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Uptake of natural and man-made radionuclides by lichens and mushrooms

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Summary. Due to the large absorbing surface of the mycelium that grows in the upper parts of the soil mushrooms take up higher amounts of $137Cs$ and $40K$ than lichens. Besides these nuclides only the long-lived radionuclides 125Sb and 6°Co could be measured; but not the short-lived fission-products $144Ce$, $95Zr$ and $95Nb$ which probably decayed before absorption into the mycelium. These nuclides, however, are present in lichens because of their surface structures which enable high foliar deposition.

The $137Cs$ -content of lichens is probably due to absorption by the mycobiont and seems to be used to satisfy their potassium-requirements. Mushrooms on the other hand are characterized by a relatively stable potassium-content and a wide ranging 137Cs-content which depends on the availability in different substrates. Occasionally the natural radionuclides 238 U and 226 Ra could be detected in mushroom and lichen samples, showing no correlation with the natural radionuclide content of the soil.

Introduction

Lichens have been reported to accumulate a variety of radionuclides and non-radioactive heavy metals (Erämetsä and Yliruokanen 1971; Garty et al. 1977; Larsson 1970; Tuominen 1967; Tuominen and Jaakola 1973). Particularly the behaviour of the long-lived radionuclides $137Cs$, $90Sr$, $239Pu$ and ^{210}Pb was investigated in subarctic ecosystems where they contribute significantly to the radiation burden of people living on reindeer (caribou) meat as a result of the lichen - reindeer - man food chain (Hanson et al. 1966; Hanson 1971; Holm and Persson 1975; Kauranen and Miettinen 1969; Miettinen 1969, 1979; Persson 1973). On the other hand the property of lichens to accumulate stable metals, e.g. lead which originates from fuel combustion, suggests the use of lichens as indicators of environmental **pollu-**

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tion (Erämetsä and Yliruokanen 1971; Garty et al. 1977; Laaksovirta et al. 1969; Nieboer et al. 1978; Rao et al. 1977). The occurrence of lead isotopes in the natural radionuclide decay series prompted us to study the uptake behaviour of lichens for natural radionuclides and to investigate the suitability of lichens as indicators of natural radionuclide contamination.

Lichens represent a symbiosis of algae (blue and green algae) and fungi (mainly asco- but also basidiomycetes). Fungi, namely the edible mushrooms were reported to take up high amounts of 137Cs (Grüter 1967, 1971; Haselwandter 1978; Johnson and Nayfield 1970; Kiefer and Maushart 1965) and 9°Sr (Marah et al. 1962). Thus the accumulation properties of mushrooms were also investigated in order to evaluate the contribution of the fungal partner in the acumulation of radionuclides in the lichen-symbiosis.

In this study, the main emphasis was placed upon the accumulation of radionuclides in fungi and lichens in order (1) to determine transferfactors from substrate to fungi (transfer factors from substrate to lichens were not calculated, for radionuclide uptake in lichens is mainly by dry and wet deposition on overground parts of the lichen thallus) and (2) to reveal accumulation mechanisms in both, fungi and lichens. Initially the problem of natural radionuclides, e.g. 238 U, 226 Ra and 232 Th in an elevated natural radioactive environment, e.g. the Gastein valley (Steinhäusler 1982) was investigated. The high content of fission products in environmental samples originating from the atmospheric nuclear weapons test of the People's Republic of China on October 16, 1980 drew our attention to these man-made radionuclides as well. These may be found in the vicinity of nuclear power stations also.

Material and methods

Lichen and mushroom samples were collected at various locations in Austria, mainly in Salzburg Province and in Lower Austria, and at different times after the Chinese nuclear weapons test. The criterion for the selection of sampling sites was to cover a wide range of natural radionuclide concentrations in soils. In addition, substrate samples were taken from these sites to allow determination of transfer-factors for the most abundant radionuclides, dependent on species and substrate.

Concerning lichens a lot of specimens had to be collected to obtain a final dry weight of about 10 g. This sample volume represents a good cross-section at a given location and consisted of about 30 individual lichens, except for larger species like *Peltigera sp.* or *Lasallia pustulata* where fewer individuals give the required sample weight.

The amount of mushrooms per sample depended on the weight of the mushroom species. From taller and thus heavier species like *Boletus edulis, Leccinum scabrum, Russula* species and *Suillus* species 5 individuals were taken, whereas samples of smaller ones like *Cantharellus* species, *Flammulina velutipes* or *Gomphus clavatus* consisted of 20 or more individuals.

Altogether 250 samples were taken mainly from species as listed in the tables, but also related species of the genera *Suillus, Xerocomus, Boletus,*

Cantharellus, Russula, Lactarius and *Amanita.* As not all samples could be listed a representative selection, consisting of rather common and - except *Russula emetica -* edible mushrooms is presented in this paper.

Lichen samples were air-dried at room temperature, ground in liquid nitrogen, and then dried again at 105° C for 24 h. Mushroom samples were dried at a temperature of 105° C both before and after the grinding procedure. After the determination of the dry weight, the samples were enclosed in 20 ml plastic containers, sealed to prevent the escape of Rn, and then stored for 30 days to obtain radioactive equilibrium of ²²⁶Ra, ²²²Rn and its short-lived daughters.

The activities of natural and man-made radionuclides were measured by gamma ray-spectrometry. The gamma-spectrometry system consists of a lead-shielded Ge(Li)-detector with an anticoincidence plastic detector for additional background reduction and a lead-shielded hyperpure Ge-detector for the low energy region. All samples were measured for 80,000 s. Automatic spectrum evaluation was performed with the aid of a PDP-11/03 computer. The long-lived parents of the Uranium and Thorium decay series were determined directly as well as via their short-lived daughters to check the equilibrium conditions. The pH-values of soils were measured by using the CaClz-method (Deutsche Normen 1977). For the pH-determination at a given site, approximately 30 soil samples were taken randomly from an area of about $\bar{5}$ m², thus yielding a representative average value for this specific site.

Results and discussion

Only an illustrative selection of results is presented in Tables 1, 2 and 3. The criteria for this selection were either to cover a wide range of species or, for a particular species, to illustrate differences in collection sites or time after the last nuclear weapons test. Specific activities listed in both tables refer to the time of sampling and not to the time of the measurement. This applies also to the lower limit of detection which is defined here as the value which has a 100% uncertainty associated with it at the 95% confidence level (Sanderson 1969).

Several differences in the uptake behaviour between lichens and mushrooms could be observed: Generally mushrooms take up higher amounts of $137Cs$ and $40K$ than lichens. The extent depends on species and substrate (Tables 1, 2 and 3). High 137Cs-accumulations were measured in *Xerocomus badius, Paxillus involutus, Suillus variegatus, Rozites caperata,* and *Hydnum repandum,* whereas *Cantharellus cibarius* and *Boletus edulis* showed only little accumulation.

It is evident from Fig. 1 that the transfer of $137Cs$ from soil to fungi is correlated with the pH-value of a given soil.

Like other mineral elements the solubility and mobility of $137Cs$ increases with decreasing pH because the $137Cs$ -ions bound by clay minerals can be exchanged for hydrogen-ions. With increasing pH there is less exchange, $137Cs$ remains bound and is therefore not available for the fungus.

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* Experimental error in percent

Table 2. Radionuclide content of selected mushroom species

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* Experimental error in percent

* Experimental error in percent

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137_{Cs-transfer factors}

Fig. 1.¹³⁷Cs-content of fungi growing on various soil types, characterized by different pH-values

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Substrate (s)	pCi/g dry weight		Species (p)	pCi/g dry weight		Transfer-factor p/s	
	40 _K	$^{137}\mathrm{Cs}$		40 _K	137Cs	40 _K	$^{137}\mathrm{Cs}$
Litter (hardwood)	$0.4(31)*$	2.5(5)	Amanita rubescens Lycoperdon perlatum Russula emetica	9.1(12) 4.5(18) 5.8(27)	(3) 39.0 1.5(29) 56.0 (3)	22.7 11.5 14.5	15.9 0.6 22.8
Calcareous brown loam soil incl. twin mull	0.8 (4)	3.0 ₁ (1)	Boletus luridus Russula cyanoxantha Russula integra	3.0(7) 4.3(11) (9) 3.1	6.5 (3) 25.5 (2) 14.5 (2)	3.3 5.5 3.9	1.6 8.6 4.8
Brown soil	(3) 1.9	2.8 (2)	Boletus edulis Cantharellus cibarius Hydnum repandum Leccinum scabrum	(8) 5.6 7.6 (7) 4.4 (4) 2.8 (7)	8.4 (4) 15.8 (3) 49.4 (1) 2.9 (5)	2.9 4.0 2.3 1.5	3.0 5.6 17.6 1.0
Podsol incl. forest litter	1.3(5)	10.0(1)	Gomphus clavatus Boletus edulis Cantharellus cibarius Hydnum repandum Leccinum scabrum Paxillus involutus Rozites caperata Suillus variegatus	2.6(43) 2.6 (5) (5) 8.9 7.3 (8) 3.5 (3) 7.8 (2) 9.8(16) 3.4 (4)	3.1(32) 1.8 (5) 5.0 (6) 147.0 (1) 9.8 (1) 31.0 (1) 233.0 (1) 23.7 (1)	2,0 2.0 6.8 5.6 2.7 6.0 7.5 2.6	0.3 0.2 0.5 14.7 1.0 3.1 23.3 2.4
Podsol	$0.6\,$ (7)	4.1 (2)	Xerocomus badius Amanita rubescens	(5) 6.3 6.2 (2)	150.5 (1) 14.1 (1)	10.5 10.3	36.6 3.4
Peat soil	0.6(28)	6.2 (3)	Amanita fulva Leccinum scabrum Russula emetica Suillus variegatus	6.6(10) 2.5(7) 6.1(23) 3.3(10)	134.2 (1) 345.4 (0) 575.0 (1) 247.0 (1)	11.0 4.2 10.0 5.5	21.6 52.5 92.7 39.8
Bark	0.7(11)	0.6(10)	Flammulina velutipes	5.9 (3)	1.0 (6)	8.3	1.5

Table 4. Experimental results of the ⁴⁰K and ¹³⁷Cs content of mushrooms in $pCig^{-1}$ dry weight and the resulting transfer factors

* Experimental error in percent

The highest ¹³⁷Cs-activities, corresponding to the highest transfer-factors were measured in mushroom samples from a peatbog near Salzburg city (Table 4). This finding probably depends on the availability of $137Cs$. Normally $137Cs$ ist tightly bound by the clay minerals of the soil, thus there is only slight uptake by the mycelium (Alexander 1967). Peat soils, however, represent humus accumulations that are deficient in minerals, especially clay minerals. $137Cs$ therefore remains in a chemical form that can be taken up more readily. A cofactor for the high accumulation is given by the low pH-values of peat soils which cause the above mentioned better mobility of cations. These two facts are supposed to cause the much higher transfer-factors for $137Cs$ compared to those for mushrooms growing on other soils (for this reason we have divided Fig. I into two distinct areas).

An interesting finding were also the different transfer-factors for podsols in the Mfihl- and Waldviertel, which can be attributed to pH differences.

Fig. 2. Radionuclide concentrations as a function of soil depth (peat soil)

According to current investigations of one of the authors $(R, Türk)$, $SO₂$ imissions could cause this acidifications of soils in this particular part of the Miihlviertel (Liebenau) where the samples were taken, because lichens from that region show the typical phenotype of SO_2 -damage.

Besides 137 Cs and 40 K, also 226 Ra, 7 Be, 238 U, 60 Co and 125 Sb could be detected in mushrooms (Table 2). On the contrary, lichens contained the short-lived fission products ^{144}Ce , ^{95}Zr , ^{95}Nb which originated from the last atmospheric nuclear weapons test carried out at the Chinese testing area Lop Nor on October 16, 1980, and in almost all cases the cosmogenic nuclide ⁷Be, whereas ²³⁸U, ²²⁶Ra and ¹²⁵Sb were not detectable.

In addition to these nuclides, also small amounts of the activation product ⁶⁰Co could be detected in some samples, most probably produced in the Chinese test.

In a previous paper (Eckl et al. 1984) we have shown that different lichen species, collected at the same time in a given area, exhibit differences in accumulation properties dependent on the different ecological requirements. For the short-lived radionuclides listed in Tables I and 2, the measured activities also reflect the different times elapsed since the Chinese nuclear test. In the case of $137Cs$, however, which remains preferentially in the upper part of the soil (see Fig. 2), transfer-factors are not affected by sampling time considerations because of its long half-life.

This apparent difference in the uptake of certain radionuclides can be explained as follows: mushrooms grow in the upper soil layer and take up minerals (nutrients) from the soil solution, the nuclide composition of which is dependent on the soil type. Fission products like ^{137}Cs and ^{125}Sb

having half-lives greater than two years can migrate into the upper soil layers before decaying totally and are therefore available to the mushrooms. The short-lived fission products ¹⁴⁴Ce, ⁹⁵Zr and ⁹⁵Nb, however, decay before being incorporated. Lichens, on the contrary, can take up radionuclides with the substrate solution as well as from deposited aerosols, water vapour and rain. The enormous effective surface of a lichen thallus due to structures for vegetative reproduction such as isidiae, soralae and sorediae increase foliar deposition. This pathway explains the detected content of the shortlived fission products $144Ce$, $95Zr$ and $95Nb$. Since the mycobiont (fungus) is responsible for the uptake of water and dissolved minerals it can be assumed that it is the main source of the lichens' ¹³⁷Cs-content. Further investigations using microautoradiography will prove that assumption.

The ⁴⁰K-content of lichens is low compared to that of soil. Similar results were reported by Hanson and Eberhardt (1971), who explained it as the inability of lichens to take up potassium from the soil. Therefore $137Cs$ as congener of potassium could be accumulated to satisfy the potassium needs. This explanation does not seem to be appropriate concerning mushrooms. Independent of the substrate the ⁴⁰K-content lies within a narrow range $(5.\dot{4} \pm 2.3 \text{ pCig}^{-1}$ dry weight), the ¹³⁷Cs-content, however, shows marked fluctuations $(111.8 \pm 156.6 \text{ pCig}^{-1}$ dry weight). This finding might indicate that mushrooms attempt to satisfy their potassium requirements in any case. The high $137Cs$ -content then could be interpreted as either indiscriminate uptake of mineral elements available or specific needs of elemental Cs.

In some cases 226Ra and 238U could be detected in mushrooms collected in the Waldviertel. This part of Austria is built up by the granites of the Bohemian Mass. Like all acidic plutonites, granites contain relatively high concentrations of nuclides of the natural decay series which pass into the soil by weathering. The uptake into the mycelium is favoured by the fact that soils are mainly podsols which are rather acidic (pH lower than 4.6). As mentioned above, the solubility of mineral elements (Ra is a congener of Ca) increases with decreasing pH-values.

In order to study the vertical distribution of radionuclides in the soil, soil samples were taken on a peatbog near Salzburg city down to a depth of 30 cm. Figure 2 shows that nearly all the $137\overline{Cs}$, but also $210\overline{P}b$ was found in the upper 10 cm. The ^{137}Cs - and ^{210}Pb -content of the overlaying forest-litter (zero-value in Fig. 2) was found to be lower because these nuclides are washed out and transfered to the upper soil layers. $125Sb$ seems to be uniformly distributed. Therefore, if transfer-factors soil-mushroom (plant) are determined, the soil-sample should be taken exactly from that part of the soil in which the mycelia (roots) are growing. This, however, is additionally complicated by the difficult separation of soil and the mycelium. Thus determined transfer-factors will always represent minimum values.

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