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FLUCTUATIONS OF THE SO_2, NO_2, CO, O_3, PM10 IN WINTERTIME IN BELGRADE

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Abstract. *Concentrations of tropospheric pollutants such as SO_2, NO_2, CO, O_3, PM10, in the wintertime in 2011, in Belgrade urban area, municipality Stari grad are presented in this paper. Their daily and weekly variations have been analyzed. Traffic and nearby heating plant "Dunav", were identified as the main sources of these pollutants at the location, thus their variations are under the influence of rush-hours traffic cycles and the activity of the heating plant. The transgression of the one hour-, eight hours- and a daily- limits imposed by the law have been identified and particularly singled out the transgression of Serbian Air Quality Index (SAQI_11) limits. Correlations of the air pollutants and relevant meteorological parameters have been determined. The purpose of this research is to provide information about concentrations of pollutants and their trends in Belgrade urban area in the wintertime and to analyze influence of meteorological parameters on their constancy.*

Key words: *air pollution, urban area, limits, meteorological parameters, correlation*

1. INTRODUCTION

Due to large areas covered with various types of solid materials and the urban structure of cities, the atmosphere over urban and industrial areas acts as a generator of pollutants. Forms of the streets and tall buildings create the effects which help contaminated air to remain trapped for a long time in the lower layers, between building blocks. Combustion of fossil fuels in all aspects of the production of electrical and thermal energy and all types of transport emit numerous pollutants in the air. By building a high capacity heating plant "Dunav", which is a source of pollution, downtown Belgrade began to solve the problem of pollution of the much greater level caused by individual furnaces. Beside these, the traffic is also a significant source of pollution. The problem is that the majority of vehicles are outdated and, also, the fuel is of poor quality. In 2013 strict standards were introduced and

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the production and distribution of fuels that don't meet the requirements of Euro IV engines were abolished.

The subject of this study was to analyze pollutants of troposphere, sulfur - dioxide (SO_2), nitrogen - dioxide (NO_2), carbon - monoxide (CO), ozone (O_3), particulate matter with diameter less than 10 µm (PM_{10}), caused by human influence in urban environment. Emissions of these gases and particles affect human health. Poor air quality is linked to asthma, respiratory infections, heart diseases, lung cancer and many other diseases [1]. Soot is full of substances that can cause cancer, and reduces the sunlight that affects an overall quality of life. If appears accompanied by fog, soot can have a fatal outcome. It is estimated that global air pollution in urban areas causes up to 1.3 million deaths per year [2].

2. Methods

Used data were acquired from the Serbian Environmental Protection Agency (SEPA), which is responsible for monitoring air quality on the national level.

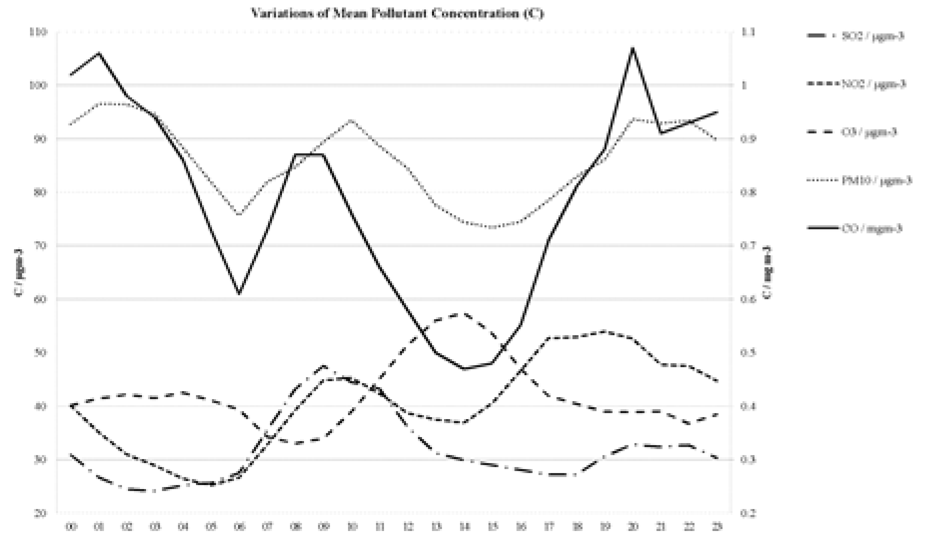
The data for this study were collected at regular one hour intervals, during January and February 2011, on the pollutants measuring station - Stari Grad. The station is located in the schoolyard of primary school "Mihailo Petrović Alas", close to the Višnjićeva street, φ = 44 ° 49 ' N and λ = 20 ° 27 ' E, at the altitude of 97 m.

Methods used for measuring the concentrations of SO_2, NO_2, CO and O_3 are in accordance with the "Regulation on the conditions and requirements for monitoring air quality" and are defined as the reference methods, while the method for measuring particulate matter, PM_{10} is not defined by this regulation. The concentration of SO_2 was measured by gas analyzer SO_2 TELEDYNE API Model 100E. This instrument uses the method of ultraviolet fluorescence regulated by standard SRPS EN 14212 standard. The concentration of NO_x was measured by gas analyzer $NO/NO_2/NO_x$ TELEDYNE API Model 200A, using hemiluminiscent method (described in SRPS EN 14211 standard). The concentration of CO was measured by gas analyzer CO TELEDYNE API Model 300A. This instrument uses non-dispersive infrared spectroscopy method (described in SRPS EN 14626 standard). For measuring the concentration of the ground level ozone, gas analyzer O_3 TELEDYNE API Model 400A was used. This instrument uses ultraviolet photometry method (described in SRPS EN 14625 standard). The concentration of PM_{10} was continuously obtained with GRIMM EDM 180 Aerosol Spectrometer device. This instrument detects light scattered from particles which are present in ambient air. The equivalence of this method is in accordance with EN 12341 standard for PM_{10}. [3]

3. Results and Discussion

3.1. Daily variations and weekly trends

Graph 1 shows processed data - arithmetic means of hourly concentrations of SO_2, NO_2, CO, O_3, PM_{10}.



Graph 1 Arithmetic means of hourly concentrations of SO_2, NO_2, CO, O_3, PM_{10}

Increased concentrations of NO_2 are common in urban areas and mainly come from road traffic vehicles. During the observed period, the concentration of NO_2 begins to rise in the morning and reaches the morning peak between 8:00 and 10:00 am. After that period the concentration slightly decreases, and with afternoon rush hours increases again, reaching the daily maximum between 17:00 and 19:00 (observed period maximum of 53.96 µg m^{-3} was also reached in this period of the day). Until the early morning concentration decreases (25.22 µg m^{-3}), and the cycle starts again. The observed daily NO_2 dynamic corresponds to the intensity of traffic, the maximum values occur during periods of morning and afternoon rush hours.

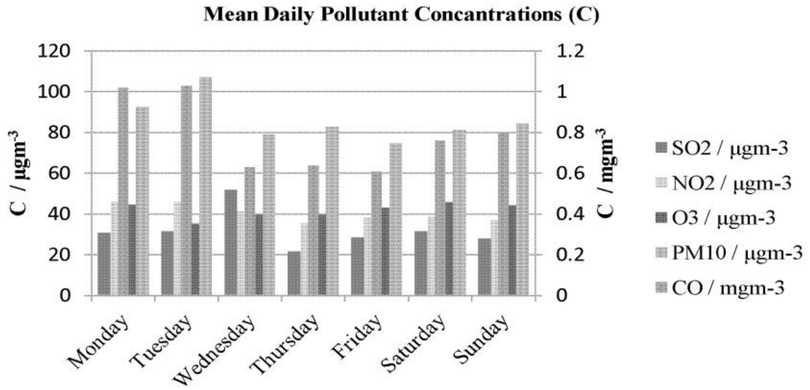
NO_2 in the atmosphere usually comes from two sources, directly from emission sources (primary pollutant) or from chemical reactions in the atmosphere (secondary pollutant) [4]. The winter NO_2 concentrations are, as expected, higher than the concentrations measured in autumn when the maximum reached 33 µg m^{-3} [5], because NO_x emissions come from energy production and distribution, which is more intense in the winter. The concentration of exhaust gases rich in unsaturated hydrocarbons increases in periods of high traffic frequency, which leads to additional formation of NO_2 from the reactions of hydrocarbon free radicals and NO emitted from traffic [6]. Due to the reduced intensity of traffic in the evening hours, concentration is declining and after midnight decreasing of the concentration accelerates.

Due to the heating of residential premises in the winter, SO_2 is always present in the air. Minimum average daily concentration of SO_2 was 24.18 µg m^{-3}. The increase of the concentration starts early in the morning with the beginning of intense operation of heating plant and constant growth continues until 9:00 when it reaches the value 47.53 µg m^{-3}. Then the first slight decrease occurs, though still maintaining high levels, and it lasts until 11:00. Higher air temperature and reducing the intensity of heating affect concentration, thus decreasing continues until 18:00. Due to sunset and lower air temperatures the heating intensify which again results in an increase of the SO_2 concentration. These higher values last until 22:00. After this period, simultaneously with the shutting down of the heating system the concentration of SO_2 decreases.

Serbian reports of the CO concentrations were first presented in 2011 [3]. The increase of the CO concentration starts in the early morning hours, at about 6:00. The highest morning value is reached during the morning rush hour between 8:00 and 9:00 am. After that the concentration decreases (0.47 mg m^{-3}). The afternoon rush hour, around 16:00, influences the concentration. It reaches its maximum value 1.07 mg m^{-3} around 19:30. After the evening decrease, the increase of concentration, with extremely high values of CO (1.06 mg m^{-3}), but also PM_{10} (96.51 μg m^{-3}), was recorded around 1:00 am. The increase in concentration that occurs after midnight may be associated with the heating of older residential buildings located in the lower parts of Dorćol, which are concentrated between Car Dušan street and railway, although some are also scattered in the courtyards between residential buildings. These facilities are quite old and of poor construction quality without adequate thermal insulation, furthermore heating is provided through individual furnaces mainly on coal. When the air temperature drops in the late evening hours the heating increases and usually leads to more intense combustion until midnight. This type of heating, which is a source of CO and PM_{10} causes the higher concentrations that were recorded around 1:00 am. Due to time required for pollution to be transported to the location of measuring station a time lag occurs. PM_{10} have a similar trend of the daily fluctuation, with a morning maximum at about 10:00 and afternoon and daily at 20:00 (93.63 μg m^{-3}) and, as mentioned, extremely high values around 1:00 am. Afternoon maximum occurs slightly later than the peak of afternoon rush hour, which may be the consequence of a location of the measuring station, one of the side streets, and the time pollutants need to reach it, as well as the increase of heating in the late afternoon. Observed similar trends of SO_2, NO_2, CO, PM_{10} were expected in urban areas in the cold part of the year, because their high values match the cycles of morning and afternoon rush hours, as well as activity of the heating plant.

Tropospheric ozone (O_3) has a typical annual trend, with higher average concentrations in the warm period of the year [7], caused by increased solar radiation, UV radiation, followed by substantially lower concentrations during the fall and winter. Same is with the daily fluctuation, with higher values being expected during daytime. Fluctuation of O_3 concentration, as it is a secondary pollutant, are changing due to influences of the photochemical reactions, transport and diffusion [5]. The increase in O_3 concentration begins in the morning, around 9:30, with the beginning of the exposure to the sun's rays, and maintains an upward trend until the early afternoon hours, when it reaches the maximum (57.43 μg m^{-3}). After 14:00, it starts to decrease, while retaining high values. Decreasing trend with slight variations continues until 22:00. Slight increase in concentration was noticed in the late evening and night, which lasted until early morning hours. Concentration begins to decrease again around 5:00 am. A decline in concentration during this period is affected by high morning's relative humidity, which dissolves O_3 and the beginning of more intense transport activity. Reaching the rush hour, emission of the NO_2 increases as well as the reactions of O_3 with NO, ozone concentration decreases and drops to a minimum value (33.05 μg m^{-3}).

Graph 2 shows weekly fluctuations of pollutants. The similarities in trends are notable. Higher values can be observed at the beginning and end of the week, which was related to the increased activity of the population in the cities. Higher values of NO_2, CO, PM_{10} are noticeable on Mondays and Tuesdays, indicating the increased intensity of traffic after the weekend.



Graph 2 Weekly fluctuations of pollutants

Due to weekends' more intense and longer periods of heating in the evening a slight increase of SO_2 and CO values appears. Concentration of SO_2 on Wednesdays (51.93 µg m^{-3}) is a significant outlier, being 62.69% higher than the weekly average. This trend was influenced by extremely high concentrations during the night and morning of the 2^{nd} of February, when the daily average exceeded the critical value defined by the law and hourly values exceeded the margin of tolerance. According to the official data of the Republic Hydrometeorological Service of Serbia, this was preceded by a period of a very cold spell. Three days in a row temperature was below -6.4°C [8]. On the 2^{nd} of February, the station Stari Grad recorded the lowest maximum temperature of the observed period, -7.29°C. These low temperatures influenced the intensity of heating, besides operating period was extended during the night. The effect of heating is particularly important since the heating plant "Dunav" is located on the NE side in the extension of the Višnjićeva street, which course is NE/ENE – SW/WSW. Furthermore, NE wind was dominant in the Višnjićeva street the night before the date when the highest concentrations were recorded. During the week, CO values tend to be high on Mondays and Tuesdays when the weekly maximum was reached (1.03 mg m^{-3}), due to increased intensity of traffic. Decrease in the concentrations begins on Wednesday and it lasts until Friday, when it reaches a weekly minimum (0.61 mg m^{-3}); downward trend doesn't continue at the weekend, but the increase of the concentration occurs, which may be the result of individual furnaces in old ground-storey buildings in the area of lower Dorćol. NO_2 values indicate a logical increase in the first two days of the week and reaches a maximum on Tuesdays (46.08 µg m^{-3}), and from Tuesday until the end of the week are in constant slight decline with minor variations.

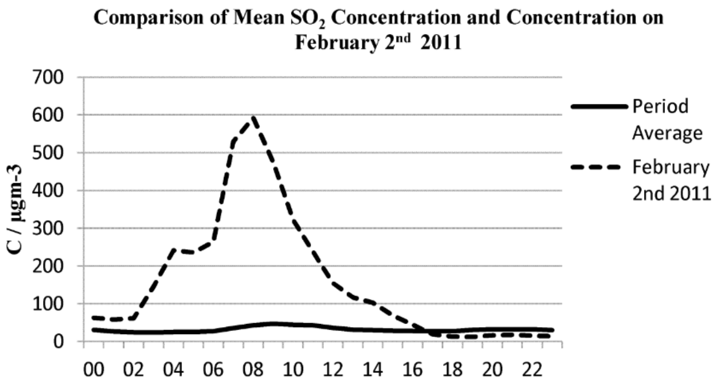
The concentration of PM_{10} on weekly basis could be an example of pollutants which were generated by all sources with a significant impact. It is noticeable that 80% of the days in the observed period were above the limit value, which indicates the constancy of the contamination with this pollutant.

The weekly concentrations of O_3 vary from higher values on Mondays and weekends and lower for the rest of the week. The concentration of O_3 was significantly influenced by a complex of factors, the presence of natural and anthropogenic precursors involved in photochemical reactions [7], insolation, in addition to this the transport from other parts of the city has a significant impact, as the positive correlation coefficient with wind speed indicates.

3.2. Violations of limited values and maximum margins of tolerance

Pursuant to 2009 Air Preservation Law (Official Gazette of the Republic of Serbia, No. 36/ 09), in 2010 Regulation on the conditions and requirements for monitoring the state of air quality [9] was passed.

Data on the emissions of SO_2 show that limit of the one-hour value (350 μg m^{-3}) was exceeded on 2nd February, Wednesday, in three consecutive hours, at 7:00, 8:00 and 9:00 am. Furthermore, the same day the limit of the average daily value (125 μg m^{-3}) was also exceeded. Tolerated one-hour value (500 μg m^{-3}) was exceeded at 7:00 and 8:00 am. The maximum value was recorded at 8:00 am, 592.65 μg m^{-3}, which means that the maximum margin of tolerance for hourly value was exceeded for 18.53 % and the limit value for 374.12%. Graph 3 shows deviation of the SO_2 on the 2nd of February comparing to the average hourly values for the observed period. According to the Regulation [9] SO_2 concentrations greater than 500 μg m^{-3} during three consecutive hours are detrimental to health, though The World Health Organization recommends that this value was not retained for longer than 10 minutes [2].



Graph 3 Comparison of mean SO_2 concentration and concentration on February 2nd 2011

According to the Regulation [9], CO values are determined by the maximum daily 8 hour mean. The limit values for CO were not exceeded in the observed period.

Exceeding the average daily limit value (85 μg m^{-3}) of NO_2 occurred from 5th to 8th February with a maximum recorded value 117.84 μg m^{-3} on the 7th of February. One-hour limit value (150 μg m^{-3}) was exceeded on the 5th of February for six hours, 7th of February for six hours with the maximum value attained at 20:00 (178.39 μg m^{-3}), and 8th of February for one hour.

Measurement of PM_{10} has shown that its value, with the limit value set by the Regulation [9] at 50 μg/m^3 and which should not be exceeded more than 35 days within a calendar year, and with tolerance value of 75 μg/m^3, has been above limit value 36 days and above tolerance value 22 days. In 2011, the value of 50 μg m^{-3} in the Stari grad was exceeded 132 times [3], which is 277.1% higher value than by law permitted exceedances each year. The maximum daily average value during this period was 244.84 μg m^{-3}, observed on the 7th of February, which is transgression of average daily limit value for 389.68% and the tolerance value for 226.45 %. However, the deadline for reaching the values from the Regulation [9] is 2016. Although maximum daily value was recorded on the 7th of February, the maximum hourly value was recorded in the early morning on the prior day, 433.59 μg m^{-3}, though this parameter was not restricted by the law.

The target value for the protection of human health, given in Regulation [9], maximum daily 8 hour mean for O_3, 120 μg m^{-3}, was not exceeded. While the recommended value of maximum daily 8 hour mean given by the World Health Organization, 100 μg m^{-3} [2], was exceeded for one day.

Table 1. shows how many times one-hour and 24-hours limits and maximum margin tolerances were exceeded during the observed period. Maximum daily eight-hour mean of CO and O_3 were not exceeded during this period.

Table 1 Number of Hourly and Daily Exceeding of limit(L) and tolerance concentration values(T)

	SO_2 /μgm^{-3}				NO_2 /μgm^{-3}			PM_{10} /μgm^{-3}			
Period	L	T	max[1]	time of measurement	L	max	measured at	L	T	max	measured at
1h	3	2	592.65	8:00 2.2.2011	13	178.39	20:00 7.2.2011	-	-	-	-
24h	1	/	159.31	2.2.2011	4	117.84	7.2.2011	36	22	244.84	7.2.2011

1 - for hourly statistics, the figure represents maximum measured value; for 24h statistics maximum daily mean value

The most frequent exceeding occurred in the daily values of PM_{10} and NO_2 hourly values, and the most drastic ones happened due to increased concentrations of SO_2 and PM_{10}, with 374.12 % for hourly SO_2 values and 389.68 % for the daily PM_{10} values. It is noticeable that the maximum daily mean values of NO_2 and PM_{10} were recorded on the same day, on Monday, the 7th of February, which were associated with very high frequency of traffic.

Air Quality Index (SAQI_11) has five classes: “excellent”, “good” and “acceptable” categories are when the values are below set limits, “polluted” above the limit values and “very polluted” above tolerated values [3]. Ozone is not subject to the standard evaluation. This way of interpreting quality status is optional and not included in the Regulation [9]. Table 2. shows the shares of air quality categories according to SAQI_11 for the observed period.

Table 2 Frequency of measured air quality, as prescribed by SAQI_11

Pollutant	Clear Air	Good Air Quality	Satisfactory Air Quality	Polluted Air	Heavily Polluted Air
SO_2	84,5%	13,3%	-	-	2,2%
NO_2	89,9%	5,3%	3,8%	1%	-
PM_{10}	-	4,4%	15,6%	31,1%	48,9%
O_3	-	-	-	-	-
CO	95,5%	4,5%	-	-	-

During the period SO_2 and PM_{10} were the ones who acceded the maximum margin tolerance values, with 48,9% for PM_{10}. Also, PM_{10} had high values for “polluted air”, 31.1% of the entire period. Air could be categorized as “excellent” in 95,5% of entire period when it comes to CO, 89,9% considering NO_2 and 84,5% of the period considering SO_2.

3.3 Correlation with meteorological parameters

Table 3. shows correlation coefficients between meteorological parameters and pollutants.

Table 3 Correlation coefficients between air pollutants and weather conditions

	SO_2	NO_2	CO	O_3	PM_{10}	T	Pa	RH	Ws	Wd	I
SO_2	1	.460	.228	-.084	.312	-.043	.107	-.217	-.135	.149	.280
NO_2		1	.708	-.299	.736	.357	.127	-.443	-.362	.093	.659
CO			1	-.368	.898	.253	.151	-.126	-.396	-.041	.587
O_3				1	-.167	-.346	.146	-.142	.301	-.140	.054
PM_{10}					1	.100	.228	-.136	-.342	-.097	.556

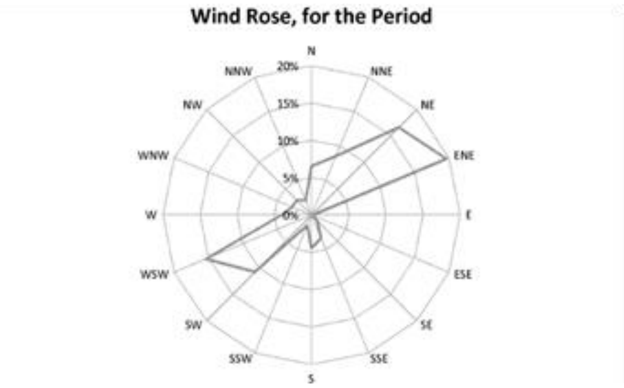
Results indicate that there were some positive correlations between SO_2, NO_2, CO, PM_{10}, whereas correlation between SO_2 and CO is not noticeable since these two have different main sources, heating plant and traffic. Moderate positive correlation between SO_2 and CO was a result of joint source individual furnaces. NO_2 and CO were in a strong positive correlation, as expected, since the main source for both compounds is mainly related to the intensity of traffic and in the addition to this heating plant and individual furnaces. PM_{10} was in a strong correlation with NO_2 and CO, indicating a close connection between increase of suspended particles with increased intensity of traffic. Weak correlation between PM_{10} and SO_2 was observed, both originating from the same source, heating plants. Positive correlations between PM_{10} and NO_2, CO, SO_2 indicate that they originate from the same sources, but one can conclude that at the observed location traffic had major impact on their concentrations. All pollutants showed some, though weak negative correlation with O_3.

Air pressure and pollutants show positive correlation, but of low significance.

Influence of relative humidity on persistence of pollutants shows very weak negative correlations, practically without significance, except for NO_2, which shows a weak negative correlation. Due to increase in relative humidity reduction of NO_2 was noticeable.

Temperature changes and fluctuations of the concentrations of NO_2, CO and PM_{10} were in the positive correlations, whereas negative correlations were noticeable with SO_2 and especially with O_3. More significant effect of temperature changes was noticed while observing NO_2 concentrations, having a weak positive correlation. This was expected because both an increase in the temperature and intense traffic are associated with daylight. Analogically, the positive correlation coefficients between insolation and all pollutants, especially NO_2, CO and PM_{10} were expected. Correlation coefficients of temperature and SO_2 concentrations indicate that between these two there is no apparent correlation. However, maximum mean daily value of SO_2 (159.31 $\mu g\ m^{-3}$) were recorded on the same day as it was recorded the lowest maximum daily temperature at the station Stari grad, -7.29°C. Also, the maximum hour value of SO_2 (592.65 $\mu g\ m^{-3}$) was recorded at the same time as the minimum temperature of the particular day, -7.9°C at 8:00. Low temperatures that lasted for days led to intense heating and this was the main source of this particular pollutant.

Wind speed is negatively correlated with all pollutants, except ozone, which indicates that wind impact led to decreasing of their concentrations. Effects of wind speed were especially noticeable on NO_2, CO, and PM_{10} concentrations. All three pollutants were generated in the surrounding area of the measuring station and originate from the same source, transport. Table 3 shows that the wind speed and fluctuation of SO_2 concentration may be regarded as there wasn't any correlation. However, as SO_2 was not being generated in the strict area of the station, wind had an effect, but on both the dilution and increasing of this compound on the location. Formation of ozone, a secondary pollutant, is determined by solar radiation. Therefore, it is being generated during daylight, which is also the period of higher temperatures. However, a negative correlation between temperature and O_3 and relatively low correlation with insolation were indicated. On the other hand, there was a positive correlation between the wind speed and O_3 concentration. One can conclude that for O_3 place of where it was being generated was of less importance, more important factor was air movement and O_3 air transport. Ozone, which was determined at the measuring station in Višnjićeva street, could only be brought by air movements that had the same course as the direction of the street. The graph No. 4 shows the wind rose, the winds that dominate Višnjićeva Street, where the station is.



Graph 4 Wind rose

Urban areas modify the direction of the wind and fit it in the paths of the streets. In this case, the values of correlations between pollutants and wind direction were as expected, in the low range. Measuring station is located in the city centre, in one of the side streets with lower traffic, and the dominant wind matches street's direction, NE/ENE - SW/ WSW.

4. CONCLUSION

In this paper the results of measuring the concentrations of tropospheric pollutants SO_2, NO_2, CO, O_3, PM_{10}, in the wintertime, in Belgrade, municipality Stari grad, registered at the measuring station in Višnjićeva street, in the street block surrounding transport infrastructure with a higher frequency of traffic were presented. In the vicinity of the site there are no big sources of pollution, such as industry plant, the only bigger polluter is the nearby heating plant complex "Dunav", built in 1987/88 and with the current installed capacity of 346 MW [10].

Daily changes of observed pollutants can be characterized as following. Concentrations of NO_2 fully meet the cycles of the rush hours when they achieve the maximum values. For SO_2 should be noted that the main source is the heating plant "Dunav", and its daily fluctuation is in the narrow correlation with intensity of heating. The concentration of CO is largely a result of traffic, and the smaller impact of the heating plant, and its maximum value occurs as a result of these sources. PM_{10} is generated from different sources, without the dominance of any of them at the given location. Fluctuations of these pollutants are typical for an urban area in the winter, because they come from two main sources, as it was indicated by their strong positive correlation coefficients. The recorded values of O_3 were as expected, increasing during daytime, but also relatively high concentrations were observed late in evening. The concentration of this secondary pollutant was highly influenced by air transport, which was indicated by its positive correlation with wind speed. Weekly trends of NO_2, CO, PM_{10} were in the close relation with increased intensity of traffic at beginning of the week. In contrast, higher weekend concentrations of CO and SO_2 were associated with more intensive and longer period of heating residential premises over weekend. High registered value of SO_2 on Wednesdays, which was for 62.69% higher value than of the weekly average was the result of high concentrations measured on the 2^{nd} of February. Concentrations of O_3 show higher values at beginning and end of week.

Exceeding of the limit values, hourly and daily averages, were noticed in SO_2, NO_2 concentrations and SO_2 also exceeded maximum margin tolerance of the hourly measurements. Exceeding of the daily mean limit values of PM_{10} were recorded during 36 days, and the tolerant values during 22 days. Limit values of CO and O_3 target values were not exceeded.

According to the report of SEPA on the state of the environment [11], based on the annual concentrations, air quality in Belgrade and the city municipality of Stari Grad, air in 2011 was characterized as excessively polluted and was categorized as the third class. Monitoring parameters for only two months it can be concluded that air pollution was at a high level and that the steps taken so far have not given acceptable results for preserving human health.

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VARIJACIJE KONCENTRACIJA SO_2, NO_2, CO, O_3 I PM10 U ZIMSKOM PERIODU U GRADU BEOGRADU

Koncentracija zagađujućih materija kao što su SO_2, NO_2, CO, O_3, PM10, u zimskom periodu u 2011. godini, u urbanom području Beograda, opština Stari Grad su prikazani u ovom radu. Njihove dnevne i nedeljne varijacije su analizirane. Saobraćaj i blizina toplane "Dunav", identifikovani su kao glavni izvori ovih zagađujućih materija na lokaciji, tako da su njihove varijacije su pod uticajem sati "Špica" (ciklusa saobraćaja) i aktivnosti toplane. Uočava se prekoračenje jednosatne, osmosatne i dnevne granične vrednisti, koje su u našem zakonodavstvu identifikovane, a posebno se izdvaja prekoračenje indeksa kvaliteta vazduha Srbije (SAQI_11). Utvrđene su korelacije vazdušnog zagađenja i relevantnih meteoroloških parametara. Cilj ovog istraživanja je da pruži informacije o koncentracijama zagađujućih materija i njihovih trendova u gradskom području Beograda u zimskom periodu i da se analizira uticaj meteoroloških parametara na postojanost koncentracija zagađujućih materija.

Ključne reči: *zagađenje vazduha, gradsko područje, granične vrednosti, meteorološki parametri, korelacija*