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ASSESSMENT OF METEOROLOGICAL EFFECTS AND OZONE VARIATION IN URBAN AREA

Abstract: The paper presents results of the measurements of the tropospheric ozone (O₃) concentration and meteorological parameters: temperature, air pressure, relative humidity, speed and wind direction. The data were collected from January 2016 to December 2016 at station located in locality Centre (Banja Luka), Republic of Srpska, Bosnia and Herzegovina. Ozone is one of the most harmful pollutants to plants and health and highly reactive secondary pollutant. The present study covers investigation of the relationship between the concentration of ozone and meteorological parameters as well as time variations of ozone concentration (by hours, months, seasons). This topic has not been studied up to now in this region, although the recent research data indicates that there is a correlation between them and previously obtained from the world's relevant scientific centres, as already cited above. Statistical analysis confirms string of rolls, which shows directional connection between tropospheric ozone and meteorological parameters, specially temperature ($r = 0.148$), air pressure ($r = -0.292$) and relative humidity ($r = -0.292$). These parameters are the most important meteorological factors influencing the variation in ozone levels during the research. The correlation ozone concentrations with speed and direction of wind is not significant, like other parameters.

Keywords: air pollution, monitoring, ozone, urban zone, meteorological parameters

Introduction

Air pollution becomes a general problem in cities and urban centres of Bosnia and Herzegovina (BiH). During the 2012 in BiH have been registered nearly 231 deaths per 100,000 people having European highest rate of death [1]. Measurements of ozone (O₃) in Banja Luka begun with the installation of the first automatic monitoring system 29th August 2005 in Republic of Srpska and BiH [2]. Similar studies have not been done on the site earlier.

Ozone is released into the atmosphere from the burning of fossil fuels [3-5]. It is considered amongst most harmful pollutants which affects to plants and health [5-10], also belongs to highly reactive secondary pollutant. It binds to plasma membranes and it alters metabolism [8]. Air pollution problem is unfortunately largely present over the Republic of Srpska (entity in BiH) [11]. The state of monitoring is still at a very low level in the Republic of Srpska and BiH, while continuous monitoring is currently carried out in cities

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and municipalities (Banja Luka, Prijedor, Brod, Gacko and Ugljevik). On the other hand, there is significant amount of published research in the field of air pollution [12-17].

Tropospheric (ground-level, surface) ozone, generated from primary atmospheric pollutants, nitrogen oxides (NO_x) and volatile organic compounds (VOCs), collectively referred to as ozone precursors, especially under intensive luminosity conditions and high temperatures [4, 10, 18-22] and pollutant emissions [23]. Recently, regulation of surface ozone in the atmosphere has become an important air quality control issue because of the rapid industrial and economic development associated with increased emission of air pollutants [23]. Ozone variation during the day scale can provide insight to the interplay of emissions, physical and chemical processes that operate on a daily cycle. Ozone levels follow the dynamic of solar radiation, resulting in higher concentrations during the daylight period [24]. Numerous studies analysed the impact of meteorological variables as well as air quality parameters [25-32], where was confirmed strongly dependence of ozone levels on meteorological parameters [19, 20], also that meteorology has an important role in ozone formation as well as transport [24]. Tremendous attention has been paid to establish relationship between surface ozone, gaseous air pollutants and primary meteorological parameters [6, 19, 33, 20]. The spatial variation of tropospheric ozone concentration reveals significant variations in the distribution of ozone precursors (NO_x and VOCs) [23] and relevant physical variable such as winds, temperatures and solar radiation.

The present study includes investigation of the relationship between the concentration of ozone and meteorological parameters, as well as time variations of ozone concentration (by hours, months, seasons) during the long period, from January 2016 until December 2016 in Banja Luka, Republic of Srpska, BiH. This topic has not been elaborated up to now in this region, although the recent research data indicates that there is a correlation between them and previously obtained from the world's relevant scientific centres, as already mention above.

Experimental section

Study area

Monitoring of ozone concentration as a contribution of pollution along with meteorological variables at locality Center in Banja Luka is the main topic of the presented paper. The measurements were made during the peak of traffic jam and high population in the city, from January 1, 2016 to December 31, 2016. As an administrative centre of the Republic of Srpska in BiH, Banja Luka is densely populated and in accordance with this potentially found to be place where a worst possible scenario of pollution is expected due to all this recognized as the convenient location for investigation. It is the second largest city in BiH, after Sarajevo, with the area of 1,239 km^2 , and population of 185,000. City is situated in a basin 164 m above sea level. The average annual temperature is 10.7 °C, average measurement value during January is 0.8 °C, whereas the average temperature in July is 21.3 °C. Typical sources of air pollution recognized in the region Banja Luka are: heating plants, traffic, foundries, metal-processing and chemical industry, and fireboxes in households, municipal waste, etc.

Sampling and analysis

Teledyne Advanced Pollution Instrumentation, Inc. (TAPI) San Diego, California, United States, model T 400 (UV Absorption analyzer) of the range 0-100 ppb/0-10 ppm,

has been used for measuring ozone concentrations. Monitoring of ozone was performed in accordance with standard BAS EN 14625:2013, as reference method [34]. The Model T400 UV Absorption analyser uses a system based on the Beer-Lambert law for measuring low ranges of ozone from ambient air. The laboratory, where research is carried out, is accredited in accordance with ISO/IEC 17025:2017 [35] standard for ozone measurement.

Presented data were recorded under condition of the ambience temperature, where simultaneously have been measured meteorological parameters: air pressure, wind speed and direction, relative humidity. At monitoring site, daily meteorological parameters have been recorded during the researching period. Measured concentration of ozone, were compared with actual values, defined by regulations regarding air quality, issues on pollution and air quality control. Based on both data along with Regulation on air quality values [36], Directive 2008/50/EC on ambient air quality and cleaner air for Europe [37] as well as standards recommended by World Health Organization (WHO) and European Union countries, an assessment of the current state will be given.

Statistical analysis

For determination of the interdependence and relationship between ozone of air quality and meteorological variables, i.e. for pollution modelling together with meteorological variables, the Excel 2016, JASP Computer software [38] and Wessa Statistics Software 1.2.1 were used [39] for statistical data processing. Descriptive statistical operations have been applied into analysing of the measured data. Correlation analysis was performed and the bagplots are shown, also, box and whiskers plots are shown for time representation of hour, day, month and seasons.

Results and discussion

Mean annual value of ozone concentration, in order to preserve human health amounts $120 \mu\text{g}/\text{m}^3$ [36], tested by sampling period time which last 8 hours, as prescribed and Directive 2008/50/EC [37]. Measured average values of ozone and meteorological parameters: speed and wind direction (*VW* and *DW*), air pressure (*P*), temperature (*T*) and relative humidity (*RH*) are shown in Table 1, together with maximal and minimal values, median, mode, standard deviation and variance.

Table 1
Statistical summary of ozone and meteorological variables

	$\text{O}_3 [\mu\text{g}/\text{m}^3]$	<i>VW</i> [m/s]	<i>DW</i> [°]	<i>P</i> [Pa]	<i>T</i> [°C]	<i>RH</i> [%]
Mean	28.68	0.3929	137.4	998.7	12.99	68.78
Median	16.66	0.2900	77.64	998.4	13.22	73.14
Mode	1.400	0.1000	70.29	995.3	16.83	89.29
Std. deviation	45.09	0.3807	113.2	8.488	8.889	17.72
Variance	2033	0.1450	12,820	72.04	79.02	314.1
Minimum	0.01	0.000	0.06	971.9	-8.69	16.80
Maximum	1042	3.290	359.8	1021	34.59	98.63
Maximum daily eight-hour mean value	$120 \mu\text{g}/\text{m}^3$	-	-	-	-	-

In a research field, it did not exceed and amounts $28.68 \mu\text{g}/\text{m}^3$ for 24 h (Table 1) and $37.72 \mu\text{g}/\text{m}^3$ for 8 h (average values for the maximum daily eight-hour mean value).

Measured average value of ozone in Banja Luka, in accordance with previous study, is $29 \mu\text{g}/\text{m}^3$ [2].

The box and whiskers plot given in Figure 1 show graphical representation of total data distribution, i.e. concentration of ozone dependence on the time of day during the measurement period. The vertical box encloses the middle 50 %. The median is the horizontal line inside the box and the cross represents the mean value. Vertical lines (whiskers), extend from each end of the box. Values that fall beyond the whiskers are plotted as individual points. Figure 1 show numerous anomalous data. All median values are lower than all mean values in Figure 1. The interquartile range is especially high during night period. Concentration of ozone is dependent on temperature and light during the day. All the atypical values that are of highest interest in the evolution of the daily ozone concentration. The concentration of ozone was the highest from 11:00 to 17:00.

Fig. 1. Mean O_3 values at certain times of the day (0-24 h)

Day variations in ozone concentration provide us with an easier understanding of processes responsible for the formation and destruction of ozone at a particular place [40]. During the early morning hours (from 6:00 to 7:00) concentration of ozone was very low. This minimum is not very pronounced in this site and it is approximately 07:00 h. Tu et al. [41] in this study states that relatively minimal values during early morning hours are typical for urban areas. Later, ozone concentration begins increasing associated with processes of rupture of the night inversion layer on one hand and photochemical reactions with NO_x on the other [24]. Maximum concentration of ozone during research was found in the period between 11 and 18 hours, which is related with the maximal intensity of solar radiation. High ozone concentrations during daytime is probably result of mixing processes in the boundary layer [42]. Duenas et al. [24] investigated the effect ozone level in correlation with the solar radiation intensity and proved it that ozone level tends to follow the solar radiation intensity, resulting in higher ozone concentrations during the daylight period. In these daily cycles, ozone level increases during daylight such as attributed to the combined effects of photochemical production of the ozone in the mixing layer and the transport from upper layers, which is favoured at noon by the convective activity

in the continental boundary layer, all activated by solar radiation [24]. Obtained results are consistent with other researches, which show that particularly in summer after 10 a.m., the ozone formation increases combining itself with NO and reducing its levels [20]. Values were reduced after 18 hours, when the lowest concentration levels are measured. The lower night ozone levels are in associate with to in situ destruction of ozone by the chemical reaction between ozone and NO [24]. Due to the formation of the night inversion layer, no significant changes occur in ozone concentrations during these hours, which is also consistent with previous research [24]. Once the night inversion layer is formed, significant variation in concentration is not expectable.

The results of hourly distribution of ozone concentrations for each season (spring, summer, autumn and winter) are represented in diagrams of box and whisker (Fig. 2). These diagrams show dependence of the concentration of ozone from the time of day during the measurement period for all season. Sorted data by season are showed on Figure 2 (spring, summer, autumn and winter), where can be seen that higher mean and median values are obtained in spring and winter. Lower average and maximal value were found during the autumn and summer. These diagrams display the day pattern of ozone with the stages previously observed in the daily cycle of ozone values. Duenas et al. [24] states in this study that intensification is essentially produced by strong and persistent inversions in the night as well as during the day due to the increase in solar radiation. Numerous anomalous data is shown on Figure 2, with the inferior ones to values reached in the spring months, whereas the high anomalous values mainly correspond to autumn and winter months. Much atypical values can be observed in the behaviour followed by the ozone concentration throughout the autumn and winter, but also during the season. They are mainly high values that coincide with and high temperature and emissions.

Although the summer level of ozone is of interest due to high temperature and speed of ozone formation (in correlation with NO_x and VOC) [6, 19, 20, 24, 43], during the survey, the ozone value was the highest recorded during winter and spring. The reason for this is emission of air pollutants and high values NO_x and VOC in Banja Luka. Its maximal concentration was recorded in the period between 14 and 17 hours, what is related with the solar radiation. Time of maximal concentration matches with the maximum of solar radiation that occur at 16 h. Episodes of ozone high concentration was found during spring and winter.

The box and whiskers plot in Figure 3 gives graphical summary of ozone concentration distribution, i.e. monthly concentrations during the measurement period. Monthly values indicates that ozone concentration is higher thorough period March-May. Significantly low concentration was found in period June-August, while during October/December is also expected low values period. Phenomenon of an maximal ozone concentration during March was caused by stratospheric-tropospheric exchange (most intense from January to April) [44]. Based on previous facts we conclude that the spring-winter maximal value of ozone concentration results from increased photochemical ozone production and significant stratospheric-tropospheric exchange events as well as high emissions other pollutants (NO_x and VOC).

Fig. 2. Mean and median ozone values at certain times of the day (0-24 h): a) spring, b) summer, c) autumn and d) winter

Each meteorological factor plays a unique role in explaining variations in ozone through its own particular response or effect [21]. Gong et al. [45] investigated the characterization of air pollution index and its affecting factors in industrial and urban areas

in Northeastern China has indicated that the influence of meteorological parameters (precipitation, temperature and pressure) on ambient air quality is significant. The relationship between ozone concentrations and other meteorological variables was analysed using the statistical tools. The results of the correlation analysis between the ozone concentration and meteorological parameters are shown for the level of significance $p < 0.001$ (Table 2). Temperature, air pressure and relative humidity are meteorological factors with relatively strongest impact to the ozone concentration level. The correlation of ozone concentrations with speed and direction of wind is not such a significant, unlike other parameters, confirmed in previous research presented by Camalier et al. [46].

Fig. 3. Graphical summary of ozone concentration distribution (January-December)

Table 2

Correlation coefficients between O₃ concentrations and meteorological variables

		Pearson		Spearman	
		<i>r</i>	<i>p</i>	<i>rho</i>	<i>p</i>
O ₃ [µg/m ³]	<i>SW</i> [m/s]	0.053 ^{***}	< 0.001	0.159 ^{***}	< 0.001
O ₃ [µg/m ³]	<i>DW</i> [Å]	0.055 ^{***}	< 0.001	0.107 ^{***}	< 0.001
O ₃ [µg/m ³]	<i>P</i> [Pa]	-0.292 ^{***}	< 0.001	-0.346 ^{***}	< 0.001
O ₃ [µg/m ³]	<i>T</i> [°C]	0.148 ^{***}	< 0.001	0.382 ^{***}	< 0.001
O ₃ [µg/m ³]	<i>RH</i> [%]	-0.292 ^{***}	< 0.001	-0.370 ^{***}	< 0.001
<i>SW</i> [m/s]	<i>DW</i> [Å]	-0.086 ^{***}	< 0.001	-0.111 ^{***}	< 0.001
<i>SW</i> [m/s]	<i>P</i> [Pa]	0.058 ^{***}	< 0.001	0.044 ^{***}	< 0.001
<i>SW</i> [m/s]	<i>T</i> [°C]	0.130 ^{***}	< 0.001	0.180 ^{***}	< 0.001
<i>SW</i> [m/s]	<i>RH</i> [%]	-0.177 ^{***}	< 0.001	-0.197 ^{***}	< 0.001
<i>DW</i> [Å]	<i>P</i> [Pa]	0.054 ^{***}	< 0.001	0.001	0.932
<i>DW</i> [Å]	<i>T</i> [°C]	0.104 ^{***}	< 0.001	0.115 ^{***}	< 0.001
<i>DW</i> [Å]	<i>RH</i> [%]	-0.077 ^{***}	< 0.001	-0.048 ^{***}	< 0.001
<i>P</i> [Pa]	<i>T</i> [°C]	-0.196 ^{***}	< 0.001	-0.193 ^{***}	< 0.001
<i>P</i> [Pa]	<i>RH</i> [%]	0.090 ^{***}	< 0.001	0.071 ^{***}	< 0.001
<i>T</i> [°C]	<i>RH</i> [%]	-0.551 ^{***}	< 0.001	-0.512 ^{***}	< 0.001

*** $p < 0.001$

We found negative correlation between ozone and air pressure ($r = -0.292$), which implies that increasing of air pressure is followed by decreasing of the ozone concentration and vice versa as shown on Figure 4a. This was also confirmed applying the Spearman's Rank Correlation Coefficient presented at Table 2 and shown on bagplot (Fig. 4a). In the bivariate case the box of the boxplot changes to a convex polygon, the bag of bagplot. In the bag are 50 percent of all points. The fence separates points within the fence from points outside, computed from increased the bag (Fig. 4a).

Fig. 4. Correlation between ozone concentrations and a) pressure, b) temperature and c) relative humidity with bagplots

Rates of chemical reactions is most sensitive to the temperature, so in accordance with this, temperature has a greatest impact to the level of ozone concentration [33]. Vasiliauskiene et al. [21] explored the effect of high temperature and strong solar radiation on ozone synthesis. Abdullah et al. [47] investigated the relationship between daily temperature maximum and maximum ground level ozone. Their results confirm that the temperature trigger formation of ozone, causing extremal concentration [21] also confirm positive linear relationship between daily maximum temperature and maximum ozone concentration [47].

From significant positive correlation ($r = 0.148$) obtained in experiment, relationship between temperature and ozone concentration is confirmed. Temperature increasing is followed by rises of the ozone concentration values, i.e. and vice versa. Correlation between temperature and ozone concentration is confirmed by the Spearman's Rank Correlation Coefficient and presented on Table 2 and Figure 4b. Janik et al. [3] also studied the connection between ozone concentrations and temperature. Results of regression analysis confirmed that ozone concentrations increased with mean air temperature, which is consistent with the results of the presented research.

Relative humidity was found as is important as a parameter with strong impact on reactivity of the system, either by affecting chain termination reactions or in the production of wet aerosols, which in turn affect the ultraviolet actinic flux. It can be a restrictive factor in the disposition of NO_2 , because high percentages of humidity favour the reaction of NO_2 with particles of Sodium chloride [24]. Camalier et al. [46] have been investigated the effect of meteorology parameters on ozone in urban areas and their use in assessing ozone trends. Their results showed that the higher humidity levels are usually associated with greater cloud abundance and atmospheric instability, while the photochemical process is slowed down and ground-level ozone is depleted.

Correlation between ozone and relative humidity found to be negative ($r = -0.230$), which implies that lowering of the ozone concentration is followed by increasing of relative humidity, and vice versa. From the data collected during the study period, ozone concentration dependence on the relative humidity is obtained and shown on Figure 3d. Correlation was also confirmed by the Spearman's Rank Correlation Coefficient and given on Table 2 and Figure 4c.

Conclusion

Despite the legal obligation, the state of monitoring is at a very low level in the Republic of Srpska and BiH with permanent requirement for improving.

Simultaneous measurements of ozone and standard meteorological parameters were performed from January 2016 up to December 2016 for one station, located at the locality Centre in Banja Luka, urban location with intensive traffic flow and relative proximity of heating plants and local industry. Statistical analysis of ozone and meteorological parameters allow us to analyse variations of ozone concentrations. Analysis represents the changes observed during the time of measurement under typical temperate climate conditions. The analysis was performed in terms of hourly, monthly, and seasonal cycles.

Mean annual value for ozone in a research area and is $28.68 \mu\text{g}/\text{m}^3$ for 24 h and $37.72 \mu\text{g}/\text{m}^3$ for 8 h (average values for the maximal daily eight-hour mean value). All over the test area, average annual value is not exceeded. Statistical analysis confirms string of rolls which show direct connection between ozone and meteorological variables.

From daily averages it was found that the lowest ozone concentration occurs in the early morning hours while the peak happened in the afternoon. The seasonal ozone maximum occurs in spring and winter, while otherwise minimum in autumn.

It is necessary to strengthen and expand the air quality monitoring network in Banja Luka, Republic of Srpska and BiH in order to monitor the quality of the air and the impact on the health of the population. It is necessary to provide reliable time series data on pollutants and quality control related to monitoring.

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