LONG TERM STUDY OF Cs-137 CONCENTRATIONS IN LICHENS AND MOSSES*

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Several studies carried out after the Chernobyl nuclear accident in 1986 showed that lichens and mosses are suitable bioindicators of the fall-out, given their long life expectancy. Cs-137 activities were measured in different mosses, epigeic and epiphytic lichens, collected from three sampling sites from Koppl, in the Salzburg province and the effective and biological half-time of Cs-137 were determined. The concentrations of Cs-137 ranged from 1765±13 to 7345±33 Bq kg^{-1} dry weight in selected lichens (*Cladonia fimbriata, Cladonia squamosa, Pseudevernia furfuracea, Hypogymnia physodes)* and from 1145±16 to 14092±46 Bq kg^{-1} dry weight in moss species (*Leucobryum glaucum, Sphagnum papillosum, Dicranodontium denudatum, Polytrichum strictum, Sphagnum fallax*). Earlier measurements on the same lichens and mosses species, sampled from the same spots by one of the authors (R.Türk), allow us to determine the biological half-time of ^{137}Cs for these perennial plants. For the epiphytic lichen, *Pseudevernia furfuracea,* it was found a biological half-time of 12.9±0.9 years.

Key words: Cs-137, lichens, mosses, biological half-time, transfer factors.

1. INTRODUCTION

A number of studies carried out before and after the Chernobyl accident, have demonstrated that lichens and mosses can accumulate high amounts of radioactivity (Hviden and Lillegraven 1961; Eckl *et al.* 1984; Hofmann *et al.* 1987, 1993 and 1995; Papastefanou *et al.* 1988 and 1992; Sawidis *et al.* 1988; Hanson 1967). Interest in the behaviour of radionuclides in natural ecosystems was first

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developed during the 1960s, at the time of weapons testing in the atmosphere, because of the observed transfer of Cs-137 along the lichen-reindeer-man-food chain.

Gorham (1959), followed by Hviden and Lillegraven (1961), in northern Norway and Eckl *et al.* (1984 and 1986) in Austria, reported for the first time that lichens are much more efficient in accumulating radiocesium than higher plants, hence representing suitable bioindicators of the radioactive fallout. Although mosses do not constitute an important element in the forest food chain, they are considered as an important object for radiological research. Mosses are able to trap and retain a large share of wet radioactive deposition (Sawidis 1987), since (i) they do not possess roots and thus accumulate mainly in a passive way, (ii) they do not have an epidermis or a cuticle, (iii) they have a high absorbing power and (iv) a large surface-volume ratio. Dragovic (2004) found that membranes of the mosses are the primary Cs-137-binding sites at the cellular level and 26.1%-43.1% of the cesium initial activity is accumulated here. The large surface area of lichens, relative to their mass, is one of the main reasons for their relatively high capacity to accumulate radionuclides or other elements, like heavy metals. Handley and Overstreet (1968) demonstrated that the fixation of radiocesium in lichen thallium does not depend on their physiological activity, being mostly a passive phenomenon. Both lichens and mosses can be found everywhere, providing the possibility to make a complete inquiry of the interested contaminated area. As these perennial plants seem to dominate the mountainous regions, where the collection of soil samples can be difficult, it is more convenient to use them for radiological researches of these territories. Moreover, their sampling is relatively simple, unlike soil.

The radioactive cloud from the Chernobyl accident, reached the Austrian Province of Salzburg, situated in the Eastern Alps, on the 30th of April 1986, accompanied by heavy rainfall on that evening. Mainly through this washout effect, this territory became one of the most contaminated areas in Western Europe. Hence, the goal of this research was to determine the Cs-137 contamination levels in lichens and mosses from Koppl, in Salzburg province, and to study the behaviour of the long-lived Cs-137, 20 years after the Chernobyl accident. Since a reference base for the radiological situation of lichens in this area has been established shortly after the Chernobyl accident and several years after, that made possible to evaluate the biological half-life (T_b) of Cs-137 for a few species of lichens and mosses. Moreover, the transfer-factors from soil to lichens were determined in order to find a potential correlation between the Cs-137 content in mosses and the substrate on which they grow.

2. MATERIAL AND METHODS

Different species of lichens and mosses were collected at three test sites in a peatbog near Koppl, in the East of Salzburg city, Austria, in December 1986, June 1994 and May 2006. The Koppl area is a mixed forest-litter (pine and birch are predominant), at an elevation of more than 700 m above sea level, at the base of Gaisberg mountain. For the determination of the T_b of Cs-137, the sampling sites corresponded with the same sampling spots from 1986 and 1994. Thus, four lichen species were chosen for the calculation of biological half-times: the epiphytic lichens *Pseudevernia furfuracea* (from *Betula pubescens*) and *Hypogymnia physodes* (from *Betula verrucoso*), specific for the mountainous and subalpine regions, and the epigeic lichens *Cladonia squamosa* and *Cladonia fimbriata.* Regarding mosses, only the data from 1994 are available, for *Leucobrum glaucum* and *Polytrichum strictum* species.

A lot of lichen specimens had to be collected to obtain a final dry weight of about 12 g -16 g for each sample, except for *Cladonia squamosa*, where only few individuals were found. This represents a convenient sample volume for a gamma analysis, at a given location, and consisted of at least 10 individual lichens from the lower branches of the same tree. The final dry weight for each moss sample was about 3 g -7 g. A total of 4 lichen and 5 moss species were collected from the three tests given sites from Koppl in May 2006. Besides, two substrate samples were taken from the first and the second sites, 5 cm below the soil surface, allowing us to determine the transfer-factors of Cs-137 for different species of lichens.

All samples were carefully separated from the substratum, cleaned from soil or other impurities and then air-dried at room temperature. After about one week, the lichens and mosses samples were dried again at 105° C for at least 24 hours and homogenized. After the determination of the dry weight, the samples were enclosed in 100 ml or 20 ml plastic containers and measured by gamma spectrometry, using two multi-channel high purity germanium detectors (EG&G Ortec: energy resolution 2.0 keV at 1.33 MeV of Co-60; 20 % and 40% relative efficiency). The measurement time was about 24 hours, depended on sample size and Cs-137 activity concentration to obtain an uncertainty for the counting statistics less than 5%. The energies of the γ-rays were as follows: E_γ= 662 keV for Cs-137, E_γ= 1461 keV for K-40 and E_γ= 477 keV for Be-7. Gamma-Vision software was used for the automatic spectrum evaluation.

3. RESULTS AND DISCUSSION

The radiological contamination in lichens, several months after the Chernobyl accident, was carried out by Hofmann *et al.* (1986), in different places of Salzburg province. In a peatbog near Koppl, a few samples from different species of mosses and lichens were measured again in 1994. This allowed us to study the local fallout patterns of Cs-137 in an undisturbed region, where the heterogeneous topography and the higher amount of precipitations are major determinants of the radionuclides behaviour. Table 1 shows the specific activity of Cs-137, K-40 and Be-7 in several species of mosses together with epigeic and epiphytic lichens, collected on May 2006. Specific activities listed below refer to the time of sampling and not to the time of measurement.

Table 1

Specific activities of radionuclides (Bq/kg dry weight) in mosses
and lichens from Koppl-Moor, collected on May 2006

Location	**Species**	**Specific activity in lichens and mosses (Bq/kg dry weight)**			**Cs-137/ K-40**
		Cs-137	**K-40**	**Be-7**	
	Epigeic lichens				
	Cladonia fimbriata	7345±33	<29**	115±30*	-
Sampling	*Cladonia squamosa*	5387±28	67±5	361±37	80.4
test site 1	**Mosses**				
	Leucobryum glaucum	14092±46	272±20	900±56	51.8
	Sphagnum papillosum	1478±13	83±7	172±21	17.8
	Epigeic lichens				
	Cladonia fimbriata	2440±35	<32**	129±23	-
	Epiphytic lichens				
Sampling	*Pseudevernia furfuracea*	1765±13	94±8	279±24	18.8
test site 2	*Hypogymnia physodes*	1923±10	77±6	128±13	25.0
	Mosses				
	Dicranodontium	1865±30	51±9	111±33*	36.5
	denudatum	3293±24	90±10	-	36.6
	Leucobryum glaucum				
	Mosses				
Sampling	*Polytrichum strictum*	1145±16	190±19	150±23	6.0
test site 3	*Sphagnum fallax*	2863±26	319±28	217±35	9.0

* Experimental error
** Activity below the detection limit of the detector

Although 20 years have passed since the nuclear catastrophe, the radiocesium contamination in non-vascular plants is still very high in the mountain region of Salzburg province.

In the present study, several differences in the uptake behaviour of lichens and mosses can be noted. For example, a large range in the activity concentrations of Cs-137 in mosses can be observed, because the highest Cs-137 content was found in one of the moss species, *Leucobryum glaucum* (14092±46 Bq/kg dry weight), while another moss species, *Politrichum strictum*, had a much lower cesium activity (1145±16 Bq/kg dry weight), caused mainly by differences in the fallout pattern, because these samples were collected from different sites. In case of *Leucobryum glaucum,* the specific activity of Cs-137 was measured in two different places and a high contamination was found at both places, but much higher in the first sampling site. It can be assumed that this moss species represents a suitable bioindicator of the radioactive fallout, even if many years have passed since the Chernobyl accident, and thus may be succesfully used in further investigations of this territory.

The measurements carried out in 2006, concluded that all the epigeic lichens were shown to be more active than the epiphytic ones. Different authors (Eckl *et al.* 1984; Kwapulinski *et al.* 1985; Attarpour *et al.* 1987) found that, in general, epigeic lichens accumulated higher Cs-137 concentrations than epiphytic lichens. The cesium content in the epiphytic lichens is mostly due to the absorption of the airborne radionuclides, unlike the epigeic lichens which can accumulate the Cs-137 from air, but also through the exchange of cesium atoms between the lithogenic substrate and lichen thallus. Moreover, epiphytic lichens are somehow protected from any kind of contamination, by the crown of the tree on which they grow. The same species of lichen (*Cladonia fimbriata*) collected from different locations (sampling sites 1 and 2), in May 2006, present various activities, reflecting differences of the microclimate where the plant grows, even if the sampling sites are not very far from each other.

Different opinions have been issued regarding the behavior of Cs-137 in non-vascular plants over time. Sawidis (1988) showed that the radiocesium level in lichens was smaller than in moss species, but Topcuoğlu *et al.* (1995) proved that this is not valid in all cases. Thus, it was concluded that only shortly after the radioactive accident, moss species presented higher Cs-137 concentrations than lichen species, but, after several years, due to differences in biological half-lifes, the proportion between the specific activities of cesium in lichens and mosses changes. As it can be seen from our results, 20 years after the Chernobyl accident, lichens have a higher storage capacity of radionuclides than mosses, but there are some moss species (like *Leucobryum glaucum*) which in certain growth conditions can present high concentration of Cs-137.

Two substrate samples have been taken from a peatbog and a podsoil included in a forest litter from Koppl in order to study whether the radiocesium transfer from soil to lichens can be determined. The transfer coefficients between soil and plants or soil-to-plant transfer factors (TF) have been determined as the ratio of the specific activity (Bq/kg) of the radionuclide in plants to that in soil. The chemical similarity of these elements (Cs-137 and K-40), gives us reasons to believe that they can be taken up through soil in the same manner and in approximately similar concentrations, depending on their availability in the surface soil. The K-40 content in lichens is very low or above the detection limit. This result is due to the lower content of K-40 in soils, but also proves the inability of lichens to take up high amounts of K-40 from the substrate they grow. In a study on Na, K, Rb, Li and Cs made by M. Greger in 2004, it was found that the uptake mechanism of Cs-137 is different from that of the other chemical similar elements. The mean value of soil-lichen TF for Cs-137, calculated for peatbog, is 14.3, in accordance to the summarized TF values for Arctic representative species of lichens, ranged from 0.13 to 22.8 (Beresford *et al.* 2005).

As mosses do not have roots, the Cs-137 content in these plants cannot be the result of absorption from soil, so the transfer factors for mosses were not calculated.

For the substrate samples collected, the Cs-137 content in peatbog was much lower than in podsoil, because in this type of soil, cesium is more available for the plant uptake. The peatbog is known to have humus accumulations deficient in minerals, especially clay minerals, and a low pH value, but the podsoil is rather acidic, with a pH value lower than 4.6. The availability of cesium is inversely proportional to the clay content of the soil, because in a soil abundant in clay minerals the cation exchange capacity of the humus is very low and Cs-137 remains bound (Cummings *et al.* 1969).

Table 2

Specific activity of Cs-137 in substrate and lichens and the soil-plant transfer factors

Location	**Specific activity in soil (Bq/kg dry weight)**			**Species**	**Specific activity in lichens (Bq/kg dry weight)**			**Transfer factors**
	Cs-137	**K-40**	**Be-7**		**Cs-137**	**K-40**	**Be-7**	**Cs-137**
Sampling site 1 (peatbog)	445±3	28±2	201±7	*Cladonia fimbriata*	7345±33	<29**	115±30	16.5
				Cladonia squamosa	5387±28	67±5	361±37	12.1
Sampling site 2 (podsoil)	832±3	14±1	10±2*	*Cladonia fimbriata*	2440±35	<32**	129±23	2.9

* Experimental error
** Activity below the detection limit of the detector

As the cosmic ray product Be-7 is rather uniformly distributed, variations in its concentrations for lichens and mosses reflect different local rainfall intensities, because of the continuous wet deposition of this short-lived radionuclide ($T_{1/2}$=53 days) onto the plants and the soil surface. As the Koppl area is at an altitude of about 700 m above sea level, rain and snow occur quite frequently all along the line and therefore Be-7 could be detected in almost all samples.

As no significant radioactive material was transported into the stratosphere after the Chernobyl accident, this provided the unique opportunity to study the biological half-lives for lichen and moss samples collected from an undisturbed area from Koppl. Biological half-life, also termed as ecological half-life, residence time or environmental half-life, refers to the time it takes to reduce the amount of a deposited element to half its initial value by natural processes. The effective half-life is the time taken for the amount of a specified radionuclide in the body to decrease to half of its initial value as a result of both radioactive decay and natural elimination.

On the data basis of three sampling periods, the effective half-life of Cs-137 in *Pseudevernia furfuracea* was calculated as a linear regression of the mean values. The starting point of the regression was set in 1986, since this corresponds to the year of the first sampling and also of the radioactive fallout. The fraction removed from the body per unit time, k, is related to the biological half-time, T_b, in the same manner as the radioactive decay constant is related to the physical half-life and is given by $k = \ln(2)/T_b$. The data of the cesium specific activity were ln-transformed to decrease the impact of inhomogeneous variance.

The effective half-time is the reciprocal of the sum of the reciprocals of the physical and biological half-times. Thus, the effective half-life T_{eff} is given by:

$$T_{eff} = \frac{T_{\frac{1}{2}} \cdot T_b}{T_{\frac{1}{2}} + T_b} \qquad [1]$$

For the epiphytic lichen *Pseudevernia furfuracea,* the value of T_b for cesium was estimated as 12.9±0.8 years. A decrease of Cs-137 content is obvious with time (through the physical decay), but leaching effects due to rain and snow accelerates this decrease. The difference between the theoretical and the measured values for the Cs-137 activity in this lichen species is quite high.

For the other studied species of lichens and mosses, only two collecting data were available and the effective half-life of Cs-137 was determined by the exponential function:

$$A = A_0 \cdot e^{-\frac{\ln(2)}{Teff} \cdot t} \qquad [2]$$

where: A_0- the initial cesium activity,

A - the activity after several years, and

t - the time elapsed since the reference data.

Table 3 shows the cesium concentration for selective species of lichens and mosses collected in December 1986, June 1994 and March 2006, as well as the effective half-life (T_{eff}) and the biological half-life (T_b) of Cs-137.

While epigeic lichens showed a higher content of Cs-137 than the epiphytic ones, the biological half-life of *Cladonia squamosa* (15.1±0.3 years) is much higher than those of the epiphytic lichens.

Table 3

Effective and biological half-time of Cs-137 in lichens and mosses

Species	**Specific activity of Cs-137 in Dec. 1986 (Bq/kg dry weight)***	**Specific activity of Cs-137 in June 1994 (Bq/kg dry weight)****	**Specific activity of Cs-137 in Mar. 2006 (Bq/kg dry weight)**	**Effective half-time, T_{eff} (years)**	**Biological half-time, T_b (years)**
Epigeic lichens					
Cladonia squamosa	21400±214	-	5387±28	10.1±0.1	15.1±0.3
Epiphitic lichens					
Hypogymnia physodes	13203±264	-	1932±10	7.2±0.1	9.4±0.2
Pseudevernia furfuracea	**7005±140**	**3701±50**	**1765±13**	**9.1±0.4**	**12.9±0.9**
Mosses					
Leucobryum glaucum (site 2)	-	6302±30	3293±24	12.9±0.2	22.5±0.7
Polytrichum strictum	-	3500±280	1145±16	7.4±0.6	9.9±1.1

* W. Hofmann *et al.*, Health Physics Society 1993;
** Gastbergger, M. *et al.*, Symposium on Radiation Protection in Neighbouring Countries in Central Europe, 1995

Most of the values found in the literature for the biological half-life of Cs-137 in different lichen species are lower than 10 years (Papastefanou *et al.* 1992; Ellis and Smith 1987; Heinrich *et al.* 1999), but all the authors made an estimation of T_b on a shorter scale of the sampling time. For *Pseudevernia furfuracea* and *Hypogymnia physodes* collected in Romania, the T_b were reported to be 2.5±0.7 and 3.05±0.7 years, respectively, for the period from 1986 to 1992 (Mócsy & Bartók, 1994). Probably some of the highest values, estimated until now (Liden and Gustafsson found T_b =17 years in lichens) can be explained by an unaccounted continuous radionuclide influx or by the resuspension of radionuclides from the soil surface, if biological half-lives were determined shortly after the Chernobyl accident.

In the present study, we calculated the half-lives from samples which were taken 12 and 20 years after the nuclear disaster. Since Cs-137 penetrated into deeper soil layers within the first few years after the accident, resuspension is not likely to significantly influence the determination of the biological half-life. Only one of the moss species (*Leucobryum glaucum*) exhibits a very high value of T_b, 22.5±0.7 years, likely due to its morphological characteristics or growth rate and in a lesser degree to its natural ecosystem, because the other moss species collected from the same place had a residence time of only 9.9±1.1 years. A reliable value for the biological half-time in mosses and lichens could be obtain if more values of the cesium concentration had been determined since 1986.

The estimation of the residence time of long-lived radionuclides in lichens and mosses is important for mineral cycling studies in natural ecosystems, especially in case of edible species, because of their important role in the food chain. Although many uncertainties have to be considered regarding the biological half-life of Cs-137 in perennial plants since it reportedly varies with meteorological and environmental conditions (heavy rains through the washout effect), but it also depends on the species pattern and the growth rate (an overgrowth in the biomass surface, through dilution).

4. CONCLUSIONS

Although 20 years have been passed since the Chernobyl nuclear accident, the cesium levels found in lichens and mosses from the mountain region of Salzburg province are still very high. Once again, these non-vascular plants seem to be suitable and inexpensive bioindicators of the radioactive fallout. Many differences could be observed between different species of lichens and mosses, mainly due to the different contamination of the selected sampling sites, but also due to the meteorological conditions, the morphological characteristics or the growth rate. The moss species *Leucobryum glaucum,* showed the highest activity of Cs-137 (14092±46 Bq/kg dry weight), but we could not conclude that all mosses have a higher capacity for radionuclide storage than lichens. As the same moss had the highest biological half-life for Cs-137 (22.5±0.7 years), among the studied species, but also one of the highest value found in literature, it can be assumed that it can be successfully used in further investigations of this territory.

All values estimated for the biological half-live of the studied species were very high in this mountain area at Koppl, mainly due to the shorter vegetation period at a higher altitude, but it can not be considered as a ‘fixed value’ over time, even in the same region and under similar conditions.

The K-40 concentrations in lichen species were very low or below the detection limit and also much lower than the Cs-137 activities for the same samples. These results are due to the different uptake mechanisms between Cs-137 and K-40, even if K-40 is known to be a congener of Cs-137, but also due to the K-40 deficiency in soil samples.

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