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VARIATIONS OF PM_{10} MASS CONCENTRATIONS AND CORRELATIONS WITH OTHER POLLUTANTS IN BELGRADE URBAN AREA*

In this paper, we present the PM_{10} levels measured at an urban residential background site in New Belgrade, in Omladinskih Brigada Street, at 15 m height (roof). The aerosol samples were collected using a European standard sampler, in four seasonal campaigns conducted in autumn: Nov. 13-Dec. 03, 2007, winter: Feb. 07-28, 2008, spring: May 06-28, 2008 and summer: July 17--August 15, 2008. The results were compared with PM_{10} mass concentrations measured with a Horiba automatic station at street level, at the same sampling site and at three more sites within the Belgrade municipal monitoring network. The results show that the PM_{10} values in the Belgrade urban area were high during autumn and winter campaigns (heating season) with a number of samples exceeding the 24-h limit value of 50 $\mu g\ m^{-3}$. On the roof station, a maximum daily value of 209 $\mu g\ m^{-3}$ was measured in the autumn campaign, with 14 values out of 20 measurements exceeding the 24-h limit. In winter, 14 out of 19 measurements exceeded the limit, with a maximum value of 196 $\mu g\ m^{-3}$. During the spring campaign, the number of exceedances was three out of 22. All values during the summer campaign were below 50 $\mu g\ m^{-3}$. The roof station equipped with the European Standard instrument showed systematically higher values than the street-level automatic monitor. PM_{10} values at all sites followed the same trend. The highest concentrations at all monitoring sites were observed during the autumn, Nov. 20-25, 2007, and winter, Feb. 19-23, 2008.

Key words: PM_{10}; urban air pollution; monitoring; gravimetric measurements.

Airborne particulate matter in urban air has been in the center of recent concerns, especially due to its adverse effects on human health. Many epidemiologic studies underlined the health significance of respirable particles, PM_{10} (particles with aerodynamic diameter smaller than 10μm) [1]. These studies have demonstrated that acute and chronic health effects are related to the inhalable PM_{10} exposure in the urban environment [2,3]. The main adverse health effects were evident during episodes of extremely high levels of air pollution which moved the authorities to initiate extensive air pollution control contributing to the establishment and adoption of regulations for air quality improvement [4].

Particulate matter levels in urban areas are influenced by several factors: human activities, urban geometry, meteorological conditions and regional and long-range transport of pollutants. Main sources of suspended particles in urban areas are traffic, local heating, re-suspended dust from the roads and industrial activities. Large point sources such as power plants may also influence the urban air quality, with high contribution in specific episodes, and adding to the background levels. Meteorological conditions, such as wind speed, wind direction, temperature, relative humidity and atmospheric stability, play an important role in dispersion of PM in the air.

Atmospheric particulate matter is a key parameter in air quality management throughout the world. According to the Directive 1999/30/EC [5], the Member States are required to reduce the emission of particles in urban areas in order to ensure that the health--based concentrations of PM_{10} do not exceed limit va-

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lues. This Directive has recently been replaced by a new European Directive 2008/50 [6], but limit values for PM_{10} remained the same. The first health-based limit value is set at 50 μg m^{-3} as a daily average (may be exceeded up to 35 days a year) and the second is set 40 μg m^{-3} as an annual average.

The regulations on EU level have led to significant improvements in the EU (WHO, 2002) [7]. Their implementation in Serbia is, however, not complete so far. New methodology for sampling and analysis is to be adopted and established within the existing air quality monitoring network [8].

Since 2002, the suspended particles PM_{10} and $PM_{2.5}$ in aerosols and their physical and chemical characterization were subject of several research projects in the Belgrade urban area. The first air quality assessment included attempts to specify the emission sources using statistical methods and modeling, and to assess the impact of meteorological conditions. The main pollution sources were identified to be traffic and local heating units [9]. The average PM_{10} biannual concentrations were 68.4 μg m^{-3}. PM_{10} levels and levels of CO, NO, NO_2 and SO_2 showed high correlation.

During the autumn of 2005, behavior of different pollutants, O_3, SO_2, NO_2, CO and PM_{10}, and the impact of meteorological conditions were further investigated in the Belgrade urban area [10]. The maximal O_3 concentrations were recorded in the E wind direction, while the highest level of other pollutants, CO, NO, NO_2, PM_{10} were connected with NW, N and SW direction.

The objectives of the current study are to establish quality assurance and quality control according to sampling reference method defined by EU Directive 2008/50, and to compare PM_{10} concentrations with data from the monitoring network. One of the goals of the project is to introduce and establish standard operating procedures and quality assurance procedures in all stages of aerosol management and analysis, to use advanced analytical techniques for PM chemical characterization and finalize the research with source apportionment analysis [11]. This analysis will also contribute insight into the present situation in the Belgrade urban area, concerning the concentration levels of other gas-phase ambient air pollutants, such as SO_2, NO_x, O_3 and PM_{10} (obtained as a data from automatic monitoring network) and their correlation with PM_{10} aerosol fraction.

EXPERIMENTAL

Aerosol sampling

The sampling campaigns were performed at an urban background residential area site, in the fast developing New Belgrade, during four sampling periods: autumn campaign from November 13th to December 3th 2007, winter campaign from February 8th to February 27th 2008, spring campaign from May 6th to May 27th 2008, and summer campaign from July 17th to August 14th 2008.

The sampling site is located in Omladinskih Brigada Street, approximately 5 km from the city centre. It is placed on the roof of Medical Institute at a height of about 15 m. This position can be considered representative of average air pollution in the investigated area. An automatic station of the municipal monitoring network is situated at street level in front of the same building, and provides SO_2, NO_2, O_3 and PM_{10} data.

Aerosol sampling was conducted using a European reference low-volume sampler (Sven/Leckel LVS3) with an inlet for PM_{10} fraction collecting, with flow rate 2.3 m^3 h^{-1}. Suspended particles were collected onto 47 mm Whatman QM-A double-sided quartz fiber filters. The PM_{10} aerosol fractions were sampled on a daily basis (24 h, beginning at 7 am) with one “field blank” per week, according to the sampling reference method defined by EU Directive 1999/30/EC [6].

The automatic air monitoring station with Horiba monitors: APSA-360, APNA-360, APOA-360 for SO_2, NO_2 and O_3 measurements, respectively, and Eberline FH-62I/R air particulate monitor for PM_{10} mass concentration measurements, was placed 2 m above the ground in front of the building. The hourly readings of SO_2, NO_2, O_3 and PM_{10} concentrations were recalculated into average daily values and were used for comparison.

Gravimetric analysis

Gravimetric measurements were conducted according to EN 12341 for PM_{10} [12]. Filters were exposed in opened Petri-slides for 48 h at 20±1 °C temperature and 50±5% relative humidity in a Class 100 clean room with automatic temperature/pressure regulation. After preconditioning, the filters were weighed twice using a Precisa XR 125 SB micro-balance with 0.01 mg mass resolution and mass concentrations were calculated as average values. Two pre-fired blank quartz filters were exposed in the conditioning room all the time and their mass checked after each series of eight sample measurements. Before and after each weighing session, certified test weights of 100 and 200 mg were used for micro-balance accuracy control. Following the gravimetric measurements, the exposed filters were stored in a cool room at 4 °C until analysis.

RESULTS AND DISCUSSION

The map of Belgrade urban area with the locations of the four monitoring stations in Omladinskih Brigada Street, Gradski zavod za javno zdravlje (GZJZ), Jerneja Kopitara and Obrenovac is presented in Figure 1.

Average monthly PM_{10} mass concentrations during 2007 and 2008 at four automatic monitoring stations within the air quality monitoring network in the Belgrade urban area are given in Table 1. The monitoring station in Jerneja Kopitara is a background residential station similar in location to the station in Omladinskih Brigada Street. The monitoring station GZJZ is a typical street canyon station with traffic as a dominant pollution source. Obrenovac monitoring station is situated in the vicinity of the Obrenovac power plant at a 40 km distance from Belgrade. All intakes of the monitoring network were at the 2 m level.

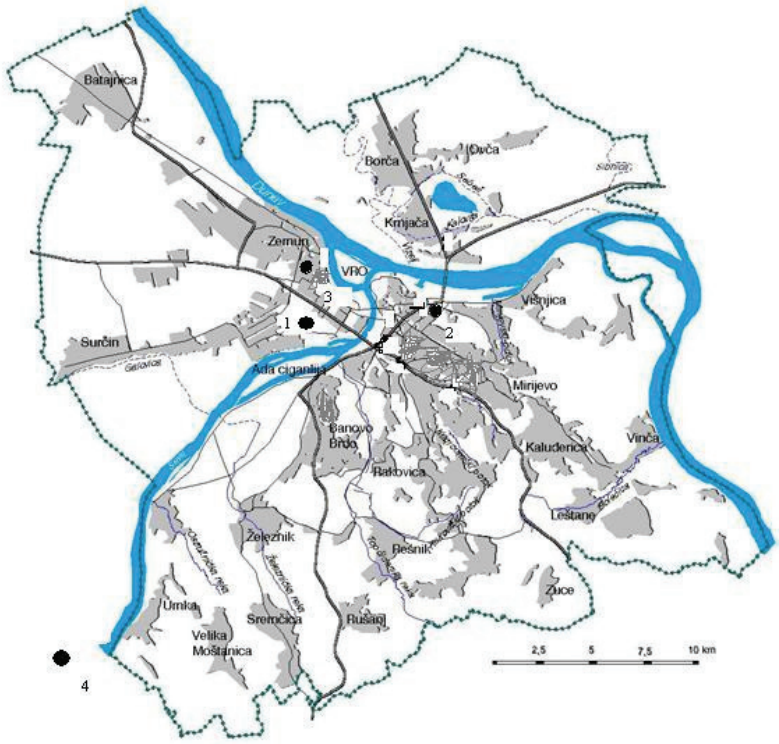

Figure 1. Belgrade urban area with the location of the monitoring stations: 1. Omladinskih Brigada, 2. Gradski zavod za javno zdravlje, 3. Jerneja Kopitara and 4. Obrenovac.

Table 1. Average monthly and annual PM_{10} mass concentrations ($\mu g/m^3$) at four monitoring stations in Belgrade during 2007 and 2008

Month	Omladinskih brigada		Obrenovac		Gradski zavod za javno zdravlje		Jerneja Kopitara	
	2007	2008	2007	2008	2007	2008	2007	2008
Jan.	-	56.0	43.1	60.0	73.4	76.7	-	94.2
Feb.	-	73.2	34.7	52.9	50.4	73.2	-	91.8
Mar.	-	30.9	33.5	26.3	40.9	34.0	32.9	39.3
Apr.	38.4	29.5	40.8	31.5	42.8	33.8	64.5	35.9
May	27.8	22.2	25.8	33.4	38.3	37.1	32.1	38.1
June	36.1	27.8	32.9	26.3	42.8	29.5	37.8	31.8
July	35.3	32.7	35.2	25.7	35.7	25.2	44.5	25.8
Aug.	34.0	28.5	32.7	31.4	36.0	31.4	37.0	31.6
Sept.	36.3	25.7	29.6	30.2	36.8	34.3	35.5	34.6
Oct.	51.4	45.3	41.6	53.7	53.8	52.5	54.2	62.6
Nov.	54.7	41.8	50.4	48.7	61.2	48.9	76.4	60.2
Dec.	60.6	25.9	52.1	26.4	64.3	37.7	76.1	34.0
Year	41.6	36.6	37.7	37.2	48.0	42.9	49.1	48.3

Mass concentrations of PM_{10} fraction, obtained daily in four seasonal campaigns during 2007/2008 in our study, show significant variations from day to day, often exceeding 50 μg m^{-3} during the autumn (14 out of 20 values, for PM_{10}) and winter (14 out of 19 values, for PM_{10}). The concentrations in the spring and summer campaigns conducted during non-heating seasons, show less exceedances of PM_{10} (3 out of 22 values in spring and all values during summer were below 50 μg m^{-3}). This is a direct result of a combination of higher emission strengths of most sources in the heating season, and atmospheric conditions (inversion layers, low temperature, air mass stagnation), and is consistent with other European data [13-15].

The concentrations of PM_{10} at all the stations are highly correlated, with the lowest correlation being between the street canyon GZZZ station and the power plant Obrenovac (0.78), the GZZZ being mostly influenced by local traffic that is not so important at Obrenovac. The highest correlation (0.98) was between the roof and the street level concentrations at Omladinskih Brigada. The roof station shows systematically higher concentrations than the street level monitor, with an average of 12 μg m^{-3}. This may be due to the differences in placement, and also due to differences in instrumentation, and should be further investigated. However, it should be noticed that these results are only indicative and should be treated with caution during policy evaluation and development, as the instruments were not intercalibrated prior to the experiment. Those instruments are calibrated separately (not in the field measurements).

For collecting meteorological data, a PMP 124A portable meteorological station (AMES) was mounted on the roof of Medical Institute at a height of about 15 m together with a PM_{10} low-volume sampler. PM_{10} concentrations during autumn and winter campaigns (Figure 2a) were the highest when the prevailing wind direction was south-west (SW). The possible explanation could be the impact of coal fired power plants (Nikola Tesla A and B) situated 30 km south-west from Belgrade. The other important prevailing wind direction concerning maximal PM_{10} concentrations was E-ESE, in the direction of the steel industry complex in Smederevo (40 km from Belgrade).

In the spring and summer campaigns, correlation between PM_{10} concentration (that are several times lower than autumn and winter concentrations) and prevailing wind direction is different (Figure 2b). There are four prevailing wind directions during the non-heating period: north (N), west (W), SW-SSW and ESE instead of only two (SW-SSW and ESE) during the heating period.

Table 2 shows the average PM_{10} concentrations per three week period (duration of seasonal campaign) at site Webiopatr-Omladinskih Brigada (with Leckel samplers) and four automatic monitoring sites (with Horiba automatic station) for PM_{10}: Omladinskih Brigada, GZJZ, Jerneja Kopitara and Obrenovac. Table 2 shows high PM_{10} concentrations in a range of 81-111 μg m^{-3} (during autumn campaign), 74-98 μg m^{-3} (during winter campaign), 24-42 μg m^{-3} (during spring campaign) and 28-32 μg m^{-3} (during summer campaign) in the Belgrade urban area. The PM_{10} concen-

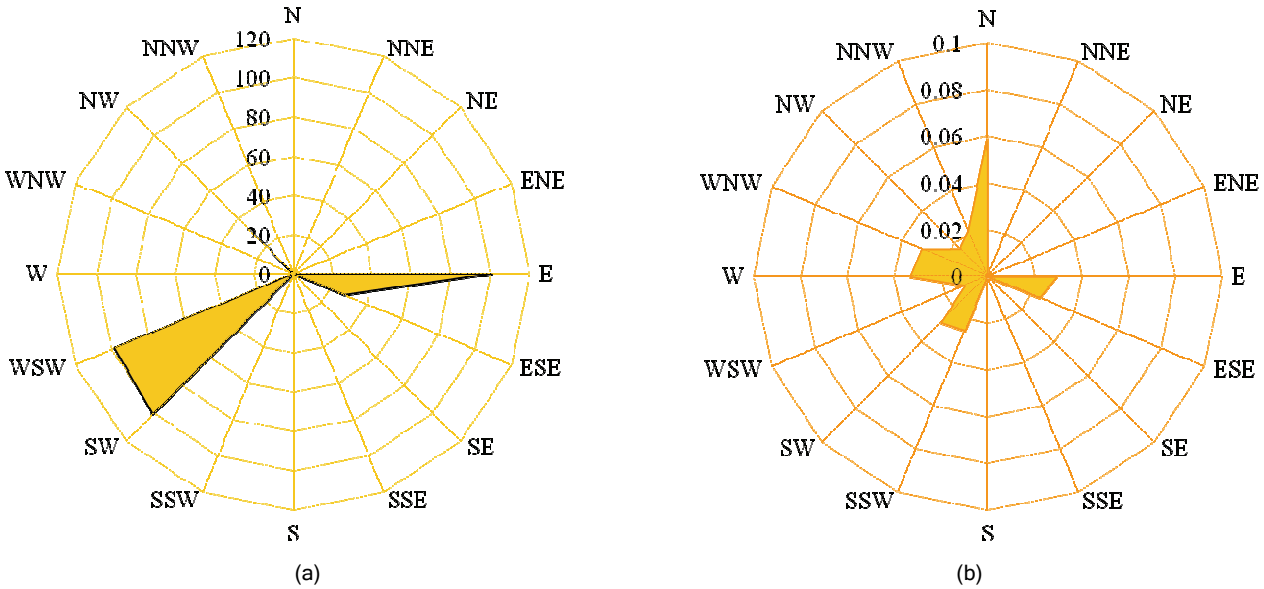

Figure 2. a) Average PM_{10} daily concentrations for prevailing wind direction during a) heating period (autumn and winter campaigns) and b) non/heating period (spring and summer campaigns).

trations at Obrenovac monitoring site were 68, 54, 33 and 29 μg m^{-3} during autumn, winter, spring and summer campaign, respectively. During autumn and winter campaigns the average relative humidity was higher (76.4 and 64.4 %) than in the spring and summer periods (56.6 and 59.0%). Average temperatures were 4.3, 5.2, 19.4 and 23.9 °C in the autumn, winter, spring and summer campaign, respectively. These seasonal differences in PM_{10} mass concentrations could be linked with the operation of central heating and with different meteorological conditions that prevail during different seasons. As reported above, higher PM_{10} concentrations are observed during the more humid period of the year, a fact that could be attributed to the production of secondary aerosols that is enhanced in humid environments. In a previous study that also refers to the Belgrade urban area, a seasonal variation in dust levels was also detected. Higher PM_{10} and $PM_{2.5}$ concentrations were recorded during the heating period and were linked to more intense traffic, combustion processes and meteorological conditions such as temperature inversions and stagnation of air masses [15].

The PM_{10} concentrations exceed 50 μg m^{-3} at all monitoring sites during the autumn and winter period [9,16]. They are extremely high during autumn/winter campaign especially at Omladinskih Brigada and Jerneja Kopitara sites. The close similarity in PM_{10} values for Omladinskih Brigada and Jerneja Kopitara can be explained by proximity of the sites and similar characteristics of their location.

The PM_{10} annual concentrations in Belgrade (Table 3) and some other European cities [17-21] (Rome and Athens) exceed the annual limit value of 40 μg m^{-3} [6]. The annual mean PM_{10} concentration in urban air in Belgrade for 2004-2006 (50 μg m^{-3}) [22] was within the range of some European cities (29.5--67.8 μg m^{-3}) with similar population density and significant traffic density.

Correlations between the PM_{10} levels and concentrations of gaseous pollutants at the Omladinskih Brigada are shown in Figures 3 and 4. The correlations between PM_{10} mass concentrations from the roof site (Webiopatr) at the and gas-phase ambient air pollutants SO_2 and NO_x, O_3 and $*PM_{10}$ daily concentrations calculated from hourly readings of the automatic air monitoring station at street level at the same site are illustrated for the first two campaigns (autumn and winter) and the last two campaigns (spring and summer) in Figures 3 and 4.

A similar trend between parameters presented in Figures 3 and 4 may be noticed. Corresponding Pearson's coefficients of regression R^2 for NO_x, SO_2, and $*PM_{10}$ related to PM_{10} measured on the roof, were: 0.88; 0.67 and 0.97 in autumn and 0.80; 0.77 and 0.88 in winter sampling period, respectively. O_3 concentration was not available at the monitoring site during autumn and winter periods.

Pearson's coefficients of regression, R^2, for NO_x, SO_2, O_3 and $*PM_{10}$ at street level related to PM_{10} at the roof level were: 0.60; 0.46; 0.07 and 0.79 in the spring and 0.40; 0.53; 0.29 and 0.61 in the summer sampling period, respectively. The lack of correlation between O_3 concentration and PM_{10} concentration was expected and already published for the Belgrade urban area [10]. The lowest values of all pollutants were observed during the spring campaign.

CONCLUSION

There are significant seasonal variations in PM_{10} mass concentration values between the heating (autumn and winter) and non-heating (spring and summer) periods during 2007/08. Correlations of PM_{10} mass concentrations obtained by the reference instrument located at the rooftop with concentrations of gas-phase ambient air pollutants SO_2, NO_2, O_3 and mass concentration of $*PM_{10}$ obtained by automatic

Table 2. Average PM_{10} concentrations during measuring campaigns

PM_{10} Concentration, μg m^{-3}	Autumn	Winter	Spring	Summer
Webiopatr-Omladinskih Brigada	95	89	40	32
Omladinskih Brigada	76	74	24	28
Gradski zavod za javno zdravlje	81	78	37	29
Jerneja Kopitara	111	98	42	30
Obrenovac	68	54	33	29

Table 3. Annual mean PM_{10} concentrations in urban air in Belgrade and some European cities

	City							
	Belgrade	Vienna	Ljubljana	Rome	Prague	Athens	Madrid	Lisbon
Year	2004-2006	2002	2001	2004	2001	2001	2002	2002
PM_{10} Concentration, μg m^{-3}	50.0	30	29.5	61.4	34	67.8	33.3	32

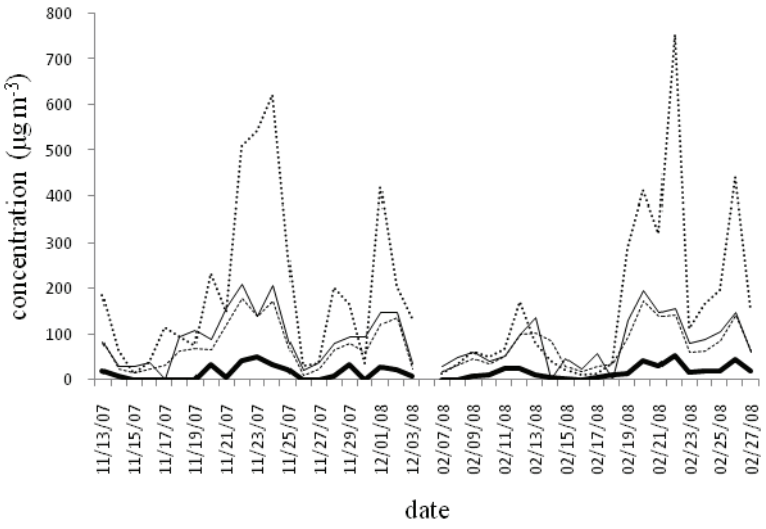

*Figure 3. Correlations between concentrations of PM_{10} (—) determined in this work and concentrations of NO_x (--), SO_2 (▬) and *PM_{10} (·····) obtained at automatic air-monitoring station during autumn and winter campaigns at Omladinskih Brigada sampling site.*

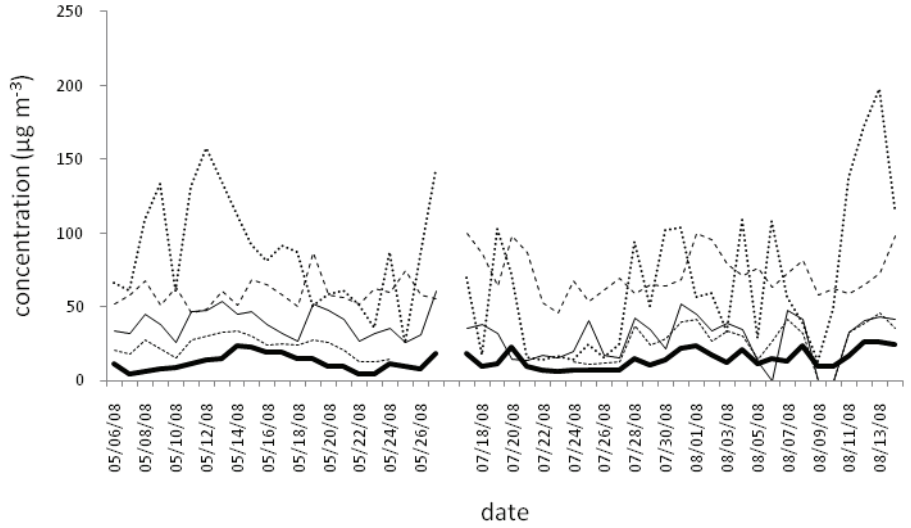

*Figure 4. Correlations between concentrations of PM_{10} (—) determined in this work and concentrations of NO_x (--), SO_2 (▬) and *PM_{10} (·····) obtained at automatic air-monitoring station during spring and summer campaigns at Omladinskih Brigada sampling site.*

air-monitoring station at street level have shown the same trend during the campaigns, with the lowest values of all air pollutants concentrations in the spring period. There was a significant systematic difference between the rooftop and the street level instrument, possibly attributable both to placement and instrumentation differences, but a formal comparison of instrumentation was not done in this work.

The mass concentration of PM_{10} aerosol fractions obtained here agree with previous results on the air quality in Belgrade urban area and with other results in the region, showing higher concentration levels in the autumn and winter, with a significant number of exceedances of the 24-h limit value.

PM_{10} concentrations during the autumn and winter campaigns were the highest when prevailing wind direction was south-west (SW) in direction of coal fired power plants and E-ESE, in the direction of steel industry complex in Smederevo while spring and summer prevailing wind directions were: north (N), west (W), SW-SSW and ESE.

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NAUČNI RAD

VARIJACIJE PM_{10} KONCENTRACIJA I NJIHOVA KORELACIJA SA KONCENTRACIJAMA OSTALIH ZAGAĐIVAČA U URBANOJ SREDINI U BEOGRADU

U ovom radu su prezentovani rezultati merenja PM_{10} na urbanom background *mernom mestu u stambenoj zoni u ulici Omladinskih Brigada u Novom Beogradu, koje je smešteno na krovu zgrade, na 15 m visine. Za uzorkovanje tokom četiri merne kampanje (jesenja od 13.11.2007. do 03.12.2007., zimska od 08.12.2008. do 27.02.2008., prolećna od 06.05.2008. do 27.05.2008. i letnja od 17.07.2008. do 14.08.2008.) su korišćeni referentni uzorkivači u skladu sa Evropskim standardom. Ovi rezultati su zatim upoređeni sa PM_{10} koncentracijom izmerenom Horiba automatskom stanicom na istom mernom mestu u nivou zemlje kao i sa koncentracijama iz još tri automatske stanice iz mreže redovnog gradskog monitoringa. Rezultati pokazuju da su PM_{10} vrednosti u urbanom vazduhu u Beogradu bile visoke tokom jesenjeg i zimskog perioda (grejna sezona) sa velikim brojem vrednosti koje prevazilaze graničnu vrednost od 50 μg m^{-3}. Na stanici na krovu zgrade, maksimalna dnevna koncentracija tokom jesenje kampanje iznosi 209 μg m^{-3} pri čemu 14 od 20 vrednosti prevazilazi granični 24-h limit. Tokom zimske kampanje, 14 od 19 vrednosti prevazilazi granični limit od 50 μg m^{-3} sa maksimalnom vrednošću od 196 μg m^{-3}. U toku prolećnog perioda uzorkovanja samo 3 vrednosti od ukupno 22 su premašile dozvoljenu graničnu vrednost dok su sve vrednosti tokom letnje kampanje su bile ispod graničnog limita od 50 μg m^{-3}. Stanica koja se nalazi na krovu zgrade i koja koristi referentne standardne uzorkivače pokazuje veće vrednosti u odnosu na stanicu smeštenu u nivou zemlje na istom mernom mestu koja koristi automatski monitor za merenje. PM_{10} masene koncentracije na svim mernim mestima imaju isti trend. Najviše masene koncentracije na svim mernim mestima su izmerene tokom periodaod 20-25 novembra 2007. u jesenjem periodu uzorkovanja i od 19-23 februara 2008. tokom zimske kampanje.*

Ključne reči: PM_{10}; urbano zagađenje vazduha; monitoring; gravimetrijska merenja.