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NATURAL AND ARTIFICIAL RADIONUCLIDES IN MOSS SAMPLES FROM THE REGION OF NORTHEN 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. Ninetyfive 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 ⁷Be ranged from 69 to 1280 Bq kg⁻¹, and the concentrations of ¹³⁷Cs ranged from 0 to 425 Bq kg⁻¹. The concentrations of ²¹⁰Pb were between 147 and 1920 Bq kg⁻¹ and for ⁴⁰K were between 120 and 750 Bq kg⁻¹. 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 ⁷Be and ²¹⁰Pb has been found.

1. 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.

Naturally occurring radionuclides ⁷Be and ²¹⁰Pb together with the artificial ¹³⁷Cs 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. ⁷Be is formed by spallation reaction between cosmic rays and nuclei of oxygen and nitrogen in the stratosphere and upper troposphere [6]. After

production, the ⁷Be atoms are attached to aerosol particles and the fate of ⁷Be 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 ⁷Be pathway. The production rate of cosmogenic isotopes depends on the intensity of the geomagnetic field. The concentrations of ⁷Be in surface air are decreasing with the increase of latitude [6].

The radionuclide ²¹⁰Pb is widely found in the terrestrial environment and it is present in the atmosphere due to the decay of ²²²Rn diffusing from the ground. The presence of ²¹⁰Pb in the atmosphere depends on the emanation rate of ²²²Rn. 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 ²¹⁰Pb 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 ¹³⁷Cs was mostly released in the atmosphere during atmospheric nuclear weapon tests and the Chernobyl nuclear accident. After that, there were no other significant ¹³⁷Cs emissions, and the atmospheric ¹³⁷Cs was exposed to physical decay as well as wet and dry deposition [6]. In recent years the Fukushima accident contributed to the release of ¹³⁷Cs in the atmosphere but with minor influence in regions far from Japan.

The aim of this study is to measure activities of the radionuclides ¹³⁷Cs, ⁷Be, ²¹⁰Pb and ⁴⁰K 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.

2. SAMPLING AND MEASUREMENTS

Ninetyfive (95) samples of *Hypnum Cupressiforme* Hedw. were collected in the region of Northern Greece. The sampling sites were located from 39.97° North to 41.65° North and from 20.97° East to 26.26° 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 ²¹⁰Pb. 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 ⁷Be line up to 5% was achieved and statistical uncertainty of 46.5 keV ²¹⁰Pb 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].

3. RESULTS AND DISCUSSION

In the gamma spectrum several prominent peaks appeared. After the subtraction of the background and peak analysis, the intensities of $^7\mathrm{Be}$ (477.6 keV), $^{210}\mathrm{Pb}$ (46.5 keV), $^{40}\mathrm{K}$ (1460.8 keV) and $^{137}\mathrm{Cs}$ (661.6 keV) were obtained. The activity concentrations of the radionuclides $^{137}\mathrm{Cs}$, $^7\mathrm{Be}$, $^{210}\mathrm{Pb}$ and $^{40}\mathrm{K}$ were calculated and are presented in Table 1. The mean value of each radioisotope is presented in the last column of Table 1. The lowest measured value of $^7\mathrm{Be}$ activity per unit mass of dry plant material is 69 Bq kg⁻¹ and the highest one is 1280 Bq kg⁻¹. The activity concentrations of $^{137}\mathrm{Cs}$ range from 0 to 425 Bq kg⁻¹. The concentrations of $^{210}\mathrm{Pb}$ are between 147 and 1920 Bq kg⁻¹ and for $^{40}\mathrm{K}$ range between 120 Bq kg⁻¹ to 750 Bq kg⁻¹.

In period from 2000-2009 there were several surveys in Serbia, Belarus and Slovakia. For comparison, results from these surveys are in Table 2.

Table 1. Activity concentrations of the radioisotopes ¹³⁷Cs, ⁷Be, ²¹⁰Pb and ⁴⁰K in Bq kg⁻¹ in moss samples collected from the region of Northern Greece during the summer of 2016

Radionuclide	Min Bq kg ⁻¹	Max Bq kg ⁻¹	mean value Bq kg ⁻¹	
⁷ Be	69 ± 14	1280 ± 100	388 ± 194	
¹³⁷ Cs	0	425 ± 27	35 ± 1	
²¹⁰ Pb	147 ± 19	1920 ± 100	817 ± 344	
⁴⁰ K	120 ± 11	750 ± 40	269 ± 138	

Table 2. Activity concentrations in Bq kg⁻¹ of the radioisotopes ¹³⁷Cs, ⁷Be and ²¹⁰Pb in moss samples from surveys in Serbia, Belarus and Slovakia

	Serbia 2007 [9]	Serbia [5]	Belarus 2000-2009	Slovakia 2000-2009 [10]	Serbia October 2005 [9]	Serbia summer 2008 [5]
Radionuclide	Mean value Bq kg ⁻¹	Mean value Bq kg ⁻¹	Range (mean value) Bq kg ⁻¹	Range (mean value) Bq kg ⁻¹	Range (mean value) Bq kg ⁻¹	Range Bq kg ⁻¹
⁷ Be					95-360(195)	201-920
¹³⁷ Cs	34	8.9	5-4833(34)	0.7-103		
²¹⁰ Pb			141-575(312)	330-1521(771)		347-885
⁴⁰ K					(281)	

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). ⁷Be and ²¹⁰Pb activity concentrations are higher in moss samples from the ground surface and rocks than those near roots. ¹³⁷Cs concentrations are higher in mosses collected near roots and rocks than those collected on the ground surface. ⁴⁰K concentrations are higher in mosses collected from branches and near roots than those collected from rocks. The ratio of the activity concentrations of ⁷Be 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 ⁷Be should be found in mosses collected in open areas and not under trees.

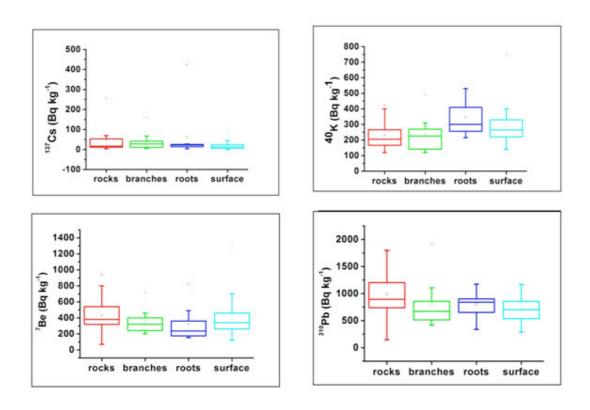


Figure 1. Differences in the activity concentrations of the radioisotopes ¹³⁷Cs, ⁷Be, ²¹⁰Pb and ⁴⁰K in moss samples collected from different surface types

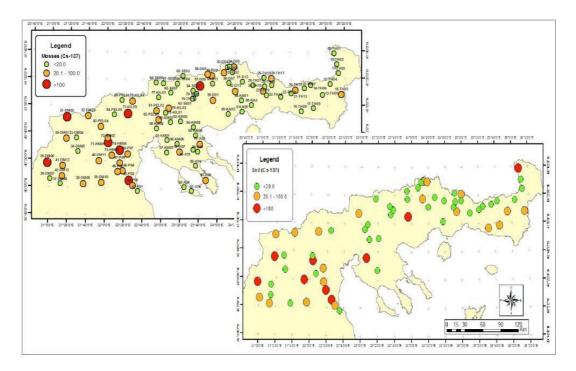


Figure 2. The activity concentrations of ¹³⁷Cs in moss samples and in soil samples collected from the same sites in Northern Greece. There is a good correlation between the concentrations of ¹³⁷Cs in soil and in moss samples

There is no correlation between the cosmogenic radionuclide ⁷Be and the natural occurring radionuclide in the Earth's crust ⁴⁰K. The long-living radionuclide ⁴⁰K was transferred to mosses due to the re-suspension and the decayed plant matter covering mosses [9]. No correlation between the concentrations of ⁷Be, ²¹⁰Pb and ¹³⁷Cs has been found. No special variances in the concentrations of ²¹⁰Pb and ⁷Be due to different altitudes or meteorological conditions have been observed. The majority of ²¹⁰Pb in mosses has arrived through aerosol deposition (e.g. dust that contains ²³⁸U daughters). The activity of ²¹⁰Pb 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 ¹³⁷Cs at the ground level in Northern Greece. The regions, in which the activity concentrations of ¹³⁷Cs in moss samples are high, are in a good correlation with the regions where the activity concentrations of ¹³⁷Cs in soil samples are high and were also characterized as highly contaminated right after the Chernobyl accident (Fig. 2). The majority of ¹³⁷Cs in mosses has arrived through aerosol deposition (re-suspension).

4. CONCLUSIONS

Mosses can be used as a sampling medium for the detection of ¹³⁷Cs, ⁷Be, ²¹⁰Pb and ⁴⁰K 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 ⁷Be and ²¹⁰Pb activity in mosses provide information about aerosols deposition and help tracking their

pathway. The concentrations of ⁷Be in mosses in Greece are higher than those in Serbia, due to the different latitude and probably due to the different existing meteorological conditions. The activity concentration of ²¹⁰Pb depends on the different soil properties and structure. ¹³⁷Cs concentration depends on how the air masses from the Chernobyl accident moved in 1986 and after that which areas were more contaminated. ¹³⁷Cs and ⁴⁰K arrive at mosses via dust particles from the surrounding soil (re-suspension) as well as the majority of ²¹⁰Pb and ⁷Be is accumulated in mosses through aerosols deposition.

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PRIRODNI I PROIZVEDENI RADIONUKLIDI U UZORCIMA MAHOVINA IZ REGIONA SEVERNE GRČKE

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SADRŽAJ

Mahovine svoje nutrijente direktno dobijaju iz padavina i suvom depozicijom i mogu se koristiti za monitoring depozicije radionuklida iz vazduha. Devedesetpet uzoraka mahovina (Hypnum Cupressiforme) je sakupljeno u regionu Severne Grčke tokom kraja leta 2016. godine. Posle pripreme, mahovine su merene na niskofonskom HPGe detektoru relativne efikasnosti 32%. Izmerene koncentracije aktivnosti ⁷Be su bile od 69 do 1280 Bq kg⁻¹, aktivnosti ¹³⁷Cs od 0 do 425 Bq kg⁻¹, aktivnosti ²¹⁰Pb od 147 do 1920 Bq kg⁻¹, a ⁴⁰K od 120 do 750 Bq kg⁻¹. Primećene su razlike u koncentracijama aktivnosti radionuklida u mahovinama koje su sakupljene sa zemlje, kamena, grana stabala i blizu korena stabla. Između ⁷Be i ²¹⁰Pb nije pronađena nikakva korelacija.