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Temporal Variation of Be-7 Concentration in the Surface Air at the Belgrade – Kumodraž Location

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Concentrations of Be-7 in surface air samples were continuously measured, using a low-background HPGe spectrometer at Belgrade (Kumodraž), Serbia. Weekly data collected over a 12 month period, from July 2008 to June 2009, are presented and discussed. The measured values of Be-7 activity concentrations in surface air samples, ranged from 1.9 mBq/m^3 to 10.2 mBq/m^3, show increase in summer season.

Key words: **radioactivity, radionuclides, air, concentration, gamma spectroscopy.**

Introduction

NUCLEAR weapon tests conducted in the atmosphere and releases of radioactive material from nuclear facilities are the main causes of man-made radioactive contamination in the human environment. Once released to the atmosphere, long-range atmospheric transport processes can cause widespread distribution of radioactive matter, although it may, as in the case of the Chernobyl accident, originate from a single point. The resulting fallout consisting of short- and long-lived radionuclides, eventually affects humans either directly or indirectly by entering the food chain through plants and animals. In both cases, fallout causes a health hazard to the population, either through direct irradiation or consumption of contaminated foodstuffs.

Potential danger of inhalation of radioactive particles must be determined and controlled using the levels and types of radioactivity. Law and local regulations [1, 2] regulate the content of radioactive particles in the air, which is allowed to be present in air samples from the natural environment and working areas. Monitoring of air radioactivity presents a control of daily values of the activity of radionuclide in air samples. The knowledge of natural radionuclide concentration in air is essential, not only because exposure to natural radionuclides in the greatest percentage contributes to radiation exposure [3], but also because this information contributes to the study of atmospheric circulation of air masses [4].

Be-7 is formed by the spallation reaction between cosmic rays and nuclei of oxygen and nitrogen in the stratosphere and upper troposphere. Almost immediately after production, Be-7 atoms are attached to aerosol particles and follow their pathways. Considering that aerosol particles contain most of the air pollutants, Be-7 can be used as a tracer in atmospheric tmospheric paths and deposition ways of atmospheric micro- particles [5, 6]. It is usually located in the form of molecules BeO or $Be(OH)_2$. Most of the Be-7, approximately 70%, is produced in the stratosphere and a smaller amount originates from the troposphere. In the stratosphere the aerosols beryllium are kept approximately a year, but the retention time of beryllium in the troposphere is about six weeks.

It is well known that the production of Be-7 depends on the intensity of cosmic rays hitting the upper layers of the atmosphere. Seasonal variations of Be-7 concentrations in the lower atmosphere are influenced by the periodical thinning of the tropopause when the exchange between the stratosphere and the troposphere is more effective [6]. The wet scavenging, the vertical mixing and the horizontal atmospheric transport could be the factors controlling surface Be-7 concentrations [7].

Because cosmogenic nuclides are produced in the atmosphere when cosmic rays strike it, their production rate is varied by the solar modulation of galactic cosmic rays invading the heliosphere, which is controlled by the solar magnetic field and, in turn, by solar activity. Hence, daily Be-7 concentrations were continuously observed in the surface air since January 2000 in the 23rd solar cycle, at which time the solar activity was at its maximum. The observation location was Yamagata, northern Japan which has an altitude of 168.33 m above the sea level. Although the half-life of Be-7 is 53 d shorter than the half-lives of C-14 and Be-10, it is reliable when used to investigate time variations shorter than 1 y for phenomena like air-mass motion. Moreover, Be-7 is a well-known atmospheric tracer that is unaffected by artificial effects such as atomic bomb experiments [8]. In particular, the temporal trends of the recent solar activity could be determined by simply measuring the daily Be-7 concentrations in the ambient air.

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The average concentration per month of Be-7 in Europe is usually several mBq/m^3. For example, in Bratislava (Slovak Republic), the average monthly concentrations in the period from 1977 to 1990 are in the range from 0.7 to 8.69 mBq/m^3 while in Thessaloniki (Greece) the average monthly concentration of Be-7 is in the range 1-10 mBq/m^3. The listed countries are located at similar geographical latitude [9, 10].

This paper shows the results of the measurements of radioactivity of Be-7 in the surface air samples, where the sampling occurred in the period from 2008 to 2009 at the location Belgrade, Kumodraž.

Materials and methods

Be-7 data used in this study were obtained from our own measurements carried out in Belgrade while the measurements were performed by the Laboratory for Chemical-Nuclear Protection and Applied Spectrometry, Military Technical Institute. The location of the sampling station is at Kumodraž, in the area of Belgrade city.

A high volume air sampler (DH-604EV.2 F&J Speciality Products, Inc) and a cellulosic filter (FJ213340) were used for aerosol collection. Air sampling was carried out with two digital samplers which can supply the flow of air from 15 to 120 m^3/h. The air was drawn through the filter 1.770 mm thick, with 65% filtration efficiency on the DOP test, at a rate of approximately. The efficiency of the filters is given by the manufacturer based on tests, and one of them is the DOP test. The test means the efficiency of the filter surface of 100 cm^2 for the filtration of an air or gas flow rate of 32 L/min for dioctylphthalate particles of approximately 0.3 μm in size in the concentration of 100 μg/L [11]. The air samples were taken at the height of 124 cm from the ground.

The cellulosic filter paper FP213340 was chosen as the most appropriate for sampling. Its characteristics are a good compromise between the efficiency of collection and the fall of pressure, i.e., resistance to air flow. For the optimization of time measuring and measuring uncertainty it was found that is necessary to have at least 5000 m^3 for the control of th radionuclide content in the samples of air, but in case of emergency events a lower volume of air is enough.

The initial air flow was 50 ± 5 m^3/h, the temperature was in the range from 3 to 34ºC, with the fluctuation of daily and night temperatures and weather conditions characteristic for the period of time from July 2008 to June 2009. During a month three samples were sampled and the average value of these measurements was presented. The filter paper, 10.2 cm in diameter, with a sample of 5000 m^3 collected air, was measured directly and was analyzed by the gamma spectroscopic method [12].

Be-7 activity collected on the filter was measured through the characteristic gamma-ray emission associated with the transitional electron capture decay of this radionuclide. The gamma spectroscopic analysis was performed using HPGe, whose characteristics are given in Table 1. The concentration of Be-7 in the surface air was determined by direct comparison with the Be-7 reference standard and using the half-life, the sampled air volume and the time delay in sampling and measurement. The intensity of 477.6 keV gamma peak was used to get the final result of the concentration of Be-7 in the surface air (in Bq/m^3). The monthly average concentrations were calculated as an arithmetic mean. The overall uncertainty in individual measurements was generally less than 10%.

The radioactive source for energy calibration was a mix gamma standard type MBSS-2 in the Marinelli geometry of 1000 mL, which was developed by the "Inspectorate for Ionizing radiation Czech Metrological Institute" and which contains the following radioactive isotopes: Am-241, Cd-109, Co-57, Ce-139, Ba-133, Sn-113, Sr-85, Cs-137, Y-88, Mn-54 and Co-60.

Considering the specific geometry of measurement, the efficiency calibration using the surface gamma standards was done. The standard was made by applying 55 drops of solution of respective volume of 10 μL on the paper in the hexagonal form. The solution was prepared by dilution of IAEA reference material that contains the following radioactive isotopes: Am-241, Cd-109, Co-57, Ce-139, Hg-203, Sn-113, Sr-85, Cs-137, Y-88 and Co-60. The specific activities of these radionuclides were 164.94 Bq/mL; 250.52 Bq/mL; 3.81 Bq/mL; 0.53 Bq/mL; 0.00 Bq/mL; 0.81 Bq/mL; 0.02 Bq/mL; 143.54 Bq/mL; 1.12 Bq/mL; 130.83 Bq/mL on October 28th 2008, respectively.

The samples were measured over the time of 255000 s.

Table. 1. Main specification High - Purity Germanium Coaxial Detector System

Model	ORTEC GEM50
Cryostat Configuration	SV-GEM
Detector Diameter and Length	(69.7 × 58.9) mm
Absorbing Layers	Aluminium – 1.50 mm Inactive Germanium - 700 μm
Recommended Operating Bias	+ 3700 V
Resolution (FWHM) at 1.33 MeV, Co-60	1,90 keV (Warranted) 1.78 keV (Measured - MTI)*
Peak-to-Compton Ratio, Co-60	66:1 (Warranted) 76:1 (Measured - ORTEC)*
Relative Efficiency at 1.33 MeV, Co-60	50% (Warranted) 59.2% (Measured - ORTEC)*
Peak Shape (FWTM/FWHM),Co-60	1,90 (Warranted) 1.90 (Measured - ORTEC)*
Resolution (FWHM) at 122 keV, Co-57	0,90 keV (Warranted) 0.76 keV (Measured - MTI)*

** Amplifier time constant - 6 μs*

Results and discussion

Fig.1 gives the total radionuclide content of Be-7 in the air samples onto the filter paper FP213340, when the initial air flow is 50 ± 5 m^3/h, the temperature is in the interval of day and night fluctuations from 3 to 34ºC and the layer from which the samples are taken is at a height of 1.24 m from the ground.

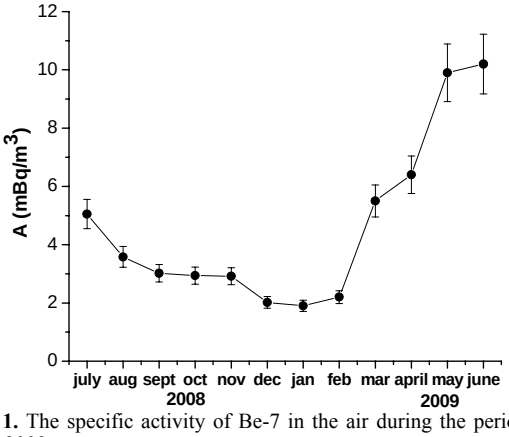

Figure 1. The specific activity of Be-7 in the air during the period from 2008 to 2009

The specific activity of cosmogenic Be-7 was in the range from 1.9 mBq/m^3 to 10.2 mBq/m^3. This agrees with published values characteristic for the city of Belgrade and the environment [13]. The measured Be-7 specific activity showed seasonal behaviour with the highest values usually measured in June. It was shown that the concentrations of Be-7 in the surface air are sensitive to the phase of the sunspot cycle. Probable explanations for these monthly variations, in addition to solar activity, were variations in temperature and rainfall, as well as meteorological factors. Lower concentrations typically occur at sunspot maximum due to a decrease in the galactic cosmic-ray flux to Earth. This is a global phenomenon [14]. They suggested that these variations were caused by rainfall anomalies during El Nin˜o/Southern Oscillation (ENSO). However, these explanations require further investigation.

Conclusion

Method of gamma spectroscopy was used for systematic monitoring of specific activities of the cosmogenic radionuclide Be-7 in the surface air samples at the location Kumodraž, Belgrade, over the period from July 2008 to June 2009. The measured Be-7 activity concentrations showed seasonal behaviour with the highest values usually measured in July. Despite the constant presence of radioactive matter in the Belgrade air during the observational period, the activity concentration values never exceeded legal limits.

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Praćenje koncentracije Be-7 u površinskom sloju vazduha na lokaciji Beograd - Kumodraž

U radu su predstavljeni rezultati kontinualnog praćenja sadržaja radionuklida kosmogenog porekla Be-7 u prizemnom sloju vazduha. Eksperiment je vršen na lokaciji Kumodraž u okolini Beograda, u vremenskom periodu od jula 2008. do juna 2009. godine uz korišćenje poluprovodničkog germanijumskog HPGe detektora visoke čistoće. Izmerene aktivnosti Be-7 su bile u intervalu od 1.9 mBq/m^3 do 10.2 mBq/m^3 i maksimumom u letnjem periodu.

Ključne reći: **radioaktivnost, radionuklidi, vazduh, koncentracija, gama spektrometrija.**

Наблюдение над концентрацией Be-7 в поверхностном фоне воздуха в районе Кумодраж-Белград

В настоящей работе представлены результаты непрерывного наблюдения содержания радионуклида космогоного происхождения Be-7 в заднем низком фоне воздуха. Эксперимент проведён в районе Кумодража в окрестности Белграда, в периоде с июля 2008-ого по июнь 2009-ого года, при помощи полупроводникового HPG детектора из германия высокой чистоты. Измеренные радиоактивности Be-7 находились в интервале с 1.9 mBq/m³ по 10.2 mBq/m³ и летом достигли максимального значения.

Kly~evwe slova: **радиоактивность, радионуклиды, воздух, концентрация, гамма-спектрометрия.**

Le contrôle de la concentration du Be -7 dans la couche d'air superficielle sur la localité de Belgrade – Kumodraž

Ce papier présente les résultats des mesurages continus de la quantité du radionuclide cosmogène Be-7 dans la couche de l'air près du sol. L'essai a été réalisé sur la localité Kumodraž , aux environs de Belgrade, pendant la période du mois de juillet 2008 jusqu'au juin 2009, a l'aide du détecteur semi conducteur de germanium HPGe de haute pureté. Les activités du Be-7 mesuré étaient dans l' intervalle de 1,9 mBq/m³ jusqu'à 10,2 mBq/m³ alors que le maximum était constaté pendant l'été.

Mots clés: **radioactivité, radionuclide, air, concentration, spectrométrie gamma.**