GAMMA RADIOACTIVITY OF THE JANUARY 2013 RAINFALL IN OSTRAVA

GAMA RADIOAKTIVITA DEŠTĚ V LEDNU 2013 V OSTRAVĚ

Petr ALEXA[1], Petra KOLIČOVÁ[1], Radim UHLÁŘ[2]

[1] Institute of Physics and Institute of Clean Technologies, VŠB-Technical University of Ostrava, 17. listopadu 15, 708 33 Ostrava, Czech Republic, tel. (+420) 59 732 3100 e-mail petr.alexa@vsb.cz

[2] Institute of Physics, VŠB-Technical University of Ostrava,VŠB-Technical University of Ostrava, 17. listopadu 15, 708 33 Ostrava, Czech Republic, tel. (+420) 59 732 4481 e-mail radim.uhlar@vsb.cz

Abstract

Gamma radioactivity of an 8 mm thick ice layer formed after a modest rain on 21 January 2013 in the VŠB-Technical University of Ostrava campus was analyzed and gamma-ray emissions from primordial ^{40}K have been found. Cosmogenic ^{7}Be produced mainly in galactic cosmic-ray spallation processes on atmospheric nitrogen and oxygen and ^{226}Ra, ^{235}U and ^{137}Cs have been also observed.

Abstrakt

Při analýze gama radioaktivity 8 mm silné vrstvy ledu, která se vytvořila při dešťových srážkách v areálu VŠB-Technické univerzity Ostrava 21.1.2013, bylo detekováno gama záření z izotopu ^{40}K. Rovněž bylo pozorováno kosmogenní ^{7}Be vznikající především v tříštivých reakcích galaktického kosmického záření s atmosférickým dusíkem a kyslíkem a také ^{226}Ra, ^{235}U and ^{137}Cs.

Key words: gamma radioactivity, air pollution, natural radioisotopes, cosmic-ray spallation

1 INTRODUCTION

Naturally occurring radioactive isotopes are practically ubiquitous on Earth and may become even concentrated as a result of human activities (nuclear industry, coal and petroleum industry, water treatment, etc.) [1, 2]. According to their origin, they can be divided into four groups: primordial isotopes (present since the formation of Earth), members of radioactive decay chains (radioactive products of primordial isotopes ^{232}Th, ^{235}U and ^{238}U), cosmogenic isotopes (produced in nuclear reactions of cosmic radiation with the gaseous and particulate constituents of the Earth atmosphere), and human produced isotopes (released in nuclear accidents, nuclear-weapon tests and controlled low-activity emissions from nuclear objects as power plants, research and medical facilities, etc.) [3]. Especially primordial ^{40}K and ^{235}U, members of the ^{232}Th chain (^{208}Tl, ^{212}Pb, ^{228}Ac) and ^{238}U chain (^{210}Pb, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{234}Th), cosmogenic ^{7}Be and human produced caesium isotopes ^{134}Cs and ^{137}Cs belong to the naturally occurring radioactive isotopes that can be detected in gamma spectrometry [4].

A positive correlation between the air pollution and the specific activity of radioisotopes in air is a well-established phenomenon [5]. In the atmosphere, radioisotopes associate with aerosol particles. The activity size distribution of ^{7}Be and ^{210}Pb aerosols peaks mostly in submicron region [6, 7]. Recently correlations between the ^{7}Be and PM_{10} concentrations have been reported from a 14-year monitoring study in Taiwan [8].

The present study was carried out in Ostrava, the most polluted area of the Czech Republic [9, 10], where the concentrations of the PM_{10} air-borne dust (particulate matter containing particles of a diameter < 10 μm) usually exceed 100 μg/m^3 in winter. Gamma spectra of two samples of an ice layer formed after a modest rain in January 2013 are analyzed to identify present radioisotopes and a possible health impact depending on the annual effective dose due to the presence of the identified radioisotopes in air is discussed.

2 EXPERIMENTAL PROCEDURE

On 21 January 2013, PM_{10} concentrations in Ostrava reached 200 – 300 μg/m^3 [11]. The unfavourable meteorological conditions were partially ceased by a modest rain (0.4 mm/h maximum) at the same day. The rain got immediately frozen in contact with a very cold earth surface. In the campus of the VŠB-Technical University of Ostrava in Ostrava-Poruba (GPS 49º49'59.498'' N, 18º9'42.994'' E), the formed ice layer thickness reached approx. 8 mm. In Fig. 1, the correlation between the rain deposit per hour in Ostrava-Poruba and the air-borne dust (PM_{10}) average concentration in the Ostrava region (for the measuring stations in Ostrava-Zábřeh, Ostrava-

Přívoz, Ostrava-Fifejdy and Studénka) during January 21, 2013 [12] can be clearly seen. One can also see two peaks in the rain deposit per hour between 2-6 p.m. and 1-6 a.m. of Central European Time (CET) that resulted in the ice layer formation. The clouds lower boundary increased from 50-100 m towards 300 m during the day [12].

The ice samples collected in the campus of the VŠB-Technical University of Ostrava were kept at room temperature to melt the ice. Approx. 1 ml of nitrogen acid was then added to the water samples to decrease their pH value below 2 [13]. The samples were put into two Marinelli beakers. One of the Marinelli beakers was completely filled with the sample (620 ml), sealed and left for 40 days to reach the secular equilibrium for radium measurements (sample A). The second Marinelli beaker (460 ml) was not sealed (sample B).

Gamma spectra of all samples were accumulated using the Canberra spectroscopic system (semiconductor germanium detector GC3018, multichannel spectrometer DSA1000, spectrometric software Genie 2000 [14], massive 100 mm Pb + 1 mm Cd + 1 mm Cu shielding). Specifically, the gamma spectrum of the sample A was recorded after 40 days and the gamma spectrum of the sample B within a few days after its preparation. The ^{152}Eu standard source (EG1) provided by Eurostandard, the Czech Republic, was used for energy calibration. Two water solutions in Marinelli beakers (460 ml and 620 ml) were then prepared from the standard ^{152}Eu solution (325 Bq/ml) provided by the National Radiation Protection Institute, the Czech Republic, for efficiency calibration. All gamma spectra were corrected for laboratory background.

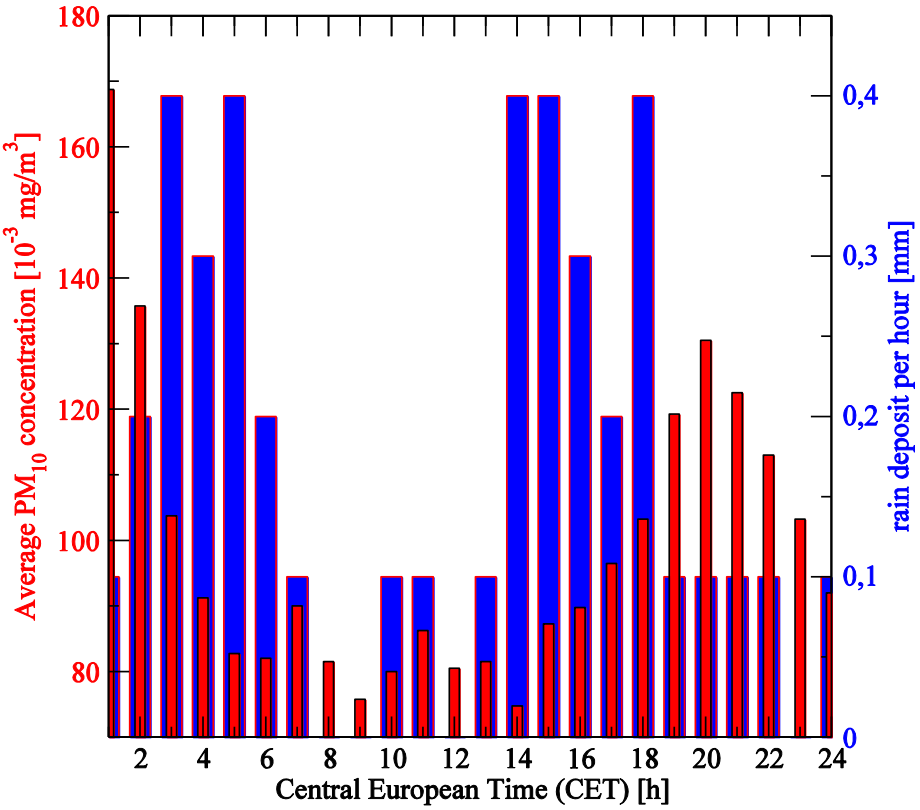

Fig. 1 Rain deposit per hour and PM_{10} average concentration on January 21, 2013 in Ostrava.

3 RESULTS AND DISCUSSION

In Fig. 2, an example of the obtained gamma spectrum is shown (sample B and background). The most important identified radioisotopes and their volume activities including standard uncertainties are summarized in Tab. 1. In the sample, B besides the expected primordial ^{40}K that is the most abundant natural radioisotope (observed gamma emissions from ^{226}Ra decay products, ^{214}Pb, ^{214}Bi, and from ^{228}Th decay product, ^{212}Pb are comparable with the laboratory background), the cosmogenic ^{7}Be was observed (see Fig. 1, the ^{7}Be peak at 478 keV clearly identified in the sample B gamma spectrum and not found in the background) [4]. ^{7}Be is produced mainly in stratosphere and upper troposphere in the spallation process on nitrogen and oxygen induced by high-energy galactic cosmic rays (protons and neutrons) and extraordinary intense solar energetic particles (protons) during great coronal mass ejections. The average production rate of ^{7}Be in the upper troposphere at 11 km was estimated to 0.02 ^{7}Be/cm^2/s [15]. It is known that ^{7}Be like man-made pollutants attaches itself to atmospheric dust particles and enters the earth surface during rain events [16]. In the sample A, the measured ^{7}Be volume activity was lower due to the ^{7}Be decay, the decay-corrected value (1.72±0.22) Bq/l is in agreement with the value obtained from the sample B within standard uncertainties. The volume activities of ^{40}K in both samples agree within standard uncertainties, too.

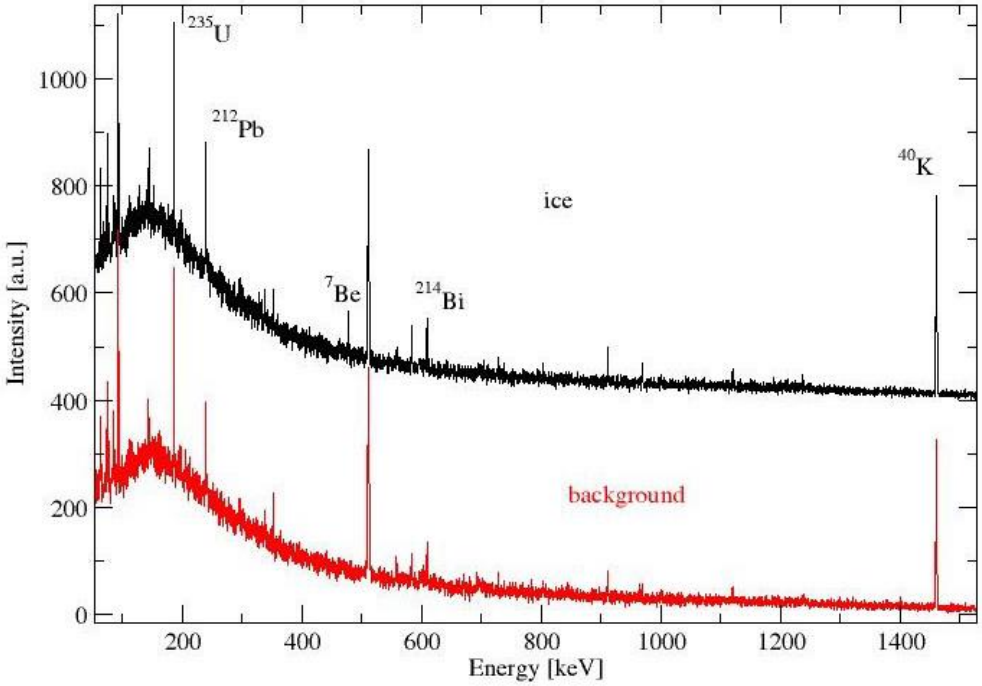

Fig. 2 Gamma spectrum of the sample B (Marinelli beaker 460 ml, 63.2 h detection time) shifted 200 units upwards compared to the laboratory background.

In the sample A, two additional peaks have been observed due to a dry deposit (dust contamination) on the ice layer between its formation on January 21, 2013 and the sample A collection on January 24, 2013. A complex peak at 186 keV can be ascribed to a mutual interference between the ^{235}U (185.7 keV) and ^{226}Ra (186.2 keV) peaks. Assuming that the ^{226}Ra is in radioactive equilibrium with its parent ^{238}U and that the ^{235}U/^{238}U isotopic ratio equals the expected natural value, the counts in the 186 keV peak can be mathematically apportioned between ^{226}Ra and ^{235}U [4] (for the resulting ^{226}Ra and ^{235}U volume activities see Tab. 1). A single peak at 662 keV indicates the presence of the post-Chernobyl and post-Fukushima isotope ^{137}Cs [17, 18].

From the measured volume activities of ^{7}Be and ^{40}K in the sample B (no dust contamination), volume activities in air can be estimated (see Tab. 1) assuming homogenous distribution of radioisotopes in air up to 100 m above the earth surface and approximately one half of the radioisotopes captured during the rainfall (supported by the observed decrease of the average PM_{10} concentration, see Fig. 1). The obtained volume activities in air are below the clearance levels for ^{7}Be (1,8 kBq/m^3) and ^{40}K (47 Bq/m^3) [19]. The level of the volume activity of ^{40}K in air corresponds to the individual effective inhalation dose of 2 μSv/y max for adults and 2.7 μSv/y for infants if comparable unfavourable meteorological conditions occur in approx. 50 days per year. The effective inhalation dose is computed from the inhalation conversion factors h_{inh} = 2,1x10^{-9} Sv/Bq and 2,4x10^{-8} Sv/Bq and the total air inhalation per year 8500 m^3 and 1000 m^3 for adults and infants, respectively [19]. The obtained effective inhalation doses represent less than 0.3 % of the annual effective dose limit of 1 mSv [19].

Tab. 1 Radioisotopes identified in the samples, energies of emitted gamma radiation and volume activities in the samples and in air. The obtained decrease of ^{7}Be volume activity is in agreement with the ^{7}Be halflife.

Sample	Radioisotope	Halflife	Gamma energies [keV]	Volume activity in samples [Bq/l]	Volume activity in air [Bq/m^3]
A	^{7}Be	53,2 d	478	1,13±0,24 (after 40 d)	
B	^{7}Be			1,38±0,43	0,203±0,064
A	^{40}K	1,28 x 10^9 y	1461	4,37±0,77	
B	^{40}K			5,58±0,84	0,82±0,13
A	^{226}Ra	1600 y	186	1,22±0,58	
A	^{235}U	7,04 x 10^8 y	186	0,057±0,027	
A	^{137}Cs	30,08 y	662	0,248±0,034	

4 CONCLUSIONS

The measured volume activities of the melted ice samples from the modest rainfall on January 21, 2013 revealed slightly increased volume activities of ^{7}Be and ^{40}K. The highest estimated annual effective inhalation dose was obtained for ^{40}K. The value is less than 0.3 % of the annual effective dose limit for population, i.e. it presents no health risk.

The detection of the cosmogenic radioisotope ^{7}Be that has been suggested as an independent indicator of air pollution [16] seems to be the most important result of the present measurement. Since rainfalls can clean the atmosphere (washout of the aerosols present below the clouds), an increased ^{7}Be specific activity (volume activity in rain samples and in air and surface activity due to a deposition on the Earth surface) in winter is expected at high PM_{10} and $PM_{2,5}$ air-borne dust concentrations as observed in this study. It is also supported by a recent study of correlations between ^{7}Be and PM_{10} concentrations from a 14-year air monitoring in Taiwan [20]. Seasonal variations of the ^{7}Be specific activity have also been observed [21]. They are ascribed to intrusion of stratospheric ^{7}Be-rich masses into the troposphere in late spring and summer. On the other hand, potential correlations between ^{7}Be wet deposition (in individual rain events) and basic rain characteristics as total precipitation, precipitation rate and the elapsed time between two rain events, have been extensively studied by different authors with divergent results [22], e.g. correlations between the ^{7}Be specific activity and the elapsed time between two subsequent rain events were reported in [21], whereas in [22] no correlations were found. The enhanced ^{7}Be specific activities due to an anomalous transport of air masses over the continent in the upper troposphere and the lower stratosphere where ^{7}Be is formed cannot be also excluded in any season [21].

To clarify the significance of the above mentioned factors and to examine their correlations to the ^{7}Be volume activity in rain, we propose to measure the ^{7}Be volume activity in individual rain events on the roof of the main building of the Technical University of Ostrava for a period of at least one year. The chosen location in the European PM_{10} hot spot in Ostrava [9, 10] will enable to test the correlations in a wide range of PM_{10} and $PM_{2,5}$ values. A collector on the roof of a high building minimizes surface dust contamination. Moreover, an automatic meteorological station of the Institute of Geoinformatics of the Technical University of Ostrava that measures relevant meteorological data every 5 minutes including total precipitation and precipitation rate is located on the same roof. It provides information about precipitation rate evolution during individual rain events that has never been used in analysis of correlations between ^{7}Be volume activity and basic rain characteristics.

Acknowledgements

The article was created within the project of the Institute of Clean Technologies for Mining and Utilization of Raw Materials for Energy Use, Reg. No. CZ.1.05/2.1.00/03.0082, supported by the Research and Development for Innovations Operational Program financed by the Structural Funds of the European Union and from the means of the state budget of the Czech Republic.

REFERENCES

[1] PESCATORE, C., MENON, S., Q. J. Nucl. Med. 44 (2000) 310.

[2] MAZUR, G., MAZUR, P., Nukleonika 55 (2010) 243.

[3] ATWOOD, D. A. (ed.), Radionuclides in the Environment, Wiley, New York, 2010.

[4] GILMORE, G. Practical Gamma-ray Spectrometry, Wiley, New York, 2011.

[5] SZARLOWICZ, K. et al., J. Radioanal. Nucl. Ch. 298 (2013) 1323.

[6] MARLEY, N.A. et al., Aerosol Sci. Tech. 32 (2000) 569.

[7] PAPASTEFANOU, C., Aerosol Air. Qual. Res. 9 (2009) 187.

[8] CHAO, J.H., CHIU, Y.J., LEE, H.P., LEE, M.C., Appl. Radiat. Isotopes 78 (2013), 82.

[9] KALIČÁKOVÁ, Z. et al., J. Phys. Conf. Ser. 429 (2013) 012005.

[10] SRAM, R.J. et al., Epidemiology 22 (2011) S232.

[11] Czech Hydrometeorological Institute Ostrava-Poruba, Data sets from the PM_{10} monitors in Ostrava region, 21.1.2013.

[12] Czech Hydrometeorological Institute Ostrava-Poruba, Data sets of temperature and rain deposit measurements in Ostrava-Poruba, 21.1.2013.

[13] Czech Version of the International Standard ISO 10703:2007.

[14] GENIE 2000, Software Manual, Canberra Inc., 2001.

[15] DUTKIEWICZ, V.A., HUSAIN, L., J. Geophys. Res. 90 (1985) 5783.

[16] KLADKO, D., PROSPERO, J., J. Geophys. Res.: Oceans 116 (2011) 6629.

[17] SUCHARA, I., RULÍK, P., HŮLKA, J., PILÁTOVÁ, H., Sci. Total Environ. 409 (2011) 1927.

[18] RULÍK, P. et al., Radiat. Prot. Dosimetry (2014), doi: 10.1093/rpd/ncu154.

[19] Decree of the State Office for Nuclear Safety No. 307/2002 Coll. on Radiation Protection.

[20] CHAO, J.H., CHIU, Y.J., LEE, H.P., LEE, M.C., Appl. Radiat. Isotopes 78 (2013) 82.

[21] CAILLET, S., ARPAGAUS, P., MONNA, F., DOMINIK, J., J. Environ. Radioactiv. 53 (2001) 241.

[22] AYUB, J.J. et al., J. Environ. Radioactiv. 100 (2009) 997.

RESUMÉ

21.1.2013 dosáhly koncentrace polétavého prachu (PM10) v Ostravě hodnot 200–300 μg/m^3 [11]. Nepříznivé rozptylové podmínky ukončil mírný déšť (max. 0,4 mm/h), který v areálu VŠB-TUO v Ostravě-Porubě vytvořil vrstvu ledu silnou cca 8 mm. Pokles koncentrace polétavého prachu v korelaci s intenzitou dešťových srážek v průběhu dne dokládají hodinová měření koncentrací PM10 ze 4 měřicích stanic na Ostravsku [1] a hodinová měření dešťových srážek v pobočce ČHMÚ v Ostravě-Porubě [12]. Vzorky ledu sebrané do 24.1.2013 byly po roztátí konzervovány kyselinou dusičnou a umístěny do dvou Marinelliho nádob (620 ml a 460 ml).

Gama spektra studovaných vzorků byla naměřena pomocí spektrometrického systému firmy Canberra (polovodičový germaniový detektor umístěný v masivním olověném stínění, mnohokanálový analyzátor a spektrometrický software Genie 2000). Pro energetickou a účinnostní kalibraci byly použity etalony ^{152}Eu. Ve vzorcích byly detekovány izotopy ^{40}K s objemovou aktivitou 6 Bq/l a ^{7}Be s počáteční objemovou aktivitou 1,4 Bq/l. Kosmogenní radionuklid ^{7}Be vzniká především ve stratosféře a horní troposféře v tříštivých reakcích galaktického a solárního kosmického záření s atmosférickým kyslíkem a dusíkem. ^{7}Be se zachytává na prachových částicích a dešťovými srážkami se dostává na zemský povrch [15,16].

Z naměřených objemových aktivit ve studovaných vzorcích byly odhadnuty objemové aktivity detekovaných radioizotopů ve vzduchu, které jsou vesměs nízké, pod uvolňovacími úrovněmi platnými pro vypouštění radioaktivních látek do životního prostředí [19]. Maximální roční efektivní dávka pro jednotlivce způsobená inhalací přítomných radioizotopů nepřesáhne 2 μSv za předpokladu, že srovnatelné rozptylové podmínky se vyskytují po 50 dnů v roce.

Hlavním výsledkem je detekce kosmogenního radioizotopu ^{7}Be, který byl navržen jako nezávislý indikátor znečištění ovzduší [16]. Proto by bylo vhodné monitorovat aktivitu ^{7}Be v dešťových srážkách na Ostravsku v průběhu roku a studovat její závislost na koncentraci polétavého prachu a srážkových úhrnech.