Journal of Environmental Radioactivity xxx (2016) 1–7

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Gamma emitters in atmospheric precipitation in Krakow (Southern Poland) during the years 2005–2015

Jerzy W. Mietelski [a, *], Ewa Nalichowska [a], Ewa Tomankiewicz [a], Kamil Brudecki [a], Paweł Janowski [b], Renata Kierepko [a]

[a] The Henryk Niewodniczanski Institute of Nuclear Physics, Polish Academy of Sciences (IFJ PAN), Radzikowskiego 152, 31-342 Krakow, Poland
[b] Faculty of Physics and Applied Computer Sciences, AGH University of Science and Technology. Al. Mickiewicza 30, 30-059 Krakow, Poland

ARTICLE INFO

Article history:
Received 27 October 2015
Received in revised form
18 January 2016
Accepted 11 February 2016
Available online xxx

Keywords:
Radioactivity of air
Gamma emitters
Cosmogenic radionuclides
Radiocesium
Resuspension
Wet and dry precipitation
Radionuclides' washout

ABSTRACT

The results of the sum of dry and wet activity deposition for naturally occurring ^{7}Be, ^{210}Pb, ^{40}K, ^{22}Na and anthropogenic ^{137}Cs radionuclides in Krakow (Southern Poland) for the samples collected over 10 years (from August 2005 to July 2015) are presented and discussed. The radionuclides were determined using low background gamma spectrometry with HPGe detectors. Additionally, in this paper there are shown the results of activity concentrations in water from air precipitation for ^{7}Be, ^{210}Pb, ^{22}Na, ^{40}K and ^{137}Cs radioisotopes from the period of 7 years (from August 2008 to July 2015). For all these series the statistical analysis including Spearman correlations, effects of seasonal variation and multiple regression models were conducted. After excluding two months from 2011 affected by the Fukushima accident, high Spearman correlation factors ($R > 0.5$) for activity deposition were noticed for the pair of the cosmogenic radionuclides ^{7}Be and ^{22}Na ($R = 0.508$) and between ^{210}Pb and ^{7}Be ($R = 0.570$). High correlation was noted between activity deposition and amount of precipitation for ^{7}Be ($R = 0.677$). The seasonal correlations between ^{7}Be–^{22}Na, ^{40}K–^{137}Cs, ^{210}Pb–^{137}Cs and ^{7}Be–^{210}Pb were investigated and the highest correlation coefficient $R = 0.731$ for the ^{40}K–^{137}Cs pair was in the spring season. High correlations were observed also between ^{210}Pb and ^{7}Be for autumn ($R = 0.594$), ^{40}K–^{137}Cs in summer ($R = 0.582$), ^{7}Be–^{22}Na in spring ($R = 0.635$) and ^{210}Pb–^{137}Cs in autumn ($R = 0.672$). The multiple regression approach showed the interesting difference in scavenging mechanisms of cosmogenic and terrestrial radionuclides. According to that model, the deposition of cosmogenic nuclides was noticeably related to the amount of precipitation, while the deposition of terrestrial radionuclides did not show such dependence.

1. Introduction

The main sources of radionuclides present in atmospheric precipitation are: soil resuspension, sea spray, industrial emissions, particles from burnt biomass and fallout from stratospheric aerosols rich in cosmogenic radionuclides produced there and in the upper troposphere (Lal and Suess, 1968; Eisenbud and Gesell, 1997). The content of radionuclides in air precipitation can vary a lot due to the facts that contributions of all these sources are not constant in time and various atmospheric processes influence on the propagation of the radioisotopes traces. Because of the different possible scavenging mechanisms, generally the dry and wet depositions have different radionuclide compositions. However, due to the technical limitation we decided to collect sum of dry and wet deposition, averaged on monthly basis.

Activity concentrations of radionuclides in the atmospheric precipitation in resulting depositions were expected to vary in a wide range. Our research was oriented to measure activities of two cosmogenic radionuclides: ^{7}Be, ^{22}Na, two natural terrestrial radionuclides: ^{210}Pb, ^{40}K, and one artificial nuclide: ^{137}Cs. These radionuclides were chosen due to their wide–range usability as physicochemical markers of different environmental processes taking place in atmospheric, aquatic and soil systems (Mabit et al., 2008; Su and Huh, 2003; Walling and He, 1998; Tsabaris et al., 2015). Moreover studies of the long – term environmental behaviour of these radioisotopes including statistical analyses of their correlations with the meteorological data represent a very important input to the radioecological modeling both for nuclear accidents and non-emergency situations (Baklanov and Sorensen,

* Corresponding author.
E-mail address: jerzy.mietelski@ifj.edu.pl (J.W. Mietelski).

http://dx.doi.org/10.1016/j.jenvrad.2016.02.010
0265-931X/

2001; Sportisse, 2007; Christoudias and Lelieveld, 2013; Mazur et al., 2014; Heikkila, 2007). Long-term monitoring of the activity deposition fluxes especially of ^{7}Be, ^{210}Pb and ^{137}Cs, which represent all the types of radioactivity sources in the atmosphere, is also described and discussed in many works (Alonso-Hernandez et al., 2006; Pham et al., 2013; Baskaran et al., 1993; Beks et al., 1998; Dueñas et al., 2011).

The goal of this study was to analyze long-term relationships between terrestrial, artificial and cosmogenic nuclides in air precipitation in non-emergency situations. Therefore in this paper the data for radionuclides, which existed only during the passage of Fukushima cloud such as ^{131}I, ^{132}I/^{132}Te, ^{134}Cs, ^{136}Cs are not presented. Additionally in some analyses all data from April and May 2011 were excluded. The overall output for months from April to October 2011 was already published elsewhere [Mietelski et al., 2014].

The results shown in this article are published for the first time as a whole set, although investigation on ^{137}Cs from years 2005–2010 was presented in MSc thesis [Janowski, 2010]. In addition for years 2005–2007 radiochemical analyses for Pu isotopes were carried out and the results were published in PhD thesis [Kierepko, 2010] and in the article [Kierepko et al., 2009].

2. Material and method

2.1. Sampling site

The sampling site is a standalone station at the Institute of Nuclear Physics (IFJ PAN) in Bronowice, a north-western suburbia of Krakow, Southern Poland. The station is located at the elevation of 215 m above sea level, longitude 19° 58′ E, latitude 50° 04′ N. The average yearly precipitation for Krakow (measured since 1863) is 679 mm [Twardosz, 2000], with 47% of rainy and snowy days unevenly distributed in a year. 41% of air precipitation occurs during three months of summer. The daily average precipitation is 1.8 mm, ranging from 0.7 in winter to 4.4 mm in summer (Twardosz, 2000).

2.2. Sampling device

Samples were collected at stainless steel tray having an area of 2.28 m^2 and connected via plastic tube to a plastic (Polyethylene) barrel with a volume of 160 L. The size of the tray was designed to enable measurements of Pu isotopes using alpha spectrometry [Mietelski et al., 1998]. Water was collected monthly. Usually the volume of collected monthly precipitation did not exceed 160 L, with an exception of few months during spring and summer.

The station was protected from adverse weather conditions. The tray was equipped with a de-icing system, using a low-power electric heater operated manually. An additional electric heater was placed inside the station. Both heaters were used in winter. The barrels were housed inside of a metal container, and isolated by a 10 cm layer of Styrofoam to prevent loses from evaporation in hot summers. The whole installation is shown in Fig. 1. The terrain around was open and covered with grass (at least 30 m in each direction) until October 2012. Then, due to start of construction works for new building at this site, the station was moved 200 m westward, and there the surrounding was cover partially with concrete and low bushes were growing about 10 m apart from west. Behind bushes there is a small park area.

2.3. Sample treatment

The collected water firstly was acidifying by adding 50 mL of concentrated nitric acid, then it was transferred in portions into stainless steel vessel of 10 L volume and evaporated to less than 1 L. This solution was carefully transferred into a 1 L glass vessel, with washing using concentrated nitric acid. Walls and bottoms of the empty barrel and vessel were carefully wiped using ash-free filtration papers and 1 M nitric acid, the papers were dried, ashed in 400 °C overnight, and added to the original solution. Then the sample was again evaporated down to about 100 mL and transferred to plastic vessel (5 cm diameter, 120 mL volume), were it was eventually gently evaporated to dryness under infrared lamp.

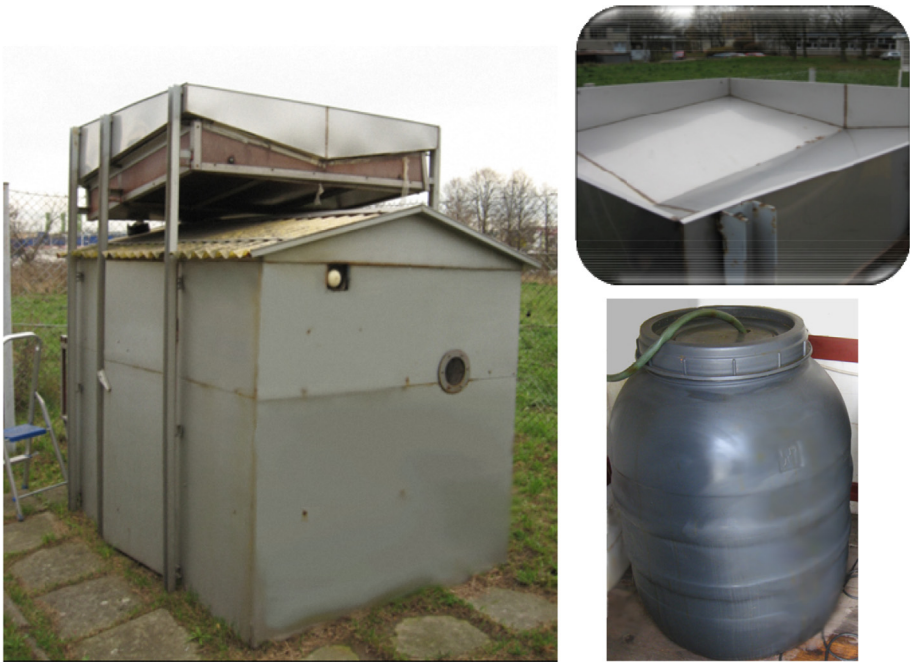

Fig. 1. Photo of air precipitation collector designed at IFJ PAN in Krakow and used to collect samples for present paper (left). Right top – the view on stainless steel trap, Right bottom – 160 L plastic barrel used for storing the sample. .

Please cite this article in press as: Mietelski, J.W., et al., Gamma emitters in atmospheric precipitation in Krakow (Southern Poland) during the years 2005–2015, Journal of Environmental Radioactivity (2016), http://dx.doi.org/10.1016/j.jenvrad.2016.02.010