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Cesium ($^{137}$Cs and $^{133}$Cs), Potassium and Rubidium in Macromycete Fungi and Sphagnum Plants

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1. Introduction

1.1 Cesium ($^{137}$Cs and $^{133}$Cs), potassium and rubidium in macromycete fungi

Radioesium ($^{137}$Cs) released in the environment as result of nuclear weapons tests in the 1950s and 1960s, and later due to the Chernobyl accident in 1986, is still a critical fission product because of its long half-life of 30 years and its high fission yield. The study of the cesium radioisotope $^{137}$Cs is important, as production and emission rates are much higher than other radioisotopes. This chapter comprises results obtained in several experiments in Swedish forest ecosystems and aims to discuss the behavior of cesium isotopes ($^{137}$Cs and $^{133}$Cs) and their counterparts potassium (K) and rubidium (Rb) in the “soil-fungi-plants transfer” system. The chapter consists of two parts: one mainly dealing with $^{137}$Cs, $^{133}$Cs, K and Rb in forest soil and macromycete fungi, and the other with the same isotopes in separate segments of Sphagnum plants.

The bioavailability of radionuclides controls the ultimate exposure of living organisms and the ambient environment to these contaminants. Consequently, conceptually and methodologically, the understanding of bioavailability of radionuclides is a key issue in the field of radioecology. Soil-fungi-plants transfer is the first step by which $^{137}$Cs enters food chains.

1.1.1 The role of fungi in $^{137}$Cs transfer in the forest

The availability of radionuclides ($^{137}$Cs in particular) in soils of different ecosystems is to a large extent regulated by various vascular plants and fungal species. Thus, the behavior of $^{137}$Cs in forest ecosystems differs substantially from other ecosystems, foremost due to the abundance of fungal mycelia in soil, which contribute to the persistence of the Chernobyl radioesium in the upper horizons of forest soils (Vinichuk & Johanson, 2003). Both saprotrophic and mycorrhizal fungi have key roles in nutrient and carbon cycling processes in forest soils. The mycelium of soil fungi has a central role in breaking down organic matter
and in the uptake of nutrients from soil into plants via the formation of symbiotic mycorrhizal associations (Read & Perez-Moreno, 2003). The fungi facilitate nutrient uptake into the host plant, both as a consequence of the physical geometry of the mycelium and by the ability of the fungi to mobilize nutrients from organic substrates through the action of extracellular catabolic enzymes (Leake & Read, 1997). In addition to acquiring essential macronutrients, mycorrhizal fungi are efficient at taking-up and accumulating microelements (Smith & Read, 1997), this ability results in the accumulation of non-essential elements and radionuclides, particularly $^{137}\text{Cs}$ and can have important consequences for the retention, mobility and availability of these elements in forest ecosystems (Steiner et al., 2002).

Although fungal biomass, in comparison to plant biomass, is relatively low in forest soil (Dighton et al., 1991; Tanesaka et al., 1993), many fungal species accumulate more $^{137}\text{Cs}$ than vascular plants do and $^{137}\text{Cs}$ activity concentrations in many fungi are 10 to 100 times higher than in plants (Rosén et al., 2011). Fungi (particularly sporocarps) accumulate $^{137}\text{Cs}$ against a background of low $^{137}\text{Cs}$ activity concentrations, thus, the contribution of fungi to $^{137}\text{Cs}$ cycling in forest systems is substantial.

Fungi are important in radiocesium migration in nutrient poor and organic rich soils of forest systems (Rafferty et al., 1997). In organic matter, the presence of single strains of saprotrophic fungi considerably enhances the retention of Cs in organic systems (Parekh et al., 2008): $\approx 70\%$ of the Cs spike is strongly (irreversibly) bound (remains non-extractable) compared to only $\approx 10\%$ in abiotic (sterilized) systems. Fungal mycelium may act as a sink for radiocesium (Dighton et al., 1991; Olsen et al., 1990), as it contains 20–30% $^{137}\text{Cs}$ in soil inventories, and as much as 40% of radiocesium can leached from irradiated samples compared to control samples (Guillitte et al., 1994). Mycelium in upper organic soil layers may contain up to 50% of the total $^{137}\text{Cs}$ located within the upper 0-10 cm layers of Swedish and Ukrainian forest soils (Vinichuk & Johanson, 2003). In terms of the total radiocesium within a forest ecosystem, fungal sporocarps contain a small part of activity and may only account for about 0.5% (McGee et al., 2000) or even less $\approx 0.01$ to 0.1% (Nikolova et al., 1997) of the total radiocesium deposited within a forest ecosystem. However, these estimates are based on the assumption radionuclide concentration in fungal sporocarps is similar to that of the fungal parts of mycorrhize (Nikolova et al., 1997). The activity concentration in sporocarps is probably higher than in the mycelium (Vinichuk & Johanson, 2003, 2004) and sporocarps constitute only about 1% of the total mycelia biomass in a forest ecosystem. Due to the high levels of $^{137}\text{Cs}$ in sporocarps, their contribution to the internal dose in man may be high through consumption of edible mushrooms (Kalač, 2001). Consequently, the consumption of sporocarps of edible fungi (Skuterud et al., 1997) or of game animals that consumed large quantities of fungi with high $^{137}\text{Cs}$ contents (Johanson & Bergström, 1994) represents an important pathway by which $^{137}\text{Cs}$ enters the human food system.

The $^{137}\text{Cs}$ activity concentration in edible fungi species has not decreased over the last 20 years ($\text{Suillus variegatus}$) or significantly increased ($\text{Cantharellus} \text{spp.}$) (Mascanzoni, 2009; Rosén et al., 2011).

1.1.2 $^{137}\text{Cs}$, $^{133}\text{Cs}$ and alkali metals in fungi

Although fungi are important for $^{137}\text{Cs}$ uptake and migration in forest systems and since the Chernobyl accident, fungal species may contain high concentrations of radiocesium, the reasons and mechanisms for the magnitude higher concentration of radiocesium in fungi...
Cesium ($^{137}$Cs and $^{133}$Cs), Potassium and Rubidium in Macromycete Fungi and Sphagnum Plants

than in plants remains unclear (Kuwahara et al., 1998; Bystrzejewska-Piotrowska & Bazala, 2008). In addition to radiocesium, fungi effectively accumulate potassium (K), rubidium (Rb) and stable cesium ($^{133}$Cs) (Gaso et al., 2000) and the concentrations of $^{137}$Cs, $^{133}$Cs and Rb in fungal sporocarps can be one order of magnitude higher than in plants growing in the same forest (Vinichuk et al., 2010b).

The chemical behavior of the alkali metals, K, Rb and $^{133}$Cs, can be expected to be similar to $^{137}$Cs, due to similarities in their physicochemical properties, e.g. valence and ion diameter (Enghag, 2000). Potassium is a macronutrient and an obligatory component of living cells, which depend on K+ uptake and K+ flux to grow and maintain life. In radioecology cesium is assumed to behave similarly to potassium. At the cellular level, K is accumulated within cells and is the most important ion for creating membrane potential and excitability. Myttenaere et al. (1993) summarize the relationship between radiocesium and K in forests and suggest the possible use of K as an analogue for predicting radiocesium behavior.

Generally, $^{137}$Cs is positively associated with K concentration across plant species in an undisturbed forest ecosystem, which suggests $^{137}$Cs, stable $^{133}$Cs and K are assimilated in a similar way and the elements pass through the biological cycle together (Chao et al., 2008). Cs influx into cells and its use of K transporters is reviewed by White & Broadley (2000) and potassium transport in fungi is reviewed by Rodríguez-Navarro (2000).

Rubidium is another rarely studied alkali metal, which may be an essential trace element for organisms, including fungi. However, there is scarce information on the concentrations and distribution of Rb in fungi and its behavior in food webs originating in the forest. Rubidium is often used in studies on K uptake and appears to emulate K to a high degree (Marschner, 1995): both K and Rb have the same uptake kinetics and compete for transport along concentration gradients in different compartments of soil and organisms (Rodríguez-Navarro, 2000). The concentrations of K, Rb and $^{133}$Cs have been analyzed in fungal sporocarps (Baeza et al., 2005; Vinichuk et al., 2010b; 2011) and a relation between the uptake of Cs and K has been found (Bystrzejewska-Piotrowska & Bazal, 2008). Cesium uptake in fungi is affected by the presence of K and Rb and the presence of $^{133}$Cs (Gyuricza et al., 2010; Terada et al., 1998). Although in fungal sporocarps, the relationships between these alkali metals and $^{137}$Cs when taken up by fungi and their underlying mechanisms are insufficiently understood, as Cs does not always have high correlation with K and it is suggested there is an alternative pathway for Cs uptake into fungal cells (Yoshida & Muramatsu, 1998).

The correlations between $^{137}$Cs and these alkali metals suggest the mechanism of fungal uptake of $^{133}$Cs and $^{137}$Cs is different from K and that Rb has an intermediate behavior between K and $^{133}$Cs (Yoshida & Muramatsu, 1998). However, this interpretation is based on a few sporocarp analyses from each species, and comprised different ectomycorrhizal and saprotrophic fungal species. Although fungal accumulation of $^{133}$Cs is reported as species-dependent, there are few detailed studies of individual species (Gillet & Crout, 2000). The variation in $^{137}$Cs levels within the same genotype of fungal sporocarps can be as large as the variation among different genotypes (Dahlberg et al., 1997).

Another way to interpret and understand the uptake and relations between $^{137}$Cs, $^{133}$Cs, K and Rb in fungi is to use the isotopic (atom) ratio $^{137}$Cs/$^{133}$Cs. Chemically, $^{133}$Cs and $^{137}$Cs are the same, but the atom abundance and isotopic disequilibrium differ. Among other factors, uptake of $^{133}$Cs and $^{137}$Cs by fungi depends on whether equilibrium between the two isotopes is achieved. An attainment of equilibrium between stable $^{133}$Cs and $^{137}$Cs in the
bioavailable fraction of soils within forest ecosystems is reported Karadeniz & Yaprack (2007) but in cultivated soils, equilibrium between fallout $^{137}\text{Cs}$ and stable $^{133}\text{Cs}$ among exchangeable, organic bound and strongly bound fractions has not reached, even though most $^{137}\text{Cs}$ was deposited on the soils more than 20 years before (Tsukada, 2006).

The important roles fungi play in nutrient uptake in forest soils, in particular its role in $^{137}\text{Cs}$ transfer between soil and fungi, requires better understanding of the mechanisms involved. Although transfer of radioactive cesium from soils to plants through fungi is well researched, there is still limited knowledge on natural stable $^{133}\text{Cs}$ and other alkali metals (K and Rb) and the potential role as a predictor for radiocesium behavior, and less is known about the relationships between $^{133}\text{Cs}$ and other alkali metals (K and Rb) during uptake by fungi. To explore mechanisms governing the uptake of radionuclides ($^{137}\text{Cs}$) data on uptake of stable isotopes of alkali metals (K, Rb, $^{133}\text{Cs}$) by fungal species, and the behavior of the three alkali metals K, Rb and $^{133}\text{Cs}$ in bulk soil, fungal mycelium and sporocarps are required. Therefore, an attempt was made to quantify the uptake and distribution of the alkali metals in the soil–mycelium–sporocarp compartments and to study the relationships between K, Rb and $^{133}\text{Cs}$ in the various transfer steps. Additionally, the sporocarps of ectomycorrhizal fungi *Suillus variegatus* were analyzed to determine whether i) Cs ($^{133}\text{Cs}$ and $^{137}\text{Cs}$) uptake was correlated with K uptake; ii) intraspecific correlation of these alkali metals and $^{137}\text{Cs}$ activity concentrations in sporocarps was higher within, rather than among different fungal species; and, iii) the genotypic origin of sporocarps affected uptake and correlation.

Substantial research in this area has been conducted in Sweden after the fallout from nuclear weapons tests and the Chernobyl accident. Some results are published in a series of several articles in collaboration with Profs K.J. Johanson, H. Rydin and Dr. A. Taylor (Vinichuk et al., 2004; 2010a; 2010b; 2011). This chapter aims to summarize the acquired knowledge from studies in Sweden and to place them in a larger context. The results are summarized and discussed and address the issues of K, Rb and $^{133}\text{Cs}$ concentrations in soil fractions and fungal compartments (Section 1.3); concentration ratios of K, Rb and $^{133}\text{Cs}$ in soil fractions and fungi (Section 1.4); relationships between K, Rb and $^{133}\text{Cs}$ in soil and fungi (Section 1.5); the isotopic (atom) ratios $^{137}\text{Cs}/\text{K}$, $^{137}\text{Cs}/\text{Rb}$ and $^{137}\text{Cs}/^{133}\text{Cs}$ in fungal species (Section 1.6); K, Rb and Cs ($^{137}\text{Cs}$ and $^{133}\text{Cs}$) in sporocarps of a single species (Section 1.7); mechanisms of $^{137}\text{Cs}$ and alkali metal uptake by fungi (Section 1.8); Cs ($^{137}\text{Cs}$ and $^{133}\text{Cs}$), K and Rb in *Sphagnum* plants (Section 2); distribution of Cs ($^{137}\text{Cs}$ and $^{133}\text{Cs}$), K and Rb within *Sphagnum* plants (Section 2.3); mass concentration and isotopic (atom) ratios between $^{137}\text{Cs}$, K, Rb and $^{133}\text{Cs}$ in segments of *Sphagnum* plants (Section 2.4); relationships between $^{137}\text{Cs}$, K, Rb and $^{133}\text{Cs}$, in segments of *Sphagnum* plants (Section 2.5); mechanisms of $^{137}\text{Cs}$ and alkali metal uptake by *Sphagnum* plants (Section 2.6); and conclusions from the Swedish studies (Section 3). Before presenting and discussing results a short description of study area, study design and methods used is presented (section 1.2).

1.2 Study area, study design and methods for results presented

1.2.1 Study area

The K, Rb and $^{133}\text{Cs}$ concentrations in soil fractions and fungal compartments were studied in an area located in a forest ecosystem on the east coast of central Sweden (60°22'N, 18°13'E). The soil was a sandy or clayey till and the humus mainly occurred in the form of mull. A more detailed description of the study area is presented by Vinichuk et al. (2010b).
Sporocarps of ectomycorrhizal fungi *Suillus variegatus* was studied in an area located about 40 km north-west of Uppsala in central Sweden (N 60°08’; E 17°10’). The forest is located on moraine and is dominated by Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*), with inserts of deciduous trees, primarily birch (*Betula pendula* and *Betula pubescens*). The field layer consisted mainly of the dwarf shrubs bilberry (*Vaccinium myrtillus* L.), lingonberry (*Vaccinium vitis-idaea* L.) and heather (*Calluna vulgaris* L.): for details about the area and sampling see Dahlberg et al. (1997).

### 1.2.2 Study design

For studies of K, Rb and $^{133}$Cs concentrations in soil fractions and fungal compartments, samples of soil and fungal sporocarps were collected from 10 sampling plots during September to November 2003. Four replicate soil samples were taken, with a cylindrical steel tube with a diameter of 5.7 cm, from around and directly underneath the fungal sporocarps (an area of about 0.5 m$^2$) and within each 10 m$^2$ area to a depth of 10 cm. Soil cores were divided horizontally into two 5-cm thick layers. Sporocarps of 12 different fungal species were collected and identified to species level, and the $^{137}$Cs activity concentration in fresh material was determined. The sporocarps were dried at 35°C to constant weight and concentrations of $^{133}$Cs, K and Rb were determined.

A selection of dried sporocarps of *S. variegatus* ($n=51$), retained from a study by Dahlberg et al. (1997) on the relationship between $^{137}$Cs activity concentrations and genotype identification, was used. The sporocarps were collected once a week during sporocarp season (end of August through September) in 1994 and were taken from five sampling sites (100 to 1600 m$^2$ in size) within an area of about 1 km$^2$. Eight genotypes with 2 to 8 sporocarps each were tested (in total 32 sporocarps) and are referred to here as individual genotypes. Sporocarps within genotypes were spatially separated by up to 10-12m. All genotypes were used for the estimation of correlation coefficients, but only genotypes with at least four sporocarps were included in the alkali metal analysis. In addition, 19 individual sporocarps with unknown genotype (i.e. not tested for genotype identity) were included: these sporocarps consisted of both the same and different genotypes. The combined set of sporocarps refers to all sporocarps: for further details about the sampling and identification of genotypes see Dahlberg et al. (1997). The $^{137}$Cs activity concentration values corrected to sampling date and expressed as kBq kg$^{-1}$ dry weight (DW) for each sporocarp, as reported by Dahlberg et al. (1997), were used.

### 1.2.3 Methods

For the studies of K, Rb and $^{133}$Cs concentrations in soil fractions and fungal compartments, fungal mycelia were separated from the soil samples (30–50 g, 0–5 cm layer depth) under a dissection microscope (magnification X64) with forceps and by adding small amounts of distilled water to disperse the soil. The prepared fraction of mycelium (30–60 mg DW g$^{-1}$ soil) was not identified to determine of the mycelia extracted from the soil samples and the sporocarps belonged to the same species, as it assumed a majority of the prepared mycelia belonged to the same species as the nearby sporocarps. The method for mycelium preparation is described in Vinichuk & Johanson (2003). Mycelium samples were dried at 35°C to constant weight for determination of K, Rb and $^{133}$Cs.
The soil samples (0–5 cm layer) were partitioned by the method described in Gorbán & Clegg (1996). First, soil was gently sieved through a 2 mm mesh giving a bulk soil fraction. The remaining soil aggregates containing roots were further crumbled and gently squeezed between the fingers: this was called the rhizosphere fraction. The residue (finest roots with adhering soil particles) was called the soil–root interface fraction. Nine samples of bulk soil fraction and mycelium, 12 samples of fungal sporocarps, and six samples of rhizosphere and soil–root interface fraction were analyzed for K, Rb and $^{133}$Cs.

The $^{137}$Cs activity concentrations in the bulk soil samples and sporocarps were determined with calibrated HP-Ge detectors, corrected to sampling date and expressed as Bq kg$^{-1}$ DW. The measuring time employed provided a statistical error ranging between 5 and 10%. For element analyses, a 2.5 g portion of each sample was analyzed by inductively coupled plasma in the laboratories of ALS Scandinavia (Luleå, Sweden) with recoveries 97–101% for K; 97.5–99.4% for Rb, and 93.7–102.5% for $^{133}$Cs. For soil, CRM SO-2 (heavy metals in soil) was used which had no certified values for K, Rb or $^{133}$Cs. Element concentrations in the analyzed fractions are reported as mg kg$^{-1}$ DW.

For element analyses (K, Rb and $^{133}$Cs) of $S$. variegatus sporocarps, aliquots of about 0.3 g of each sample were analyzed by the same technique. Element concentrations are reported as mg kg$^{-1}$ DW and the isotopic ratio of $^{137}$Cs/$^{133}$Cs was calculated with Equations 1 and 2 (Chao et al., 2008):

$$\frac{^{137}\text{Cs}}{^{133}\text{Cs}} = \frac{A}{C} \times \frac{\lambda}{a} \times N \times 10^3 \quad (1)$$

where: $A$ is the $^{137}$Cs radioactivity (Bq kg$^{-1}$); $\lambda$ is the disintegration rate of $^{137}$Cs 7.25 x10$^{-10}$ s$^{-1}$; $a$ is the atomic weight of cesium (132.9); $N$ is the Avogadro number, which is 6.02 x10$^{23}$; and, $^{133}$C and C are the $^{133}$Cs concentration (mg g$^{-1}$). Eq. (1) can be simplified to Eq. 2:

$$\frac{^{137}\text{Cs}}{^{133}\text{Cs}} = 3.05 \times 10^{-10} \times \frac{A}{C} \quad (2)$$

where: $A$ is the $^{137}$Cs activity concentration in Bq kg$^{-1}$ and $^{133}$C is the $^{133}$Cs concentration in mg kg$^{-1}$. Thus, the units of the isotope ratio are dimensionless.

Relationships between K, Rb, $^{133}$Cs and $^{137}$Cs concentrations in different fractions and sporocarps of $S$. variegatus were identified by Pearson correlation coefficients. Correlation coefficients were analyzed in five separate sets of samples: in four sets, all samples had known genotype identity, and in the last set, there was a combined set of samples containing both genotypes that had been tested by somatic incompatibility sporocarps and genotypes that had not been tested. Correlation analyses for genotypes with three or less sporocarps were omitted. All statistical analyses were run with Minitab® 15.1.1.0. (© 2007 Minitab Inc.) software, with level of significance of 5% (0.05), 1% (0.01) and 0.1% (0.001).

### 1.3 K, Rb and $^{133}$Cs concentrations in soil fractions and fungal compartments

K, Rb and $^{133}$Cs concentrations values in soil fractions and fungal compartments are necessary for calculating the concentration ratio at each step of its transfer in the soil-fungi system, differences in the uptake between elements and the relationships. This in turn will be the main reason for the different K, Rb and $^{133}$Cs concentrations observed in sporocarps of various fungal species. Concentrations of K, Rb and $^{133}$Cs in bulk soil were not significantly different from those in the rhizosphere, although the values for all three elements were slightly higher in the rhizosphere fraction (Table 1).
Cesium (\(^{137}\)Cs and \(^{133}\)Cs), Potassium and Rubidium in Macromycete Fungi and Sphagnum Plants

<table>
<thead>
<tr>
<th>Element</th>
<th>Bulk soil</th>
<th>Rhizosphere</th>
<th>Soil root-interface</th>
<th>Fungal mycelium</th>
<th>Fruit bodies</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>642.6 (214.6)a</td>
<td>899.3 (301.4)a</td>
<td>3215 (842.8)b</td>
<td>2 867(727.5)b</td>
<td>43 415 (20 436)b</td>
</tr>
<tr>
<td>Rb</td>
<td>3.9 (2.7)a</td>
<td>5.4 (4.4)a</td>
<td>6.8 (1.7)a</td>
<td>13.8 (6.9)b</td>
<td>253.9 (273.6)b</td>
</tr>
<tr>
<td>(^{133})Cs</td>
<td>0.3 (0.2)a</td>
<td>0.4 (0.3)a</td>
<td>0.2 (0.05)a</td>
<td>0.8 (0.8) a</td>
<td>5.65 (7.1)b</td>
</tr>
</tbody>
</table>

*Means within rows with different letters (a or b) are significantly different (p < 0.001).

Table 1. Mean concentrations of K, Rb and \(^{133}\)Cs (mg kg\(^{-1}\) DW (standard deviation)) in soil fractions and fungi.

Potassium concentrations were higher in both the soil-root interface and fungal mycelium fractions than in the bulk soil and rhizosphere fraction. A comparison of K, Rb and \(^{133}\)Cs concentrations revealed fungal sporocarps accumulated much greater amounts of these elements than mycelium. For example, K concentrations in fungal sporocarps collected from the same plots where soil samples and mycelium were extracted were about 15 times higher than K concentrations found in mycelium. The concentrations of Rb in fungal sporocarps were about 18-fold higher than in corresponding fungal mycelium, and those of \(^{133}\)Cs were about 7-fold higher (Table 1).

Thus, potassium concentration increased in the order bulk soil<rhizosphere<fungal mycelium<soil-root interface<fungal sporocarps and was higher in the soil-root interface fraction and fungi than in bulk soil. The high concentrations of K in fungal sporocarps may reflect a demand for this element as a major cation in osmoregulation and that K is an important element in regulating the productivity of sporophore formation in fungi (Tyler, 1982).

Rb in mycelium was 3.5-fold higher than in bulk soil and 2.5-fold higher than in rhizosphere, and concentrations increased in the order bulk soil<rhizosphere<soil-root interface<fungal mycelium<fungal sporocarps. The concentrations of Rb were slightly higher in the soil-root interface fraction than in bulk soil; thus, fungi appeared to have higher preference for this element, as the accumulation of Rb by fungi, and especially fungal sporocarps, was pronounced. Rubidium concentrations in sporocarps were more than one order of magnitude higher than those in mycelium extracted from soil of the same plots where fungal sporocarps were sampled. The ability of fungi to accumulate Rb is documented: mushrooms accumulate at least one order of magnitude higher concentrations of Rb than plants growing in the same forest (Yoshida & Muramatsu, 1998).

Concentrations of stable cesium varied considerably among samples but no significant differences were found among the different fractions analyzed. Cesium concentrations increased in the order soil-root interface<bulk soil<rhizosphere<fungal mycelium<fungal sporocarps, and were only significantly higher in fungal sporocarps, compared with bulk soil. Stable \(^{133}\)Cs was generally evenly distributed within bulk soil, rhizosphere and soil-root interface fractions, indicating no \(^{133}\)Cs enrichment in those forest compartments. However, \(^{133}\)Cs concentrations in sporocarps were nearly one order of magnitude higher than those found in soil mycelium.

Radioactive \(^{137}\)Cs presented similar to \(^{133}\)Cs behavior, where \(^{137}\)Cs activity increased in the order soil<mycelium<fungal sporocarps (Vinichuk & Johanson, 2003; Vinichuk et al., 2004). The differences between fungal species in their preferences for uptake of \(^{137}\)Cs or stable \(^{133}\)Cs appear to reflect the location of the fungal mycelium relative to that of cesium within the soil profile (Rühm et al., 1997). Unlike \(^{137}\)Cs, stable \(^{133}\)Cs originates from soil; therefore, the
amount of unavailable $^{133}$Cs, compared to the total amount of $^{133}$Cs, in soil presumably higher than that of $^{137}$Cs. As a result, stable $^{133}$Cs is considered less available for uptake as it is contained in mineral compounds and is difficult for fungi or plants to access: the concentration ratio of stable $^{133}$Cs in mushrooms is lower than for $^{137}$Cs (Yoshida & Muramatsu, 1998). The differing behavior of the natural and radioactive forms of $^{133}$Cs may derive from their disequilibrium in the ecosystem (Horyna & Řanad, 1988).

1.4 Concentration ratios of K, Rb and $^{133}$Cs in soil fractions and fungi

The concept of concentration ratios (CR, defined as concentration of the element (mg kg$^{-1}$ DW) in a specific fraction or fungi divided by concentration of the element (mg kg$^{-1}$ DW) in bulk soil) is widely used to quantify the transfer of radionuclides from soil to plants/fungi. This approach allows the estimation of differences in uptake of elements. The elements concentration ratio data followed a similar pattern, but the enrichment of all three elements in fungal material was more evident, particularly in the sporocarps (Table 2).

<table>
<thead>
<tr>
<th>Element</th>
<th>Rhizosphere</th>
<th>Soil root-interface</th>
<th>Fungal mycelium</th>
<th>Fruit bodies</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>1.7 (0.4)</td>
<td>6.1 (1.9)</td>
<td>5.1 (1.4)</td>
<td>68.9 (23.1)</td>
</tr>
<tr>
<td>Rb</td>
<td>1.3 (0.4)</td>
<td>2.7 (1.1)</td>
<td>3.9 (1.1)</td>
<td>121.7 (172.2)</td>
</tr>
<tr>
<td>Cs</td>
<td>1.1 (0.5)</td>
<td>0.8 (0.3)</td>
<td>2.1 (0.9)</td>
<td>39.7 (67.6)</td>
</tr>
</tbody>
</table>

Table 2. Concentration ratios CR (defined as concentration of the element (mg kg$^{-1}$ DW) in the specific fraction divided by concentration of the element (mg kg$^{-1}$ DW) in bulk soil) (mean values (standard deviation)).

Thus, for all three alkali metals studied, the levels of K, Rb, $^{133}$Cs and $^{137}$Cs in sporocarps were at least one order of magnitude higher than those in fungal mycelium (Table 2). The concentration ratios for each element varied considerably between the species sampled. The saprotrophic fungus Hypholoma capnoides had the lowest values and the mycorrhizal fungus Sarcodon imbricatus had the highest. Sporocarp:bulk soil concentration ratios are presented in Table 3. Sarcodon imbricatus accumulates nearly 100 000 Bq kg$^{-1}$ of $^{137}$Cs, giving TF values (defined as $^{137}$Cs activity concentration (Bq kg$^{-1}$ DW) in fungi divided by $^{137}$Cs deposition (kBq m$^{-2}$)) about 22 (Vinichuk & Johanson, 2003). The sporocarps of Sarcodon imbricatus had distinctively high concentration ratios of Rb and $^{133}$Cs than other species analyzed. The mycorrhizal fungus Cantharellus tubaeformis, is another species showing relatively high concentration ratios, particularly for K and Rb. Cantharellus tubaeformis accumulates several tens of thousands Bq kg$^{-1}$ of $^{137}$Cs (Kammerer et al., 1994). Among those with moderate concentration ratios for each element are Boletus edulis, Tricholoma equestre, Lactarius scrobiculatus and Cortinarius spp.

Thus, the levels of K, Rb, $^{133}$Cs and $^{137}$Cs in sporocarps were at least one order of magnitude higher than those in fungal mycelium indicating biomagnification through the food web in forest ecosystems.
Cesium ($^{137}$Cs and $^{133}$Cs), Potassium and Rubidium in Macromycete Fungi and Sphagnum Plants

<table>
<thead>
<tr>
<th>Plot</th>
<th>Species</th>
<th>K</th>
<th>Rb</th>
<th>$^{133}$Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>Boletus edulis</td>
<td>62.7</td>
<td>77.4</td>
<td>37.4</td>
</tr>
<tr>
<td>6</td>
<td>Cantharellus tubaeformis</td>
<td>104.7</td>
<td>109.7</td>
<td>15.5</td>
</tr>
<tr>
<td>7</td>
<td>Cortinarius armeniacus</td>
<td>67.5</td>
<td>69.6</td>
<td>19.2</td>
</tr>
<tr>
<td>5</td>
<td>C. odorifer</td>
<td>71.8</td>
<td>70.9</td>
<td>34.7</td>
</tr>
<tr>
<td>8</td>
<td>C. spp.</td>
<td>90.9</td>
<td>157.2</td>
<td>14.8</td>
</tr>
<tr>
<td>8-10</td>
<td>Hypholoma capnoideas</td>
<td>26.6</td>
<td>13.1</td>
<td>6.9</td>
</tr>
<tr>
<td>1</td>
<td>Lactarius deterrinus</td>
<td>29.9</td>
<td>17.2</td>
<td>2.6</td>
</tr>
<tr>
<td>3</td>
<td>L. scrobiculatus</td>
<td>67.8</td>
<td>26.2</td>
<td>3.7</td>
</tr>
<tr>
<td>6</td>
<td>L. trivialis</td>
<td>77.5</td>
<td>126.9</td>
<td>52.2</td>
</tr>
<tr>
<td>5-7</td>
<td>Sarcodon imbricatus</td>
<td>101.7</td>
<td>675.7</td>
<td>258.8</td>
</tr>
<tr>
<td>2</td>
<td>Suillus granulates</td>
<td>58.6</td>
<td>41.4</td>
<td>14.7</td>
</tr>
<tr>
<td>10-11</td>
<td>Tricholoma equestre</td>
<td>66.6</td>
<td>75.4</td>
<td>15.4</td>
</tr>
</tbody>
</table>

$^{1}$Saprophyte, all other analyzed fungal species are ectomycorrhizal

Table 3. Element concentration ratios (mg kg$^{-1}$ DW in fungi)/(mg kg$^{-1}$ DW in bulk soil) in fungi for fungal sporocarps.

1.5 Relationships between K, Rb and $^{133}$Cs in soil and fungi

Although correlation analysis may be not definitive, it is a useful approach for elucidating similarities or differences in uptake mechanisms of cesium ($^{137}$Cs and $^{133}$Cs), K and Rb: close correlation between elements indicates similarities in their uptake mechanisms. No significant correlations between K in soil and in either mycelium ($r=0.452$, ns) or in sporocarps ($r=0.338$, ns) has been identified and sporocarp Rb and $^{133}$Cs concentrations were unrelated to soil concentrations, however, in mycelium both elements were correlated with soil concentrations (Rb: $r=0.856$, $p=0.003$; Cs: $r=0.804$, $p=0.009$). There was a close positive correlation ($r=0.946$, $p=0.001$) between the K:Rb ratio in soil and in fungal mycelium (Figure 1b) and this relationship was also apparent between soil and sporocarps, but was weak and not significant ($r=0.602$, ns: Figure 1b).

The K:$^{133}$Cs ratio in soil and fungal components had a different pattern: the K:Cs ratio in mycelium was closely positively correlated ($r=0.883$, $p=0.01$) to the K:$^{133}$Cs ratio in soil (Figure 1a), but was relatively weakly and non-significantly correlated to soil in fungal sporocarps. No significant correlations were found between the concentrations of the three elements in fungi, soil pH or soil organic matter content (data not shown). The competition between K, Rb and $^{133}$Cs in the various transfer steps was investigated in an attempt to estimate the relationships between the concentrations of these three elements in soil, mycelium and fungal sporocarps. The lack of a significant correlation between K in soil and in either mycelium or sporocarps indicated a demand for essential K in fungi, regardless of the concentration of this element in soil. Regardless of fungal species, K concentration in fungi appears to be controlled within a narrow range, (Yoshida & Muramatsu, 1998), and supports the claim K uptake by fungi is self-regulated by the internal nutritional requirements of the fungus (Baeza et al., 2004).
The relationships observed between K:Rb and K:Cs ratios in fungal sporocarps and soil mycelia, with respect to the soil in which they were growing (Figure 1), also indicated differences in uptake of these alkali metals by fungi. Although correlation analyses is not the best tool for analyzing the uptake mechanism, the closest positive correlations between K:Rb ratios in fungal mycelium and in soil indicated similarities in the uptake mechanism of these two elements by fungi, although the relationships between K:Cs ratios in soil mycelium and in soil were less pronounced. These findings were in good agreement with the suggestion by Yoshida & Muramatsu (1998) that there might be an alternative pathway for Cs uptake into cells and the mechanism of Cs uptake by fungi could be similar to that for Rb, as Cs does not show a good correlation with K. The high efficiency of Rb uptake by fungi indicates Rb, but not Cs, eventually replaces essential K due to K limitation (Brown & Cummings, 2001) and Rb has the capacity to partially replace K, but Cs does not (Wallace, 1970 and references therein). Forest plants apparently discriminate between K+ and Rb+ in soils and a shortage of K+ favors the uptake of the closely related Rb+ ion (Nyholm & Tyler, 2000), whereas, increasing K+ availability in the system decreases Rb+ uptake (Drobner & Tyler, 1998). These results provided new insights into the use of transfer factors or concentration ratios.

1.6 The isotopic (atom) ratios $^{137}$Cs/K, $^{137}$Cs/Rb and $^{137}$Cs/$^{133}$Cs in fungal species

The isotopic ratios of $^{137}$Cs/K, $^{137}$Cs/Rb and $^{137}$Cs/$^{133}$Cs in the fungal sporocarps belonging to different species were used to interpret the distribution of $^{137}$Cs and the alkali metals in fungi and to provide better understanding of its uptake mechanisms. Measurements of trace levels of stable Cs could be another way of obtaining information about the biological behavior of Cs. To obtain better estimates, the isotopic ratios for fungal sporocarps in this
study (Vinichuk et al., 2010b) were calculated and compared with estimates calculated in similar studies by Yoshida & Muramatsu (1998). Mean values of isotopic ratios of $^{137}\text{Cs}/\text{K}$, $^{137}\text{Cs}/\text{Rb}$ and $^{137}\text{Cs}/^{133}\text{Cs}$ in the fungal sporocarps, and range and correlation coefficients between concentration ratios $^{137}\text{Cs}/^{133}\text{Cs}$ and $\text{K}$, $\text{Rb}$ and $^{133}\text{Cs}$ are presented in Table 4.

<table>
<thead>
<tr>
<th>Data set</th>
<th>n</th>
<th>$^{137}\text{Cs}/\text{K}$</th>
<th>$^{137}\text{Cs}/\text{Rb}$</th>
<th>$^{137}\text{Cs}/^{133}\text{Cs}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vinichuk et al. (2010b), Sweden</td>
<td>12</td>
<td>14.4(1.54–45.4)$\times10^{-13}$</td>
<td>7.8(0.55–30.9)$\times10^{-10}$</td>
<td>4.9(0.30–15.1)$\times10^{-8}$</td>
</tr>
<tr>
<td>Yoshida &amp; Muramatsu (1998), Japan</td>
<td>29</td>
<td>5.2(0.15–23.0)$\times10^{-16}$</td>
<td>3.4(0.14–18.2)$\times10^{-13}$</td>
<td>4.1(1.53–5.94)$\times10^{-9}$</td>
</tr>
</tbody>
</table>

Table 4. Isotopic (atom) ratios of $^{137}\text{Cs}/\text{K}$, $^{137}\text{Cs}/\text{Rb}$, $^{137}\text{Cs}/^{133}\text{Cs}$, correlation coefficients between isotopic ratios $^{137}\text{Cs}/^{133}\text{Cs}$ and mass concentrations of $\text{K}$, $\text{Rb}$ and $^{133}\text{Cs}$ in fungal sporocarps (n = number of sporocarps analyzed).

The activity concentrations of $^{137}\text{Cs}$ in fungal sporocarps were about 13 to 16 orders of magnitude lower than mass concentrations of $\text{K}$, 10 to 13 orders of magnitude lower than mass concentrations for $\text{Rb}$, and 8 to 9 orders of magnitude lower than mass concentrations for $^{133}\text{Cs}$. Isotopic (atom) ratios in the fungal sporocarps collected in Sweden were two-three orders of magnitude narrower than those collected in Japan, which reflected the level of $^{137}\text{Cs}$ concentrations in mushrooms: the median value for all fungi species was 4151 Bq kg$^{-1}$ DW in Swedish forests and 135 Bq kg$^{-1}$ DW in Japanese forests. Isotopic (atom) ratios of $^{137}\text{Cs}/\text{K}$, $^{137}\text{Cs}/\text{Rb}$, $^{137}\text{Cs}/^{133}\text{Cs}$ were variable in both datasets and appeared independent of specific species of fungi. These ratios might reflect the isotopic ratios of soil horizons from which radiocesium is predominantly taken up and be a possible source of the variability in isotopic ratios in fungal fruit bodies. Rühm et al. (1997) used the isotopic ratio $^{134}\text{Cs}/^{137}\text{Cs}$ to localize mycelia of fungal species in situ; alternatively, the isotopic (atom) ratio $^{137}\text{Cs}/^{133}\text{Cs}$ can be used to localize fungal mycelia in situ. However, this approach is only applicable for organic soil layers, which contain virtually no or very little clay mineral to which cesium can bind. The isotopic ratios $^{137}\text{Cs}/^{133}\text{Cs}$ in fruit bodies of fungi were similar to those found in organic soil layers of forest soil (Rühm et al., 1997; Karadeniz & Yaparak, 2007).

The relationships observed between the concentration ratios $^{137}\text{Cs}/^{133}\text{Cs}$ and $\text{K}$, $\text{Rb}$ and $^{133}\text{Cs}$ in fungal sporocarps also varied widely and were inconsistent (Table 4). The concentration of $\text{K}$, $\text{Rb}$ and $^{133}\text{Cs}$ in sporocarps appeared independent of the $^{137}\text{Cs}/^{133}\text{Cs}$ isotopic ratio, suggesting differences in uptake of these alkali metals by fungi and complex interactions between fungi, their host and the environment.
1.7 K, Rb and Cs (\(^{137}\)Cs and \(^{133}\)Cs) in sporocarps of a single species

Most results presented in this Chapter are already published (Vinichuk et al., 2011), and are based on sporocarp analysis of different ectomycorrhizal and saprotrophic fungal species. Fungal accumulation of \(^{137}\)Cs is suggested to be species-dependent, thus, \(^{137}\)Cs activity concentration and mass concentration of K, Rb and \(^{133}\)Cs in fungal sporocarps belonging to the mycorrhizal fungus \(S.\ variegatus\) were analyzed. \(S.\ variegatus\) form mycorrhiza with Scots pine and predominantly occur in sandy, acidic soils and have a marked ability to accumulate radiocesium (Dahlberg et al., 1997): as this is an edible mushroom, high radiocesium contents present some concern with regard to human consumption.

The concentrations of K (range 22.2-52.1 g kg\(^{-1}\)) and Rb (range 0.22-0.65 g kg\(^{-1}\)) in sporocarps of \(S.\ variegatus\) varied in relatively narrow ranges, whereas, the mass concentration of \(^{133}\)Cs had a range of 15.8 to 150.9 kBq kg\(^{-1}\). Both \(^{133}\)Cs and \(^{137}\)Cs had wider ranges than K or Rb within sporocarps from the same genotype or across the combined set of sporocarps (Table 5). The mean of the \(^{137}\)Cs/\(^{133}\)Cs isotopic ratio in the combined set of sporocarps was 2.5 x \(10^{-7}\) (range 8.3 x \(10^{-8}\) and 4.4 x \(10^{-7}\)). The \(^{137}\)Cs/\(^{133}\)Cs isotopic ratios from identified genotypes were site-genotype dependent: the ratio values of genotypes at site 4 were about two-times higher than the ratios of genotypes at site 2 (Table 6).

### Table 5. Potassium, rubidium and cesium (\(^{133}\)Cs) mass concentrations and \(^{137}\)Cs activity concentrations in sporocarps of \(S.\ variegatus\) (DW) from identified and unknown genotypes, where \(n = \) number of sporocarps of each genotype analyzed, \(M = \) mean, \(SD = \) standard deviation, \(CV = \) coefficient of variation.

<table>
<thead>
<tr>
<th>Site-genotype</th>
<th>(n)</th>
<th>K (g kg(^{-1}))</th>
<th>Rb (g kg(^{-1}))</th>
<th>(^{133})Cs (mg kg(^{-1}))</th>
<th>(^{137})Cs (kBq kg(^{-1}))</th>
<th>(%)</th>
<th>(%)</th>
<th>(%)</th>
<th>(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Site Genotype</td>
<td></td>
<td>M</td>
<td>SD</td>
<td>CV</td>
<td>M</td>
<td>SD</td>
<td>CV</td>
<td>M</td>
<td>SD</td>
</tr>
<tr>
<td>Sporocarps with identified genotypes</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2-1</td>
<td>8</td>
<td>30.6</td>
<td>8.06</td>
<td>26.4</td>
<td>0.47</td>
<td>0.12</td>
<td>24.7</td>
<td>12.1</td>
<td>4.23</td>
</tr>
<tr>
<td>2-2</td>
<td>6</td>
<td>28.0</td>
<td>6.99</td>
<td>25.0</td>
<td>0.50</td>
<td>0.07</td>
<td>13.8</td>
<td>16.6</td>
<td>2.19</td>
</tr>
<tr>
<td>4-3</td>
<td>4</td>
<td>28.5</td>
<td>2.13</td>
<td>7.5</td>
<td>0.39</td>
<td>0.16</td>
<td>4.0</td>
<td>6.6</td>
<td>0.44</td>
</tr>
<tr>
<td>4-4</td>
<td>3</td>
<td>33.6</td>
<td>8.60</td>
<td></td>
<td>0.30</td>
<td>0.04</td>
<td>3.0</td>
<td>0.60</td>
<td>0.0</td>
</tr>
<tr>
<td>4-5</td>
<td>2</td>
<td>38.9</td>
<td>2.40</td>
<td></td>
<td>0.36</td>
<td>0.02</td>
<td>3.8</td>
<td>0.04</td>
<td>0.0</td>
</tr>
<tr>
<td>4-6</td>
<td>2</td>
<td>35.2</td>
<td>8.84</td>
<td></td>
<td>0.37</td>
<td>0.11</td>
<td>3.7</td>
<td>2.16</td>
<td>-</td>
</tr>
<tr>
<td>7-7</td>
<td>5</td>
<td>33.7</td>
<td>5.79</td>
<td>17.2</td>
<td>0.34</td>
<td>0.06</td>
<td>17.9</td>
<td>6.7</td>
<td>0.80</td>
</tr>
<tr>
<td>6-8</td>
<td>2</td>
<td>25.4</td>
<td>1.34</td>
<td></td>
<td>0.31</td>
<td>0.03</td>
<td>8.7</td>
<td>2.16</td>
<td>-</td>
</tr>
<tr>
<td>Sporocarps with unknown genotypes</td>
<td>19</td>
<td>33.4</td>
<td>6.69</td>
<td>20.0</td>
<td>0.38</td>
<td>0.08</td>
<td>20.3</td>
<td>7.7</td>
<td>1.97</td>
</tr>
<tr>
<td>Combined set of sporocarps (identified and unknown genotypes)</td>
<td>51</td>
<td>31.9</td>
<td>6.79</td>
<td>21.3</td>
<td>0.40</td>
<td>0.09</td>
<td>23.6</td>
<td>8.7</td>
<td>4.36</td>
</tr>
</tbody>
</table>

\(^{1}\)Site numbering according to Dahlberg et al. (1997), the second figure is a running number of the study’s different genotypes.
Cesium (\(^{137}\text{Cs}\) and \(^{133}\text{Cs}\)), Potassium and Rubidium in Macromycete Fungi and Sphagnum Plants

Identification of genotypes

<table>
<thead>
<tr>
<th>Site-genotype(^1)</th>
<th>Identified genotypes</th>
<th>Unidentified genotypes</th>
<th>Combined set of sporocarps</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2-1 2-2 4-3 4-4 4-5 4-6 7-7 6-8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>M</td>
<td>1.67 1.43 3.16 3.95 2.86 2.43 3.27 2.24 2.62 2.50</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CV (%)</td>
<td>97.1 36.4 10.4 5.1 78.1 29.5 9.2 3.9 20.0 34.6</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^1\)Site numbering according to Dahlberg et al. (1997), the second figure is a running number of the study’s different genotypes.

Table 6. \(^{137}\text{Cs}/^{133}\text{Cs}\) isotopic (atom) ratios in sporocarps of \(S. \text{variegatus}\) from identified genotypes, with unknown genetic belonging, and the two combined groupings, \(x10^{-7}\). M = mean, CV = coefficient of variation.

Similarly, in results obtained from a previous study (Vinichuk et al. 2004) the concentrations of K in sporocarps of \(S. \text{variegatus}\) were not related to the concentrations of \(^{137}\text{Cs}\) (\(r=0.103\)) or \(^{133}\text{Cs}\) (\(r=-0.066\)) in the combined data set (Figure 2: c, b). In contrast, the concentrations of K and Rb were significantly correlated in the combined dataset (\(r=0.505\), Figure 2: a).

Rubidium was strongly correlated with stable \(^{133}\text{Cs}\) (\(r=0.746\)) and moderately correlated with \(^{137}\text{Cs}\) (\(r=0.440\)) and K (\(r=0.505\); Figure 2: d, e, a). Both \(^{133}\text{Cs}\) and \(^{137}\text{Cs}\) were significantly correlated in the combined dataset (Figure 2: f).

The \(^{137}\text{Cs}/^{133}\text{Cs}\) isotopic ratio in the combined dataset was not correlated to K concentration, but correlated moderately and negatively with both \(^{133}\text{Cs}\) (\(r=-0.636\)) and Rb (\(r=-0.500\)) concentrations (Figure 3: a, c, b).

Thus, the study of \(S. \text{variegatus}\) revealed no significant correlations between \(^{133}\text{Cs}\) mass concentration or \(^{137}\text{Cs}\) activity concentration and the concentration of K in sporocarps, either within the whole population or among the genotypes.

Potassium, \(^{133}\text{Cs}\) and \(^{137}\text{Cs}\) within the four genotypes were also not correlated, with one genotype exception (Table 7). However, the exception was conditional due to a one single value. Three of four analyzed sporocarp genotypes had high correlation between K and Rb: the forth was only moderately correlated (Table 7).

However, the correlations between \(^{137}\text{Cs}\) and K and Rb and \(^{133}\text{Cs}\) in the four genotypes were inconsistent (Table 3). Potassium, Rb, \(^{133}\text{Cs}\) and \(^{137}\text{Cs}\) were correlated in genotype 2-1 (due to one single value), whereas, no or negative correlations were found between the same elements/isotopes for the other three genotypes. In two of four genotypes, the \(^{137}\text{Cs}/^{133}\text{Cs}\) isotopic ratio was not correlated with \(^{133}\text{Cs}\), K or Rb; however, there was a negative correlation with Rb in one genotype (2-2) and positive correlation with \(^{133}\text{Cs}\) in another (4-3) (Table 7).

Data obtained for \(S. \text{variegatus}\) supported results from earlier studies (Ismail, 1994; Yoshida & Muramatsu, 1998) on different species of fungi, suggesting cesium (\(^{137}\text{Cs}\) and \(^{133}\text{Cs}\) and K are not correlated in mushrooms. Thus, correlation analysis may be a useful, although not definitive, approach for elucidating similarities or differences in uptake mechanisms of cesium (\(^{137}\text{Cs}\) and \(^{133}\text{Cs}\) and K.

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Fig. 2. Relationship between $^{137}$Cs and K, Rb and $^{133}$Cs concentrations in sporocarps in the combined set of all $S$. variegatus sporocarps (a-f). K:Rb (a); K:$^{133}$Cs (b); K:$^{137}$Cs (c); Rb:$^{133}$Cs (d); Rb:$^{137}$Cs (e); and, $^{133}$Cs:$^{137}$Cs (f). *** p<0.001

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Fig. 3. Relationship between the $^{137}\text{Cs}/^{133}\text{Cs}$ isotopic (atom) ratios ($\times 10^{-7}$) and K, Rb and $^{133}\text{Cs}$ mass concentrations in the combined set of $S$. variegatus sporocarps, (a) $^{137}\text{Cs}/^{133}\text{Cs}:\text{K}$; (b) $^{137}\text{Cs}/^{133}\text{Cs}:\text{Rb}$; and, (c) $^{137}\text{Cs}/^{133}\text{Cs}:^{133}\text{Cs}$. *** $p=0.001$
Table 7. Correlation coefficients between concentrations of potassium, rubidium and cesium (\(^{133}\text{Cs}\) and \(^{137}\text{Cs}\)) in genotypes of \(S.\ variegatus\) with more than four sporocarps analyzed\(^1\).

<table>
<thead>
<tr>
<th>Genotype 2-1 (8 sporocarps)</th>
<th>(^{137}\text{Cs})</th>
<th>K</th>
<th>Rb</th>
<th>(^{133}\text{Cs})</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>0.502</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rb</td>
<td>0.626*</td>
<td>0.966***</td>
<td>0.745*</td>
<td>0.837**</td>
</tr>
<tr>
<td>(^{133}\text{Cs})/(^{137}\text{Cs})</td>
<td>−0.172</td>
<td>−0.058</td>
<td>0.240</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Genotype 2-2 (6 sporocarps)</th>
<th>(^{137}\text{Cs})</th>
<th>K</th>
<th>Rb</th>
<th>(^{133}\text{Cs})</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>−0.472</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rb</td>
<td>−0.658</td>
<td>0.928**</td>
<td>−0.138</td>
<td>0.159</td>
</tr>
<tr>
<td>(^{133}\text{Cs})/(^{137}\text{Cs})</td>
<td>−0.263</td>
<td>−0.352</td>
<td>−0.608</td>
<td>−0.586</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Genotype 4-3 (4 sporocarps)</th>
<th>(^{137}\text{Cs})</th>
<th>K</th>
<th>Rb</th>
<th>(^{133}\text{Cs})</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>−0.531</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rb</td>
<td>0.177</td>
<td>0.696</td>
<td>−0.569</td>
<td>0.182</td>
</tr>
<tr>
<td>(^{133}\text{Cs})/(^{137}\text{Cs})</td>
<td>−0.488</td>
<td>0.163</td>
<td>0.930</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Genotype 7-7 (5 sporocarps)</th>
<th>(^{137}\text{Cs})</th>
<th>K</th>
<th>Rb</th>
<th>(^{133}\text{Cs})</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>−0.562</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rb</td>
<td>−0.472</td>
<td>0.987**</td>
<td>−0.528</td>
<td>−0.404</td>
</tr>
<tr>
<td>(^{133}\text{Cs})/(^{137}\text{Cs})</td>
<td>−0.115</td>
<td>−0.135</td>
<td>−0.345</td>
<td></td>
</tr>
</tbody>
</table>

\(^* p=0.05; \ ** p=0.01; \ *** p=0.001\)

The concentration of K in sporocarps appeared independent of the \(^{137}\text{Cs}/^{133}\text{Cs}\) isotopic ratio in both the whole population (Figure 3) and among the genotypes, with one exception (Table 7). The absence of correlation between \(^{137}\text{Cs}\) (or \(^{133}\text{Cs}\)) and K in fungi may be due to the incorporation of K being self-regulated by the nutritional requirements of the fungus, whereas, incorporation of \(^{137}\text{Cs}\) is not self-regulated by the fungus (Baeza et al., 2004). Although K and cesium (\(^{133}\text{Cs}\) and \(^{137}\text{Cs}\)) concentrations did not correlate within \(S.\ variegatus\), both K\(^+\) and Cs\(^+\) ions may compete for uptake by fungi. In experiments under controlled conditions and with sterile medium (Bystrzejewska-Piotrowska & Bazala, 2008), the competition between Cs\(^+\) and K\(^+\) depends on Cs\(^+\) concentration in the growth medium and on the path of Cs\(^+\) uptake. In studies of Cs uptake by hyphae of basidiomycete \(H.\ vinosophyllum\) when grown on a simulated medium (Ban-Nai et al., 2005), the addition of monovalent cations of K\(^+\), Rb\(^+\), and NH\(_4\)\(^+\) reduced uptake of Cs. In addition, radiocesium transport by arbuscular mycorrhizal (AM) fungi decreases if K concentration increases in a compartment accessible only to AM (Gyuricza et al., 2010), and a higher Cs:K ratio in the nutrient solution increases uptake of Cs by ectomycorrhizal seedlings (Brunner et al., 1996). A noticeable (20-60\%) and long-lasting (at least 17 years) reduction in \(^{133}\text{Cs}\) activity concentration in fungal sporocarps \textit{in situ} due to a single K fertilization of 100 kg ha\(^{-1}\) in a Scots pine forest is reported by Rosén et al., (2011).
The relation between $^{137}\text{Cs}$ and K, and Rb and $^{133}\text{Cs}$ within S. variegatus (Figure 2) was similar to an earlier report on different species of fungi (Yoshida & Muramatsu, 1998). Rubidium concentration in sporocarps was positively correlated with $^{133}\text{Cs}$ and $^{137}\text{Cs}$, but generally negatively correlated with $^{137}\text{Cs}/^{133}\text{Cs}$ isotopic ratio, i.e. a narrower $^{137}\text{Cs}/^{133}\text{Cs}$ ratio in sporocarps resulted in higher Rb uptake by fungi. This ratio may reflect the soil layers explored by the mycelia (Rühm et al., 1997), as fungi have a higher affinity for Rb than for K and cesium (Ban-Nai et al., 2005; Yoshida & Muramatsu, 1998), and Rb concentrations in sporocarps can be more than one order of magnitude greater than in mycelium extracted as fungal sporocarps from soil of the same plots (Vinichuk et al., 2011).

Soil mycelia always consist of numerous fungal species and the intraspecific relationships between soil mycelia and sporocarps has not yet been estimated; however, the development of molecular methods with the ability to mass sequence environmental samples in combination with quantitative PCR may now enable such analysis to be conducted.

Mass concentration of $^{133}\text{Cs}$ and activity concentration of $^{137}\text{Cs}$ have different relations in fungal sporocarps: in three of four genotypes, there was a high correlation, two of which were significant ($r=0.908**$ and $r=0.979$), and there was no correlation in the fourth genotype ($r=0.263$, Table 7), whereas, correlation between $^{133}\text{Cs}$ and $^{137}\text{Cs}$ within the whole population was only moderate ($r=0.605***$ Figure 2). In terms of $^{137}\text{Cs}$ and $^{133}\text{Cs}$ behavior, there would be no biochemical differentiation, but there could be differences in atom abundance and isotopic disequilibrium within the system. Fungi have large spatiotemporal variation in $^{133}\text{Cs}$ and $^{137}\text{Cs}$ content in sporocarps of the same species and different species (de Meijer et al., 1988), and the variation in K, Rb, $^{133}\text{Cs}$ and $^{137}\text{Cs}$ concentrations within a single genotype appeared similar, or lower, than the variation within all genotypes. The results for $^{137}\text{Cs}$ and alkali elements in a set of samples of S. variegatus, collected during the same season and consisting of sporocarps from both different and the same genotype, indicated the variability in concentrations was similar to different fungal species collected in Japan over three years (Yoshida & Muramatsu, 1998).

The relatively narrow range in K and Rb variation and the higher $^{133}\text{Cs}$ and $^{137}\text{Cs}$ variations might be due to different mechanisms being involved. The differences in correlation coefficients between $^{137}\text{Cs}$ and the alkali metals varied among and within the genotypes of S. variegatus, suggesting both interspecific and intrapopulation variation in the uptake of K, Rb, stable $^{133}\text{Cs}$ and, $^{137}\text{Cs}$ and, their relationships could be explained by factors other than genotype identity. The variability in $^{137}\text{Cs}$ transfer depends on the sampling location of fungal sporocarps (Gillett & Crout, 2000), for S. variegatus, these interaction factors might include the spatial pattern of soil chemical parameters, heterogeneity of $^{137}\text{Cs}$ fallout, mycelia location, and heterogeneity due to abiotic and biotic interactions increasing over time (Dahlberg et al., 1997).

Within the combined set of sporocarps the concentration of Rb and $^{137}\text{Cs}$ activity concentration in S. variegatus sporocarps were normally distributed but the frequency distribution of $^{133}\text{Cs}$ and K was not: asymmetry of $^{137}\text{Cs}$ frequency distributions is reported in other fungal species (Baeza et al., 2004; Gaso et al., 1998; Ismail, 1994). According to Gillett & Crout (2000), the frequency distribution of $^{137}\text{Cs}$ appears species dependent: high accumulating species tend to be normally distributed and low accumulating species tend to be log-normally distributed. However, lognormal distribution is almost the default for concentration of radionuclides and is unlikely to be a species-specific phenomenon, as it also occurs in soil concentrations, which implies normal distribution would not be expected, even if large set of samples were analyzed.
1.8 Mechanisms of $^{137}$Cs and alkali metal uptake by fungi

Generally, little is known about the mechanisms involved in the uptake and retention of radionuclides by fungi. Studies of uptake mechanisms and affinity for alkali metals in fungi are scarce, but some results are reviewed by Rodríguez-Navarro (2000). Compared to plants, fungal fruit bodies can be characterized by high $^{137}$Cs, $^{133}$Cs and Rb concentrations and low calcium (Ca) and strontium (Sr) concentrations. In a laboratory experiment with the wood-inhabiting mushroom *Pleurotus ostreatus* (Fr.) Kummer Y-l (Terada et al., 1998), $^{137}$Cs uptake by mycelia decreased with increasing of $^{133}$Cs, K or Rb concentration in the media, and K uptake by mycelia decreased with increasing of $^{133}$Cs concentration. In an experiment with pure cultures of mycorrhizal fungi (Olsen et al., 1990) some species had preference for Cs over K and in the experiments with yeast (Conway & Duggan, 1958), K had preference over Cs and the affinity for alkali metal uptake decreased in the order K$^+$ < Rb$^+$ < Cs$^+$ followed by Na$^+$ and Li$^+$, with a relative ratio of 100:42:7:4:0.5. Fungi (mycelium and sporocarps) have a higher affinity for uptake of Rb and K to Cs, and based on the CR values for fungal sporocarps (Table 3), alkali metal can be ranked in the order Rb$^+$ > K$^+$ > Cs$^+$, with a relative ratio of 100:57:32, which is within the range of 100:88:50 derived by Yoshida & Muramatsu (1998).

The affinity for an alkali metal depends on the nutritional status of the organism, which at least partly explains differences reported between field experiments and laboratory experiments with a good nutrient supply. The mycorrhizal species *Sarcodon imbricatus* was found to be the most efficient in accumulating K, Rb and Cs, which was in agreement with results obtained by Tyler (1982), where a mean CR for Rb in litter decomposing fungus *Collybia peronata* was reported to be 41, and the mean CR for Rb in *Amanita rubescens*, which is mycorrhizal with several tree species, was above 100. However, lower 40K content for mycorrhizal species is reported by Römmelt et al. (1990), which means mycorrhizal species do not necessarily accumulate alkali metals more efficiently than saprotrophic ones.

Accumulation of stable and radioactive cesium by fungi is apparently species-dependent but is affected by local environmental conditions. According to de Meijer et al. (1988), the variation in concentrations of stable and radioactive cesium in fungi of the same species is generally larger than the variation between different species and the variation in $^{137}$Cs levels within the same genet of *S. varegatus* is as large as within non-genet populations of the species (Dahlberg et al., 1997), suggesting both interspecific and intrapopulation variation in the uptake of K, Rb, stable $^{133}$Cs and $^{137}$Cs, and that their relationships can be explained by factors other than genotype identity (Vinichuk et al., 2011). There is about two orders of magnitude variation in Cs uptake, with the highest CR value in e.g. *S. imbricatus* (256) and the lowest in *Lactarius deterrimus* (2.6), although other studies (Seeger & Schweinshaut, 1981) report the highest accumulation of stable Cs is in *Cortinarius* sp.

2. Cs ($^{137}$Cs and $^{133}$Cs), K and Rb in Sphagnum plants

2.1 Introduction

Peatlands are areas where remains of plant litter have accumulated under water-logging as a result of anoxic conditions and low degradability of the plant material. They are generally nutrient-poor habitats, particularly temperate and boreal bogs in the northern hemisphere, in which peat formation builds a dome isolating the vegetation from the surrounding groundwater. Hence, bogs are ombrotrophic, i.e. all water and nutrient supply to the vegetation is from aerial dust and precipitation, resulting in an extremely nutrient-
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poor ecosystem often formed and dominated by peat mosses (\textit{Sphagnum}). \textit{Sphagnum}-dominated peatlands with some groundwater inflow (i.e. weakly minerotrophic ‘poor fens’) are almost as nutrient poor and acid as true bogs. \textit{Sphagnum} plants absorb and retain substantial amounts of fallout-derived radiocesium, and some attention has been given to the transfer of the radioactive cesium isotope \(^{137}\text{Cs}\) within raised bogs (Bunzl & Kracke, 1989; Rosén et al., 2009), and relatively high \(^{137}\text{Cs}\) bioavailability to bog vegetation and mosses in particular are found (Bunzl & Kracke, 1989).

The transfer of \(^{137}\text{Cs}\) within a peatland ecosystem is different from that in forest or on agricultural land. In soils with high clay content, there is low bioavailability and low vertical migration rate of radiocesium due to binding to some clay minerals (Cornell, 1993). In nutrient-poor but organic-matter-rich forest soils, the vertical migration rate of \(^{137}\text{Cs}\) is also low, but bioavailability is often high, particularly for mycorrhizal fungi (Olsen et al., 1990; Vinichuk & Johansson, 2003; Vinichuk et al., 2004; 2005). In forests and pastures, extensive fungal mycelium counteracts the downward transport of \(^{137}\text{Cs}\) by an upward translocation flux (Rafferty et al., 2000); this results in a slow net downward transport of \(^{137}\text{Cs}\) in the soil profile.

In peatlands, \(^{137}\text{Cs}\) appears to move through advection in peat water (review by Turetsky et al., 2004). Small amounts of clay mineral in the peat reduce Cs mobility (MacKenzie et al., 1997), but most \textit{Sphagnum} peat is virtually clay mineral free organic matter. In wet parts of open peatlands that lack fungal mycelium, the downward migration of \(^{137}\text{Cs}\) in the \textit{Sphagnum} layers is expected to be faster than in forest soil and Cs is continuously translocated towards the growing apex of the \textit{Sphagnum} shoots, where it is accumulated. Some attempts have been made to investigate whether \(^{137}\text{Cs}\) is associated with essential biomacromolecules in mosses and to determine the \(^{137}\text{Cs}\) distribution among intracellular moss compartments (Dragović et al., 2004).

The chemical behavior of radiocesium is expected to be similar to that of stable \(^{133}\text{Cs}\) and other alkali metals, i.e. K, Rb, which have similar physicochemical properties. Moreover, stable \(^{133}\text{Cs}\) usually provides a useful analogy for observing long-term variation and transfer parameters of \(^{137}\text{Cs}\) in a specific environment, particularly in peatlands that are cut off from an input of stable Cs from the mineral soil. As the relationship between K and Rb in fungi is not clearly understood, whether Cs follows the same pathways as K in \textit{Sphagnum} is also unclear.

Thus, the \(^{137}\text{Cs}\) activity concentration and mass concentration of K, Rb and \(^{133}\text{Cs}\) was analyzed within individual \textit{Sphagnum} plants (down to 20 cm depth) growing on a peatland in eastern central Sweden and its distribution in the uppermost capitulum and subapical segments of \textit{Sphagnum} mosses were compared to determine the possible mechanisms involved in radiocesium uptake and retention within \textit{Sphagnum} plants.

Additionally, the isotopic (atom) ratios of \(^{137}\text{Cs}/\text{K}, ^{137}\text{Cs}/\text{Rb} and ^{137}\text{Cs}/^{133}\text{Cs}\) within individual \textit{Sphagnum} plants were recorded for determining the distribution of \(^{137}\text{Cs}\) and alkali metal, and to obtain a better understanding of the uptake mechanisms and the biological behavior of \(^{137}\text{Cs}\) in nutrient-poor \textit{Sphagnum} dominated ecosystem. There are few studies on the influence of alkali metals (K, Rb, \(^{133}\text{Cs}\)) on \(^{137}\text{Cs}\) distribution and cycling processes in peatlands.

Plant species growing on peat have varying degree capacities for influencing uptake and binding of radionuclides, but no systematic study has covered all the dominant species of \textit{Sphagnum} peatlands their competition for radionuclides and nutrients. The important role of \textit{Sphagnum} mosses in mineral nutrient turnover in nutrient-poor ecosystems, in particular
their role in $^{137}$Cs uptake and binding, necessitates a clear understanding of the mechanisms involved.

The general aim was to gain better insight into mechanisms governing the uptake of both radionuclides ($^{137}$Cs) and stable isotopes of alkali metals (K, Rb, $^{133}$Cs) by Sphagnum mosses. The specific aim was to compare the distribution of $^{137}$Cs, K, Rb and $^{133}$Cs in the uppermost capitulum and subapical segments of Sphagnum mosses to be able to discuss the possible mechanisms involved in radiocesium uptake and retention within Sphagnum plants. Most results obtained in this study are published in collaboration with Prof. H. Rydin (Vinichuk et al., 2010a).

2.2 Study area and methods

2.2.1 Study area

The study area was a small peatland (Palsjömossen) within a coniferous forest in eastern central Sweden, about 35 km NW of Uppsala (60°03′40″N, 17°07′47″E): the peatland area sampled was open and Sphagnum-dominated (Figure 4). A weak minerotrophic influence was indicated by the dominance of Sphagnum papillosum, and the presence of Carex rostrata, Carex pauciflora and Menyanthes trifoliata (fen indicators in the region). The area had scattered hummocks, mostly built by Sphagnum fuscum, and was dominated by dwarf-shrubs such as Andromeda polifolia, Calluna vulgaris, Empetrum nigrum and Vaccinium oxycoccos. Sampling was within a 25 m$^2$ low, flat ‘lawn community’ (Rydin & Jeglum, 2006) totally covered by $^{137}$Cs

2.2.2 Methods

Samples of individual Sphagnum shoots that held together down to 20 cm were randomly collected in 2007 (May and September) and 2008 (July, August and September). Thirteen samples of Sphagnum plants were collected and analyzed; three in 2007 and 10 sets in 2008. Each sample consisted of approximately 20–60 individual Sphagnum plants (mostly S. papillosum, in a few cases S. angustifolium or S. magellanicum). In the laboratory, the fresh, individual, erect and tightly interwoven Sphagnum plants were sectioned into 1 cm (0–10) or 2 cm (10–20 cm) long segments down to 20 cm from the growing apex. The $^{137}$Cs activity concentrations were measured in fresh Sphagnum segments. Thereafter, the samples were dried at 40°C to constant weight and analyzed for K, Rb and $^{133}$Cs. The activity concentration (Bq kg$^{-1}$) of $^{137}$Cs in plant samples was determined by calibrated HP Ge detectors. Statistical error due to the random process of decay ranged between 5 and 10%. Plant material was measured in different geometries filled up, except a few samples that contained about 1 g of dry material. All $^{137}$Cs activity concentrations were recalculated to the sampling date and expressed on a dry mass basis. The analysis of Sphagnum segments for K, Rb and Cs was by a combination of ICP-AES and ICP-SFMS techniques at ALS Scandinavia AB. For K concentration determination, ICP-AES was used and for $^{133}$Cs and Rb, ICP-SFMS was used. The detection limits were 200 mg kg$^{-1}$ for K, 0.04 mg kg$^{-1}$ for $^{133}$Cs and 0.008 mg kg$^{-1}$ for Rb. The isotopic (atom) ratio of $^{137}$Cs/$^{133}$Cs was calculated with Equations 1 and 2 (Chao et al., 2008). Relationships between K, Rb and $^{133}$Cs concentrations in different Sphagnum segments were determined by Pearson
correlation coefficients. All statistical analyses were with Minitab (© 2007 Minitab Inc.) software.

Fig. 4. The study area of peatland, Palsjömossen: Sphagnum-dominated bog.

2.3 Distribution of Cs (\(^{137}\)Cs and \(^{133}\)Cs), K and Rb within Sphagnum plants
Concentration values of Cs (\(^{137}\)Cs and \(^{133}\)Cs) and neighboring alkali counterparts K and Rb in different segments of plant provide information on differences in their uptake, distribution and relationships. The averaged \(^{137}\)Cs activity concentrations in Sphagnum segments are presented in Figure 5a. Within the upper 10 cm from the capitulum, \(^{137}\)Cs activity concentration in Sphagnum plants was about 3350 Bq kg\(^{-1}\), with relatively small variations. Below 10-12 cm, the activity gradually declined with depth and in the lowest segments of Sphagnum, \(^{137}\)Cs activity concentrations was about 1370 Bq kg\(^{-1}\).

For individual samples, K concentrations ranged between 508 and 4970 mg kg\(^{-1}\) (mean 3096); Rb ranged between 2.4 and 31.4 mg kg\(^{-1}\) (mean 18.9) and \(^{133}\)Cs ranged between 0.046 and 0.363 mg kg\(^{-1}\) (mean 0.204): averaged concentrations of K, Rb and \(^{133}\)Cs in Sphagnum segments are presented in Figure 5b. Concentrations of Rb and \(^{133}\)Cs were constant in the upper 0-10 cm segments of Sphagnum moss and gradually declined in the lower parts of the plant length; whereas, the concentration of K decreased with increasing depth below 5 cm. Generally, the distribution of all three alkali metals was similar to \(^{137}\)Cs, but with a weaker increase of Rb towards the surface. The \(^{137}\)Cs activity concentrations had the highest coefficient of variation (standard deviation divided by the
mean) in *Sphagnum* (43%). The coefficients of variation were 35% for K, 35% for Rb and 37% for $^{133}$Cs concentrations.

Two important features should be mentioned when discussing distributions of K, Rb, $^{133}$Cs and $^{137}$Cs in a *Sphagnum*-dominated peatland. Firstly, this type of peatland is extremely nutrient-poor, where only a few plant and fungal species producing small fruit bodies can grow and no mycorrhiza, except ericoid mycorrhiza, exists. Secondly, the upper part of the stratigraphy is composed of living *Sphagnum* cells that selectively absorb mineral ions from the surrounding water, and the binding of K, Rb and $^{133}$Cs can be at exchange sites both outside and inside the cell.

![Graph](image)

Fig. 5. $^{137}$Cs and alkali metals in *Sphagnum*: (a) average $^{137}$Cs activity concentration (kBq kg$^{-1}$) in *Sphagnum* segments (+/− SE, n = 13); (b) average concentrations of K (scale values should be multiplied by 10$^3$), Rb (x10$^3$) and $^{133}$Cs (x10$^{-1}$) (mg kg$^{-1}$) in *Sphagnum* segments (+/− SE, n=4).

The distribution of $^{137}$Cs within *Sphagnum* plants was similar to stable K, Rb and $^{133}$Cs. The $^{137}$Cs activity concentrations and K, Rb and $^{133}$Cs concentrations were always highest in the uppermost 0-10 cm segments of *Sphagnum* (in the capitula and the subapical segments) and gradually decreased in older parts of plant. Such distribution could be interpreted as dependent on the living cells of capitula and living green segments in the upper part of *Sphagnum*. Similar patterns of K distribution within *Sphagnum* plants are reported (Hájek, 2008). $^{137}$Cs is taken up and relocated by *Sphagnum* plants in similar ways to the stable alkali metals, as the ratios between K, Rb, Cs and $^{137}$Cs in *Sphagnum* segments (Figure 6) were
much the same down to about 16 cm, and displayed a slightly different pattern in the lower part of the plant.

2.4 Mass concentration and isotopic (atom) ratios between $^{133}$Cs, K, Rb and $^{137}$Cs, in segments of Sphagnum plants

Ratios between mass concentrations of all three alkali metals and $^{137}$Cs activity concentrations, i.e. $^{133}$Cs/$^{137}$Cs; K/$^{137}$Cs, Rb/$^{137}$Cs and $^{133}$Cs/$^{137}$Cs, were constant through the upper part (0-16 cm) of Sphagnum plants (Figure 6). The ratio K/Rb was higher in the uppermost (0-2 cm) and the lowest (18-20 cm) parts of the plant (Figure 6).

![Fig. 6. Ratios between K:$^{137}$Cs, Rb:$^{137}$Cs, K:Rb and Cs:$^{137}$Cs](image)

However, the isotopic (atom) ratios between $^{137}$Cs activity concentrations and mass concentrations of alkali metals, i.e. $^{137}$Cs/K, $^{137}$Cs/Rb and $^{137}$Cs/$^{133}$Cs, had distinctively different pattern of distribution through the upper part (0-20 cm) of Sphagnum plants (Figure 7). The $^{137}$Cs/K ratio was relatively narrow through the upper part (0-16 cm) of Sphagnum plants and wider with increasing depth, whereas, the $^{137}$Cs/$^{133}$Cs ratio was fairly constant through the upper part (0-12 cm) of Sphagnum plants and becomes narrower in the lower (14-20 cm) parts. The $^{137}$Cs/Rb ratio was constant through the middle part (4-16 cm) of Sphagnum plants and somewhat narrower in the uppermost (0-4 cm) and lowest (16-20 cm) parts (Figure 7).

The distribution of the isotopic (atom) ratios between $^{137}$Cs activity concentrations and mass concentrations of alkali metals K and Rb through the upper part (0-20 cm) of Sphagnum plants are probably conditioned by at least three processes: physical decay of $^{137}$Cs atoms
with time; attainment of equilibrium between stable $^{133}\text{Cs}$ and $^{137}\text{Cs}$ in the bioavailable fraction of peat soil; and, relation between cesium ($^{133}\text{Cs}$ and $^{137}\text{Cs}$), K and Rb when taken up by the Sphagnum plant.

Fig. 7. Isotopic (atom) ratios $^{137}\text{Cs}/^1\text{K}$ (scale values should be multiplied by $10^{-12}$), $^{137}\text{Cs}/^\text{Rb}$ ($x10^{-09}$), and $^{137}\text{Cs}/^{133}\text{Cs}$ ($x10^{-07}$) in Sphagnum segments. Calculations based on $^{137}\text{Cs}$ activity concentrations and mass concentrations of K, Rb $^{133}\text{Cs}$ (Eq. 2) (mean values, $n=4$ for each of $^{137}\text{Cs}$, K, Rb and $^{133}\text{Cs}$).

### 2.5 Relationships between $^{133}\text{Cs}$, K, Rb and $^{137}\text{Cs}$ in segments of Sphagnum plants

Relationships between $^{133}\text{Cs}$, K, Rb and $^{137}\text{Cs}$ in separate segments of Sphagnum plants is a tool allowing future investigate its uptake mechanism. There were close positive correlations between K, Rb and $^{133}\text{Cs}$ mass concentrations and $^{137}\text{Cs}$ activity concentrations in Sphagnum segments (Table 8). Correlation between $^{137}\text{Cs}$ activity concentrations and Rb mass concentrations ($r=0.950$; $p=0.001$) and correlation between K and Rb mass concentrations ($r=0.952$; $p=0.001$) in 10-20 cm length of Sphagnum plants were highest, but $^{137}\text{Cs}$ and K had a weaker correlation only when the upper 0-10 cm part of Sphagnum plants were analyzed ($r=0.562$; $p=0.001$). $^{137}\text{Cs}/^{133}\text{Cs}$ isotope (atom) ratios and mass concentrations of alkali metals (K, Rb and $^{133}\text{Cs}$) were not or negatively correlated (Table 8).

The marked decrease in $^{137}\text{Cs}$ activity concentration below 14 cm (Figure 5a) raises the question as to at what depth the 1986 Chernobyl horizon was when the sampling was done. A peat core was sampled in May 2003 at Åkerlänna Römosse, an open bog about 14 km SW of Pålsjömossen, by van der Linden et al. (2008). Detailed dating by $^{14}\text{C}$ wiggle-matching indicated the Chernobyl horizon was then at a depth of 17 cm. Depth-age data estimated a linear annual peat increment of 1.3 cm yr$^{-1}$ over the last decade ($R^2=0.998$), indicating the Chernobyl horizon would be at about 23 cm deep when the $^{137}\text{Cs}$ sampling was done in 2007-08. Even if there are uncertainties in applying data from different peatlands, the Chernobyl horizon should be at, or below, the lowest segments sampled. Thus, an upward migration of $^{137}\text{Cs}$ was obvious, but no downward migration could be tested in the study.
The relatively unchanged $^{137}\text{Cs}/K$, $^{137}\text{Cs}/\text{Rb}$ and $^{137}\text{Cs}/^{133}\text{Cs}$ isotopic (atom) ratios in the upper 0-14 cm part of Sphagnum plant and the noticeable widening below 14-16 cm supported this assumption. An upward migration of $^{137}\text{Cs}$ has been observed in earlier studies (Rosén et al., 2009); similarly, most $^{137}\text{Cs}$ from the nuclear bomb tests from 1963 was retained in the top few cm of Sphagnum peat 20 years after, but there was also a lower peak at the level where the 1963 peat was laid down (Clymo, 1983): Cladonia lichens also retain high activity concentrations in the shoot apices.

<table>
<thead>
<tr>
<th></th>
<th>$^{137}\text{Cs}$</th>
<th>K</th>
<th>Rb</th>
<th>$^{133}\text{Cs}$</th>
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<tr>
<td>0-10 cm length</td>
<td></td>
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<tr>
<td>K</td>
<td>0.562***</td>
<td></td>
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<tr>
<td>Rb</td>
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<td>0.792***</td>
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<td>10-20 cm length</td>
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<tr>
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<tr>
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<tr>
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<td>-</td>
<td>0.122</td>
<td>0.219</td>
<td>-0.401</td>
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</table>

Table 8. Correlation coefficients between concentrations of potassium, rubidium and cesium ($^{133}\text{Cs}$ and $^{137}\text{Cs}$) in Sphagnum segments (** p=0.001).

2.6 Mechanisms of $^{137}\text{Cs}$ and alkali metal uptake by Sphagnum plants

Presumably, $^{137}\text{Cs}$ is bound within capitula, living green segments and dead brown segments of Sphagnum plants. According to Gstoettner and Fisher (1997), the uptake of some metals (Cd, Cr, and Zn) in Sphagnum papillosum is a passive process as they living and dead moss accumulate metal equally. For a wide range of bryophytes, Dragović et al. (2004) found $^{137}\text{Cs}$ was primarily bound by cation exchange, with only a few percent occurring in biomolecules. Sphagnum mosses have remarkably high cation exchange capacity (Clymo, 1963), and according to Russell (1988), a high surface activity of Sphagnum is related to its high cation exchange capacity, which ranges between 90-140 meq/100 g. In a water saturated peat moss layer, water washes (1 L de-ionised water added to a column of about 1.4 L volume) removed about 60% of K from Sphagnum (Porter B. Orr, 1975), indicating this element was held on cation exchange sites. In turn, the desiccation of living moss usually causes cation leakage from cell cytoplasm, during which most of the effused K$^+$ is retained on the exchange sites and reutilized during recovery after rewetting (Brown & Brümelis, 1996; Bates, 1997).

However, this is not necessarily the case for $^{137}\text{Cs}$, as $^{137}\text{Cs}$ has a weaker correlation with K, especially in the uppermost parts of the plant, which means $^{137}\text{Cs}$ uptake might be somewhat different from that of K. Even within the same segments of the plant, $^{137}\text{Cs}$ activity concentrations has higher variation than K concentration. An even stronger decoupling between $^{137}\text{Cs}$ and K is observed in the forest moss Pleurozium schreberi, in which $^{137}\text{Cs}$ is retained to a higher degree in senescent parts (Mattsson & Lidén, 1975). However, close correlations, were found between Rb and $^{137}\text{Cs}$, which suggests similarities in their
uptake and relocation: these observations complied with results reported for fungi (Vinichuk et al., 2010b; 2011).

Some lower parts of Sphagnum plants are still alive and able to create new shoots (Högström, 1997), however, although still connected to the capitulum, much of lower stem is dead. Thus, the decrease of $^{137}$Cs activity concentration in plant segments below 10 cm indicates a release of the radionuclide from the dying lower part of Sphagnum and internal translocation to the capitulum.

The mechanism of radiocesium and alkali metal relocation within Sphagnum is probably the same active translocation as described for metabolites by Rydin & Clymo (1989). Although external buoyancy-driven transport (Rappoldt et al., 2003) could redistribute $^{137}$Cs, field evidence suggests buoyancy creates a downward migration of K (Adema et al., 2006); thus, this mechanism appears unlikely. Likewise, a passive downwash and upwash (Clymo & Mackay, 1987) cannot explain accumulation towards the surface.

3. Conclusions from the Swedish studies

The concentrations of the three stable alkali elements K, Rb and $^{133}$Cs and the activity concentration of $^{137}$Cs were determined in various components of Swedish forests – bulk soil, rhizosphere, soil–root interface fraction, fungal mycelium and fungal sporocarps. The soil–root interface fraction was distinctly enriched with K and Rb, compared with bulk soil. Potassium concentration increased in the order bulk soil < rhizosphere < fungal mycelium < soil–root interface < fungal sporocarps, whereas, Rb concentration increased in the order bulk soil < rhizosphere < soil–root interface < fungal mycelium < fungal sporocarps. Cesium was generally evenly distributed within bulk soil, rhizosphere and soil–root interface fractions, indicating no $^{133}$Cs enrichment in these forest compartments.

The uptake of K, Rb and $^{133}$Cs during the entire transfer process between soil and sporocarps occurred against a concentration gradient. For all three alkali metals, the levels of K, Rb and $^{133}$Cs were at least one order of magnitude higher in sporocarps than in fungal mycelium. Potassium uptake appeared to be regulated by fungal nutritional demands for this element and fungi had a higher preference for uptake of Rb and K than for Cs. According to their efficiency of uptake by fungi, the three elements may be ranked in the order $\text{Rb}^+ > \text{K}^+ > \text{Cs}^+$, with a relative ratio 100:57:32. Although the mechanism of Cs uptake by fungi could be similar to that of Rb, uptake mechanism for K appeared to be different. The variability in isotopic (atom) ratios of $^{137}$Cs/K, $^{137}$Cs/Rb and $^{137}$Cs/$^{133}$Cs in the fungal sporocarps suggested they were independent on specific species of fungi. The relationships observed between concentration ratios $^{137}$Cs/$^{133}$Cs and K, Rb and $^{133}$Cs in fungal sporocarps also varied widely and were inconsistent. The concentration of K, Rb and $^{133}$Cs in sporocarps appeared independent of the $^{137}$Cs/$^{133}$Cs isotopic ratio.

The study of S. variegatus sporocarps sampled within 1 km² forest area with high $^{137}$Cs fallout from the Chernobyl accident confirmed $^{137}$Cs and $^{137}$Cs uptake is not correlated with uptake of K; whereas, the uptake of Rb is closely related to the uptake of $^{133}$Cs. Furthermore, the variability in $^{137}$Cs and alkali metals (K, Rb and $^{133}$Cs) among genotypes in local populations of S. variegatus is high and the variation appears to be in the same range as found in species collected at different localities. The variations in concentrations of K, Rb and $^{133}$Cs and $^{137}$Cs activity concentration in sporocarps of S. variegatus appear to be influenced more by local environmental factors than by genetic differences among fungal genotypes.

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For *Sphagnum* the distribution of $^{137}$Cs can be driven by several processes: cation exchange is important and gives similar patterns for monovalent cations; uptake/retention in living cells; and downwash and upwash by water outside the plants. However, the most important mechanism is internal translocation to active tissue and the apex, which can explain the accumulation in the top layer of the mosses.

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5. References


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The book Radioisotopes - Applications in Physical Sciences is divided into three sections namely: Radioisotopes and Some Physical Aspects, Radioisotopes in Environment and Radioisotopes in Power System Space Applications. Section I contains nine chapters on radioisotopes and production and their various applications in some physical and chemical processes. In Section II, ten chapters on the applications of radioisotopes in environment have been added. The interesting articles related to soil, water, environmental dosimetry/tracer and composition analyzer etc. are worth reading. Section III has three chapters on the use of radioisotopes in power systems which generate electrical power by converting heat released from the nuclear decay of radioactive isotopes. The system has to be flown in space for space exploration and radioisotopes can be a good alternative for heat-to-electrical energy conversion. The reader will very much benefit from the chapters presented in this section.

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