RAP CONFERENCE PROCEEDINGS, VOL. 7, PP. 57–57, 2022
ISSN 2737-9973 (ONLINE) | DOI: 10.37392/RAPPROC.2022.13
RAP-PROCEEDINGS.ORG

MONITORING COSMOGENIC AND TERRESTRIAL RADIONUCLIDES IN GROUND LEVEL AIR SAMPLES BY GAMMA SPECTROMETRY IN ALBANIA

Erjon Spahiu[1], Irma Bërdufi[2], Manjola Shyti[2*], Florinda Cfarku[2]

[1]Department of Physics, Faculty of Natural Sciences, University of Tirana, Tirana, Albania
[2]Institute of Applied Nuclear Physics, University of Tirana, Tirana, Albania

Abstract. *The activity concentrations of ^{7}Be, ^{210}Pb, ^{40}K and ^{137}Cs in ground level air at the monitoring station in Tirana, Albania were determined during the period from January 2021 to January 2022. To perform a routine air radioactivity monitoring, we used a typical aerosol sampling station located at the Institute of Applied Nuclear Physics in Tirana not only for routine air radioactivity monitoring, but also to monitor the air in the institute from the radiation protection point of view because in the institute are located the temporary radioactive waste site, ^{137}Cs source used in the secondary standard dosimetry laboratory and ^{137}Cs irradiation source. Activities in all aerosol samples are measured by gamma spectrometer with High Purity Germanium detector (HPGe). The cylinder geometry efficiency curve generated by Canberra's Laboratory Sourceless Calibration Software (LabSOCS) was used to analyze the air filters. The obtained results show the activity concentrations of cosmogenic ^{7}Be ranged from 2.38 to 6.82 mBq m^{-3} with a maximum in the spring/summer period. The activity concentrations for ^{210}Pb were in the range 0.37 to 1.27 mBq m^{-3}. The activity concentrations of anthropogenic ^{137}Cs in ground level air was observed only in three air filters in the range 0.30–6.01 μBq m^{-3}. The monitoring is done for the first time in Albania, providing us the data of cosmogenic and terrestrial radionuclides in ground level air. This study will continue also in the future in order to see the variation of radionuclides during the years.*

Keywords: *air filter, cosmogenic radionuclides, aerosol samples, HPGe gamma-ray spectrometry*

1. INTRODUCTION

The work presented in this paper describes the detection of artificial and natural radionuclides in the ground-level atmosphere by means of continuous monitoring of the radioactivity collected on a filter. This monitoring is done for the first time in Albania, providing us the data for the assessment of the environmental impact of radioactivity from anthropogenic sources compared with natural ones. Mainly, the presence of cosmogenic radionuclides in the Earth's atmosphere are produced by the secondary cosmic rays in the stratosphere or troposphere. The typical monitored radionuclides in ground level air are cosmogenic ^{7}Be, natural ^{210}Pb, ^{40}K and anthropogenic ^{137}Cs [1-2].

Cosmogenic ^{7}Be is formed primarily from the cosmic ray spallation of oxygen and nitrogen in the stratosphere, troposphere, and surface of the earth. ^{210}Pb is produced in the lower troposphere and may rapidly attach to aerosol particles, which is mainly accomplished via washout by precipitation from the atmosphere [3]. ^{210}Pb its daughter of ^{222}Rn, which emanates from the earth's crust and decays in the atmosphere. ^{222}Rn is affected from different factors such as atmospheric pressure variations, seasonal variations in meteorological conditions, temperature inversions, precipitations accumulation, soil moisture and ground coverage by snow and ice. Therefore, there will be variation of activity concentrations of ^{210}Pb in ground level air [4]. For identifying and quantifying several atmospheric processes the natural radionuclides of ^{7}Be and ^{210}Pb serve as powerful tracers. ^{7}Be and ^{210}Pb can be widely used to trace the sources, transport processes, and mixing of aerosols.

Radionuclide of ^{40}K is in soil, rock and plant and can be attach to aerosol particles and ascend into the atmosphere. This effect happens more often in summer when farmer fertilize the land.

^{137}Cs is an anthropogenic radionuclide in the air, caused by nuclear weapon atmospheric tests and nuclear power plant accidents as case of Chernobyl in April of 1986.

2. EXPERIMENTAL

2.1. Sampling

The Aerosol Sampling Station (ASS-500) is located in the Institute of Applied Nuclear Physics in Tirana, Albania and it is used not only for routine air radioactivity monitoring, but also to monitor the air in the institute from the radiation protection point of view because in the institute are located the temporary radioactive waste site, ^{137}Cs source used in the secondary standard dosimetry laboratory and ^{137}Cs irradiation source. This kind of station is not applicable to detect automatically if artificial radionuclides are present on a filter during the collection period, or the increase of dose

* manjolashyti@yahoo.com

rate is the result of artificial or natural radionuclides present in air [5].

The air aerosols samples of ground level were collected during the period from January 2021 to January 2022 at the Institute of Applied Nuclear Physics (IANP) in Tirana, Albania with a typical aerosol sampler type ASS-500. In this type of sampler, we use Petryanov FPP-1.5–1.5 filters with dimensions of 0.45 m by 0.45 m. The nominal airflow rate for ASS-500 is 500 $m^3 h^{-1}$. This monitoring system works weekly, collecting aerosols from 50 000 to 80 000 m^3 of air on a single ASS-500 filter [6-7]. After that the filter is placed in a cylindrical container in order to be ready for measurement.

2.2. Gamma Spectrometry

The activity concentration of the prepared air filter samples was measured using a high-resolution gamma spectrometry system with a coaxial high purity germanium detector with a relative efficiency of 40 % and a resolution of 1.8 keV for the 1332 keV gamma ray emission of ^{60}Co. The HPGe γ-ray detector (GC4018-7500SL) was coupled with digital spectrum analyzer, DSA-1000. The detector was well shielded to minimize the γ-ray background to be able to measure low radioactivity. For analyzing of spectra was used Genie 2000 (V3.2.1) software from Canberra. Counting time interval was 86400 seconds for each air filter sample. Energy calibration was performed with a set of standard point sources, whereas the efficiency curve was obtained by LabSOCS (Laboratory Sourceless Calibration Software) [8-10]. The combined uncertainty of measured absolute efficiencies was determined to be less than 10 %. Figure 1 shows the cylinder geometry template used for efficiency calibration curve for analyzing of air filters. For calculation of Minimum Detectable Activity (MDA) Curie formula was used [11].

The calibration procedure is then cross validated through the participation in the wide-open proficiency tests [12]. Radionuclides were determined at gamma energies 477 keV for ^{7}Be, 46 keV for ^{210}Pb, 1460.8 keV for ^{40}K and 661.6 keV for ^{137}Cs. The MDA of the radionuclides in air were: 0.01 mBq m^{-3} for ^{7}Be, 0.02 mBq m^{-3} for ^{210}Pb, 0.01 mBq m^{-3} for ^{40}K, and 1.8 μBqm^{-3} for ^{137}Cs, respectively.

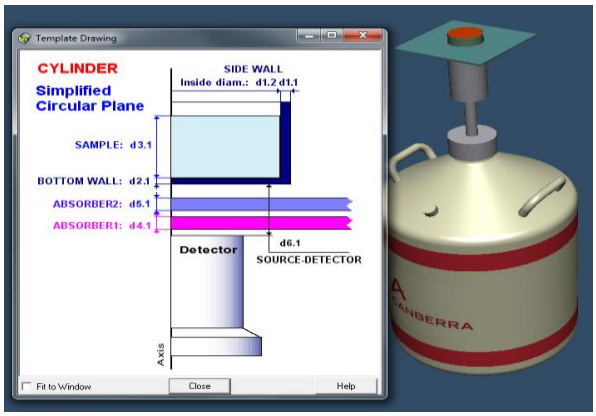

Figure 1. Cylinder geometry template used for efficiency calibration curve.

3. Results and Discussions

The levels of ^{7}Be, ^{210}Pb and ^{40}K in the analyzed samples were detected. The average monthly concentrations of these radionuclides in ground level air at IANP location in Albania during the period January 2021– January 2022 are presented in Table 1. This table provides related statistical information such as minimum and maximum values, arithmetic mean and standard deviation. These values are given in mBq m^{-3}. The activity concentrations of anthropogenic ^{137}Cs in ground level air is not presented in Table 1, because its values were below MDA and it was observed only in three air filters in the range 0.30 – 6.01 μBq m^{-3}.

Table 1. Average activity concentration of radionuclides in air filters (mBq m^{-3}) from January 2021 to January 2022.

Year 2021	**^{7}Be**	**^{210}Pb**	**^{40}K**
January 2021	2.38±0.15	0.37±0.05	0.02±0.01
February	3.29±0.20	0.53±0.08	0.05±0.01
March	2.93±0.18	0.54±0.08	0.04±0.01
April	3.55±0.22	0.45±0.06	0.09±0.03
May	5.67±0.35	0.65±0.10	0.09±0.03
June	6.82±0.43	1.19±0.17	0.17±0.03
July	5.22±0.33	1.00±0.15	0.06±0.01
August	4.77±0.30	1.17±0.17	0.02±0.01
September	5.33±0.33	1.27±0.18	0.05±0.01
October	3.29±0.21	1.09±0.16	0.03±0.01
November	3.87±0.24	0.95±0.14	0.01±0.003
December	3.45±0.22	0.65±0.10	0.02±0.01
January 2022	3.66±0.23	0.80±0.12	0.01±0.003
Min	**2.38**±0.15	**0.37**±0.05	**0.01**±0.003
Max	**6.82**±0.43	**1.27**±0.18	**0.17**±0.03

Concentrations of ^{7}Be in air were in the range of 2.38 – 6.82 mBq m^{-3} and exhibited maximum in spring/summer and minimum in winter (Figure 2). Also, the dependence of the mean monthly activity concentration of ^{7}Be versus the mean monthly temperature is given in Figure 3, reaching its higher values during the summer months. During the spring/summer season the layers of air near the ground warm up and the air climbs upwards, while the upper layers of the troposphere, which are rich in ^{7}Be descend down, near the surface of the earth, thus showing an increase in the concentration of ^{7}Be [4].

The higher values of average monthly activities of ^{210}Pb were during summer and early autumn period. Meteorological parameters such as temperature and precipitation influenced the specific activities of ^{210}Pb. The lower values of ^{210}Pb were during the winter/spring period due to rainy weeks, which reduces the concentration of radionuclides in the airborne particulate material (Figure 4). High temperatures and dry weather during the summer season, increase the amount of ^{222}Rn coming out of the ground near its surface, which is the parent radionuclide in the production of ^{210}Pb. Also, the frequent exchange of the

air layers during autumn, enriches the air layers near the ground with radon ^{222}Rn, and consequently increases the concentration of ^{210}Pb.

In the case of ^{40}K the maximum value of activity concentrations of 0.17 mBq m^{-3} were in June. ^{40}K were mainly transported to the air as resuspended particle from the soil. Its increase or decrease will depend mainly on local resuspension of soil dust and also due to industrial air pollution (Figure 5).

The temporal variation of ^{7}Be, ^{210}Pb, and ^{40}K activity concentrations in the ground level air is graphically shown.

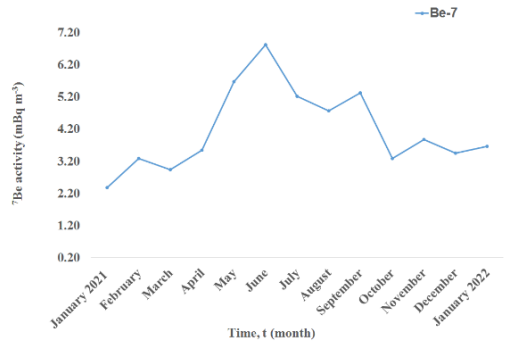

Figure 2. Temporal variation of ^{7}Be activity concentration in the ground level air.

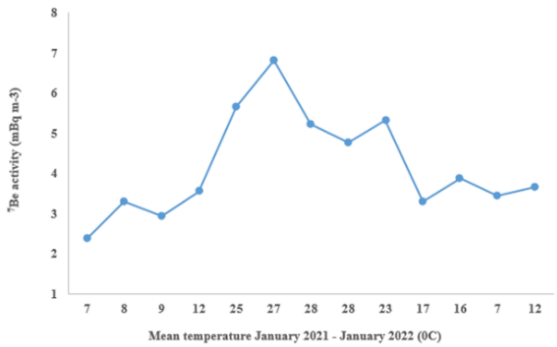

Figure 3. Mean monthly activity concentration of ^{7}Be versus temperature, T(^{o}C).

Figure 4. Temporal variation of ^{210}Pb activity concentration in the ground level air.

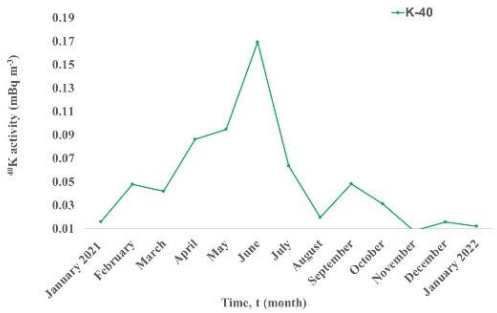

Figure 5. Temporal variation of ^{40}K activity concentration in the ground level air.

The results obtained were compared with other studies of the same type in the region. The range of activity concentrations for ^{7}Be in the air during period 2011-2012 in Belgrade (Serbia) were 1.50 – 8.80 mBq m^{-3}, for ^{210}Pb were 0.36–3.00 mBq m^{-3} and for ^{137}Cs values were below 8.00 μBq m^{-3} [13].

The range of the activity concentrations for ^{7}Be in the air for years 1987-2001 in Thessaloniki (Greece) were 0.47 – 12.70 mBq m^{-3} and for ^{210}Pb were 0.11 – 1.98 mBq m^{-3} [4].

4. Conclusion

This is the first study on air activity of cosmogenic and terrestrial radionuclides in ground level measured by gamma spectrometry in Albania. The obtained results by us in determination of activity concentrations are comparable with those reported by other investigators and show seasonal variation for ^{7}Be and ^{210}Pb.

The activity concentrations of ^{7}Be in air were in the range of 2.38 – 6.82 mBq m^{-3} and exhibited maximum in spring/summer and minimum in winter.

The range of activity concentrations for ^{210}Pb in the air were 0.37 - 1.27 mBq m^{-3}. The higher values were during summer and early autumn period. The lower values of ^{210}Pb were during the winter/spring period.

In the case of ^{40}K the maximum value of activity concentrations of 0.17 mBq m^{-3} were in June. Its increase is mainly due to air pollution in that period in the city of Tirana.

The activity concentrations of anthropogenic ^{137}Cs in ground level air were below MDA and it was observed only in three air filters in the range 0.30 – 6.01 μBq m^{-3}.

This work is continuing and all obtained data during period January 2021 – January 2022 will serve as database for the future in the monitoring of radionuclides in ground level.

References

[1] S. Bławzej, J. W. Mietelski, “Cosmogenic 22Na, 7Be and terrestrial 137Cs, 40K radionuclides in ground level air samples collected weekly in Kraków (Poland) over years 2003-2006,” *J. Radioanal. Nucl. Chem.*, vol. 300, no. 2, pp. 747 – 756, May 2014.

DOI: 10.1007/s10967-014-3049-6
PMid: 26224972
PMCid: PMC4514665

[2] D. Todorovic, D. Popovic, J. Nikolic, J. Ajtic, "Radioactivity monitoring in ground level air in Belgrade urban area," *Radiat. Prot. Dosimetry*, vol. 142, no. 2 – 4, pp. 308 – 313, Dec. 2010.
DOI: 10.1093/rpd/ncq211
PMid: 20833680

[3] D. Huang, H. Bao, T. Yu, "Temporal Variations in Radionuclide Activity (7Be and 210Pb) in Surface Aerosols at a Coastal Site in Southeastern China," *Aerosol Air Qual. Res.*, vol. 19, no. 9, pp. 1969 – 1979, Sep. 2019.
DOI: 10.4209/aaqr.2019.02.0084

[4] A. Ioannidou, M. Manolopoulou, C. Papastefanou, "Temporal changes of ^{7}Be and ^{210}Pb concentrations in surface air at temperate latitudes (40°N)," *Appl. Radiat. Isot.*, vol. 63, no. 2, pp. 277 – 284, Aug. 2005.
DOI: 10.1016/j.apradiso.2005.03.010
PMid: 15921916

[5] P. Lipinski, K. A. Isajenko, M. Biernacka, A. Zak, "Integration of Polish Monitoring Networks (ASS-500 and PMS systems)," *Nukleonika*, vol. 46, no. 4, pp. 143 – 146, Aug. 2001.
Retrieved from:
https://yadda.icm.edu.pl/baztech/element/bwmeta1.element.baztech-article-BUJ6-0006-0103?q=bwmeta1.element.baztech-volume-0029-5922-nukleonika-2001-vol 46 nr 4;5&qt=CHILDREN-STATELESS
Retrieved on: Sep. 15, 2021

[6] S. Grabowska, J. W. Mietelski, K. Kozak, P. Gaca, "Gamma Emitters on Micro-Becquerel Activity Level in Air at Kraków (Poland)," *J. Atmos. Chem.*, vol. 46, no. 2, pp. 103 – 116, Oct. 2003.
DOI: 10.1023/A:1026067614448

[7] M. Bysiek, M. Biernacka, P. Lipinski, "Radioactivity of ground-level air in Poland. Results from ASS-500 stations network," *Nukleonika*, vol. 46, no. 4, pp. 171 – 173, Sep. 2001.
Retrieved from:
http://www.ichtj.waw.pl/ichtj/nukleon/back/full/vol46_2001/v46n4p171f.pdf
Retrieved on: Sep. 15, 2021

[8] M. C. Lépy et al., "Intercomparison of efficiency transfer software for gamma-ray spectrometry," *Appl. Radiat. Isot.*, vol. 55, no. 4, pp. 493 – 503, Oct. 2001.
DOI: 10.1016/S0969-8043(01)00101-4

[9] F. L. Bronson, "Validation of the accuracy of the LabSOCS software for mathematical efficiency calibration of Ge detectors for typical laboratory samples," *J. Radioanal. Nucl. Chem.*, vol. 255, no. 1, pp. 137 – 141, Jan. 2003.
DOI: 10.1023/A:1022248318741

[10] M. Shyti, "Calibration and performance of HPGe detector for environmental radioactivity measurements using LabSOCS," *AIP Conf. Proc.*, vol. 2075, no. 1, 130012, Feb. 2019.
DOI: 10.1063/1.5091297

[11] L. E. De Geer, "Currie detection limits in gamma–ray spectroscopy," *Appl. Radiat. Isot.*, vol. 61, no. 2–3, pp. 151 – 160, Aug.-Sep. 2004.
DOI: 10.1016/j.apradiso.2004.03.037
PMid: 15177337

[12] A. Mauring, S. Patterson, B. Seslak, S. Tarjan, A. Trinkl, *IAEA-TEL-2020-03 World Wide Open Proficiency Test Exercise, Pie-charts, S-Shapes and Reported Results with Scores*, Rep. IAEA-TEL-2020-03, IAEA, Vienna, Austria, 2021.
Retrieved from:
https://nucleus.iaea.org/sites/ReferenceMaterials/Pages/Interlaboratory-Studies.aspx
Retrieved on: Nov. 10, 2021

[13] M. M. Janković et al., "Temporal concentration changes of beryllium-7 and lead-210 in ground level air level Serbia," *Chem. Ind.*, vol. 68, no. 1, pp. 83 – 88, Jan. 2014.
DOI: 10.2298/HEMIND130320031J