Airborne Be-7 and artificial Cs-137 radionuclides in mosses of Armenia

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Abstract. Moss samples collected throughout Armenia in the period of 2016-2019 served as a base for the assessment of artificial airborne Cs-137 and cosmogenic Be-7 radionuclides. The sampling campaign implemented within ICP Vegetation moss survey (35 samples) and Radioecological monitoring in the area of Armenia – REMA I and II projects (40 samples). The most widespread moss species (*Bryum argenteum, Homalothecium philippeanum, Ptilium crista-castrensis, Syntrichia ruralis*) were selected at each sampling location. At the laboratory, mosses have air-dried, manually cleaned and analyzed on a low-background gamma-ray spectrometry system (HPGe detector coupled to a multichannel analyzer, Genie 2000 software, CANBERRA). The counting time was 30,000 sec. After background correction, the activity concentrations (Bq/g) of Be-7 and Cs-137 were determined in Genie 2000 software from energy peaks of 477.60 and 661.5 keV, respectively.

Although no significant correlation (Spearman non-linear correlation) between the activity of radionuclides and altitude was found for the overall dataset, such correlation observed for samples of separate mountain ridges and massifs. Be-7 activity ranges from <MDA to 7.546 Bq/g (n=75, Valid N=34) with the mean value of 1.152 Bq/g. The highest activity for Be-7 observed in the eastern part of the country, from 1895 m a.s.l, close to the Sevan Lake. For artificial Cs-137 the activity ranges were <MDA to 0.297 Bq/g (n=75, valid N=44) with the 0.075 Bq/g mean value. The highest activity was observed in the moss from 2600 m a.s.l., in the area of highest peak of country - Aragats Mountain. The distribution of artificial Cs-137 and natural Be-7 in mosses enables the development of a Radioecological database for Armenia.

KEYWORDS: Cs-137; Be-7; Moss; Radionuclides

INTRODUCTION

Mosses are considered as one of the most commonly applied indicators for biomonitoring studies of radioactive contamination by both natural and artificial origin. Among the other plant species, mosses correspond perfectly to the criteria of biological indicators of atmospheric pollution. Their widespread occurrence in different geographical areas allows a comparison of the pollutants in diverse regions. Since mosses have no root systems they absorb nutrients via foliar uptake from precipitation or dry deposition. Hence, the capacity for the accumulation of airborne pollutants is much greater in contrast to terrestrial plants growing in the same environments. The morphologies of mosses are not affected by seasons, leading to the accumulation of pollutants throughout the year [8,11,13]. Mosses accumulate and retain variety contaminants including radionuclides, they are useful for recording spatial and contemporary deposition patterns, thereby being detectors of pollution events and provide a baseline of environmental radioactivity levels [5,10,11,14].
The regular monitoring of airborne radionuclides is a base for the detection of the radioactive dynamical changes in the environment. The distribution of fallout radionuclides in the terrestrial environment is one of the applicable study objects for environmental radioecological studies. Radionuclides migrating by atmospheric pathways removed from the atmosphere by dry or wet deposition of dust particles. The presence of fission product Cs-137 in the environment is associated mainly with past nuclear accidents and incidents [1–3], whereupon the contamination with Cs-137 became considerable due to its global spread with air migration pathways and distribution even far from emission sources. Cs-137 isotope is one of the major gamma dose-forming radionuclides along with natural primordial radionuclides. Due to the long half-life (30.2 y) and chemical similarity with potassium, Cs-137 is one of the important and hazardous radionuclides, therefore one of the main

study materials for environmental radioactivity [4,5]. There are no extensive sources of Cs-137 emissions in the last decade (after Fukushima Daiichi NPP accident), however Cs-137 is still exposed to physical decay and its deposition could be determined by atmospheric transport and accumulation in the terrestrial environment [6]. The deposition of artificial Cs-137 depends on geographic location and local climatic conditions and mostly precipitation, which amount varies by altitude [3,7,8].

Be-7 is a cosmogenic radioisotope that originates in the stratosphere and upper troposphere during the interaction of cosmic radiation and atmospheric nitrogen and oxygen. The amount of Be-7 depends on the flux of cosmic rays and meteorological conditions. Be-7 has a half-life of approximately 53 days. It is carried in the atmosphere attached to the aerosols and is deposited on the land surface by wet and dry deposition. Be-7 attaches to aerosols in the submicron size range, therefore, it is susceptible to the same transport and depositional processes as aerosols, making it a useful tracer in atmospheric studies. [8–11]. Despite two of these radionuclides have different origins, their presence in the terrestrial environment conditioned with the local geomorphology and amount of precipitations [12].

The mountain regions play important role as a barrier in the atmospheric migration processes of Cs-137 and Be-7. The Republic of Armenia positioned at the highest altitude compared with the rest of South Caucasian countries. The mean altitude is 1830 meters above sea level (m a.s.l) with the highest peak of Aragats Mountain located at 4090 m a.s.l. The highlands of Armenia are sensitive to the influence of atmospheric transfer of radionuclides and it is proved by previously identified radionuclides from the Chernobyl NPP accident [3].

The aim of current study is the 1) assessment of the activities of Cs-137 and Be-7 radionuclides in mosses of Armenia; 2) reveal the altitudinal distribution characteristics of Cs-137 and Be-7.

MATERIALS AND METHODS

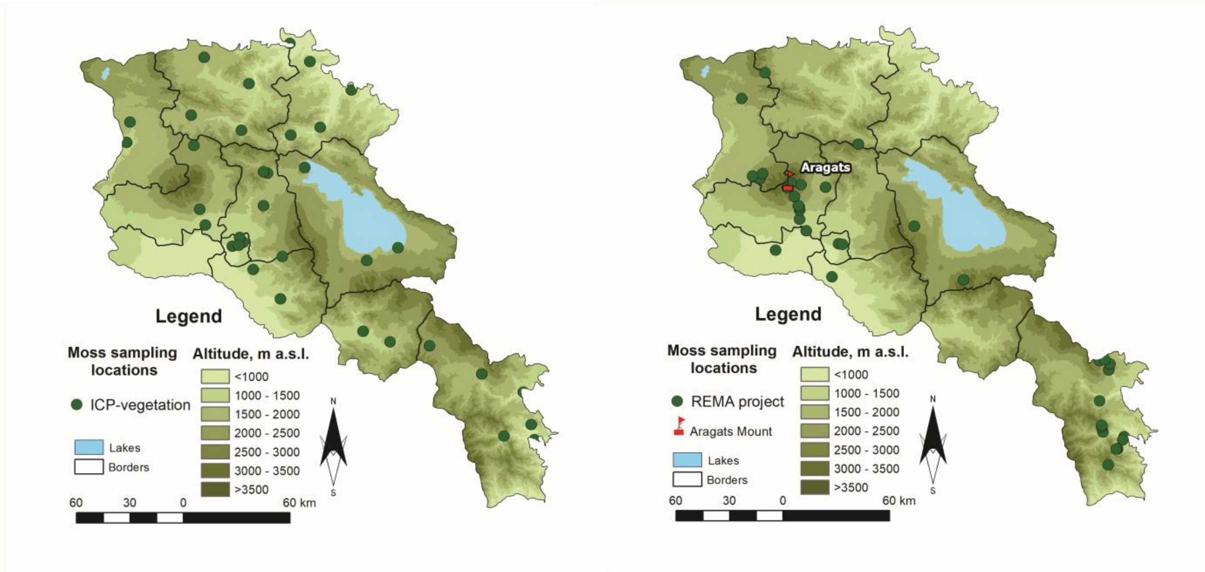
The Republic of Armenia (total area 29.743 square kilometers) lying at the south of the great mountain range of the Caucasus and northeast of the Armenian Highlands, and fronting the north-western extremity of Asia (latitudes $38^0$50' and $41^0$18', longitudes $43^0$27' and $46^0$37'). The neighboring countries are Georgia from the north, Azerbaijan from the east, Turkey from the west and southwest and Iran from the south. The territory of Armenia is mostly mountainous; the average height above sea level is 1830 m, with the lowest point of 379 m (Araks river canyon) and the highest of 4090 m (Aragats massif). The climate is continental with the dry, hot summers and cold winters [15].

35 moss samples sampled from all regions of Armenia within ICP vegetation project (UNECE) in 2016. The sampling altitude ranges were 468-2213 m a.s.l. 40 moss samples were sampled from mountain regions of Armenia during Radioecological monitoring in the area of Armenia – REMA I and II projects (2016-2020). The ranges of sampling location altitudes were 846-3200 m a.s.l (Figure 1).

The most common land cover moss species in Armenia were selected for the study (*Bryum argenteum, Homalothecium philippeanum, Ptilium crista-castrensis, Syntrichia ruralis, etc.*). Field sampling procedures were implemented according to the IAEA vegetation sampling guideline and UNECE ICP Vegetation manual [16,17].

In each sampling location the most abundant moss species were selected, samples placed in plastic bags and labeled. In the laboratory, mosses were cleaned manually to remove soil and foreign matters and air-dried at room temperature to a constant weight. The dried samples were weighted, then packed and kept in cylindrical plastic containers.

Figure 1: Moss sampling locations in Armenia, ICP vegetation and REMA projects



Radionuclides activity concentration determinations performed using gamma-ray spectrometry system consist of a high purity germanium detector with the energy resolution of 1.8 keV FWHM coupled to a DSA-1000 multichannel analyser (CANBERRA). The efficiency calibration performed by using LabSOCS (Laboratory Sourceless Calibration Software). For the gamma spectrum peak energy measurements energies of 105.3 keV of Eu-155, 1275 keV of Na-22 and 1332.5 keV of Co-60 used as reference. Genie 2000 software was used for spectrum acquisition and analysis. The gamma spectrum acquisition time for each sample was 30.000 sec. Background measurements implemented once a week using clean and empty cylindrical boxes. For laboratory quality control and assurance, an inter-laboratory comparison program was conducted with the Armenian Nuclear Forensic laboratory. After correcting for background and Compton contribution activity concentrations of Cs-137 and Be-7 were determined directly from 477.6 and 661.7 keV gamma lines respectively. Activity concentrations of Be-7 re-calculated up to initial activity using the sampling date as a reference. Data analysis and visualisation performed using IBM SPSS Statistics and ArcMap software, respectively.

RESULTS AND DISCUSSION

The following table represents the statistical parameters of Be-7 and Cs-137 (Table 1).

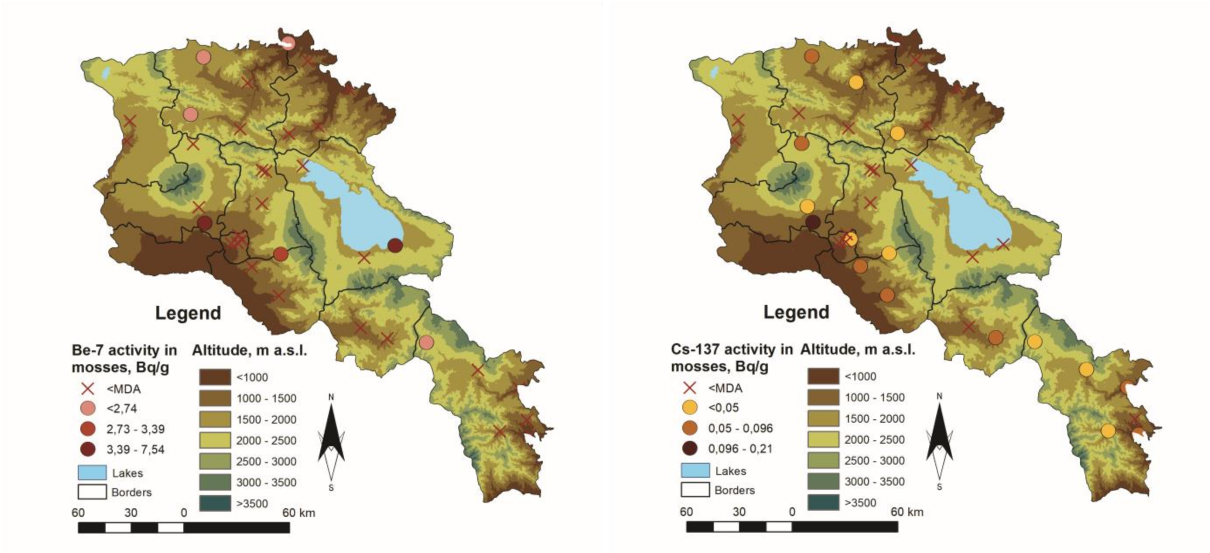
Table 1: Descriptive statistics of Be-7 and Cs-7 radionuclides in mosses

Parameter	Be-7	Cs-137
N Valid	34	44
Mean	1.15	0.075
Median	0.49	0.05
Std. Deviation	1.77	0.07
Variance	3.42	0.005
Skewness	2.73	1.36
Kurtosis	7.47	1.17
Minimum	0.07	0.005
Maximum	7.545	0.297

MDA for Be-7 0.005 Bq/g; MDA for Cs-137 – 0.001 Bq/g

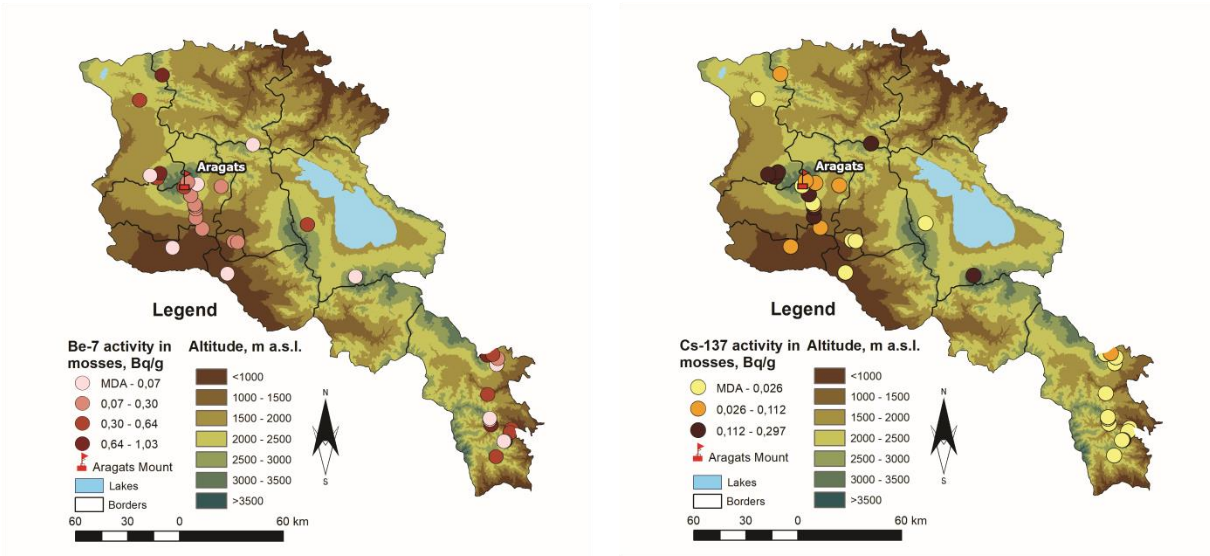
For the overall dataset Be-7 was identified only in 45% of samples (34 Valid, 41 Missing) and 59% results above MDA were recorded for Cs-137 (44 Valid, 31 Missing). The ranges for Be-7 were from <MDA to 7.54 Bq/g, with the mean value of 1.15 Bq/g. For artificial Cs-137 the activity ranges were from <MDA to 0.297 Bq/g, with the 0.075 Bq/g mean value. The distributions of Be-7 and Cs-137 in mosses from ICP project presented in Figure 2.

Figure 2: Distribution of Be-7 and Cs-137 in mosses of Armenia by altitude: ICP-vegetation data



As it is shown, the relatively higher activities for Be-7 observed in the central part and in the east of the country. The highest activity determined in the moss sample from 1895 m a.s.l, near Sevan Lake. For Cs-137 higher activities observed mainly in the west of the country. The highest activity (0.2 Bq/g) located at 1075 m a.s.l. The results of mosses from REMA project are shown in Figure 3.

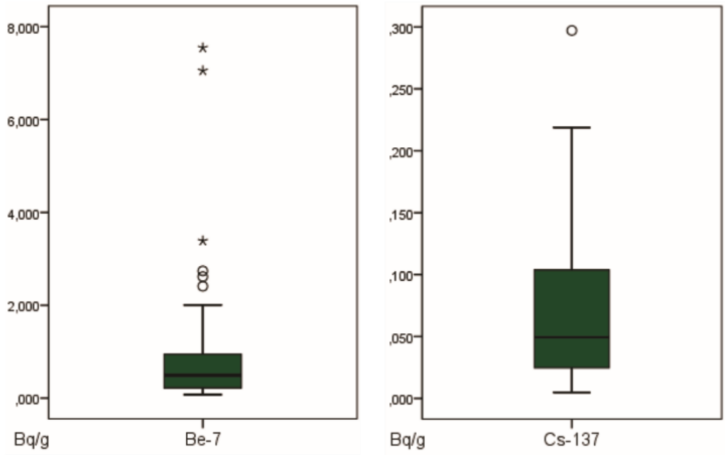
Figure 3: Distribution of Be-7 and Cs-137 in mosses of Armenia by altitude: REMA projects data



It is noticeable, that higher activities observed mainly in the north and south of the country. The highest activity of Be-7 (1.03 Bq/g) observed in the moss sample from 1417 m, from the area of Aramazd Mountain, located in the south of Armenia (3399 m a.s.l.). For Cs-137 the highest activities observed mainly in the mosses form Aragats Mountain region at the altitudes more than 2000 m a.s.l. The highest activity (0.297 Bq/kg) recorded at 2600 m a.s.l. The activity increases with the altitude in

the area of Aragats Mountain, where the moss sampling was designed by altitude with a 200 m sampling interval.
For the overall dataset, normal distribution did not observe (Shapiro-Wilk Normality test, $p<0.05$). Moreover, the log-transformation of the data did not reveal the lognormal distribution as well. The distribution affected by the presence of outliers and extreme values in both Be-7 and Cs-137 samples (Figure 4).

Figure 4: Boxplots of Be-7 and Cs-137 distribution



It is noticeable from

Figure 5 and Figure 6 there is no complete distribution pattern by altitude from radionuclides for the overall dataset. Such a pattern of distribution observed in separate Mountain ridges, for both Be-7 and Cs-137 higher activity concentrations observed there. In addition, the differences between different moss species were not significant for investigated radionuclides.
No significant correlation was observed between activity concetrations of radionuclide in mosses and the altitude (Table 2).

Figure 5: The distribution of Cs-137 in mosses by altitude

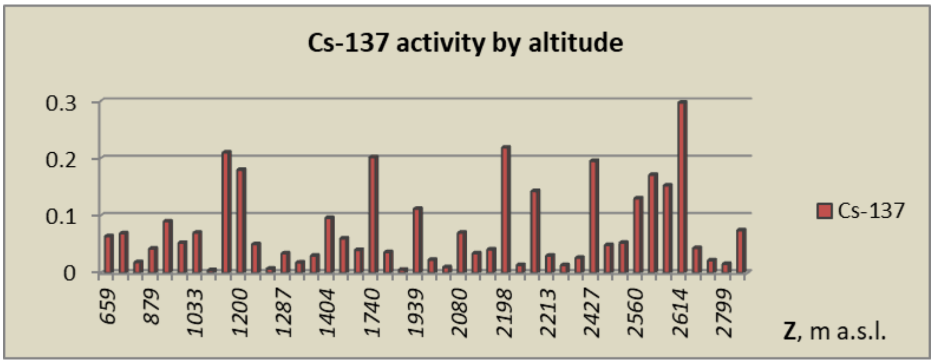


Figure 6: The distribution of Be-137 in mosses by altitude

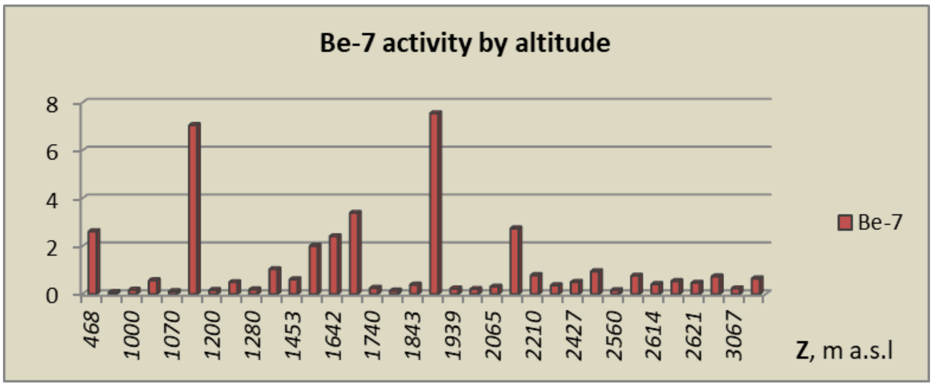


Table 2: Spearman's rho between altitude, Be-7 and Cs-7 activity concentrations

	Altitude	Be-7	Cs-137
Altitude	1.000		
Be-7	0.079 N=34	1.000	
Cs-137	0.080 N=44	0.132 N=25	1.000

Similar studies have been carried out in different regions in order to assess the accumulation of airborne radionuclides by mosses. For example, in Northern Greece the average values in different kind of ground mosses for Be-7 and Cs-137 were 388 Bq/kg and 35 Bq/kg, respectively [18]. In present study the mean values for Be-7 and Cs-137 were 1.15 Bq/g (or 1150 Bq/kg) and 0.075 Bq/g (or 75 Bq/kg), respectively. Another study compares the activity levels of airborne radionuclides in mosses from different climatic zones; Serbia and south of Thailand [6]. The average activity of Be-7 in mosses of Serbia was 314 Bq/kg and for Thailand 226 Bq/kg. The Cs-137 activity levels from the same study were different. In the south of Thailand Cs-137 was not detected in mosses. In contrast, in Serbia high levels of Cs-137 in mosses were recorded. Comparing the results from different and the current study it could be assumed that the distribution of airborne radionuclides is different due to the local geographical conditions: The observed highest values for Be-7 in Armenia are conditioned by the highest altitudinal location compared with the results of the mentioned studies. For Cs-137 the observed high activity in Armenia is also conditioned by the local geographic conditions, but it is also may be a consequence of the radioactive fallout in the Northern Hemisphere since the Cs-137 activity is still significant due to its long half-life. Hereby, mosses are perspective for studying the spatial distribution of airborne radionuclides in different geographic areas. Moreover, for fission product Cs-137 moss studies can describe the atmospheric transfer directions and re-distributions in different regions.

CONCLUSION

The main conclusion yielded from this research is that there is no significant correlation (Spearman non-linear correlation) between the activity of radionuclides in mosses and altitude for the overall dataset. High variability recorded both for Be-7 and Cs-137: >MDA (0.005) to 7.546 and >MDA (0.001) to 0.297 Bq/g, respectively. In separate sampling locations (Aragats Mountain, Aramazd Mountain) higher activities of radionuclides recorded at the highest altitudes. The maximal activity of Be-7 observed at 1895 m a.s.l, in the east of the country. For Cs-137 the highest activity was in

Aragats massif at 2600 m a.s.l. In the mountain regions the highest activities are associated with a large amount of precipitation and a higher level of radioactive fallout in highlands. Additionally, the differences between the different moss species were not significant for the accumulation of Be-7 and Cs-137. However, further investigations and more moss samples are necessary to study the accumulation rates. The current study enables the development of Radioecological database of Armenia, including radionuclides baseline activities in mosses.

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