RAD Conference Proceedings, vol. 7, pp. 20-23, 2023
ISSN 2466-4626 (online) | DOI: 10.21175/RadProc.2023.04
www.rad-proceedings.org

CONTENT OF NATURAL AND MAN-MADE RADIONUCLIDES IN ANTARCTIC MOSSES

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Abstract. *Mosses are collectors of anthropogenic pollution and are suitable as bioindicators for the content of heavy metals and radionuclides in the air. Due to their lack of a real root system, the main way nutrients are supplied to them is through air moisture. Airborne substances are absorbed and retained in the moss, even when these plants are thousands of kilometers from the source of pollution. In regard to radionuclides, mosses are very indicative. The subject of the research are mosses from Livingstone Island, South Shetland Islands, Antarctica. The presence of natural and man-made radionuclides is investigated. The presence of Natural and man-made radionuclides were determined by means of gamma-spectrometry. The content of Pb-210, U-238, Ra-226, Pb-214, Bi-214, Th-232, K-40, Cs-137 was determined. The activity of cesium-137 is between 3.5±1.2 and 11±2 Bq/kg, for Pb-210 - between 68±7 u 163±15 Bq/kg. Results from samples taken in 2012 and 2022 are compared. This allows to assess the current state of the Antarctic ecosystem and to ascertain changes over a 10-year period. The research confirms that even the most distant points on the globe are contaminated with radionuclides.*

Keywords: *radionuclides, cesium-137, lead-210, mosses, Livingstone Island, Antarctica 10*

1. Introduction

Livingstone Island is the second largest island of the South Shetland Islands, West Antarctica (Fig. 1). It is located in the Southern Ocean, 110 km northwest of the Antarctic continent and 809 km from Cape Horn, South America [1]. The island is covered almost entirely with ice and snow. Only a tenth of it remains exposed and makes it possible for the island's dominant plant species to thrive on the ice-free rocks. The low floristic diversity here is compensated by a relatively large number of species of lichens and mosses - over 50 species [2], [3]. Due to the short Antarctic summer, characterized by low temperatures (rarely exceeding 2-3°C [4] and intense light and the lack of light in winter, mosses can only grow during the summer weeks. The growth of mosses is slow. This, as well as the fact that their nutrition is mainly aerial, through dry or wet deposition of aerial aerosols, makes mosses a very good indicator of fall out pollution. Furthermore, mosses have no ability to excrete the pollution. It can be registered for a long time after absorption.

Figure 1. Map of Livingstone Island by Labajo, A., 2008

The purpose of the research is gamma spectrometric determination of the content of natural and man-made radionuclides in the most common types of mosses from the flora of Livingston Island, in organisms older than 40-50 years. This can provide information on the spread of airborne radioactive contamination from nuclear tests in the atmosphere, from nuclear accidents or as a result of routine human activity, thousands of kilometers away from the cleanest region on earth. The possibility to repeat the research 10 years later with mosses of the same species and the same territories gives the most accurate idea of the behavior of natural and artificial isotopes in the environment and possible changes in the radiological characteristics of the ecosystem. The research included analysis of the content of Pb-210, U-238, Ra-226, Pb-214, Bi-214, Th-232, K-40, Cs-137 in the collected samples.

2. Materials and Methods

The object of study were 3 of the most common moss species *Sanionia georgicouncinata* (Müll. Hal.), *Polytrichum juniperinum* (Hedw.) and *Sanionia uncinata* (Hedw.), collected in 2012 and 2022 in the area of the Bulgarian Antarctic Base "St Kliment Ohridski', the Spanish Antarctic Base 'Juan Carlos I' and Hanna Point on Livingstone Island, Antarctica. After the samples were collected, the moss was cleaned from extraneous mataerials and dried to an air-dry state. It was stored in labeled paper envelopes and thus transported almost 14,000 km. distance. Before measurement, the samples were cleaned again,

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homogenized and dried to a constant weight for 24 hours.

The radiological studies in 2012 and 2022 were conducted in an accredited laboratory of radioecology and radioisotope measurements with the same radiological equipment and the application of the same methods. This ensures absolute comparability of results.

Content of natural and man-made radionuclides was determined by gamma-spectrometric analysis. A multi-channel analyser DSA 1000 (CANBERRA) with HPGe detector, 30% efficiency and 1.8 keV resolution were used. Samples were measured in 450 ml Marinelli Beaker sample containers. The spectrum was analysed by GENIE-2000 software with measurement uncertainties less than 10%. A measurement was performed for an average time t = 120000 s (the time depends on the activity of the sample). The measurements were performed according to BDS EN ISO 18589-3 standard [5] of technogenic origin: ^{137}Cs (661.6 keV full energy peak) and naturally present: ^{238}U – by the gamma lines of ^{234}Th (63.5 and 92 keV), ^{235}U (185.7 keV), ^{226}Ra – by the gamma line at 186,2 keV after correction for ^{235}U contribution in the full energy peak at 185.7 keV, ^{210}Pb (46.5 keV), ^{232}Th – by the gamma line of ^{228}Ac at 911 keV and ^{40}K (at 1461 keV). The efficiency calibration of the system was performed using standard containing lead-210 with density 1 g/cm^3. All samples were measured in the same geometry and at the same counting conditions as the calibration cocktail. For the analysis of attenuation correction of ^{210}Pb a procedure described by Długosz-Lisiecka and Bem was performed [6]. Activity ratios of two pairs of γ-lines ^{228}Ac - 911 keV/209 keV and ^{214}Bi – 609 keV/^{214}Pb - 295 keV were calculated. The values obtained were used as an index for evaluating the attenuation correction factors of ^{210}Pb activity in samples.

3. Results and discussion

Mosses are one of the most diverse plant groups and they are distributed worldwide. They do not have a proper root system and the main way of receiving nutrients is through air moisture. Therefore, mosses are an important tool for mapping the global distribution of radionuclides after atmospheric nuclear tests, nuclear accidents and other human activities [7], [8], [9], [10].

air, radionuclides interact with aerosols. Their migration depends on the characteristics and behavior of these aerosols. The air is purified from them by precipitation at different speeds on the ground or leaf surface. Their movement then depends on air humidity, rainfall, plant uptake, solubility, plant morphology and many other factors. Radionuclides that have passed into the soil can migrate, be fixed at a certain depth, be dissolved by precipitation, or removed by precipitation or ice melting from the sparse soil layer. This depends on their solubility, on the characteristics of the soil - acidity, organic matter, presence of plant cover and others. Soil-to-plant transfer factors (TF) were calculated as the ratio of the activity of an element in moss to its activity in soil [11].

The investigated man-made radioisotopes are registered in all samples (except sample from Hannah Point, 2022, where ^{137}Cs is below the MDA). The results of their content in moss from 2012 and 2022 are given in Table 1.

Lead-210 (with a half-life of 22.3 years) is the result, in addition to the natural decay of uranium-238 from the main rock, and from a number of anthropogenic activities - uranium mining, coal burning, phosphate fertilizers, automobile gases, etc. [12], [13], [14]. After mixing in the environment of natural and anthropogenic Pb, it is difficult to trace the migration of the pollutant. In the studied samples Pb-210 activity remained high in all samples, ranging from 100±15 to 145±15 Bq/kg in 2012 and from 46±5 to 163±15 Bq/kg in 2022. The high Pb-210 values cannot be explained solely by enriched radon emanation and the release of larger areas of ice and snow during the Antarctic summer. In comparison, the content of lead-210 in soil was between 6 -12±2 Bq/kg. Transfer coefficients of lead -210 showed that only between 10 and 20% of the soil content of the radioisotope accounts for its activity in plants. Because of the summer melt, it was possible for secondary deposition of dissolved lead-210 from the soil in the water to reach the mosses and further contaminate them. The large difference between the concentration in the mosses and the soil may be an indicator of the transfer of anthropogenic pollution with the air masses. Over the ten-year period in the study areas for the lead-210,

Table 1. Content of radionuclides in Antarctic moss (2012 – 2022) in Bq/kg.

Sample from	Pb-210	U-238	Ra-226	Pb-214	Bi-214	Th-232	K-40	Cs-137
Todorini buzi 2012	145±15	<3	<3	-	-	4±1	132±13	8±1
Bulgarian Antarctic Base 2012	100±15	<3	<3	-	-	4.3±1.2	165±15	15±2
Hannah Point 2022	46±5	14±4	13±4	8±3	9±3	8±3	300±20	<2
Spain Antarctic base 2022	163±15	14±4	18±5	11±2	11±2	16±3	250±20	11±2
Todorini buzi 2022	126±10	<10	≤10	<10	<10	163±11	360±40	11±2
Bulgarian Antarctic base 2012	68±7	12±4	<10	7±3	5±2	6±2	230±20	3,5±1,2

a decrease evident was no observed (from 145±15 (2012) to 126±10 (2022) and from 100±15 (2012) to 68±7 (2022) Bq/kg).

Cesium-137 is a gamma emitter with a half-life of 30.1 years. It is obtained exclusively from man-made processes and mostly from nuclear explosions. The study of Antarctic mosses showed that cesium was found in all samples (except sample from Hannah Point, 2022, where ^{137}Cs is below the MDA). Its distribution was inhomogeneous from 3.5±1.2 to 11±2 Bq/kg in 2022. The decrease of Cs-137 activity through half-life could not be established. An expected decrease was observed in only one moss sample from BAB from 15±2 Bq/kg in 2012 to 3.5±1.2 Bq/kg in 2022. In a sample from the Todorini Buzi area, Cs-137 was recorded with 8± 1 Bq/kg in 2012 and 11±2 Bq/kg in 2022. The amount of cesium in the soil was significantly lower – between 0.5±0.1 and 6±0.2 Bq/kg. Cesium can be fixed in the soil in the layer rich in organic matter. With a deficiency of such substances and due to its solubility in water, it can be easily removed. Its amount in the soil can be many times less than that in plants. [15]. When compared with additional ours studies of the remaining representatives of the island flora: lichens, algae and grass (Poa Annua - a naturalized anthropophyte) [16], was established that mosses are the best indicator of cesium-137 contamination. In the rest of the plant species, its amount in the measured samples was respectively: ≤1, 1.0±0.4 and 2±0.5 Bq/kg. [16].

The presence of Cs-137 was not only a result of the Chernobyl accident. Its initial introduction into the biotope and biocenosis of Antarctica was recorded with a peak, as early as 1965, after a series of high-altitude nuclear weapons tests conducted by the United States in the Pacific Ocean in 1962 [17]. In 2023, during the gamma spectrometric measurement of cesium-137 in mosses, from the collection of the National Bryological Bank in Bulgaria, it was established that the concentrations of the radionuclide in samples from 1964, 1965 and 1966 significantly exceeded the amounts of cesium-137 immediately after the Chernobyl accident (unpublished research).

The first radiological tests on Livingstone Island were performed on mosses collected 11 months after the accident at the Fukushima Daiichi Nuclear Power Plant (NPP) (11.03.2011). The amount of cesium-134 and cesium-137 released as a result of the accident was equal. [18], [19]. Due to the short half-life of cesium-134 (2 years), its presence in the moss samples would indicate that the contamination from the Fukushima NPP had spread to Antarctica. Cesium-134 in the 2012 and 2022 samples was not detected.

With an area of 13.5 million km^2 and an average value of 56 Bq/m^2, the total content of the Cs-137 deposition over Antarctica is estimated to be 760 TBq, representing less than 0.08% of the total deposition of this radionuclide in the world. For research carried out at 21 stations and in different parts of Antarctica, the obtained yield for Cs-137 was less than expected [20].

The highest was the content of the natural radioisotope K-40 - from 132±13 to 165±15 Bq/kg for 2012 and from 230±20 to 360±40 Bq/kg for 2022. The amount was comparable to that in the soil - between 190±15 and 280±20 Bq/kg [15] and that in volcanic ash - 180±10 Bq/kg [21].

The uranium and thorium are in equilibrium because of the time between sampling and measurement, but in 2022, distortions in the U-238:Ra-226 ratio were observed. In the soils of the island it is respectively (7:10 and 5:13) (unpublished research), while for mosses uranium-238 is in greater concentrations. Uranium is more mobile and can be washed away more easily from the soil, by water.

4. Conclusions

Through air moisture, the radionuclides were incorporated into the moss tissues and could hardly be removed or washed-off.

For almost all radionuclides, significantly higher activities were reported in the moss samples than in the soil. This made moss a good indicator of both natural and anthropogenic radionuclide contamination.

Compared to other plant species on Livingstone Island, mosses were most indicative of cesium-137 content. The same applies to K-40, which mosses absorb and retain much better.

Many times higher values of lead-210 may be an indicator of additional anthropogenic contamination carried by the air masses.

Bi-214 and Pb-214 have comparable activities.

Acknowledgements: *This work is the result of a one-year research project for Polar scientific research for young scientists No. 70-25-60/2022 at the Center for Polar Research, financed by Sofia University "St. Kliment Ohridski", Bulgaria.*

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