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## Air Radioactivity Monitoring in Serbia

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### 1. Introduction

Today radioactivity monitoring in the environment is required not only around sites where the significant amounts of radioactive material are used or stored, but in a number of other processes as land remediation or decommissioning of nuclear plants. Since the end of the 20<sup>th</sup> century there has been an increasing awareness of the so-called technologically enhanced natural radioactivity and the need for its monitoring in a wide range of non-nuclear industries including oil and gas extraction, ceramic and glass industries, production of phosphoric acid, production of different phosphoric fertilizers and other chemical industries. Coal electric power plants as significant sources of ashes and other contaminants are included, too. Contrary to nuclear facilities or radioactive waste control these processes require measuring radionuclides at very low levels of radioactivity. This demands specific methodology and instrumentation and responding to the very real economic importance of classifying waste products from decommissioning and remediation processes. However, natural or man made, waste or industrial, environmental radioactivity monitoring requires adequate equipment calibrated in the full range of radionuclides likely to be encountered, as well as trained and experienced personnel. It is the task of national metrological institutions to provide radioactive reference materials and standards for calibration as pure isotopes, mixtures of isotopes or reference materials for particular applications and to supervise the processes of quality control and quality assurance. Besides primary standards and reference materials national metrological institutions should provide laboratory proficiency testing exercises to support radioactivity measurements within academic, industrial or research laboratories (Spasic, 1984; Spasic, 1985; Spasic, 1987a; Spasic et al. 1987).

### 2. Environmental Radioactivity Monitoring

To protect the general public, national and international regulations require that levels of radioactivity in the environment and in foodstuffs are regularly monitored. This include first, the primary standards for radionuclides to be measured and secondly, a unique range of very low activity standardized solutions (e.g. about 1 Bq per ml) based on the primary standards to be provided (Fisenne, 1993; CCEMRI, 1996). The main advantage of the solutions is that they are 'ready-to-use', thus improving the accuracy of calibration and reducing the risk of contaminating expensive analytical instrumentation. Also,

environmental monitoring laboratories are obliged to demonstrate the accuracy of their measurements, for compliance with quality assurance management systems such as ISO 17025 (Spasic-Jokic, 1998; Spasic-Jokic et al., 2002; Todorovic et al., 2003; Spasic-Jokic et al., 2006).

Methods used in the environment radioactivity monitoring include a number of highly sophisticated nuclear techniques and instrumentation such as  $^{40}\text{K}$  beta-gamma counting, electron capture gamma counting, alpha-gamma and X ray-gamma coincidence counting, tracer-efficiency counting, liquid scintillation counting, high resolution gamma ray spectrometry, re-entrant ionization chambers, gas counting and total X ray counting (CCEMRI, 1994; Quin, 1996; Spasic-Jokic, 1998; Spasic-Jokic et al., 1998).

### **2.1 Quality Assurance (QA) and Quality Control (QC) in Air Radioactivity Monitoring**

To assure the quality of the environmental radioactivity measurements it is necessary to provide optimum site location and sampling procedure, appropriate and high quality instrumentation, best operating practices, calibration procedures and frequency, appropriate data and documentation handling, statistical procedures and compatibility with national network standards. All measurements undertaken have to be underpinned by accurate calibration techniques with traceability to international standards. Thus it is necessary to design and undertake industrial and urban air and water pollution surveys to meet European Union Directive requirements, to develop and implement new measurement protocols, to carry out site and operation audits, to advise on CEN (Comité Européen de Normalisation, European Committee for Standardization) standards requirements and to research into novel techniques and instrumentation for pollution monitoring. Also, it is imperative to train local staff to carry out day-to-day monitoring functions, to design and optimize fixed air and water-quality monitoring networks, and to negotiate with local authorities and State Environment Agency (Fisenne, 1993).

Quality Assurance and Quality Control programs in radionuclide metrology deal with primary reference materials, standards, techniques and systems, secondary standards and systems, validation and demonstration of equivalence for measurement standards, infrastructures for environmental monitoring, nuclear data for applications of ionizing radiation and last but not least, the transfer of knowledge (Spasic, 1982; Spasic-Jokic et al., 2002).

### **2.2 Traceable Radionuclide Standards in Environmental Radioactivity Monitoring**

National regulations and legislation demand the highest standards for calibration and traceability to National Measurement Standards in radioactivity measurements. Nuclear site licensing, environmental impact assessments, national and international accreditation and ISO 9000 certification increasingly rely on the use of traceable low level activity radionuclide standards for spectrometers calibration and radiochemical assay validation. Low activity mixed nuclide gamma ray and single gamma ray emitters, alpha and beta particle emitters as environmental radionuclide standards are now in routine laboratory use by nuclear fuel cycle facilities, governmental regulatory agencies and reactor research centers. Wide range of low level activity mixed nuclide gamma-ray emitters, single nuclide gamma ray emitters, alpha particle and beta particle emitter radionuclide solutions, radon gas and spiked standards have been specifically developed to meet the needs of radioactivity environmental measurements. Thus, single gamma ray

radionuclide solutions and milk powder are suitable for calibration of spectrometers with either Ge or NaI detectors as they efficiently overcome problems associated with cascade summing and sample density effects. Alpha and beta particle emitter solutions are widely used as chemical yield tracers, in isotope dilution techniques, as internal standards for radiochemical assays as well as in calibration of alpha spectrometers. These solutions are also suitable for calibration of proportional counters, Geiger-Müller tubes and liquid scintillation counters. All low activity environmental radionuclide standards have to be directly traceable to national absolute primary radioactivity standards and further to meet the validation and intercomparison with other national standards laboratories organized under the auspices of the International Bureau for Weights and Measures (BIPM) in Sevre, France (Spasic, 1986; Spasic-Jokic, 1998; Quin, 1996; BNM, 1995; CCEMRI, 1991; CCEMRI, 1994; CCRMRI, 1996).

For efficiency calibration of hyper-pure Germanium gamma ray detectors mixed gamma-radionuclide standard are used, with typical activity values in Bq/g (in brackets):  $^{241}\text{Am}$  (30),  $^{139}\text{Ce}$  (9),  $^{60}\text{Co}$  (50),  $^{109}\text{Cd}$  (250),  $^{57}\text{Co}$  (7),  $^{137}\text{Cs}$  (30),  $^{203}\text{Hg}$  (30),  $^{113}\text{Sn}$  (30),  $^{85}\text{Sr}$  (50) and  $^{88}\text{Y}$  (160), and the uncertainties in the range  $\pm 0.5\%$  to  $\pm 4\%$  (at 95% confidence level). The most frequently used environmental radioactivity standards are: Strontium-90, Caesium-137, Caesium-134, Mixed Nuclides, Europium-152, Technetium-99, Americium-241, Iodine-129, Plutonium-242, Americium-243, Carbon-14, Uranium-232, Neptunium-237, Lead-210, Plutonium-239, Curium-244, Thorium-229, Uranium-238, Uranium-238, Tritium and Organically Bound Tritium (all in activity range 0.3 Bq/g - 20 kBq/g, mass 1 - 10 g nominal) (Spasic et al, 1987; Spasic-Jokic, 1998; CCEMRI, 1996). The methods and instrumentation most frequently used for verification of radioactive sources as certified reference materials are Ge spectrometers, proportional counters, digital coincidence counting (DCC), triple-to-double coincidence ratio (TDCR) and selective sampling (Fisenne, 1993; Spasic-Jokic, 1998; Spasic et al., 1987).

### 2.3 Metrological Legislative in Serbia

Officially, the system of metrological assurance in the field of ionizing radiation in Serbia was initiated in 1981 when Advisory Commission for Ionizing Radiation was formed within Federal Bureau of Measures and Precious Metals, in Belgrade. The very first task of the Commission was to prepare various acts of metrological legislation as a base of metrological assurance. That became highly urgent during and after the Chernobyl nuclear plant accident in 1986. Since than more than 60 regulatory papers, guidelines and classification schemes were prescribed by the Commission and became legally obliged (Spasic, 1984;, Spasic & Bek-Uzarov, 1986; Spasic-Jokic, 1998; Spasic, 1987b; Spasic, 1987c).

Among 64 regulatory papers 28 of them are dedicated to radionuclide metrology and to metrological assurance of the environmental radioactivity measurements. These include:

- 1) 6 classification schemes (hierarchy of standard instruments and sources) for dissemination of unit for activity of radionuclides alpha, beta and gamma emitters, issued until 1998;
- 2) 12 regulatory papers dedicated to metrological conditions for various instruments as are: semiconductor and scintillation gamma spectrometers and counters, semiconductor and scintillation alpha spectrometers and counters, GM counters,

proportional counters in various geometries, plastic and liquid scintillation counters and various ionization chambers; and

- 3) 12 guidelines for verification of instruments as in line 2).

#### **2.4 Instrumentation and Methods in Radioactivity Measurements: State of Art in Serbia**

Since 1991 the Commission for Ionizing Radiation in the Federal Bureau of Measures and Precious Metals in Belgrade has been elaborating the data base on human, equipment and laboratory resources in the environment radioactivity research area. The Working Group for Radioactivity Measurements prepared and distributed the Questioner on Instrumentation and Methods in Radioactivity Measurements which provided data on instrumentation, methods, personnel, sampling procedures and general working conditions in the institutions involved in the environmental radioactivity measurements. The Questioner was aimed to improve the Quality Assurance and Quality Control procedures. It was prepared according to Environmental Measuring Laboratory Procedures Manual "Quality Control and Quality Assurance in Radioactivity Measurements" HASL-300 (US Energy Department, New York, 1990/92) and was distributed to 15 laboratories and institutions involved in the environmental radioactivity monitoring in the country. The principal users of the results were and still are Ministries for health, science and environmental protection as well as Military, Police and other governmental offices (DPL, 1998).

The data from the Questioner on the laboratory space showed significant differences among laboratories in total working area (30 m<sup>2</sup> - 400 m<sup>2</sup>) and in number of available premises (2 - 21). All laboratories were air-conditioned and with appropriate detectors protection but only two could be considered as low-background laboratories. Staff and experts, mostly physicists, electrical engineers and chemists with MSc and PhD degree were well trained, but there was a significant shortage of well trained technicians. The number of personal differed significantly among the laboratories, too (4 - 16 per laboratory) (Spasic-Jokic et al., 1998).

Average number of samples measured per laboratory varied between 50 and 1000 (maximum 6000). As sampling network was very loosely defined, location and collecting frequency became the subject of the national Law of Radiation Protection, issued in 1994 and of other regulatory papers (Spasic-Jokic et al., 1998).

In the time when Questioner was sent the laboratories were equipped mostly with HPGe gamma spectrometers, also with NaI (Tl) and Ge(Li) detectors, liquid scintillation detectors, some flow proportional alpha-beta anti-coincidence counters and alpha semiconductor spectrometers and some nationally developed radiometric laboratories (LARA). Most of the instruments were purchased after the Chernobyl accident, in 1986. Instruments are of standard construction, with commercial lead shielding (5-10 cm). Depending of the shielding, the background counts were between 0.8 cps and 7 cps for semiconductor gamma detectors, 0.1 cps to 1.2 cps for beta and alpha radiation detectors and up to 20 cps for Na(I) detectors. To note, during the Chernobyl accident there was only one laboratory for low level activity measurements in the environmental samples, with background count of 1 cps (100 - 2000 keV) at the Department of Radiology, Faculty of Veterinary Medicine, Belgrade. Over 90 % laboratories were monitoring gamma activity, only a few of them were equipped for alpha and beta emitters monitoring.



Among natural radionuclides most commonly determined were  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$ ,  $^{220}\text{Rn}$ ,  $^7\text{Be}$ ,  $^3\text{H}$ ,  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$  and among fission products those were  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{60}\text{Co}$  (Spasic-Jokic et al., 1998).

Calibrations were performed by standard certified reference materials generally in geometry of point sources and/or Marinelli bakers. Radionuclides in mixed or pure sources were usually  $^{152}\text{Eu}$ ,  $^{133}\text{Ba}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Am} + ^{239}\text{Pu} + ^{244}\text{Cm}$ ,  $^{60}\text{Co}$ ,  $^{57}\text{Co}$ ,  $^{144}\text{Ce}$ ,  $^{54}\text{Mn}$ ,  $^{22}\text{Na}$ ,  $^{85}\text{Sr}$ ,  $^{88}\text{Y}$ ,  $^{203}\text{Hg}$ ,  $^{40}\text{K}$ , but some laboratories also used standard reference samples of sludge, soil, sand and phosphates. All laboratories use commercial sources provided from International Agency of Atomic Energy (IAEA), Hungarian National Office of Measurement (OMH), National Institute of Standards and Technology (NIST) and Amersham Co., with extended measuring uncertainty up to 5 %. Most of the laboratories participated in the first national intercomparison organized and coordinated by Federal Bureau of Measures and Precious Metals in 1992. The intercomparison pointed to some problems in sampling procedures, inappropriate calibration, and shortage of legally verification culture and non-existence of well defined and applied QC and QA programs and methods. Those problems were over-bridged by numerous intercomparisons organized at national, regional and international level and closer connection with international bodies (IAEA) and organizations in the field (Kandic et al., 1993; Spasic-Jokic et al., 1998).

### 3. Air Radioactivity Monitoring

On global scale air radioactivity monitoring provides important data for comparing environmental impact of radioactivity from anthropogenic sources to natural ones and for studying global atmospheric processes (Baeza et al., 1996; Gaggelar, 1995; Gustafson et al., 1981; Arimoto et al., 1999; Djuric & Popovic, 1994). Most of air-born radionuclides are attached to aerosols and their concentration in ground level air is mainly due to behavior of aerosols in the atmosphere. As aerosols play an important role in the atmospheric processes as carriers of atmospheric materials and available surfaces for different chemical processes, radionuclides can be used to trace their transport, as well as transport of ozone in the atmosphere (Gaffney et al., 1994; Gaffney et al., 2004; Groundel & Papastefanou, 2004). Among naturally occurring radionuclides in air, radon ( $^{222}\text{Rn}$ ) and its progenies ( $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$ ,  $^{210}\text{Po}$ ) are most significant, while  $^{131}\text{I}$ ,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are of major interest among the fission products. Most commonly radionuclides monitored in ground level air are cosmogenic  $^7\text{Be}$ , natural  $^{210}\text{Pb}$  and anthropogenic  $^{137}\text{Cs}$ .

Berillium-7 (half life 53.28 days) is produced by cosmic rays in spallation processes with light elements (N,O,C) in the upper troposphere and lower stratosphere and its annual mean concentrations are good indicator of the changes in the atmospheric production rate due to cosmic ray intensity and are correlated to stratosphere-to-troposphere exchange processes (Gaggelar, 1995; Gaffney et al., 2004). Concentrations of  $^7\text{Be}$  in air at midlatitudes exhibit maxima in spring/summers periods attributed to seasonal thinning of tropopause that allows stratospheric masses rich with  $^7\text{Be}$  to enter the troposphere (Agelaio et al., 1984; Bettoli et al., 1998; Cannizzaro et al., 1995; Hirose et al., 2004; Ioannidou & Papastefanou, 1997; Todorovic, 1997). Concentration of  $^7\text{Be}$  in air is effected by air exchange between stratosphere and troposphere, vertical mixing

within the troposphere and the washout effects (Todorovic, 1997; Gerasopoulos et al., 2003).

$^{210}\text{Pb}$  (half life 22.3 years) is produced in the decay of  $^{238}\text{U}$  into gaseous  $^{222}\text{Rn}$  (half life 3.8 days) that escapes from soils into the atmosphere where one of its short lived daughters ( $^{218}\text{Po}$ ) decays into  $^{210}\text{Pb}$  (Gaggelar, 1995; Gaffney et al., 2004; Groundel & Postendourfen, 2004; Todorovic et al., 2000). In small amounts radon and thus  $^{210}\text{Pb}$  are produced in coal combustion processes and nuclear explosions (Todorovic et al., 2007). Deposition of  $^{210}\text{Pb}$  varies with season and geographic position and variations in  $^{210}\text{Pb}$  concentration in ground level air are used to trace continental emission and to investigate aerosols transport (Peters et al., 1997; Tokieda et al., 1996).  $^{210}\text{Pb}$  concentrations exhibit maxima in autumns that may be attributed to increased radon emanation (Hirose et al., 2004).

Long lived  $^{137}\text{Cs}$  (half life 30 years) is most significant among fission products and a reliable indicator of anthropogenic pollution in the environment due to nuclear weapon atmospheric tests and nuclear power plant accidents. After the moratorium on the atmospheric nuclear weapon tests in the 1970ties and the decrease in concentrations of cesium released in the atmosphere during the Chernobyl nuclear plant accident in 1986 due to its migration into deeper layers of soils, concentrations of  $^{137}\text{Cs}$  in ground level air in the 1990ties were of order of  $\mu\text{Bq}/\text{m}^3$ . The concentration pattern was exhibiting one or two maxima in summers and winters, the summer maxima due to stratosphere to troposphere air exchange and the winter one due to soil dust resuspension in air (Cannizzaro et al., 1995; Larsen et al., 1995; Todorovic et al., 1997).

#### 4. Air Radioactivity Monitoring In the Institute Vinca, Belgrade

Air radioactivity control in the Institute for Nuclear Sciences Vinca started in the late 1950ties, after an accident at the experimental reactor. It comprised continuous monitoring of ground level air radioactivity in several sites in the immediate vicinity of the Institute, chosen to enable the control of the most probable trajectories of radioactive matter possibly released from the reactor. Two of the four control stations, at the distance of 2 and 7 km were in the direction of the dominant wind blowing towards the city of Belgrade. Air radioactivity monitoring program included continuous control of short and long lived radioactive aerosols, background spectra, radioactive gasses in the free atmosphere, wet and dry deposition, as well as logistic meteorological measurements (Todorovic et al., 1996; Todorovic et al., 2007).

##### 4.1 Materials and Methods

At the very start of the monitoring program only 4 automatic devices for aerosols total beta activity with movable filter belt and continuous counting (Friesseke-Hoepfner 59A) were used. The devices enabled measuring activity of the radionuclides in 2 different delay periods compared to the time of sampling, as 2 scintillation detectors were positioned along the belt. Sampling period was 228 minutes, while delay periods were 78 minutes and 60 hours. Thus the first measurement provided the data on the total short and long lived radionuclides in air, and the second one provided the data on the activity of long lived radionuclides only. Data on short lived radionuclides in air were obtained by simply contracting the results of the measurement.

Under normal conditions, the rate of natural radionuclides originated from radon and thoron is dominant in the measurements after 78 minutes, showing pronounced daily variations: maxima in early mornings and minimums in the afternoons. This daily pattern is closely connected with meteorological conditions in the ground level air, and the best early indication for increased level of anthropogenic radionuclides in air.

Measurements of total beta activity in air in 24-hour's aerosols samples were introduced in the late 1960ties and they completely replaced the automatic devices in 1976. The activity were determined by  $2\pi$  proportional alpha/beta counters in air voluminous samples (600 m<sup>3</sup>), taken on filters with effective diameter of 13 cm. The samples were measured 5 hours and 5 days after the sampling. Under no anthropogenic pollution, first measurement provided data on the level of radionuclide thorium-B and its daughters, and the other one on the activity of long lived radionuclides in the aerosols, originated from the nuclear tests or the reactor emission. As alpha activity data are very significant (fission products are in general beta/gamma emitters) so when increased beta activity after 5 hours is followed by an adequate increasement of alpha activity, it could be attributed to natural radionuclides. On the contrary, non proportionality of alpha and beta activity in air would undoubtedly pointed to anthropogenic origin of radioactive pollution.

Monitoring of gamma emitting radionuclides in aerosols by gamma spectrometry started in the mid 1960ties, first with an Na(Tl) detector (well-type, "3x3" crystal with 100 channel amplitude analyzer). Today three high computerized gamma spectrometric systems with HP Ge detectors are in use (see 5.1). Under normal conditions composite monthly filter samples previously ashed up to 370°C are analyzed, while when radioactivity is increased, filters could be analyzed without previous preparation.

To determine plutonium in air a method of radiochemical separation of plutonium from aerosols was developed. The process included air filters ashing at 400°C, dissolving in nitric and chlorohydrogenic acid, separation on anions resins and finally electro-deposition of plutonium on steel plates. Plutonium was than determined by alpha spectrometry. Efficiency of the method was obtained by adding a standard <sup>236</sup>Pu solution (Harwel, UKAEA) (Todorovic et al., 1996). Concerning different psychical-chemical forms of iodine that could be present in the atmosphere during episodes of iodine pollution, a special sampling procedure was developed in 1969. A complex sampler type May-pack was constructed, containing an aerosol filter for sampling iodine attached to aerosols, two coal impregnated filters for sampling large organic compounds and 6 layers of active coal for sampling lighter iodine organic compounds. The system has an efficiency of nearly 100% for all iodine compounds, and proved highly efficient during the nuclear plant accident at Chernobyl in 1986 (Todorovic et al., 1996).

#### **4.2 Air Radioactivity Monitoring at the Institute Vinca up to 1991**

The data of total beta activity in air measured at the Institute Vinca from 1961-1968 are presented as average monthly values in Fig.1. As the results are affected by the time delay between the measuring and the sampling periods the results for the 5 days delay period and for the 60 h delay period for years 1967 and 1968 are presented on the figure, too. The effects of the level of natural radionuclides (ThB) on the total beta activity measured with the time delay of 60 hours are obvious and therefore only the results obtained by measuring 24 sampling hours' aerosols with time delay of 5 days should be considered as reliable. Daily sampling and determining total beta activity in air with a 5 days delay period over the long time period enabled forming a comprehensive data base giving a reliable insight into the



fluctuations of the activities of radionuclides in air and a possibility to define a reference level for further investigations and intervention (Todorovic et al., 1996).

The results of long time trend of total beta activity in air in 1967-1991 measured in the Institute Vinca are presented in Fig. 2.

The seasonal maxima (fig. 2.) observed in spring (April, May, June) are due to the transport of air from higher to lower heights while other maxima are due to nuclear weapon tests performed up to 1980. A sharp increase in May 1986 is due to the nuclear plant accident at Chernobyl that resulted in a general increase in the level of the radionuclides activities in air in the next months, as well as to incidental increases due to soil resuspension in January 1987 (Todorovic et al., 1996; Todorovic, 1997; Todorovic et al., 1997; DM Report 96-01; Popovic & Spasic-Jokic, 2006).

Generally, the level of natural radionuclides in air is about two orders of magnitude higher than the level of anthropogenic radionuclides. Daily fluctuations in the level of natural radionuclides are pronounced: during temperature inversion in early mornings the level of natural radionuclides could be even three orders of magnitude higher than in the afternoons. Natural radioactivity is much affected by microclimate, too, presenting a seasonal pattern with maxima in late summers/early autumn's period (August - October) due to the increased radon emanation from soils (Popovic & Todorovic, 2006).

Fission product cesium-137 was detected in the atmosphere first after the nuclear bomb attacks at Hiroshima and Nagasaki, in 1945. In the period of intensive nuclear weapon tests in 1966 and 1967 the level of  $^{137}\text{Cs}$  in air measured at Vinca Institute in Belgrade was in the range of 0.1 - 0.8 mBq/m<sup>3</sup>. In the late 1970ties and early 1980ties cesium was on the limit of detection until the nuclear plant accident at Chernobyl, in April-May, 1986. Activities of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{103}\text{Ru}$  and  $^{131}\text{I}$  in daily air samples during the nuclear plant accident in Chernobyl are presented in Table 1., together with total beta activity in air. Since 1991, continuous gamma spectrometry of aerosols has been performed. Cesium-137 had specific seasonal pattern following total beta activity seasonal variations (Fig.3.) (Smiljanic et al., 1989; Todorovic et al., 1996; Todorovic et al., 1997).

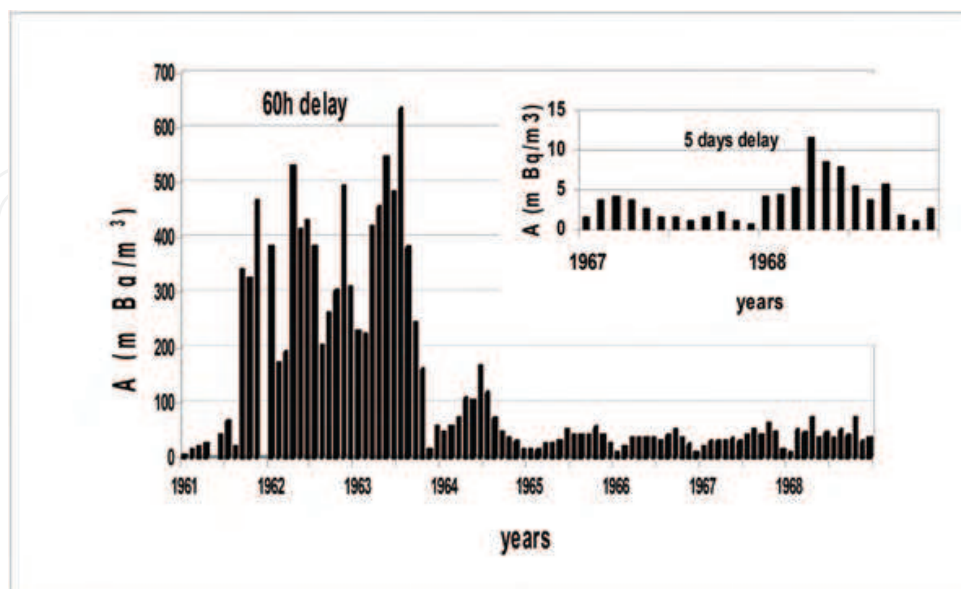


Fig. 1. Total beta activity in air 1961-1968, Institute Vinca, Belgrade (Todorovic et al., 1996)

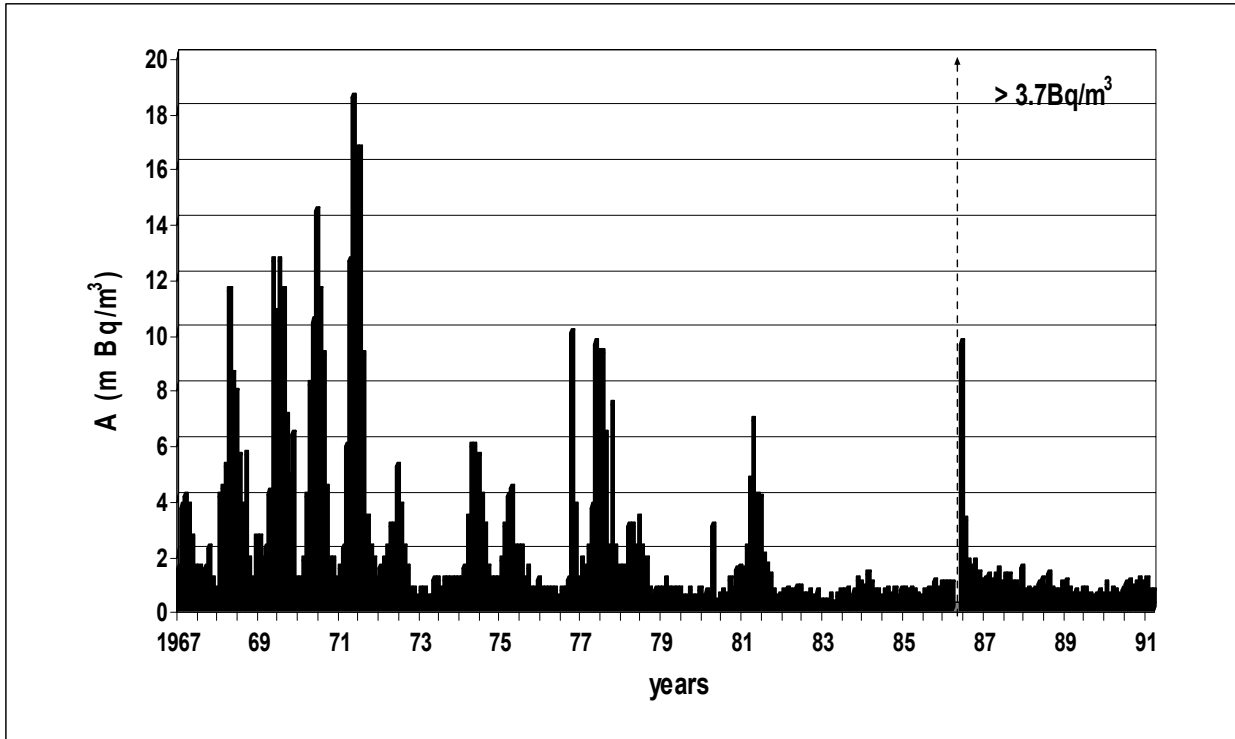


Fig. 2. Total beta activity in air, Institute Vinca 1967 -1991 (Todorovic et al., 1996)

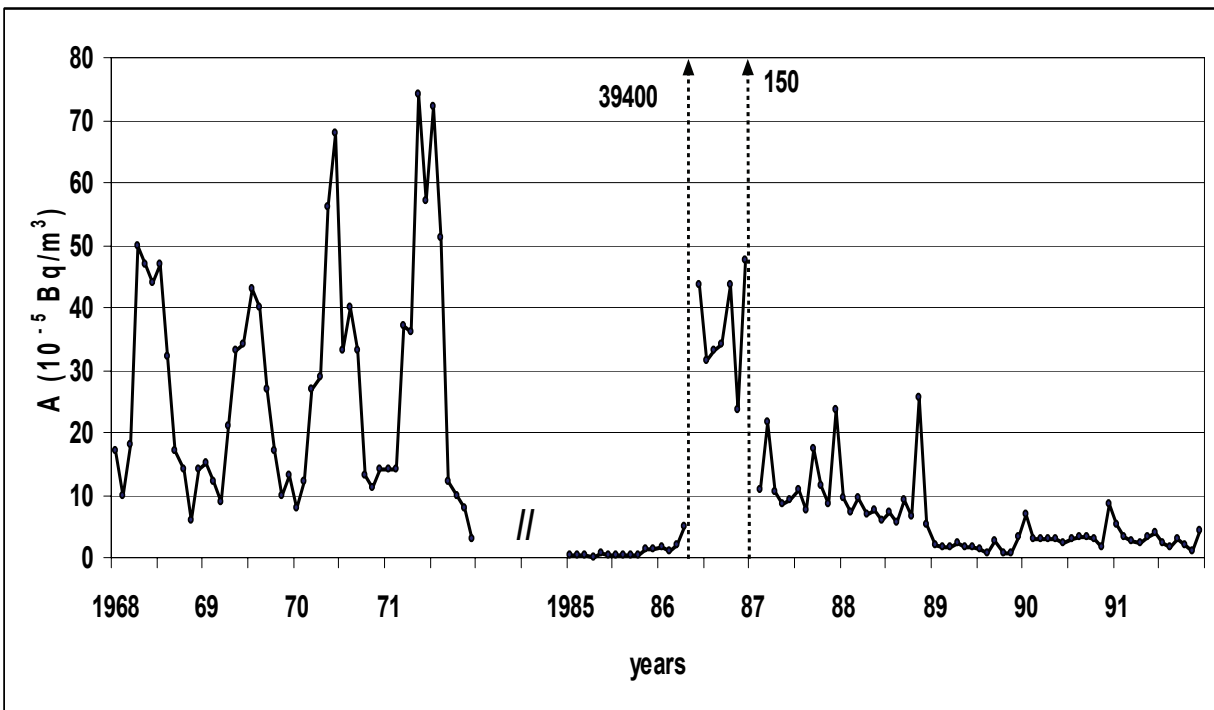


Fig. 3. <sup>137</sup>Cs in ground level air, Institute Vinca up to 1991 (Todorovic et al., 1996)

Date	Total $\beta$ activity (Bq/m <sup>3</sup> )	Activity (Bq/m <sup>3</sup> )			
		<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>131</sup> I(1.*)	<sup>103</sup> Ru(*)
May 1986.	(Bq/m <sup>3</sup> )				
01.	18.9 10 <sup>-3</sup>	22.4 10 <sup>-3</sup>	<10 <sup>-3</sup>	>38.6(2)	9.6 10 <sup>-3</sup>
02.	33.4	2.6	1.3	>20.8(2)	9.2
03.	28.5	2.9	1.4	>4.1(2)	8.1
04.	6.2	0.5	0.3	>15.0(2)	1.4
05.	2.6	0.3	0.1	7.3	0.73
06.	17.7	1.9	1.0	8.2	9.1
07.	12.9	1.5	0.8	3.8	7.7
08.	6.3	0.6	0.3	4.6	4.5
09.	4.2	0.4	0.2	1.5	3.4
10.	1.3	4.9 10 <sup>-3</sup>	<10 <sup>-3</sup>	1.0	20.3 10 <sup>-3</sup>
11.	23.4 10 <sup>-3</sup>	5.9 10 <sup>-3</sup>	3.2 10 <sup>-3</sup>	0.5	41.5 10 <sup>-3</sup>
12.	28.0 10 <sup>-3</sup>	2.7 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.2	15.3 10 <sup>-3</sup>
13.	10.4 10 <sup>-3</sup>	1.2 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.1	16.3 10 <sup>-3</sup>
14.	11.1 10 <sup>-3</sup>	0.5 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.3	17.6 10 <sup>-3</sup>
15.	20.9 10 <sup>-3</sup>	7.7 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.2	51.1 10 <sup>-3</sup>
16.	29.5 10 <sup>-3</sup>	5.8 10 <sup>-3</sup>	1.9 10 <sup>-3</sup>	0.1	59.6 10 <sup>-3</sup>
17.	13.6 10 <sup>-3</sup>	0.7 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.1	11.8 10 <sup>-3</sup>
18.	7.7 10 <sup>-3</sup>	0.8 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.1	9.6 10 <sup>-3</sup>
19.	7.0 10 <sup>-3</sup>	1.2 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.1	30.6 10 <sup>-3</sup>
20.	9.7 10 <sup>-3</sup>	3.5 10 <sup>-3</sup>	<10 <sup>-3</sup>	0.1	39.8 10 <sup>-3</sup>
21.	17.8 10 <sup>-3</sup>	<10 <sup>-3</sup>	<10 <sup>-3</sup>	<0.1	17.5 10 <sup>-3</sup>
22.	14.3 10 <sup>-3</sup>	1.2 10 <sup>-3</sup>	<10 <sup>-3</sup>	<0.1	15.3 10 <sup>-3</sup>

(1) maximum daily activities (30 minutes sampling period), (2) sampling on coal filter

\* NaJ(Tl) detector, well-type.

Table 1. Average daily air radioactivity in May 1986, Institute Vinca (Smiljanic et al., 1989)

In May 1986, only about 30% of the total iodine released in the atmosphere could be detected in the aerosols filters. Other 36% of elemental iodine and less active organic

compounds was detected by coal impregnated filter and the rest 34% of active organic compounds by active coal (Todorovic et al., 1996; Smiljanic et al., 1989; Andradi, 1986).

The data on the activity of plutonium in air in the period of intensive nuclear weapon tests in 1965-1969 are presented in Table 2. The results are in agreement with the reported data on plutonium in air on the same latitudes (Todorovic et al., 1996).

Year	Month	$^{239,240}\text{Pu}$	$^{238}\text{Pu}$
1965	March	$13.1 \pm 1.3^*$	$(0.5)^{***}$
	April	$9.7 \pm 1.3$	n.d.**
	April	$7.6 \pm 0.6$	$(0.2)^{***}$
	May	$16.7 \pm 1.1$	n.d.**
1967	July	$2.1 \pm 0.3$	$1.0 \pm 0.2$
	August	$1.6 \pm 0.4$	$0.9 \pm 0.4$
	October	$0.7 \pm 0.2$	$(0.3)^{***}$
	October	$0.8 \pm 0.7$	$(0.4)^{***}$
	November	$(0.3)^{***}$	$(0.1)^{***}$
	November	$(0.4)^{***}$	$(0.2)^{***}$
	December	$0.5 \pm 0.3$	$(0.3)^{***}$
	Jan.Feb.Mar.	$(1.5)^{***}$	n.d.**
	Apr.May.June	$2.9 \pm 0.6$	$1.0 \pm 0.14$
	July.Aug.Sep.	$2.2 \pm 0.3$	$1.1 \pm 0.2$
	Oct.Nov.Dec.	$1.5 \pm 0.4$	$0.7 \pm 0.4$
1969	Apr.May June	$1.5 \pm 0.4$	$0.6 \pm 0.4$
	Oct.Nov.Dec.	$0.8 \pm 0.2$	$(0.1)^{***}$

\* 95% uncertainty level, \*\* non detected, \*\*\* values with low uncertainty level due to short counting period about 24 hours or less

Table 2. Activity of  $^{239,240}\text{Pu}$  ( $\text{mBq}/\text{m}^3$ ) and  $^{238}\text{Pu}$  ( $\text{mBq}/\text{m}^3$ ) in air 1965-1969, Institute Vinca, Belgrade (Todorovic et al., 1996)

Daily activity of plutonium in air during nuclear accident at Chernobyl, from May, 1<sup>st</sup> -15<sup>th</sup>, 1986 were in the range of  $0.4 - 10.6 \times 10^{-6} \text{ Bq/m}^3$  for  $^{239,240}\text{Pu}$  and  $0.3 - 5.7 \times 10^{-6} \text{ Bq/m}^3$  for  $^{238}\text{Pu}$ , with a maximum on May the 5<sup>th</sup> (Todorovic et al., 1996).

#### 4.3 Air radioactivity monitoring at the Institute Vinca in 1991-2006

Monitoring of  $^7\text{Be}$  in ground level air in the Institute Vinca started in 1991. Average monthly concentrations of  $^7\text{Be}$  in air up to 2006 were in the range of  $2.0\text{-}7.0 \text{ mBq/m}^3$  and exhibited one/two maxima in summer/early autumn and a minimum in winter (Fig.4), corresponding to the values measured in Europe and elsewhere (Gaffney et al., 1994; Groundsel & Postendurfen, 2004; Hernandez et al., 2005; Ioanidou & Papastefanou, 1997; Iskihawa et al., 1995). The maxima were correlated with the increment of temperature, while the minimums were linearly correlated with precipitation. Sharp increase of  $^7\text{Be}$  concentrations in air in 2001 and 2003 was probably due to increased stratosphere-to-troposphere exchange (Hernandez et al., 2005; Todorovic et al., 1997; Todorovic et al., 2000; Todorovic et al., 2005). Concentrations of  $^{137}\text{Cs}$  in air in the same period were in the range of  $0.1 - 8.5 \times 10^{-5} \text{ Bq/m}^3$  with a maximum in spring/summers and one in winter due to local resuspension effects (Fig.5). We should note that since 1989,  $^{137}\text{Cs}$  concentrations were decreasing and in 1998 obtained the level before the Chernobyl accident (Todorovic et al., 2005; Todorovic et al., 2007). Activity of  $^{210}\text{Pb}$  in ground level air has been measured since 1985 and was in the range of  $0.1 - 31.7 \times 10^{-4} \text{ Bq/m}^3$ , with a maximum in early/late autumns (Fig.6) (Todorovic et al., 1997; Todorovic et al., 1999; Todorovic et al., 2000; Todorovic et al., 2002; Todorovic et al., 2005). This corresponds to the values reported by other authors (Arimoto et al., 1999; Duenas et al., 2004; Gaffney et al., 1994; Ionadiou & Papastefanou, 1997). Higher values of  $^{210}\text{Pb}$  measured in Belgrade air in some periods are probably due to the anthropogenic sources (heavy traffic run by leaded gasoline and city heating plants run by crude oil and coal). The maxima are due to increased radon emanation from soils (Todorovic et al., 2005).

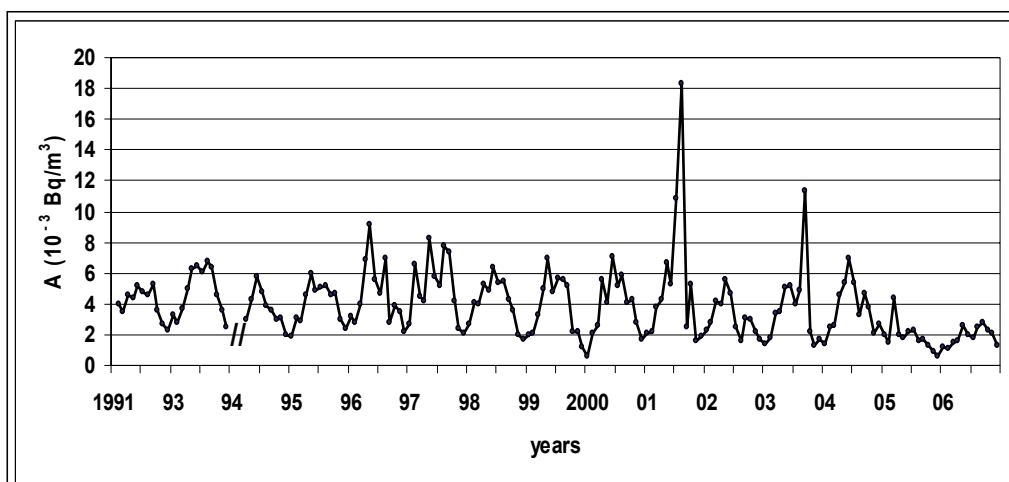


Fig. 4. Average monthly concentrations of  $^7\text{Be}$  in air (Institute Vinca, 1991-2006)



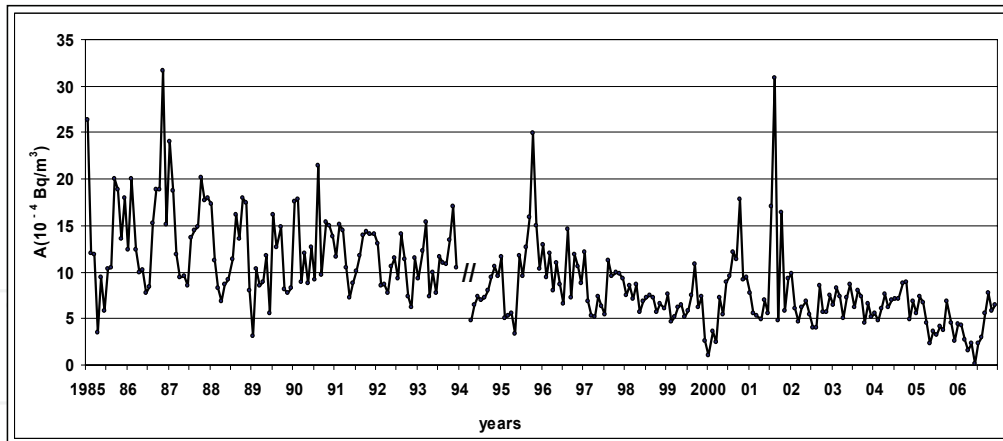


Fig. 5. Average monthly concentrations of  $^{210}\text{Pb}$  in air (Institute Vinca, 1985-2006)

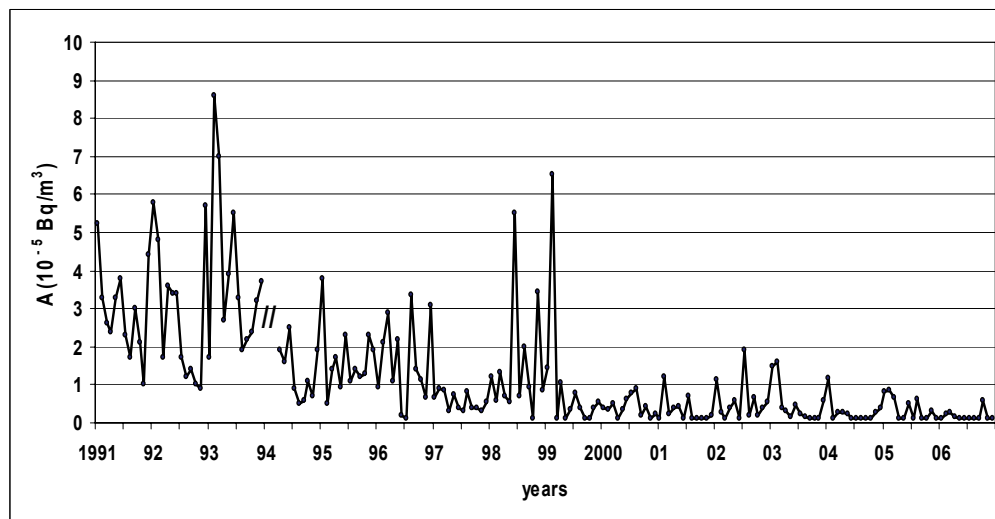


Fig. 6. Average monthly concentrations of  $^{137}\text{Cs}$  in air (Institute Vinca, 1991 -2006)

## 5. Air Radioactivity Monitoring Program in Belgrade Central City Area

Due to high population density, heavy traffic and industrial plants located in the outskirts urban areas especially central city areas are exposed to severe air pollution. In the last decade, air radioactivity monitoring in urban areas is a part of the pollution monitoring program in most of the European countries. In Serbia, there are many studies on the natural and anthropogenic radionuclides in ground level air but still there is no comprehensive database on the air radioactivity in the central city areas and the monitoring program started only recently (Manic et al., 2006; Popovic et al., 1996; Popovic et al., 1996a; Popovic et al., 2000; Todorovic et al., 2007; Popovic et al., 2000a).

### 5.1 Site, Materials and Methods

The city of Belgrade (44°47'N, 20°32'E; 205 m a.s.l) is located in West-Central Serbia on the junction of the rivers Sava and Danube and has about 2 millions inhabitants. The climate is moderate continental. In the last decades, severe air pollution in the form of aerosol smog occurred in the city center during winters due to increasingly heavy traffic and household

heating. Leaded gasoline is still in use and city heating plants and domestic heating plants use natural gas, crude oil and coal.

Aerosol for radionuclides study were collected on the representative "black spots" in very center of the city. Samples were collected on filter papers (FILTRAK/Whatman 41/DDR, 15 cm diameter, relative efficiency for deposited dust 80%) by constant flow rate samplers (average air flow 20 m<sup>3</sup>/h, average daily volume 600 m<sup>3</sup>), ashed at temperatures below 400°C and a monthly composite sample containing 30-31 daily filters was formed (average volume 15x10<sup>3</sup> m<sup>3</sup>). The samples were measured in small metallic containers.

The activity of the radionuclides was determined on 3 HPGe detectors (Canberra, Ortec relative efficiency 23%, 20%, 18% respectively, resolution 1.89 keV at 1332 keV) by standard gamma spectrometry. The two detectors (vertical, coaxial) were placed in a shielding cage of Pb bricks (width 10 cm) with layers of Cu (3 mm) and Fe (6 mm - 10mm). The third detector has the commercial shielding; it is a reverse-electrode Ge, with a thin Be window (Todorovic et al., 1994).

Energy calibration (100 keV-2000 keV) was performed with a set of standard point sources (Coffret d'etalon gamma ECGS-2, Sacle, France) containing <sup>133</sup>Ba, <sup>57,60</sup>Co and <sup>137</sup>Cs (10<sup>3</sup> Bq - 10<sup>4</sup> Bq, 25.11.1987). Geometric efficiency was determined with IAEA-083 (AIR4) stimulated air filter (spiked with solution of <sup>60</sup>Co: 2160 Bq/filter, <sup>133</sup>Ba: 846 Bq/filter, <sup>137</sup>Cs: 1182 Bq/filter and <sup>210</sup>Pb: 151 Bq/filter, uncertainty 5%, 1.1.86). Another reference radioactive material was also used: an aerosol powder matrix in plastic box (11 g, with <sup>137</sup>Cs: 207 Bq, <sup>54</sup>Mn: 14 Bq, <sup>65</sup>Zn: 16 Bq, <sup>57</sup>Co: 5 Bq, <sup>40</sup>K: 7 Bq; 09.03.1988) (ZND89). These are standard calibration procedures applied in air radioactivity monitoring in the Vinca Institute.

Radionuclides were determined at gamma energies 661.6 keV for <sup>137</sup>Cs, 477 keV for <sup>7</sup>Be and 46 keV for <sup>210</sup>Pb. Minimum detectable concentrations, were derived from the lower limit of detection as  $LLD = k^2 \pm 2LC$ , where  $k$  is the coefficient of normal distribution corresponding to the confidence level of 95%, while  $LC$  is the critical level depending on background photo peak counts. Minimum detectable concentrations (MDC) of the radionuclides in air were: 1.0 μBq/m<sup>3</sup> for <sup>137</sup>Cs, 10 μBq/m<sup>3</sup> for <sup>7</sup>Be and 20 μBq/m<sup>3</sup> for <sup>210</sup>Pb.

Counting time intervals were from 150 ks - 250 ks. Background spectrum integral mean count was 1.7 cps. Data were statistically analyzed on IBM/PS2 by SPECTRAN-AT and Genie 2000 programe. Total standard error of the method (including relative errors in geometric efficiency estimation, photopeak counts estimation, sample volume determination, etc.) was estimated below 25%.

Precipitation data for the period were obtained from the Department of Meteorology, Institute of Nuclear Sciences Vinca (DM Report 96-01).

## 5.2 Monitoring of <sup>137</sup>Cs, <sup>7</sup>Be and <sup>210</sup>Pb in Air in Belgrade Central City Area

Monitoring program on radionuclides in ground level air in Belgrade central city area started in 2002. The concentrations of radionuclides in air on different sites within the city differed significantly (45% for <sup>7</sup>Be and <sup>210</sup>Pb and about 80% for <sup>137</sup>Cs), but the overall seasonal variations pattern was similar to one obtained at Institute Vinca, outside the city. The average concentration values were (0.4 - 13.5) x10<sup>-3</sup> Bq/m<sup>3</sup> for <sup>7</sup>Be, (0.1 - 1.50) x10<sup>-5</sup> Bq/m<sup>3</sup> for <sup>137</sup>Cs and (0.1 - 11.0)x10<sup>-4</sup> Bq/m<sup>3</sup> for <sup>210</sup>Pb. The lowest concentrations were measured at sites exposed to good ventilation, while higher ones were measured at sites with dense vegetation and poor ventilation. Since 2004, monitoring of air radioactivity continued on a site with heavy traffic and one of the most polluted areas in the city (Popovic

et al.,2000a; Todorovic et al., 2002; Todorovic et al., 2005; Todorovic et al., 2007; Todorovic et al. 2005a).

The average monthly concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in air measured at the site in central city area, in 2004-2006 are presented in Fig.7 and Fig.8. The results for  $^{137}\text{Cs}$  concentrations in air are not presented, as more than 80% of the values were below the lower limit of detection that was  $1 \times 10^{-6} \text{ Bq/m}^3$  (Todorovic et al., 2007).

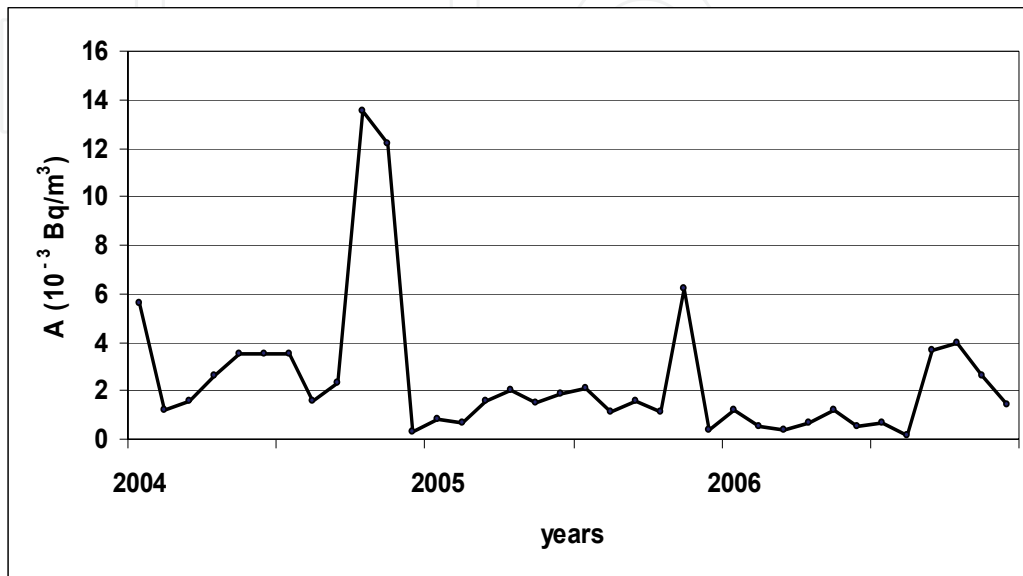


Fig. 7. Average monthly concentrations of  $^7\text{Be}$  in Belgrade air (2004-2006) (Todorovic et al.,2007)

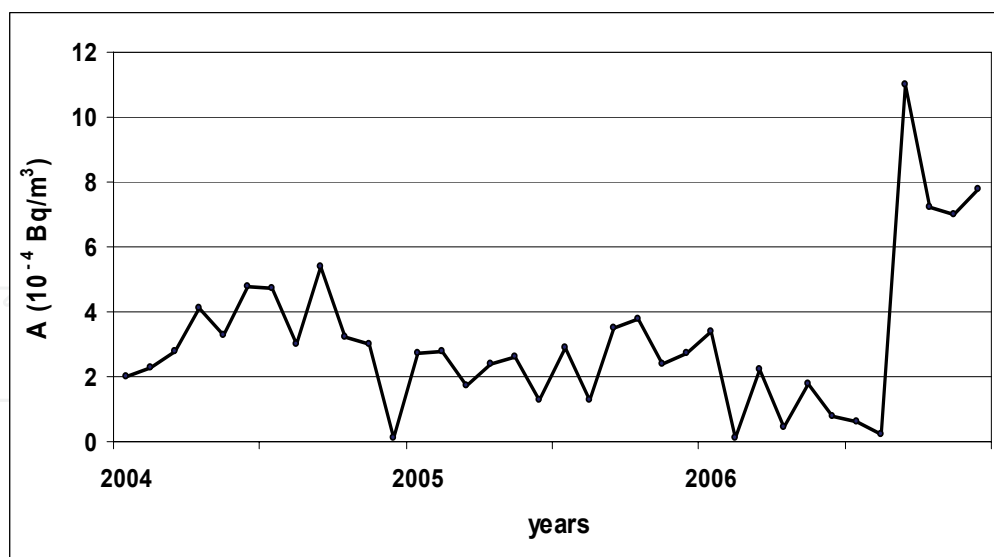


Fig. 8. Average monthly concentrations of  $^{210}\text{Pb}$  in Belgrade air (2004-2006) (Todorovic et al., 2007)

The concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  measured in air in 2004- 2006 in Belgrade center city area were in the range of values measured in the same period at Institute Vinca, but the pattern of seasonal variations was different. This could be attributed to local climate conditions and

short sampling period. Both  $^7\text{Be}$  and  $^{210}\text{Pb}$  exhibited seasonal variations with maxima in (late) autumns. The pattern of  $^7\text{Be}$  variations is more pronounced than the one of  $^{210}\text{Pb}$  variations, what is probably due to the vicinity of the highway and, apart from the different origin of the radionuclides, to the short period of sampling. Concentrations of both radionuclides are very low in winters, as the result of precipitation and snow coverage effects (Popovic et al., 2000; Todorovic et al., 2007). Extremely low values of radionuclides concentrations in air are often due to technical problems (deposition of dust within the pumping system, air flow suffocation, etc), but in general sampling procedure is the main cause of errors in air radioactivity monitoring and control.

## 6. Conclusion

It can be concluded that monitoring natural and anthropogenic radionuclides in ground level air provide important information on the content of radionuclides due to their origin, weather and climate conditions (rate of precipitation, washout effects, temperature differences and inversion, wind direction and intensity, troposphere-to-stratosphere transport and exchange, etc). The local topology, as well as anthropogenic factors such as the effects of traffic and heating plants are highly important, especially when monitoring radionuclides in air in urban areas. Sampling procedures, adequate measuring techniques and calibration are essential for providing reliable results and data. National and international standards regarding sampling sites and measuring laboratories network are to be precisely applied so as to provide high quality assurance and quality control in the air radioactivity monitoring system.

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