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Determination of the concentration activity of radiolead ^{210}Pb in mosses of Bosnia and Herzegovina

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ABSTRACT

The radioactive gas radon ^{222}Rn, through a series of short-lived radionuclides, decomposes into a long-lived isotope of lead ^{210}Pb, which is classified as one of the most dangerous chemotoxic and radiotoxic elements. Natural aerosols containing ^{210}Pb lead isotope are always present in the atmosphere, which is a global problem for human health. This paper investigates the contamination levels of moss radiolead ^{210}Pb obtained from 92 locations in Bosnia and Herzegovina. Gamaspectrometric analysis of the collected samples was performed using three high resolution purity germanium detectors with 35, 23 and 70% relative efficiency. In all tested samples of moss contained ^{210}Pb, with concentrations of ^{210}Pb ranged from 10 to 2000 Bq/kg dry weight in selected moss species.

INTRODUCTION

Natural radionuclides, which are present in the environment, are the main sources of human exposure to radiation, and represent the basic level of ionizing radiation. Radon is one of the most biologically significant radioisotopes in nature because along with its decay products, it makes a major contribution to human exposure to natural radiation sources. Due to the longest half-life (3.825 days) and isotopic abundance, the primary contributor to exposure to population radiation from natural radon isotopes is ^{222}Rn.

The major sources of ions in the near-Earth atmosphere, with variable relative contributions depending on altitude and latitude, are radon isotopes, cosmic rays, and terrestrial gamma radiation (UNSCEAR, 2008). Radon decay products are in most cases positively charged ions, 80 to 85% of them, and are chemically active. They interact with the negative ions present in the atmosphere and in the process of charge exchange their coupling to natural aerosols occurs (UNSCEAR, 2000). This creates naturally occurring radioactive aerosols. Of the four natural radioactive isotopes of lead: ^{210}Pb, ^{211}Pb, ^{212}Pb and ^{214}Pb, the presence of lead isotope ^{210}Pb in the atmosphere is most significant due to its radiotoxicity and isotopic abundance, which is caused by a half-life of 22.26 years. The largest part of ^{210}Pb binds to aerosols whose dimensions are of the order of several hundred nanometers (Suzuki et al., 1999). Otherwise, there are four stable isotopes in natural lead: ^{204}Pb (1.48%), ^{206}Pb (23.6%), ^{207}Pb (22.6%) and ^{208}Pb (52.3%) (Los Alamos National Laboratory, 2016). The ^{204}Pb lead is the primordial from the solar nebula and serves as a constant reservoir reference isotope, while the other three isotopes are end products of the decay of natural radioactive elements of the radioactive series.

The composition and structure of the soil define its porosity and absorption power for various radioactive elements (Gržetić and Jelenković, 1995). Radon diffusion within the soil matrix depends significantly on these factors. For the certain chemical composition of the soil and even with a certain configuration of the roads within

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the soil, radon cannot diffuse, so its decay products remain in the soil. Therefore, the long-lived ^{210}Pb lead isotope in soil samples and plants growing on that soil has two sources: ^{210}Pb generated *in situ* from its parent ^{226}Ra in which the ^{222}Rn precursor did not reach the atmosphere ("supported"). Another contribution is the so-called ^{210}Pb excess ("unsupported" or "excess"), $^{210}Pb_{exc}$, arising from the decay of ^{222}Rn in the atmosphere, which is removed from the atmosphere by precipitation or dry deposition due to gravitational forces, while retaining the vegetation, and gently penetrates the soil. However, during rainy periods, the ^{210}Pb isotope reached by surface wet and dry aerosol deposition onto the soil surface, rinsing with surface water into watercourses.

Lead can be introduced into the human body by inhalation of aerosols, ingestion of contaminated food and water, as well as contact through the skin. Depending on the accumulated dose, ^{210}Pb lead, as an alpha-radioactive nuclide, is extremely dangerous when absorbed by the human body, as it can lead to various hematologic, immunological, reproductive disorders, as well as an increased incidence of benign and malignant neoplasms in various tissues and organs (Counis, 1998). According to the physicochemical properties, Pb^{2+} ions can be easily replaced by Ca^{2+} ions in calcified tissues (bones and teeth), as well as in various soluble complexes of this metal with bioligands in biological fluids and tissues.

In order to obtain a more detailed and reliable distribution of areas with increased radioactivity, and because of the inability to use conventional techniques, biomonitors are used which, due to their wide distribution, are very suitable for monitoring the level of radioactivity in the environment. Bioindicators are plant or animal organisms that accumulate chemical elements or compounds from the environment and indicate exposure to the biota. The use of bioindicators can verify the occurrence of radionuclide concentrations, determine their spatial and temporal distribution, monitor local, regional and sub-regional levels of pollution, and monitor their long-term environmental effects (Wolterbeek, 2002). Simplification and reduction of radiomonitoring costs are also significant, with a high level of reliability of measurement results. Because of all these advantages, this biological method has become an adjunct to radiomonitoring carried out by nuclear measuring methods.

Mosses are most commonly used in monitoring atmospheric pollution (Szczepaniak and Biziuk, 2003). Mosses have been confirmed as a reliable link in the biomonitoring of global radionuclide deposition, primarily in examining the effects of nuclear tests in the atmosphere, as well as in assessing the consequences of the Chernobyl nuclear accident in 1986 (Marović, et al., 2008). Due to their widespread distribution and easy collection, as well as their ability to accumulate all types of radionuclides present in the atmosphere and in precipitation, mosses are considered to be the most suitable bioindicators. Unlike other taller plants, mosses do not have conductive tissues, so they are also called non-vascular plants (Nikolić, 2013). These are non-vascular plants, which means they have no conductive system in the form of phloem and xylem, to settle with water and nutrients. Therefore, all mosses can absorb water throughout the surface of the body. Thus, the vegetative body of moss is not differentiated into three basic parts: root, stem and leaf, but analogous entities exist. The role of the root is performed by rhizoids, stem kauloids, and leaf phyloids. Rhizoids are thin, tube-like hairs that secure the moss body to the substrate. Most mosses are ectohydric, meaning they absorb water and inorganic matter directly from the atmosphere rather than from soil or substrate. However, some studies of moss as a sensitive bioindicator show that resorption from soil is not a negligible process and therefore a less important source of soil contamination (Klos et al., 2012). This process is primarily conditioned by the optimal humidity of the entire surface layer on which the moss grows.

Ecological heterogeneity of the area of Bosnia and Herzegovina, geomorphological and hydrological diversity, specific geological past, diversity of eco-climate, have caused the rich wildlife on its territory. Southeast Europe is biologically the least explored area of Europe. Bryophytes flora of the region accommodates hornworts, 267 liverworts and 897 moss species (Sabovljević et al., 2011). Within this, knowledge of mosses in Bosnia and Herzegovina is meager in comparison with data from Western Europe countries. The report for the Convention on Biological Diversity of Bosnia and Herzegovina according to the area of Bosnia and Herzegovina is characterized by a high diversity of mosses (Redžić et al., 2008). Bryophytes B&H had counted 565 species from 52 genera and 187 families, which fall into two classes, the most diverse types of families are Pottiaceae (71), Bryaceae (55) and Brahytheciaceae (42) (Redžić et al., 2008). Figure 1 shows the positions of measuring points in the territory of Bosnia and Herzegovina.

MATERIALS AND METHODS

Mosses in these studies were sampled on 92 localities from Bosnia and Herzegovina (Figure 1). Different types of mosess habitats were chosen: land, lignohumus in forest and meadow ecosystems, trees, limestone, ultrabasic and silicate rocks and cliffs (Figure 2). Several species are sampled in spring habitats, streams and rocky barriers. For reasons of representativeness of the samples, at each location were taken about five samples of mosses from five different places, which were located on the surface of approximately 30 m^2, followed by mixing samples from each location in one composite sample. This procedure is performed at all locations where it was possible, due to the configuration of the terrain and to the

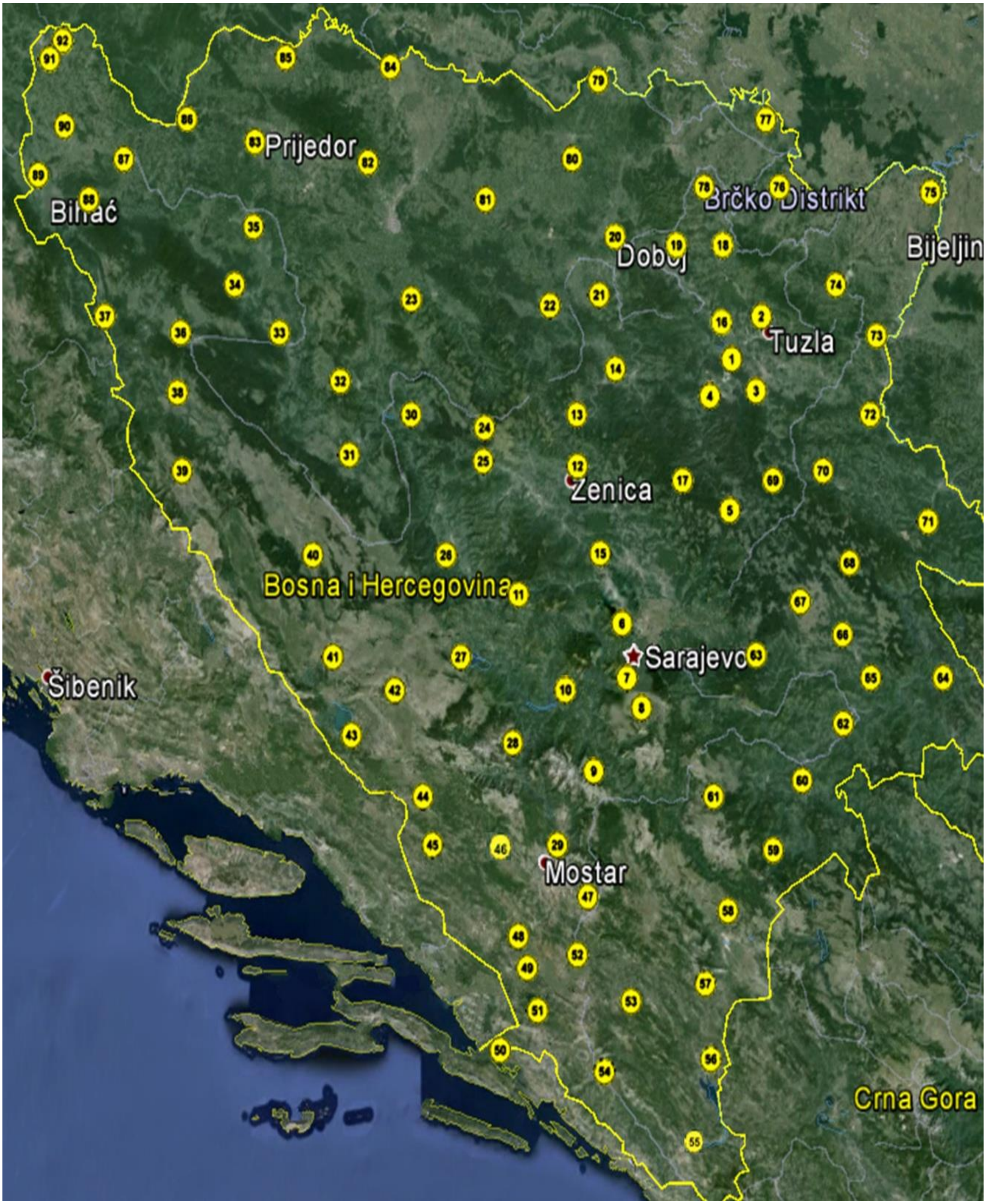


Figure 1. Positions of measurement points on the territory of Bosnia and Herzegovina.

Figure 2. Moss samples collected to: **a,** from the ground; **b,** from the tree wood; **c,** from the stone.

distribution of mosses. In the case of urban areas, a larger surface area for sampling where used, due to the lower cover of mosses. A particular problem for the sampling of mosses was minefields. In Bosnia and Herzegovina is still about 1417 mined sites, which in total have endangered the lives of more than half a million people or about 15% of the total population in Bosnia and Herzegovina. Experts warn that the number of mine sites in Bosnia and Herzegovina is much higher, because it is practically impossible to trace each mine or to trace some other unexploded ordnance that is still remaining after the last conflict in Bosnia and Herzegovina. Identification of moss in these studies was performed using a optical microscope (Motic) and following literature keys (Smith, 2004; Pavletić 1968).

Moss sample preparation was performed in the Laboratory of dosimetry and radiation protection (LDRP) of the Faculty of Natural Sciences and Mathematics, University of Tuzla (Figure 3). 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 mosses samples were dried again at 105°C for at least 24 h and homogenized. Low radioactivity samples can be effectively measured in Marinelli-type containers or in cylindrical containers of certain volumes. In the performed measurements on three gamma spectrometers, these two geometries were used. With the ground powder from the samples were filled the plastic 500 mL Marinelli beakers and cylindrical vessels with a height of 60 mm and a diameter of 70 mm,

Figure 3. Preparation of the moss samples (LDDRP) at University of Tuzla.

in order to be compact and filled with an equal volume of the whole container and thus has received the uniform packing density. The pots were then closed with the beeswax and aged for 30 days (before the start of measurement) to establish radioactive equilibrium. The background spectrum was recorded regularly after or before the sample counting, with an empty cylindrical polyethylene vessel, and plastic Marinelli beaker.

There are two dominant methods used to determine the specific activity of lead isotope ^{210}Pb: alpha spectrometry and gamma spectrometry. In these studies, the gamma spectrometric method was used. Performed by gamma spectrometric measurements of ^{210}Pb activities were carried out according to standard ASTM C 1402-04 Standard Guide for High Resolution Gamma Ray Spectrometry. Three HPGe detectors of the following characteristics were used in the study:

- Canberra HPGe system, GR7023-7500SL model, the energy range of 3 keV to 10 MeV, the relative efficiency 70%, the energy resolution at 1.33 MeV 60Co is 2.30 keV FWHM (full width at half maximum).
- Canberra HPGe system, model GC3518, the relative efficiency 35%, the energy resolution at 1.33 MeV ^{60}Co is 1.77 keV FWHM.
- Canberra HPGe system, model GC 2018-7500, the relative efficiency 20%, the energy resolution at 1.33 MeV ^{60}Co is 1.8 keV FWHM.

In order to calibrate and to check the efficiency of the detectors certified calibration standards and reference materials were used. The laboratory for calibration of □□-spectrometers and for measurements of activity of radioactive sources -□ emitters, Department of Nuclear Physics, University of Novi Sad - did the calibration of gamma-spectrometers on the efficiency, using the method of ASTM E 181-98/03 and (ASTM 2003), and using certified standards: a) ^{152}Eu, Marinelli geometry, from a manufacturer of Framatome, France, Model EU152EGR115, with an activity of 32.5 Bq [uncertainty of measurement was: 5%, the level of confidence was: 95% (at 2σ)], and b) ^{241}Am, point source - Standard source, from a manufacturer of Framatome, France, Model 9CH03EGSA20, with an activity of 437 Bq (measurement uncertainty 3.5% at 2σ) (Adrovic et al., 2017). The spectra were analysed using the program GENIE 2000 (Canberra, 2013).

Based on the intensity of the gamma line at 46.539 MeV that were recorded in the measured spectra, the specific activity of the lead isotope ^{210}Pb in the samples (*A*) was calculated using the equation:

$$A = \frac{N}{m \cdot t \cdot P_{\gamma} \cdot \varepsilon} \qquad (1)$$

where *N* is count rate of the sample, *m* – mass (kg) of the sample, *t* – counting time (s), P_{γ} – probability of gamma decay (%), and ε – detector efficiency (%).

RESULTS AND DISCUSSION

The results of gamma spectrometric analysis of the concentration of ^{210}Pb in moss samples collected from 92 sites throughout Bosnia and Herzegovina are shown in Table 1. The ^{210}Pb lead isotope was detected in all moss samples analyzed. The concentrations of ^{210}Pb in the moss samples at the investigated sites were in the range of (10 - 2000) Bq/kg. The mean specific activity of ^{210}Pb of analyzed moss samples in Bosnia and Herzegovina was 413 Bq/kg. The highest activity level of ^{210}Pb of 2000 Bq/kg was detected in moss *Polytrichum commune* L. from site No. 57 Gacko (the sea elevation of 1048 m).

The lowest activity level of ^{210}Pb, 10 Bq/kg, was measured in a sample of *Hypnum cupressiforme* moss, from site No.13, Nemila (the sea elevation of 228 m). Special attention in the research is paid to the

Table 1. The specific activities of ^{210}Pb (Bq/kg dry weight) in moss samples from the territory Bosnia and Herzegovina.

S/N	Location	Coordinates	Species	^{210}Pb
1	Modrac	44°29’27.26“ N, 18°30’17.04“ E	*Brachythecium capillaceum*	400 ± 82
2	Tuzla	44°32’22.20“ N, 18°41’13.11“ E	*Callicladium haldanianum*	513 ± 99
3	Živinice	44°27’19.53“ N, 18°41’42.70“ E	*Brachytecium albicans*	279 ± 54
4	Banovići	44°24’37.29“ N, 18°27’03.90“ E	*Brachytecium albicans*	160 ± 45
5	Olovo	44°05’41.26’’ N, 18°33’07.76“ E	*Hypnum cupressiforme*	710 ± 70
6	Sarajevo	43°52’59.97“ N, 18°23’01.14“ E	*Brachythecium capillaceum*	240 ± 51
7	Igman	43°46’05.27“ N, 18°19’26.99“ E	*Brachythecium mildeanum*	< 10
8	Bjelašnica	43°42’41.64“ N, 18°18’13.30“ E	*Bryum capillare*	300 ± 65
9	Boračko j.	43°33’11.06“ N, 18°02’08.48“ E	*Brachythecium capillaceum*	120 ± 20
10	Ivan-Sedlo	43°44’23.20“ N, 18°02’22.83“ E	*Brachythecium mildeanum*	< 60
11	Fojnica	43°57’50.16“ N, 17°53’38.07“ E	*Grimmia pulvinata*	1100 ± 90
12	Zenica	44°12’06.32“ N, 17°54’41.75“ E	*Brachitechium rutabutum*	560 ± 62
13	Nemila	44°19’45.90“ N, 17°55’43.12“ E	*Hypnum cupressiforme*	10 ± 3
14	Zavidovići	44°26’04.70“ N, 18°09’27.52“ E	*Callicladium haldanianum*	180 ± 38
15	Visoko	43°58’48.87“ N, 18°10’42.98“ E	*Brachytecium albicans*	200 ± 41
16	Lukavac	44°32’27.08“ N, 18°31’57.35“ E	*Brachytecium velutinum*	342 ± 61
17	Vareš	44°08’48.15“ N, 18°19’24.88“ E	*Brachitechium rutabutum*	580 ± 70
18	Srebrenik	44°42’59.40“ N, 18°27’21.38“ E	*Brachytecium velutinum*	187 ± 42
19	Gračanica	44°42’17.29“ N, 18°20’29.39“ E	*Callicladium haldanianum*	120 ± 28
20	Doboj	44°43’53.13“ N, 18°02’45.61“ E	*Hypnum cupressiforme*	45 ± 7
21	Tešanj	44°36’46.98“ N, 17°59’34.98“ E	*Bryum capillare*	660 ± 74
22	Teslić	44°37’27.49“ N, 17°51’14.89“ E	*Brachythecium capillaceum*	247 ± 45
23	Kotor Varoš	44°36’53.54“ N, 17°34’34.08“ E	*Hypnum cupressiforme*	330 ± 70
24	Vlašić	44°19’10.03“ N, 17°34’15.03“ E	*Bryum capillare*	< 40
25	Travnik	44°26’04.70“ N, 17°39’35.23“ E	*Grimmia pulvinata*	760 ± 60
26	Bugojno	44°02’24.60“ N, 17°26’36.48“ E	*Brachythecium capillaceum*	440 ± 91
27	Ramsko jezero	43°49’02.46“ N, 17°31’43.03“ E	*Polytrichum commune* L.	430 ± 40
28	Jablanica	43°39’16.35“ N, 17°45’18.37“ E	*Brachythecium capillaceum*	< 20
29	Mostar	43°22’02.04“ N, 17°50’11.88“ E	*Grimmia pulvinata*	493 ± 88
30	Jajce	44°20’26.62“ N, 17°16’30.94“ E	*Hypnum cupressiforme*	582 ± 110
31	Šipovo	44°17’28.35“ N, 17°05’56.12“E	*Callicladium haldanianum*	< 20
32	Mrkonjić Grad	44°24’27.68“ N, 16°45’27.50“ E	*Callicladium haldanianum*	474 ± 78
33	Ključ	44°31’52.73“ N, 17°45’18.37“ E	*Grimmia pulvinata*	688 ± 136
34	Sanica	44°37’05.57“ N, 16°39’25.94“ E	*Brachythecium capillaceum*	455 ± 52
35	Sanski Most	44°45’29.93“ N, 16°40’11.27“ E	*Brachytecium albicans*	530 ± 85
36	Bos. Petrovac	44°33’46.89“ N, 16°04’53.63“ E	*Bryum capillare*	900 ± 187
37	Kulen Vakuf	44°21’58.92“ N, 16°23’15.91“ E	*Atrichum undulatum*	610 ±50
38	Drvar	44°10’50.05“ N, 16°21’03.66“ E	*Eurinchyum hians*	460 ± 70
39	Bosansko Gra.	44°02’25.60“ N, 16°51’35.05“ E	*Brachytecium albicans*	820 ± 80
40	Glamoč	44°02’25.60“ N, 16°51’35.05“ E	*Hygrohypnum eugyrium*	300 ± 71
41	Livno	43°49’49.13“ N, 17°00’32.29“ E	*Polytrichum strictum*	653±87
42	Tomislavgrad	43°43’12.84“ N,17°12’49.79“ E	*Brachythecium capillaceum*	135 ± 25
43	Buško jezero	43°42’23.72“ N, 16°58’01.37“ E	*Hygrohypnum eugyrium*	113 ± 21
44	Posušje	43°27’54.63“ N, 17°19’44.63“ E	*Grimmia pulvinata*	432 ±53
45	Grude	43°21’28.82“ N, 17°25’01.93“ E	*Hypnum cupressiforme*	95 ± 21
46	Široki Brijeg	43°23’03.05“ N, 17°35’39.26“ E	*Callicladium haldanianum*	300 ±50
47	Blagaj	43°15’32.65“ N, 17°52’58.43“ E	*Brachytecium albicans*	800 ± 150
48	Međugorje	43°11’58.75“ N, 17°41’00.03“ E	*Hypnum cupressiforme*	598 ± 97
49	Čapljina	43°06’26.94“ N, 17°42’38.95“ E	*Callicladium haldanianum*	200 ± 44
50	Neum	42°55’46.79“ N, 17°36’40.66“ E	*Cratoneuron filicinum*	460 ± 40
51	Huntovo blato	43°03’51.42“ N, 17°45’14.39“ E	*Eurinchyum hians*	260 ± 56

Table 1. Contd.

52	Stolac	43°04’56.81“ N, 17°57’09.09“ E	*Hypnum cupressiforme*	271 ± 52
53	Ljubinje	42°57’49.20“ N, 18°94’04.29“ E	*Bryum capillare*	620 ± 160
54	Popovo polje	42°50’53.52“ N, 17°58’40.21“ E	*Brachythecium populeum*	411± 49
55	Trebinje	42°41’47.82“ N, 18°20’46.25“ E	*Brachytecium albicans*	300 ± 64
56	Bileće	42°52’42.53“ N, 18°25’22.09“ E	*Hypnum cupressiforme*	340 ± 40
57	Gacko	43°09’41.46“ N, 18°33’11.80“ E	*Polytrichum commune* L.	2000 ± 200
58	Čemerno	43°13’50.17“ N, 18°35’00.43“ E	*Bryum capillare*	220 ± 36
59	Tjentište	43°20’40.86’’ N, 18°23’21.71“ E	*Pylaisia polyantha.*	390 ± 40
60	Foča	43°29’48.04“ N, 18°45’58.35“ E	*Eucladium angustifolium*	430 ± 40
61	Kalinovik	43°30’05.96“ N, 18°27’09.90“ E	*Brachythecium mildeanum*	560 ± 51
62	Goražde	43°40’09.64“ N, 18°59’40.43“ E	*Brachytecium albicans*	499 ± 45
63	Pale	43°48’56.71“ N, 18°34’38.36“ E	*Hypnum cupressiforme*	1000 ± 90
64	Višegrad	43°46’58.91“ N, 19°17’12.18“ E	*Brachythecium mildeanum*	128 ± 22
65	Rogatica	43°49’27.28“ N, 18°57’37.96“ E	*Callicladium haldanianum*	100 ± 13
66	Podromanija	43°52’09.92“ N, 18°51’04.91“ E	*Brachytecium albicans*	242 ± 27
67	Sokolac	43°56’09.18“ N, 18°48’28.24“ E	*Brachytecium albicans*	410 ± 38
68	Han Pijesak	44°04’58.35“ N, 18°56’52.33“ E	*Brachythecium mildeanum*	510 ± 55
69	Kladanj	44°13’46.67“ N, 18°41’57.22“ E	*Hypnum cupressiforme*	394 ± 41
70	Tišča	44°13’11.38“ N, 18°53’43.29“ E	*Bryum capillare*	470 ± 40
71	Srebrenica	44°06’05.97“ N, 19°17’56.91“ E	*Plagiomnium undulatum*	530 ± 53
72	Zvornik	44°22’35.10“ N, 19°05’44.84“ E	*Brachytecium albicans*	318 ± 40
73	Kozluk	44°29’56.84“ N, 18°97’19.91“ E	*Atrichum undulatum*	430 ± 51
74	Ugljevik	44°41’36.66“ N, 19°00’06.91“ E	*Brachythecium rutabulum*	572 ± 60
75	Dvorovi	44°48’25.67“ N, 19°15’41.32“ E	*Hypnum cupressiforme*	361 ± 42
76	Brčko	44°49’17.85“ N, 18°46’34.25“ E	*Plagiomnium undulatum*	310 ± 39
77	Orašje	45°02’19.28“ N, 18°41’29.88“ E	*Callicladium haldanianum*	303 ± 32
78	Gradačac	44°52’38.26“ N, 18°25’26.76“ E	*Brachytecium albicans*	910 ± 41
79	Bosanski Brod	45°08’56.26“ N, 18°00’05.83“ E	*Hypnum cupressiforme*	446 ± 35
80	Derventa	44°58’46.30“ N, 17°54’03.56“ E	*Callicladium haldanianum*	560 ± 50
81	Prnjavor	44°52’01.63“ N, 17°39’42.34“ E	*Brachytecium velutinum*	463 ± 42
82	Potkozarje	44°54’49.02“ N, 17°02’50.67“ E	*Atrichum undulatum*	325 ± 41
83	Prijedor	44°58’35.20“ N, 16°42’14.47“ E	*Hypnum andoi*	399 ± 42
84	Gradiška	45°08’40.26“ N, 17°14’01.20“ E	*Polytrichum commune* L.	412 ± 40
85	Bosanska Dub	45°10’51.21“ N, 16°47’15.64“ E	*Pylaisia polyantha*	487 ± 51
86	Novi Grad	45°02’52.18“ N, 16°22’48.07“ E	*Callicladium haldanianum*	399 ± 40
87	Bosanska Kru	44°52’52.63“ N, 16°08’30.46“ E	*Brachythecium mildeanum*	1000 ± 91
88	Bihać	44°48’45.10“ N, 15°52’15.80“ E	*Plagiomnium undulatum*	322 ± 35
89	Izarčić	44°52’43.98“ N, 15°47’52.04“ E	*Brachytecium albicans*	109 ± 16
90	Cazin	44°58’06.26“ N, 15°56’41.71“ E	*Polytrichum commune* L.	521 ± 55
91	Velika Klad	45°10’56.51“ N, 15°47’58.60“ E	*Brachytecium albicans*	443 ± 66
92	Ponikve	45°12’53.74“ N, 15°50’42.74“ E	*Brachytecium albicans*	544 ± 42

consideration of lead isotope pollution of ^{210}Pb of anthropogenic origin, which is a consequence of technologically modified natural radioactivity. Namely, various technological and physical processing procedures, as well as certain ways of using materials containing natural radionuclides (NORM), obtain materials of increased natural radioactivity (TENORM-Technologically Enhanced Naturally Occurring Radio-active Materials). This primarily refers to fly ash from the chimneys of thermal power plants, sediment ash and slag, which are emitted in huge quantities into the atmosphere and disposed of in landfills. Anthropogenic sources include emissions from various steel and non-ferrous metallurgy plants, the petrochemical industry, smelters, industrial and individual combustion plants, as well as exhaust gases from traffic. Moss samples, collected in the impact zone of coal-fired power plants (CFPP) Tuzla (No. 2), CFPP Gacko (No. 57), CFPP

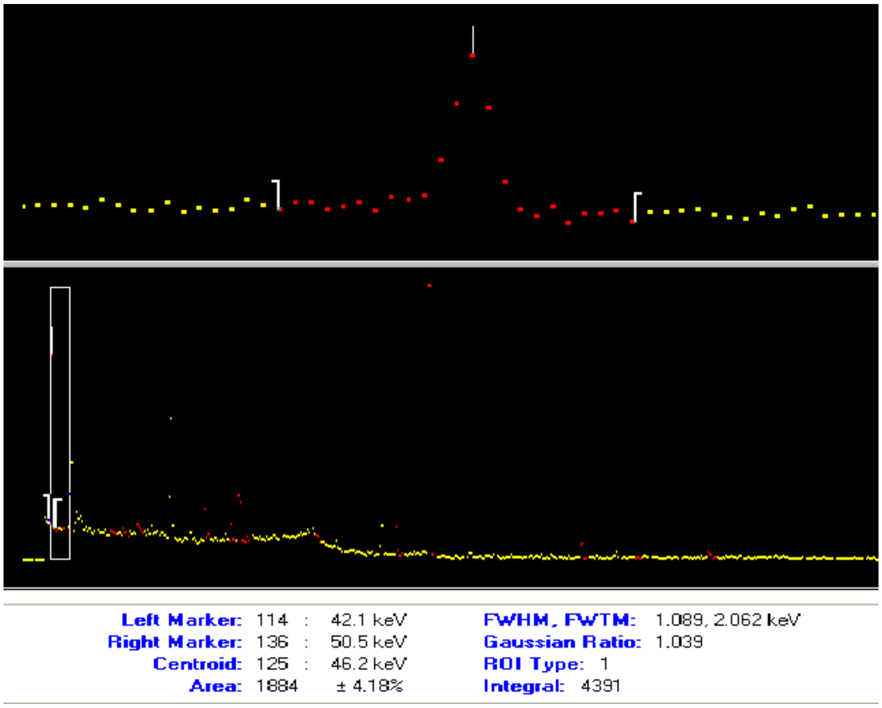
Figure 4. Coal-fired power plants (CFPP) Gacko, measuring point No 57.
Source: Center for Environment – Gacko.

Ugljevik (No. 74), and from the area of industrial cities Zenica (No. 12) and Lukavac (No 16), had the following mean values of lead isotope activity ^{210}Pb: (513 ± 99) Bq/kg, (572 ± 60) Bq/kg, (2000 ± 200) Bq/kg, (342 ± 61) Bq/kg and (560 ± 62) Bq/kg, respectively. Figure 4 shows the measuring point where the mosses with the highest concentration of lead isotope ^{210}Pb were sampled, while Figure 5 shows gamma spectrum of sample No. 57 with a focuse on the specific activity. In this area, in open pits, coal is produced and extracted, as well as its combustion in the strong CFPP Gacko, which results in enormous environmental pollution.

On the other hand, in the places: Fojnica (No. 11), Bosanska Krupa (No. 87) and Pale (No. 63), which are considered ecologically clean areas, and are relatively far from large industrial plants, the specific activity of ^{210}Pb and amounted to (1100 ± 90) Bq/kg, (1000 ± 91) Bq/kg and (1000 ± 90) Bq/kg, respectively. The reasons for these amounts of specific activity of ^{210}Pb in mosses should be sought in the increased exhalation of ^{222} Rn from the surface soil and the processes of deposition of its decomposition products, as well as the frequency of movement of regional air masses. The largest part of the generated atoms of the ^{210}Pb lead isotope binds to aerosols whose dimensions are of the order of several hundred nanometers (Suzuki et al., 1999). Aerosols that are present in the atmosphere can be transported by moving air masses to distances of several hundred, even over a thousand kilometers from the source of pollution, before being removed from the atmosphere in the processes of dry and wet deposition. Figure 6 shows the frequencies of specific activity of lead ^{210}Pb in moss of Bosnia and Herzegovina.

Table 1 shows that the spatial distribution of ^{210}Pb activity is markedly uneven. Activity levels of ^{210}Pb in moss samples varied from place to place. This indicates that its distribution and deposition are functions of different meteorological conditions as well as different emanations of radon from the soil. Namely, in the open air space, radon gas comes from the soil with a flux density that depends on the geological composition and altitude of the soil, the dispersion in the atmosphere, which is all related to meteorological conditions. Exhaling from the earth's surface into the free atmosphere, radon dissipates in it under the influence of vertical convection and turbulent mixing, due to which its concentration decreases with height. Radon reaches the atmosphere by diffusion through a system of pores and cavities in the soil. For a given concentration of radon in the soil, the concentration of this gas emanating into the atmosphere depends on the permeability of the soil, humidity and meteorological parameters (soil and air temperature, atmospheric pressure, wind speed and blowing direction). Due to its relatively long half-life (3.82 days), radon can stay in the atmosphere for a relatively long time before it decays. In this way, it participates in turbulent transmission through the atmosphere and can reach its higher layers and travel great distances. For these reasons, the reliability of the detection of the ^{222}Rn generation site, and thus its long-lived decay product of the lead isotope ^{210}Pb, is a complex scientific problem.

Likewise, the processes of leaching and displacement of ^{210}Pb could have led to a recognizable uneven distribution of this radionuclide even in one area. The



Left Marker: 114 : 42.1 keV
Right Marker: 136 : 50.5 keV
Centroid: 125 : 46.2 keV
Area: 1884 ± 4.18%

FWHM, FWTM: 1.089, 2.062 keV
Gaussian Ratio: 1.039
ROI Type: 1
Integral: 4391

Figure 5. Gamma spectrum of sample No. 57, with a focused peak on the 46.2 keV line of lead ^{210}Pb (specific activity of ^{210}Pb in this sample was 2000 Bq/kg), measured on a Canberra HPGe system, model GC3518.

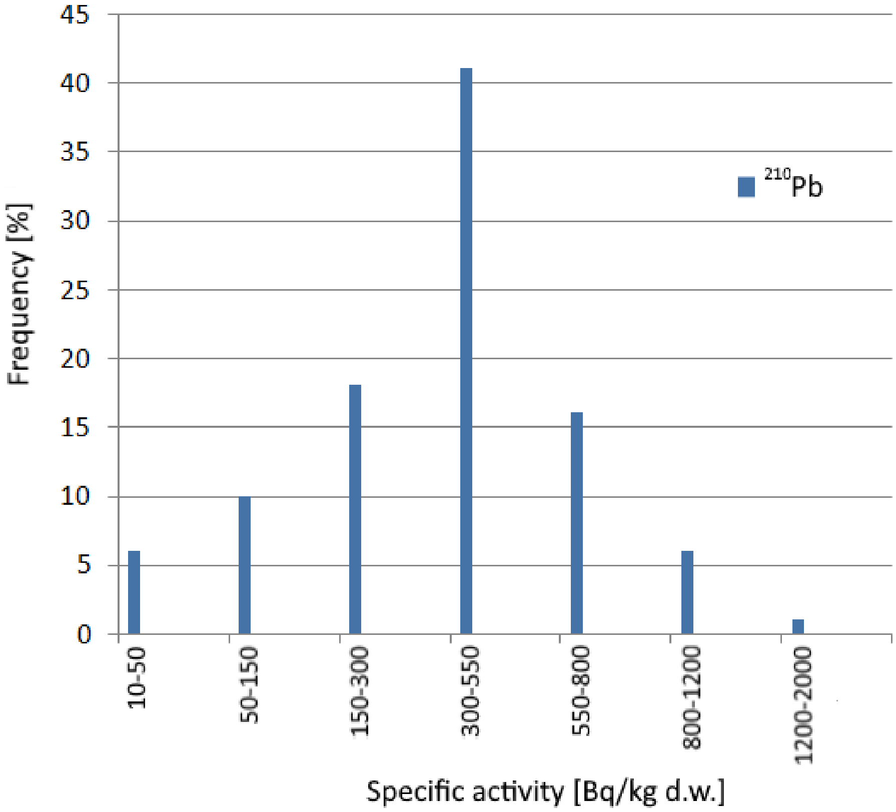


Figure 6. Frequency of ^{210}Pb specific activity [%] in moss in Bosnia and Herzegovina.

dependence of the degree of accumulation of lead ^{210}Pb on the type of moss has been noticed, which is understandable, considering the diversity of their biological characteristics, such as structure, size, reproduction, ecology, etc. Based on the results of gamma-spectrometric analysis, it can be concluded that in most locations in Bosnia and Herzegovina the largest "collectors" of this radiotoxic moss pollutant are: *P. commune* L., *Grimmia pulvinata*, *Bryum capillare, H. cupressiforme, Callicladium haldanianum*.

At all measuring points where moss samples were taken, measurements of the concentration of external radon activity and the strength of the equivalent dose of gamma radiation were performed at the same time. These measurements were performed using Alpha GUARD PQ 2000 PRO and Gamma Tracer, professional measuring systems of Bertin Instruments Corporation (developed at Genitron Instruments-Frankfurt). Mean dose rates of gamma radiation in the air at these locations ranged from 81 to 151 nGy/h. These data are directly related to the activity concentrations of ^{210}Pb, as a long-lived descendant of radon decay, because the gamma rays ^{214}Bi and ^{214}Pb are by far the largest component and the largest energy in the uranium series. Gamma radiation, under normal conditions, varies from 20 to 1100 nGy/h worldwide (UNSCEAR, 2000).

Measurements of the concentration of external radon activity were performed at all 92 locations where moss sampling was performed (Adrovic et al., 2017). The time interval of measurements ranged from 3 to 6 h, and in some locations even longer, depending on the meteorological conditions in the field. At some locations, where the weather conditions (intense winds) changed during the measurement, the measurement was performed at intervals. The AlphaGuard PQ 2000 PRO measuring system is placed 1 m above the ground. The mean values of concentrations of radon activity at the investigated localities were in the range of 15 to 38 Bq/m^3.

In the investigated samples of moss from certain locations, there is a moderate to good correlation between the concentration of radon activity and the concentration of activity of its long-lived offspring, the lead isotope ^{210}Pb. A good correlation of these variables was observed in moss samples sampled in bays and valleys, wooded areas, and in locations less exposed to winds. However, in a number of investigated locations there is no correlation of these quantities, primarily due to the fact that deviations in radon concentration are unpredictable and depend on a large number of meteorological variables. This primarily refers to atmospheric pressure, temperature and humidity, which, in addition to geological conditions, predominantly determine the levels of radon in the air, and thus the amount of deposited radioactive lead ^{210}Pb on the soil surface. These were mainly the reasons why the Pearson correlation coefficient between the concentration of radon activity and the concentration of lead isotope activity ^{210}Pb in moss samples gives a relatively weak correlation of $r_{\mathrm{Rn,\,Pb}} = 0.38$.

Conclusion

Due to its high radiotoxicity and potential radiological impact on human health, the lead isotope ^{210}Pb is one of the most important radionuclides in the environmental load. For this reason, in the last few decades, great attention has been paid to it all over the world. In this paper, the levels of contamination of mosses with ^{210}Pb radio lead, which were obtained from 92 locations in the territory of Bosnia and Herzegovina, were investigated. In all tested samples of moss contained ^{210}Pb, with concentrations of ^{210}Pb ranged from 10 to 2000 Bq/kg dry weight in selected moss species. Increased levels of ^{210}Pb in some samples of moss are mainly a consequence of direct and indirect anthropogenic activities, but also in increased amounts of exhaled ^{222}Rn from the surface soil, as well as the frequency of movement of regional air masses. Thanks to a number of characteristic morphological and physiological properties, mosses have a great power of accumulation of pollutants. Even with large amounts of contaminants, there is no damage to their organism or disturbances in the reproductive cycle, the reasons are that these plants have become the basic bioindicators of radio pollution of ecosystems.

On the other hand, because mosses have a long life, those contaminated with higher amounts of radionuclides represent an important component in the natural and artificial increase in levels of activity in the environment over a longer period of time. Due to the radiation safety of the population, it is necessary to further monitor the activity levels of these bioindicator species in the same localities.

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