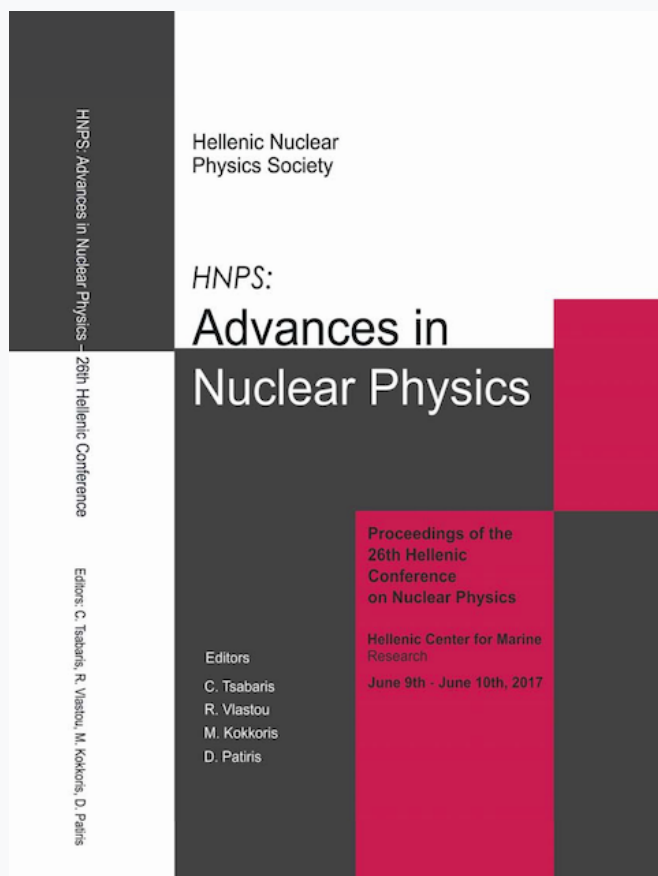


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## Natural and artificial radionuclides in moss samples from the region of Northern Greece

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**Abstract** Terrestrial mosses obtain most of their nutrients directly from precipitation and dry deposition, and they can be used for monitoring of airborne radionuclide depositions. Ninety five samples of *Hypnum Cupressiforme* were collected in Northern Greece during the end of summer 2016. After the preparation, mosses were measured in a low-background HPGe detector with relative efficiency 32%. The activity concentrations of <sup>7</sup>Be ranged from 69 to 1280 Bq kg<sup>-1</sup>, and the concentrations of <sup>137</sup>Cs ranged from 0 to 425 Bq kg<sup>-1</sup>. The concentrations of <sup>210</sup>Pb were between 147 and 1920 Bq kg<sup>-1</sup> and for <sup>40</sup>K were between 120 and 750 Bq kg<sup>-1</sup>. Differences have been observed in the activity concentrations between the mosses collected from ground surface, rocks, branches and near roots. Finally, no correlation between the concentrations of <sup>7</sup>Be and <sup>210</sup>Pb has been found.

**Keywords** moss technique, airborne radionuclides, bio-monitoring, <sup>137</sup>Cs

## INTRODUCTION

Using biological indicators is a quite interesting and reliable method for detecting radioactive contamination of an ecosystem [1]. Mosses have been used as bioindicators of atmospheric deposition of radionuclides and for atmospheric assessments since the late 1960s in Scandinavian countries [2, 3, 4]. Carpet-forming moss species can be used as biomonitors providing a number of advantages. Mosses obtain most of their nutrients directly from precipitation and dry deposition. The absence or strong reduction of the cuticle and thin leaves allows easy uptake from the atmosphere. Lack of an elaborate rooting system also means that uptake from the substrate is normally insignificant. These properties make mosses an ideal sampling medium for metals and airborne radionuclides deposited from the atmosphere, as they are accumulated by the moss, producing concentrations much higher than those in the original wet or dry deposition [5, 6]. Another advantage of moss technique is that the sample collection is so simple, that a high sampling density can be achieved, in contrast to the conventional precipitation analysis and the air sampling. High resolution gamma spectrometry measurements can be carried out with the moss technique, without any chemical treatment of the samples.

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Naturally occurring radionuclides  $^7\text{Be}$  and  $^{210}\text{Pb}$  together with the artificial  $^{137}\text{Cs}$  are a useful tool in studying the environmental processes. It is expected that the content of radionuclides that are deposited from the atmosphere can vary from place to place, depending on several factors.  $^7\text{Be}$  is formed by spallation reaction between cosmic rays and nuclei of oxygen and nitrogen in the stratosphere and upper troposphere [6]. After production, the  $^7\text{Be}$  atoms are attached to aerosol particles and the fate of  $^7\text{Be}$  will become the fate of the carrier aerosols. Since aerosol particles contain most of the air pollutants, the transport of the latter can be investigated by tracking the  $^7\text{Be}$  pathway. The production rate of cosmogenic isotopes depends on the intensity of the geomagnetic field. The concentrations of  $^7\text{Be}$  in surface air are decreasing with the increase of latitude [6].

The radionuclide  $^{210}\text{Pb}$  is widely found in the terrestrial environment and it is present in the atmosphere due to the decay of  $^{222}\text{Rn}$  diffusing from the ground. The presence of  $^{210}\text{Pb}$  in the atmosphere depends on the emanation rate of  $^{222}\text{Rn}$ . The latter depends on different factors such as the geological properties of the ground, the ability of radon to leak from the ground and enters in the atmosphere and the conditions of the ground surface layer (the humidity of the soil, the presence or no of the snow cover, the thickness of the frozen soil crust [7]. Once  $^{210}\text{Pb}$  is formed in the atmosphere, it is attached to aerosol particles and follows their path. So it can be used for tracking the aerosols deposition and their residence time in the atmosphere.

The artificial radionuclide  $^{137}\text{Cs}$  was mostly released in the atmosphere during atmospheric nuclear weapon tests and the Chernobyl nuclear accident. After that, there were no other significant  $^{137}\text{Cs}$  emissions, and the atmospheric  $^{137}\text{Cs}$  was exposed to physical decay as well as wet and dry deposition [6]. In recent years the Fukushima accident contributed to the release of  $^{137}\text{Cs}$  in the atmosphere but with minor influence in regions far from Japan.

The aim of this study is to measure activities of the radionuclides  $^{137}\text{Cs}$ ,  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  in mosses and investigate their possible variabilities over different places in Northern Greece. The different meteorological conditions, the wind direction and precipitation can influence the deposition of airborne radionuclides, as well as their activities in mosses.

## EXPERIMENTAL DETAILS

Ninety five (95) samples of *Hypnum Cupressiforme* Hedw. were collected in the region of Northern Greece. The sampling sites were located from  $39.97^\circ$  North to  $41.65^\circ$  North and from  $20.97^\circ$  East to  $26.26^\circ$  East. The samples were collected from different altitudes, from 30 m to 1450 m above the mean sea level. All samples of fresh plant material were collected in a short time interval during the end of summer 2016. During the sampling there was no rain, thus avoiding the exposure to additional precipitation and extra component of airborne radionuclides by rain washing of the atmosphere [6]. The regions from where samples were collected were open regions far from treetops in most of the sampling sites. All the sampling sites were selected avoiding possible contact of mosses with surface water. All the samples were collected according to the instructions of the Protocol of the European Survey ICP



Vegetation [8], in which Greece is the first time that is included. After sampling, mosses were dried at 105°C for 2 hours to a constant weight. Soil and all other mechanical impurities were removed manually. After the preparation, each sample of mosses were put in two cylindrical plastic containers, diameter 67 mm and height 31 mm. The minimum mass of dry moss was 8.9 g and the maximum was 58 g.

Low-background extended range HPGe detector equipped with a Be window was used in order to get evidence about  $^{210}\text{Pb}$ . Relative efficiency of detector is 32%. Background count was reduced by passive shield (18 cm of lead, 1 mm of tin and 1.5 mm of copper) which does not interfere with gamma photons emitted from samples. Gamma spectrum of each sample was collected until statistical uncertainty of the 477.6 keV  $^7\text{Be}$  line up to 5% was achieved and statistical uncertainty of 46.5 keV  $^{210}\text{Pb}$  line was up to 5%. The detection efficiency was established using NIST Standard Reference Material 4350B (Columbia River sediment) packed in same geometry. Accuracy of efficiency calibration was tested using IAEA source made from dry grass [6].

## RESULTS AND DISCUSSION

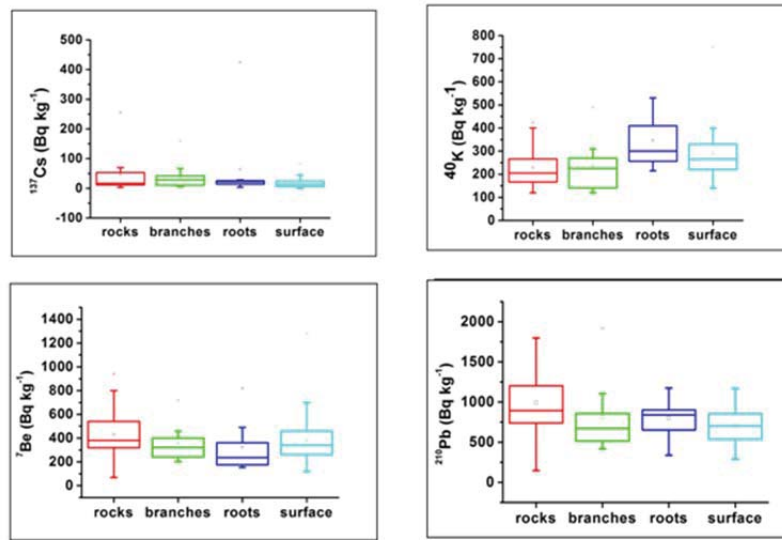
In the gamma spectrum several prominent peaks appeared. After the subtraction of the background and peak analysis, the intensities of  $^7\text{Be}$  (477.6 keV),  $^{210}\text{Pb}$  (46.5 keV),  $^{40}\text{K}$  (1460.8 keV) and  $^{137}\text{Cs}$  (661.6 keV) were obtained. The activity concentrations of the radionuclides  $^{137}\text{Cs}$ ,  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  were calculated and are presented in Table 1. The mean value of each radioisotope is presented in the last column of Table 1.

Radionuclide	Min Bq kg <sup>-1</sup>	Max Bq kg <sup>-1</sup>	mean value Bq kg <sup>-1</sup>
$^7\text{Be}$	69 ± 14	1280 ± 100	388 ± 194
$^{137}\text{Cs}$	0	425 ± 27	35 ± 1
$^{210}\text{Pb}$	147 ± 19	1920 ± 100	817 ± 344
$^{40}\text{K}$	120 ± 11	750 ± 40	269 ± 138

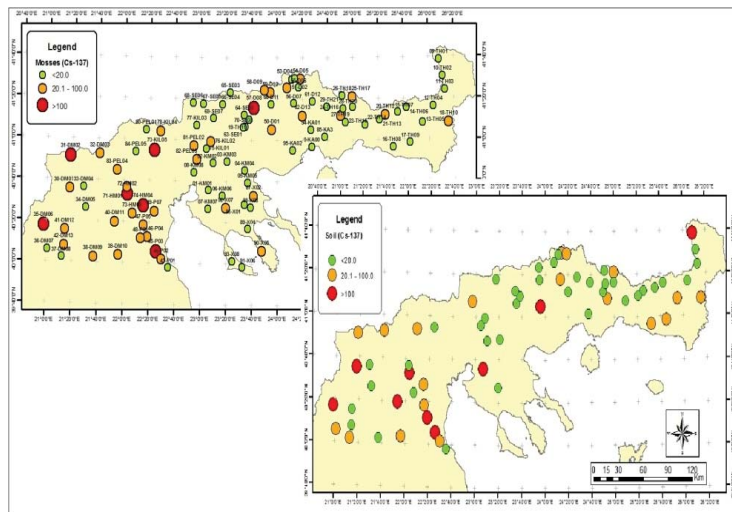
**Table 1.** Activity concentrations of the radioisotopes  $^{137}\text{Cs}$ ,  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  in Bq kg<sup>-1</sup> in moss samples collected from the region of Northern Greece during the summer of 2016.

Differences have been observed in the activity concentrations between mosses collected from different surface types such as ground surface, rocks, branches and roots (Fig.1).  $^7\text{Be}$  and  $^{210}\text{Pb}$  activity concentrations are higher in moss samples from the ground surface and rocks than those near roots.  $^{137}\text{Cs}$  concentrations are higher in mosses collected near roots and rocks than those collected on the ground surface.  $^{40}\text{K}$  concentrations are higher in mosses collected from branches and near roots than those collected from rocks. The ratio of the activity concentrations of  $^7\text{Be}$  between mosses collected from the surface and those collected near roots is 1.5. This ratio is logical, if someone can expect that the majority of  $^7\text{Be}$  should be found in mosses collected in open areas and not under trees.





**Fig. 1.** Differences in the activity concentrations of the radioisotopes <sup>137</sup>Cs, <sup>7</sup>Be, <sup>210</sup>Pb and <sup>40</sup>K in moss samples collected from different surface types.



**Fig. 2.** The activity concentrations of <sup>137</sup>Cs in moss samples and in soil samples collected from the same sites in Northern Greece. There is a good correlation between the concentrations of <sup>137</sup>Cs in soil and in moss samples.

There is no correlation between the cosmogenic radionuclide <sup>7</sup>Be and the natural occurring radionuclide in the Earth's crust <sup>40</sup>K. The long-living radionuclide <sup>40</sup>K was transferred to mosses due to the re-suspension and the decayed plant matter covering mosses [9]. No correlation between the concentrations of <sup>7</sup>Be, <sup>210</sup>Pb and <sup>137</sup>Cs has been found. No special variances in the concentrations of <sup>210</sup>Pb and <sup>7</sup>Be due to different altitudes or meteorological conditions have been observed. The majority of <sup>210</sup>Pb in mosses has arrived through aerosol deposition (e.g. dust that contains <sup>238</sup>U daughters). The activity of <sup>210</sup>Pb in mosses can vary from region to region due to the different soil structure.



The coincidence of heavy rainfall events during May 1986 with the passage of air masses from the Chernobyl area to Northern Greece had as a result high concentrations of  $^{137}\text{Cs}$  at the ground level in Northern Greece. The regions, in which the activity concentrations of  $^{137}\text{Cs}$  in moss samples are high, are in a good correlation with the regions where the activity concentrations of  $^{137}\text{Cs}$  in soil samples are high and were also characterized as highly contaminated right after the Chernobyl accident (Fig. 2). The majority of  $^{137}\text{Cs}$  in mosses has arrived through aerosol deposition (re-suspension).

## CONCLUSIONS

Mosses can be used as a sampling medium for the detection of  $^{137}\text{Cs}$ ,  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  radionuclides accumulated through some period, covering a high density of sampling points in differently sized areas [9]. A big number of sampling sites (95) was covered and the information obtained using mosses as biomonitors, provide a detailed spatial distribution of the above radionuclides over Northern Greece. Mapping of  $^7\text{Be}$  and  $^{210}\text{Pb}$  activity in mosses provide information about aerosols deposition and help tracking their pathway. The activity concentration of  $^{210}\text{Pb}$  depends on the different soil properties and structure.  $^{137}\text{Cs}$  concentration depends on how the air masses from the Chernobyl accident moved in 1986 and after that which areas were more contaminated.  $^{137}\text{Cs}$  and  $^{40}\text{K}$  arrive at mosses via dust particles from the surrounding soil (re-suspension) as well as the majority of  $^{210}\text{Pb}$  and  $^7\text{Be}$  is accumulated in mosses through aerosols deposition.

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