjeno ekološko vrijeme boravka ^{137}Cs za mahovine je oko 978 dana, za travu oko 126 dana za prvi period nakon nesreće u Černobilu, dok je ekološko vrijeme boravka ^{137}Cs u gljivama procijenjeno na oko 5865 dana (16,1 godina). Procijenjen je rizik na hrvatsku populaciju procjenom kolektivne efektivne doze od unosa radiocezija gljivama (35 mSv godišnje). Konzumacija gljiva ne predstavlja kritičan put u transferu radiocezija od oborina do čovjeka nakon nuklearne nesreće u Černobilu.