Assessment of Concentration Levels of Particulate Matters (PM_{10}, TSP and BS) in the Area of Zrenjanin, Vojvodina, Serbia

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Concentration level of PM_{10}, total suspended particles and black smoke in the period from 2005 – 2007 has been analyzed in the medium-sized city of Zrenjanin. The research included analysis, mathematical regression and comparison of daily concentrations of PM_{10}, TSP and BS and seasonal variation, with evaluation of the influence of meteorological parameters. Dominant source of particle pollution in Zrenjanin has been identified. Research includes correlation between BS and TSP on one side, and PM_{10} on the other side, as basis for potential calculation of PM_{10} in other cities, which have data only on BS. Studies of particle pollution in settlements of Vojvodina are scarce. This analysis has been conducted for the first time on the territory of AP Vojvodina. The results of the study support the scenario, that traditional, individual heating in settlements may represent an important factor of air pollution.

Keywords: PM_{10}, black smoke, total suspended particles, meteorological parameters

Particulate matters (PM) consist of mixture of solid and liquid fine particles of organic and inorganic substances, suspended in air, and, as complex system, negatively affect human organism, more than any other pollutants. The size, chemical composition, and atmospheric lifetime, depend on the particulate forming processes. Basic components of particulate matters are sulfates, nitrates, ammonium, sodium-chloride, carbon, mineral dust and water. PMs are present in the air in various shapes and sizes, ranging from less than 10 nanometers to 10 micrometers in aerodynamic diameter (ad) [19]. These diameters represent the continuum from a few molecules, up to the size where particles can no longer be carried by a gas. Concentration level of PM in ambient air is, mainly, quantified through measurement of concentration of PM_{10} (particles with ad<10μm) or $PM_{2.5}$ (ad<2.5μm). $PM_{2.5}$ is strongly correlated with PM_{10}, since $PM_{2.5}$ in background air, in a medium-sized city, such as Zrenjanin, is likely to be within potential future air quality objectives [8].

PM, through inhalation, penetrate in the human organism and are being deposited in the respiratory system. Penetration and deposition of particles in human respiratory system depend on the size and shape of particles and defense mechanism of the respiratory system. Due to the size of the airborne particle they penetrate in the deepest part of the lungs ($PM_{2.5}$ – respirable fraction, PM_{10} – thoracic fraction).

In the late '70s and early '80s, results showed that exposure to high concentrations of particulate matters may negatively affect human health. Studies conducted in the mid '80s showed that longer exposure to particulate matters increases the risk of respiratory system diseases, changes in the lung function and other negative health effects. Newer studies are focused on health effects caused by short-term and acute exposure to particulate matters. Both studies point out on the fact that chronic and acute exposure to particulate matters cause significant effects on human health, even though toxicological mechanism for these effects are less familiar [9].

Considering the fact that suspended matters substantially contribute to degradation of the quality of ambient air in the urban environment, the possibility to predict the concentration of particulate matters is significant, in order to raise public awareness, as well as to manage the quality of ambient air [12].

Systematic measurement of concentration of PM_{10}, on the territory of AP Vojvodina (area of 21.506 km^2), still, does not exist. The reasons for undeveloped network for PM monitoring in Serbia are severe economic crisis during '90s due to political instability and international isolation of Serbia during recent conflicts in the region.

The aim of the research is to define the source, with dominant influence on the concentration levels of PM_{10}, and to recognize mutual functional correlation, based on available data from preliminary information systems of Autonomous Province of Vojvodina, in order to project concentration of PM_{10} in other cities, which have data available only for BS for long period of time.

Since BS can be considered as surrogate for concentrations of PM [3], analysis and evaluation with mathematical modeling of concentrations of particle pollutants of various sizes of radius (BS, TSP and PM_{10}) in Zrenjanin area was conducted in this research.

Legal framework for ambient air quality monitoring and limit values in Serbia

Each country has its own standards for evaluation of the air pollution. In Europe, EU standards are in power for EU member countries, then NAAQS (National Ambient Air Quality Standards) in USA, or WHO-AQGs [5]. Legal framework in the Republic of Serbia is defined through the Law on Environmental Protection ("Official Gazette RS", no. 135/04). The Law regulates integral system of

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environmental protection, which ensures the human right to live and grow in healthy environment as well as balance between economic development and preservation of the environment in the Republic of Serbia. Due to the fact that the area and section of ambient air protection was not completely regulated in Serbian legislation by the time this paper was prepared, limit values (LV) defined in the old Serbian Law have been used. Basis for air quality monitoring, and its protection from pollution was defined within Bylaws (Articles 18.-23.) of the old Law on environmental protection ("Official Gazette RS", no. 66/91, 83/92, 53/93, 67/93, 48/94, 53/95). Other bylaws and regulations within this field were:

- Bylaw on determination of Air Quality Monitoring Programme, adopted by the Government for the period of 2 years, and defines the network of stations for ambient air quality control (basic network of meteorological and urban stations), the exact number of monitoring sites by settlements and the type of monitored pollutants;
- Regulation on limit values, methods for measurements of ambient air quality, criteria for establishment of monitoring sites and data collection ("Official Gazette RS", no. 54/92, 30/99, and 19/2006);
- Regulation on limit values of emission, types and deadlines for measurements, and data collection ("Official Gazette RS", no. 30/97, 35/97).

New Law on Ambient air protection ("Official Gazette RS", no. 36/09) and Bylaw on monitoring conditions and air quality requirements ("Official Gazette RS", no. 11/10) have been adopted in the meantime introducing new limit values in accordance with EU requirements.

Air Quality Guideline (World Health Organization) has set the new guideline for particulate matters, aiming to achieve the lowest possible concentrations.

Table 1 gives overview of limit values for concentrations of $PM_{2.5}$, PM_{10}, BS and TSP defined by Regulation of RS, EU Directive and WHO recommendation.

On the territory of Vojvodina (north part of Serbia, [11] bordering Hungary, Romania, Croatia), monitoring of the concentration of various pollutants in ambient air has been conducted occasionally and unsystematically during the past years.

Monitoring site

Zrenjanin is a city and municipality, located in central part of Vojvodina province. It is the administrative center of the Central Banat District of Serbia.

Zrenjanin is located on the western side of Banat's loess plateau, on the confluence of channeled river Begej into former river bed of Tisa river. The area terrain of the municipality is part of the Pannonian plain. City of Zrenjanin lies on 20°23' eastern longitude and 45°23' northern latitude, in the center of the Serbian part of the Banat region, on the river banks of Begej and Tisa. Altitude of the Zrenjanin is 80 m, and on the territory of the city it ranges from 77 – 97 m.

According to the Nation Census (2002), the city population was 79.773, while the Zrenjanin municipality had 132.051 inhabitants.

This locality was selected for this study, because Zrenjanin is a typical city for this region. It is the third biggest city in the region, and located in the central part of Vojvodina province, typical agricultural area. It represents developing city with constant population increase, and redevelopment of industry, devastated significantly during '90's crisis. Zrenjanin, also, represents the city with typical structure of heating in the region. On its territory, around 26.500 (92.3 %) households, partly in domestic heating (19.000 households), and partly through remote heating system (7.500 households), use natural gas, as main energy product.

Available data, gained by measurements of concentrations of PM_{10}, BS and TSP on two monitoring sites in Zrenjanin, located next to each other, on approximately 200 m distance have been used in this paper. Monitoring site (1) used for measurements of concentration of PM_{10} and meteorological parameters was in vicinity of heavy-traffic street. On the north side there were maximum three-story buildings, while on southeastern side, on approximately 1 km distance, there were radiator production plant, meat processing plant and power plant, and about 500 m on south, there was corn processing industry (fig. 1). Monitoring site (2), used for measurement of concentrations of BS and TSP was located in the surrounding of ground residential objects on the north, and park on the east side, while on the south and west side, it was surrounded by, maximum three-story objects.

Physical distance among monitoring sites represented limiting factor, which have had influence on impossibility to define general predictions on broader area, which would be the basic hypothesis in experimental approach of the study of concentration of particulate matters (PM_{10}, BS and TSP).

Materials and methods

Results gained in the period from 2005 – 2007 have been used in this research. Accredited institutions and laboratories conducted measurements by passive sampling methods and by collecting data from automatic stations for ambient air quality [14-16]. Methods for analysis of ambient air samples have been defined within the Regulation on limit values, methods for measurements of ambient air quality, criteria for establishment of monitoring sites and data collection:

- air samples containing BS pass through white paper filter (Whitman No. 1) during 24 h, by the speed of 0.5 l min^{-1}. Chemical analysis was conducted by reflectometric method.
- TSP collecting was conducted with AT 2000 device for sampling of airborne particles in the air, under the pressure gradient (50 m^3 h^{-1} with maximum subpressure of 200 mbar), using paper filter (Glass microfiber GF/A Whatman,

Table 1
LIMIT VALUES (μgm^{-3}) BY SERBIA'S REGULATION, EU DIRECTIVE AND WHO RECOMMENDATION

	RS				EU Directive		WHO	
	$PM_{2.5}$	PM_{10}	BS	TSP*	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}
1 hour	-	-	150		-			
24 hours	-	50	50	120	-	50	25	50
1 year	20	40	50		20	40	10	20

*Due to the fact that limit value for TSP has not been defined in newly adopted legislation, authors have applied limit value of 120 $\mu g\ m^{-3}$ – Regulation on limit values, methods for measurements of ambient air quality, criteria for establishment of monitoring sites and data collection ("Official Gazette RS", no. 54/92, 30/99, and 19/2006).

Fig. 1. Location of monitoring sites: 1 (PM_{10} and meteorological parameters) and 2 (BS and TSP), in relation to the location of the industrial zone in Zrenjanin

diameter ϕ110 mm). Analysis was conducted by gravimetric method.

Sampling and analysis of PM_{10} have been conducted with MP101M - TEOM (Tapered Element Oscillating Microbalance) device. Sampling was conducted using vacuum pump through fiber band (glass fiber), with the air flow of 1 $m^3 h^{-1}$, depositing PM_{10} on the band.

Determination of concentration of PM_{10} has been conducted using principle of cyclic measurements, based on the decrease of the intensity of β-radiation (the source of radiation is C_{14}, with the half-life of 5000 g).

Sensor for measurement of wind speed was Tacho-anemometer C 500S/DNA 501 with rotor. Measurement of the wind speed is based on the principle of direct proportionality of angular rotor speed with cups and wind speed, and linearity of the response is independent from the air density. Range of measurement is from 0–50 m s^{-1}.

Statistical software Minitab 15 has been used for analysis and mathematical regression.

Results and discussion

First known measurements of concentrations of particulate matters and the study of the influence on human health date back to early '70s, starting from measurement of concentrations of higher diameters to measurements of concentrations of particles of very small diameters (<1μm). In Serbia, measurements of concentrations of certain pollutants (sulfur-dioxide, nitrogen-dioxide, BS and residual particles) in ambient air were not initiated until '90s. Measurement of concentration levels of TSP was conducted only as indicative measurement, for detection of presence of pollution with minimum annual data coverage of 14 % (Directive 2008/50/EEC).

Results of measurements were divided into two seasons: winter (cold) season (from 15th October till 14th April) and summer (warm) season (from 15th April till 14th October), in accordance with the beginning and end of official heating season in urban areas of medium-sized cities in Vojvodina region.

Measurement of concentration of PM_{10}, BS and TSP has been conducted in the period from 2005 – 2007 (three warm and three cold season), with total number of measurements – 674, 731, and 77 for PM_{10}, BS and TSP, respectively.

Following average values have been recorded in the period of measurement: 33.69, 8.10, and 119.20μg m^{-3} for PM_{10}, BS, and TSP, respectively. For all three parameters, extremely high maximum concentrations have been recorded, even 4 times higher than allowed for PM_{10}, around 2.5 times higher for BS, and 2 times higher for TSP.

Number of exceeds, out of total number of measurements, expressed in percentages, was 15.68, 1.50, and 48 % for PM_{10}, BS and TSP, respectively (table 2).

Daily limit value for PM_{10} in 2005 has been exceeded 41 times, which is 1.2 times (6 days) more than maximum allowed 35 days per year [4]. 73 % of exceeds of limit values for PM_{10} occurred during winter period. Average annual concentration was 36 μg m^{-3}.

During 2006, number of exceeds was 59 days (1.7 times, 23 days), with 94 % of exceeding occurring in winter period and average annual concentration of 40.5 μg m^{-3}.

During 2007, rapid drop occurred, with only 6 exceeds of daily limit value, and 16% of exceeds occurring in winter period and average annual concentration of 23.8 μg m^{-3}.

Taking into account meteorological conditions during 2005 and 2006, it can be assumed that the reason for such high number of exceeds of daily limit value for PM_{10}, during 2006, was due to slightly higher average value for wind speed during winter period of 2005 than for 2006, and the fact that in considered area ventilation was better, and retention time for PM_{10} was shorter.

Unfortunately, the data on wind direction, which would significantly point out on the direction of movement of particles, in most cases, are missing, due to the technical limits.

During 2005 and 2006, exceeding of daily limit value of 50 μg m^{-3} was recorded for BS, three times each year, and 100% occurring in winter period. Average annual concentration for 2005 was 6.8 μg m^{-3}, while during 2006, recorded average concentration value was 7.2 μg m^{-3}. In 2007, number of exceeds was 5 days, of which 100% occurred in winter time. Average annual concentration was 14.9 μg m^{-3}. All measurements for concentration of TSP were conducted during 2005, with recorded average value of 119.2 μg m^{-3}. Number of days with exceeds of LV (120 μg m^{-3}) concentration was 37, and 54 % occurred during winter period. Average annual concentration was 119.2 μg m^{-3}.

As expected, seasonal variations were significant. Presence of additional source of emission (domestic heating) during measurements in colder period is clearly recognizable on average and maximum concentrations for all observed pollutants: PM_{10}, TSP and BS (table 3).

Based on the results, it can be concluded that all maximum concentration values of pollutants have been recorded in the winter period. Figure 2 presents distribution of ratios between daily average concentration levels for BS/PM_{10}, BS/TSP, PM_{10}/TSP, in cold and warm periods from 2005 – 2007.

On figure 2 , it can be seen that the correlation between average daily concentrations for BS/PM_{10}, BS/TSP, PM_{10}/TSP is much lower in summer period, than in winter

Table 2

STATISTICAL OVERVIEW OF THE DATA IN THE PERIOD OF MEASUREMENTS (2005 – 2007) (μg m^{-3})

	PM_{10}	BS	TSP
No. measurements	676	731	77
C average	33.69	8.1	119.20
C min	2.75	0	17.00
C max	206.86	134.00	241.00
>Limit Value (LV)	106	11	37
% >LV	15.68	1.50	48.05

($P<0.001$), which is in accordance with behaviour trend of relations among PM of various diameters in other studies [8].

Median for ratios of daily values of BS/PM_{10} and BS/TSP is significantly lower than for correlation PM_{10}/TSP, even though there is substantial correlation between BS/TSP in summer period (r^2>50%). Substantial correlation has been recorded for correlation between daily concentrations of PM_{10}/TSP in both periods (>60%) (table 4 and table 5).

Based on the measurements, conducted during the period from 2005 – 2007 (divided into 2 seasons, warm and cold), regression curves of linear type have been obtained [2] (fig. 3). During the analysis of regression curves for concentration of BS with other pollutants, relatively weak functional correlation between BS/PM_{10} has been registered: r^2=12.4% in warm season and r^2=3.9% in cold season; relatively good for correlation between BS/TSP in warm season r^2=56%; and the best correlation was between PM_{10}/TSP in both seasons r^2=62% in warm season and 63% in cold season (table 5).

Weak correlation, as well as high variability of values detected within ratios of daily concentrations of BS/PM_{10} and BS/TSP could be associated with the fact that BS is characteristic indicator for specific source of environmental pollution by black particles, produced by combustion process in domestic heating. During summer, the contribution of secondary particles is higher in total concentration of PM, while the participation of BS, as primary particles, in PM, during winter, increased, due to combustion processes [8].

The slope of regression curves is different from case to case. The slope of regression curves for TSP/BS and PM_{10}/BS is always higher in summer period, which is caused by the fact that during summer, the participation and the

Table 3

SEASONAL VARIATION OF PM_{10}, TSP AND BS (μg m^3)

	Warm period			Cold period		
	PM_{10}	BS	TSP	PM_{10}	BS	TSP
No. measurements	411	328	49	263	403	28
C average	27.95	2.95	120	42.68	12.35	117.75
C min	2.75	0	43.00	6.21	0,00	17.00
C max	81.60	24.00	236	206.86	134.00	241.0
>LV	31	0	25	75	11	12

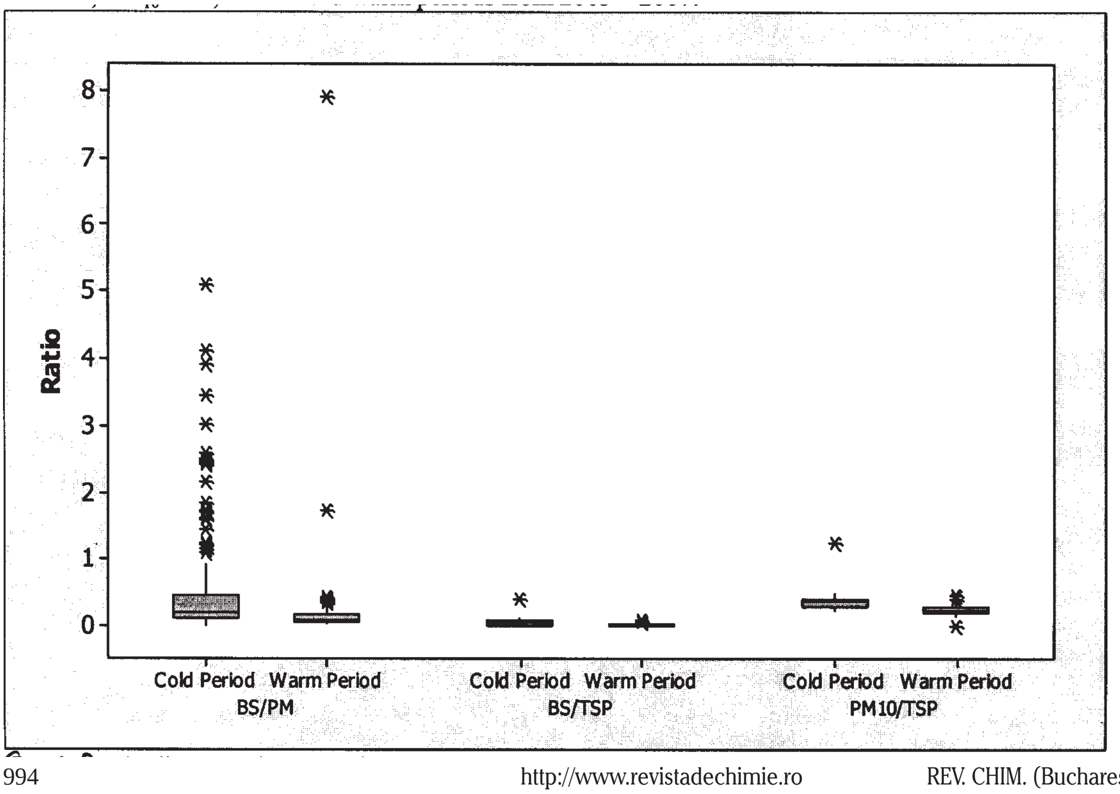


Fig. 2. Distribution of ratio BS/PM_{10}, BS/TSP, PM_{10}/TSP in summer and winter period

		Q_1	Mediana	Q_3
BS/PM_{10}	**Warm**	0,07	0,10	0,2
	Cold	0,14	0,23	o,47
BS/TSP	**Warm**	0,014	0,23	0,033
	Cold	0,016	0,067	0,10
PM_{10}/TSP	**Warm**	0,21	0,34	0,30
	Cold	0,30	0,37	0,413

Table 4
VALUES FOR MEDIAN, UPPER (Q_3) AND LOWER (Q_1) QUARTILE FOR RATIOS OF CONCENTRATIONS OF BS/PM_{10}, BS/TSP, PM_{10}/TSP IN COLD AND WARM PERIOD OF MEASUREMENTS

Regression line (warm)	R^2 (%)	Regression line (cold)	R^2 (%)
PM_{10}=2.75+0.227TSP	62	PM_{10}=14.6+0.193TSP	63
PM_{10}=25.7+1.38BS	12.4	PM_{10}=38.3+0.332BS	3.9
TSP=99.3+6.35BS	56	TSP=61.3+3.69BS	3.4

Table 5
OVERVIEW OF LINEAR EQUATIONS OF REGRESSION CURVES WITH CORRELATION COEFFICIENTS

concentration of non-black components in TSP, and PM_{10} is more dominant [1].

Interception point, in summer period has very large concentration range, from 2.75 – 99.3, and for winter period 14.6 – 61.3 (fig. 3 a-c, table 5). Raised values for interception point in winter period show that various sources of pollution influence on concentration levels, and content of PM during summer and winter period, i.e. during summer, contribution of re-suspended particulate matters in daily concentrations of PM_{10} is higher [1]. When interception points are higher than zero, this indicates on the presence of non-black and non-adsorbing components in PM_{10} fraction, which is the characteristic of particles, with size around 1μm [10].

Re-suspension effect is influenced by the relief characteristics of the area itself. Zrenjanin, as the center of mid Banat Region in the large Pannonian plain, has over 87 % of arable land, which is mostly chernozem (Pedologocial map of Vojvodina 1971). Important fact for explanation of distribution and contribution of re-suspended particulate matters in concentration of PM_{10}, is the very low percentage of afforestation of Vojvodina (less than 7 %, and in Zrenjanin even less than 3 % of total area), which makes it the least afforested and the most endangered region in Europe. In the broader surroundings of Zrenjanin, there are no topographic obstacles and heights, which would, beside forests, have substantial role in reduction of resuspended matters from arable land.

Effects of meteorological parameters on concentration levels of PM_{10}, BS and TSP

Based on available data on concentration of pollutants and meteorological parameters (wind speed), correlation between concentrations of PM_{10}, BS and TSP and wind speed has been formed (fig. 4).

The presence of U-shaped curve for correlation between particulate matters (PM_{10} and TSP) and wind speed, proves the absence of dilution effect for both of these parameters [7]. Even though, in summer period, the presence of U-shaped curve can be expected, due to higher re-suspension of dust from the ground, it is also present in winter period. In winter period higher dilution effect can be expected for PM_{10} and TSP, due to increased ventilation, i.e. concentration of PM in the air decreases as wind speed increases (fig 4a, c).

On the contrary, for BS, expected decreasing curve was noticed in winter period, but also in the summer period, which confirms that re-suspension of fine particles has not influence on concentration of BS during summer period (fig. 4b) [10].).

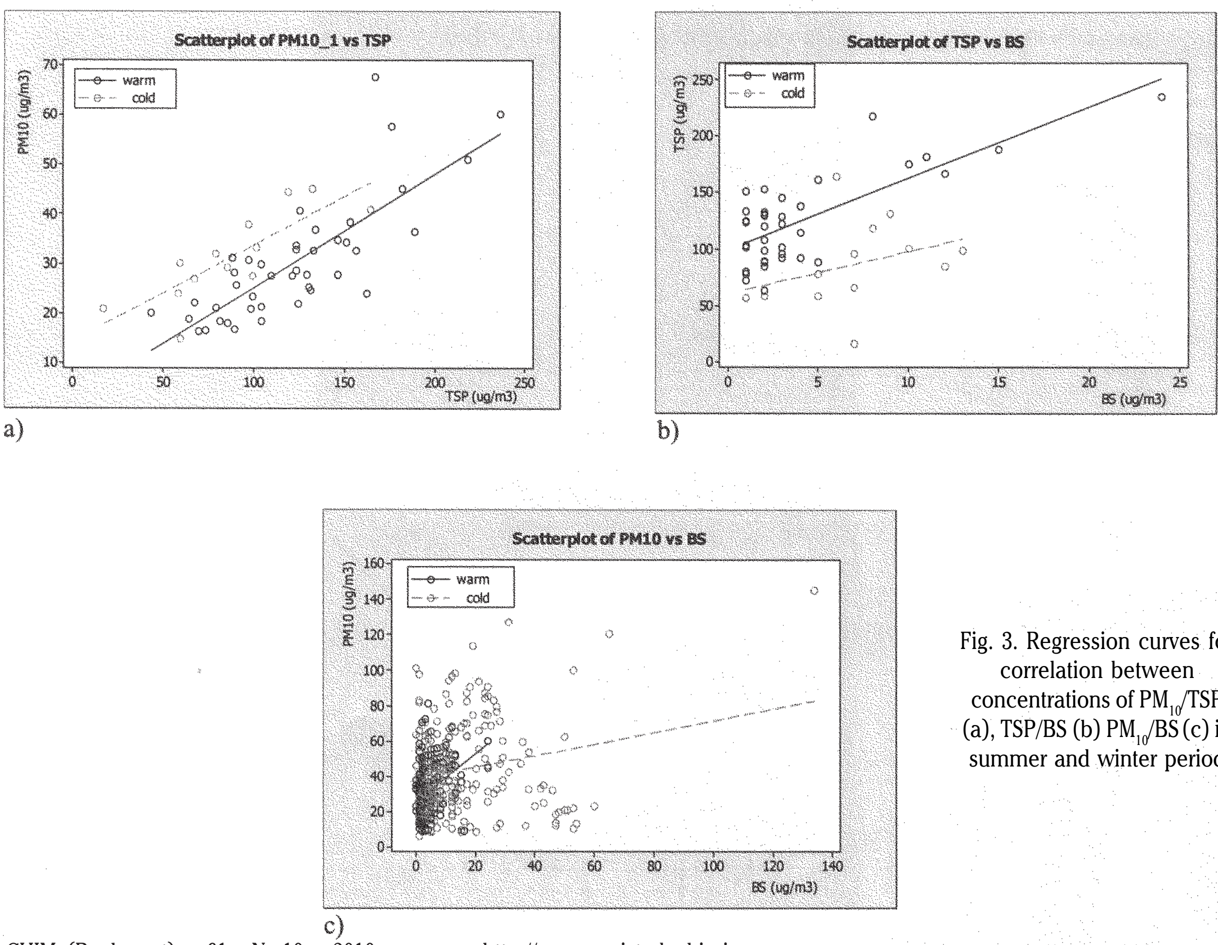
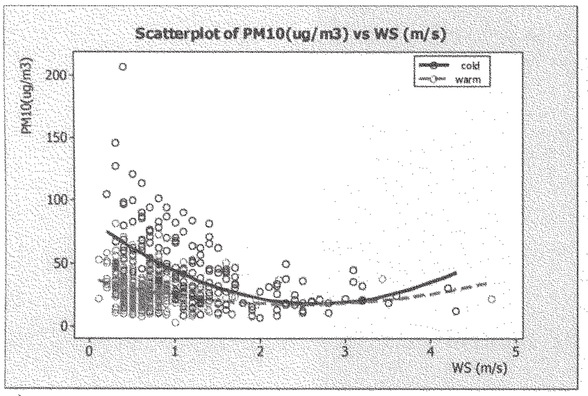
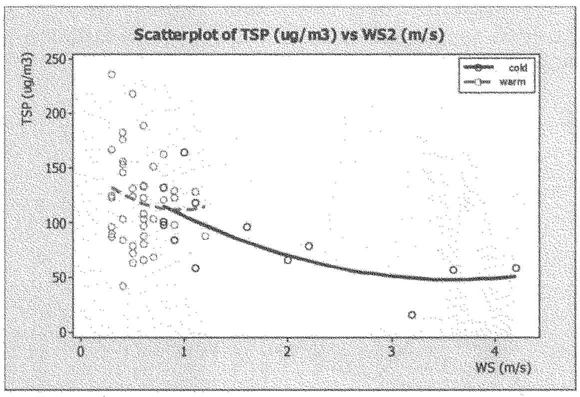


Fig. 3. Regression curves for correlation between concentrations of PM_{10}/TSP (a), TSP/BS (b) PM_{10}/BS (c) in summer and winter period



a)



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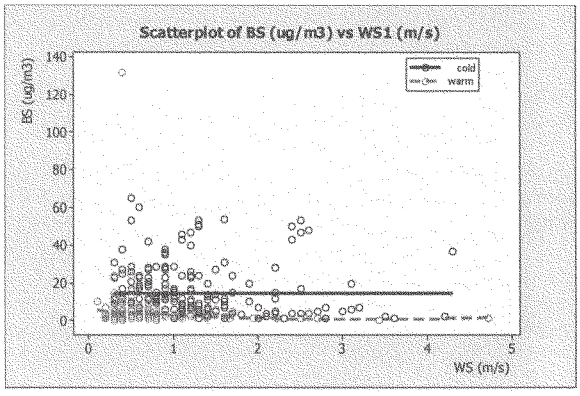

c)

Fig. 4. Correlation between concentrations of PM_{10} (a), BS (b), and TSP (c) and wind speed

Although the shape of curve is indicative, there is no significant correlation between concentration of pollutants and wind speed, according to calculated r-values ($r^2<0.03$). Our results confirm that there isn't significant correlation between PM_{10}, BS and TSP with air temperature ($r^2<0.1$).

Conclusions

Monitoring of the air quality, on the territory of AP Vojvodina, started developing in the mid '90s. Due to political isolation and severe economic crisis in Serbia during last decade of 20th century, development of monitoring of air quality did not follow the pace of development in surrounding countries in the region as well as in Eastern European countries [17]. Therefore, monitoring of airborne pollutants has been conducted unsystematically and sporadically. During the preparation of this paper legislation in this filed was not revised, and it was only partly harmonized with EU standards, therefore some of old limit values have been applied. New Law on Ambient air protection and Bylaw on monitoring conditions and air quality requirements have been adopted in the meantime, introducing new limit values in accordance with EU requirements.

Data used in the research are related to the medium-sized city of Zrenjanin, which represents typical agglomeration in the region: through geographic location, population, level of industry development and percentage of presence of natural gas in individual and remote heating systems of residential objects.

Analysis and mathematical regression of concentrations of PM_{10}, TSP and BS have been carried out aiming to determine dominant source, which contributes to higher concentrations of PM_{10}, and to project concentrations of PM_{10} in other cities, based on available data on concentration levels of BS.

Pollution and meteorological data were collected in period from 2005 – 2007 in the urban area of Zrenjanin, whose population and industrialization is constantly increasing, as well as the exposure to PM_{10}, BS and TSP. Data used in the research are divided into two seasons, warm (15th April – 14th October) and cold (16th October – 14th April) in accordance with official start and end of heating season.

Based on the analysis of results, it can be perceived that registered concentrations of PM_{10}, TSP and BS were above limit values. Due to the fact that maximum values, during the year, were recorded in the winter period, seasonal variation of concentrations of PM_{10}, BS and TSP are significant, and in accordance with trends in other countries [6].

Correlation between PM_{10}/TSP is significant for both seasons (summer – 62%, winter – 63%), while correlation between PM_{10}/BS was weak, which confirms that BS is pollutant from specific pollution source (domestic heating). However, having in mind the fact that limit values for BS have not been defined in EU Directives, historical data on BS concentration levels could not be used for characterization of ambient air pollution in this area.

Considering the results of this study, it has been concluded that air pollution by PM_{10} can generally be characterized by re-suspension phenomena. Important fact for explanation of distribution and contribution of re-suspended matters in concentration of PM_{10}, is very low level of afforestation of Pannonian plain in Vojvodina (less than 7%, and in Zrenjanin even less than 3 % of total area), which makes it the most endangered region in Europe. In the broader surroundings of Zrenjanin, there are no topographic obstacles and heights, which would, besides forests, have substantial role in reduction of re-suspended matters from arable land.

Even though it is clear that re-suspension has considerable part in PM fraction, as well as the influence on its elevated concentration, influence of other sources should not be disregarded. As already mentioned, measuring point for PM_{10} is located in the vicinity of industrial plants, which can also contribute to elevated concentration of PM_{10}, due to the characteristics of the production process: corn processing industry, which emits significant amounts of dust in the technological preparation process, and follow up processes of raw treatment; power plant and heavy traffic street, which are also potential sources of high concentration of suspended particulates. Since suspended particles are prone to long range transport, depending on the shape and size, the importance of emission of this type of pollution on longer distances in relation to measuring point (construction sites, open storages, etc) should not be understated.

Conclusion on correlation of airborne pollutants indicates that this kind of research needs to be continued in Vojvodina and Serbia and organized in accordance with EU standards, with *a priori* projected network of monitoring sites.

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