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# Beryllium-7 and $^{210}$Pb atmospheric deposition measured in moss and dependence on cumulative precipitation

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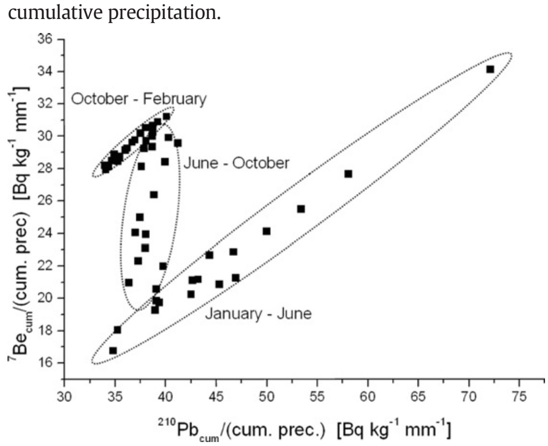
## HIGHLIGHTS

- Use of mosses in measurement of airborne radionuclides deposition was investigated
- Prior work indicated $^{7}$Be and $^{210}$Pb activities were not correlated with precipitation
- This is unusual since radionuclides moss tissues depends on depositional fluxes.
- A new method for study of $^{7}$Be and $^{210}$Pb depositional dynamics was developed
- Different seasonal regimes of $^{7}$Be deposition are more noticeable in new technique

## GRAPHICAL ABSTRACT

Correlation between cumulative activity of $^{7}$Be and $^{210}$Pb measured in moss samples normalized by the cumulative precipitation.

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## ABSTRACT

This paper focuses on analysis of the time series of $^{7}$Be and $^{210}$Pb activity measured in moss, and the amount, as well as duration of precipitation, to gain a better understanding of the possible relationships between airborne radionuclide deposition and precipitation. Here we consider whether the amount of these airborne radionuclides in moss samples is a cumulative measure of radionuclide deposition and decay, and a new approach for analyses of the relationships between precipitation and moss activity concentrations is suggested. Through these analyses it was shown that comparison of cumulative activity measured at one location using moss, normalized by values of cumulative amount or duration of precipitation, showed different regimes of airborne radionuclide deposition.


## 1. Introduction

Natural airborne radionuclides $^{7}$Be and $^{210}$Pb are frequently used as tracers in atmospheric studies (Lal and Peters 1962; Vecchi and Valli

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1997; Baskaran 2011). They have very different origins. Beryllium - 7 is primarily created in stratosphere, and upper troposphere by way of interactions of high-energy particles of cosmic radiation with nuclei of the most abundant elements in the air, nitrogen and oxygen (Lal et al. 1958). After formed, $^{7}$Be attaches to aerosol due to high chemical reactivity. Thus, $^{7}$Be follows all atmospheric paths of transport and deposition of aerosol (Papastefanou and Ioannidou 1995; Aldahan et al. 2001; Kaste et al. 2002). Soil containing Uranium, is the dominant source of $^{210}$Pb. As a member of $^{238}$U decay chain, $^{222}$Rn as a noble gas, escapes all chemical bounds and enters the atmosphere. When $^{222}$Rn decays, daughter elements are produced that have a high affinity for aerosol are produced. $^{210}$Pb, the only one long living daughter product of $^{222}$Rn with half life of 22 years, becomes involved in atmospheric transport as well as deposition processes (Baskaran and Naidu 1995; Koch et al. 1996; Preiss et al. 1996; Dibb 2007). Measurement of $^{7}$Be and $^{210}$Pb activity concentration (in Bq m$^{-3}$) in atmosphere is usually done by air sampling (Kulan et al. 2006; Cho et al. 2007; Papastefanou 2009; Carvalho et al. 2013, Yang et al. 2013). Air is collected by a calibrated vacuum pump through a filter to trap aerosol. After chosen collection time the activity concentration from filter is measured by the standard gamma spectrometry, following either some chemical treatment or in bulk. Aerosols removed from atmosphere by dry or wet deposition can be collected by deposition samplers (Wallbrink and Murray 1994, Hirose et al. 2004; Du et al. 2008; Renfro et al. 2013). After some period of time, collected content in depositional vessel is measured by gamma spectroscopy device to determine the activity the radionuclide of interest in fallout (in Bq m$^{-2}$). Temporal variations of $^{7}$Be and $^{210}$Pb activity in air, as well as in precipitation, have been a subject of numerous studies (cf. Feely et al. 1989; Azahra et al. 2003; Ioannidou et al. 2005; Yamamoto et al. 2006; Doering and Akber 2008; Kikuchi et al. 2009; Mann et al. 2011). Seasonal variations of $^{7}$Be and $^{210}$Pb in air and fallout were observed, including 11-year periodicity of $^{7}$Be concentration in the atmosphere as a consequence of solar modulation of arriving cosmic radiation (Cannizzaro et al. 2004; Papastefanou and Ioannidou 2004; Kulan et al. 2006). In some cases correlation between measured airborne radionuclide and some atmospheric parameters (precipitation) has also been found (Caillet et al. 2001; Lozano et al. 2012; Tositti et al. 2014).

Currently, terrestrial mosses are widely used in environmental monitoring (Rühling and Tyler 1973; Buse et al. 2003). Measurement of element concentrations in moss has been shown to be very suitable for studying the atmospheric deposition of heavy metals as well as other trace elements, including radionuclides (Godoy et al. 1998; Delfanti et al. 1999; Dowdall et al. 2005). Widespread occurrence, simple sampling procedure, and gamma spectroscopy measurement (without any chemical treatment of moss samples) make mosses a very useful medium in investigation and monitoring of airborne radionuclide deposition. In order to use $^{7}$Be or $^{210}$Pb effectively as a tracer (Krmar et al. 2007; Krmar et al. 2009), to study different atmospheric processes, their depositional characteristics should be known and a comprehensive understanding of their variability in the atmosphere, as well as dependence on a number of atmospheric parameters. In previously and later published papers (Ishikawa et al. 1995; Guebuem et al. 2000; Caillet et al. 2001; Ioannidou and Papastefanou 2006; Zhu and Olsen 2009) a moderate direct correlation was observed between precipitation (which washes-out the atmosphere and deposit activity) and total measured depositional fluxes of airborne radionuclides. On the contrary, concentrations of airborne radionuclide measured in near ground air usually show a week inverse correlation in respect to the precipitation amount (Likuku 2006). It can be expected because precipitation washes-out atmosphere and after rain or snow, lower concentrations of airborne activity are measured until sedimentation from higher layers of atmosphere brings new amount of radionuclides.

The main goal of this paper is to explore some techniques which can provide more insight into possible relationships between measured activities of airborne radionuclides and amount and the duration of precipitation. Krmar et al. (2009) pointed out that $^{7}$Be and $^{210}$Pb activities measured in mosses are not correlated with precipitation amount and the duration of precipitation, despite the fact that content of radionuclides in moss tissues depends on depositional fluxes. This can be explained since mosses cumulate activity, i.e., an amount of some airborne radionuclide in moss sample is cumulative measure of radionuclide deposition and decay and/or physical removing. Thus a different approach in analysis of relations between precipitation and moss activity has to be considered. Our objective for this paper is to investigate (i) the use data of cumulative precipitation and the cumulative duration of precipitation; and (ii) explore relationships between those datasets and measured values of the $^{7}$Be and $^{210}$Pb activities that provide a dataset of “cumulative activities” of airborne radionuclides in mosses.

## 2. Materials and methods

Samples of naturally growing mosses were collected from single location (~ 300 m$^{2}$) from the horizontal eaves of the Department of Biology in Novi Sad, Serbia. One sample per week was taken during 14 months. The green part of the moss that contains 3–4 annual growth segments was used for analyses. Samples were cleaned from all mechanical impurities (leaves and other plant material), and dried until constant mass was reached. Dry mosses (from 22 to 48 g) were packed in cylindrical plastic containers (0.7 mm wall thickness, diameter 67 mm, 62 mm height) and gamma radiation measured $<$3 days after sampling. A low-background HPGe detector, 36% efficiency, shielded by 16 cm of lead and 1.5 mm inner Cu shield was used. The longest sample counting time was 48 ks until statistical uncertainty of 477.6 keV $^{7}$Be line up to 5% was achieved. The next set of measurements were carried out using extended range HPGe detector equipped by Be window to get evidence about $^{210}$Pb concentration. For this isotope the detector of 32% relative efficiency, was shielded by 18 cm of lead, 1 mm of tin and 1.5 mm of copper. Measurement time was from 22 to 88 ks until statistical uncertainty of 46.5 keV $^{210}$Pb line up to 5% was obtained. Efficiency calibration was obtained using several reference materials (IAEA-372 (grass), IAEA-330 (spinach) and IAEA-447 (moss-soil)). Correction for self-attenuation effects was not necessary because of the very similar composition and density of the sample and reference materials. The daily amount of precipitation and duration of precipitation were taken from the nearest meteorological station, Rimski Šančevi, from the network of meteorological stations of Republic Hydrometeorological Service of Serbia. Detailed information about sampling location, collection of samples, measurement setup, and other details are described in Krmar et al. 2009.

Cumulative activities of $^{7}$Be and $^{210}$Pb were calculated by simple addition of successive measured values until the end of the studied period of time. Note, that cumulative moss activity has not the same physical meaning as the cumulated precipitation. The cumulated precipitation is a measure of integral amount of precipitation at some place because at the beginning of a new measurement, the empty precipitation gauge is exposed to rain or other precipitations. Thus, just a new amount of precipitation is gathered and then added to the sum of all previously measured precipitation amounts. In contrast, mosses accumulate all previous deposited activity. For that reason, fresh and continually exposed sample includes a long history of deposition and decay operating in parallel. However, “cumulative activity”, even constructed on this way, can offer some useful information that is at least comparable with information obtained by analysis of particular week measurements. Now, we consider cases comprised by the proposed method of “cumulative activity” in analysis of $^{7}$Be and $^{210}$Pb atmospheric deposition on cumulative precipitation.