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Quantitive Assessment of the Importance of the Atmospheric Environment on Air Pollutant Concentrations at Regional and Local Scales in Sofia

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Abstract. The study was carried out on the territory of the city of Sofia because of its high building density, intensive traffic and specific relief together forming preconditions for high levels of air pollutants. The study comprises data processing of 1-hour concentrations of NO, NO_2, O_3, and PM_{10}, as well as meteorological data as temperature, relative humidity, global solar radiation and wind speed for 2009 – 2015. The results underline the local character of the air pollutants in the area of "Kopitoto" locality which is confirmed by the value of R – coefficient (R = -0.04 ÷ 0.12). For the other observed areas, the regional impact on the occurrence of the air pollutants is stronger or less pronounced R = (0.81 ÷ 0.96). The sole exception is „Druzhba" district where the measured concentrations of PM_{10} expressed strong local influence (R = - 0.36) and 3/4 of the ozone concentrations have a regional influence of their occurrence (R = 0.76).The meteorological variables have the strongest impact on the monthly concentrations of NO, NO_2, and PM_{10}, respectively (47.5%), (54.2%) and (49.1%), whereas O_3 concentrations expressed the strongest dependence on meteorological variables at diurnal scale (68.3%).

Key words: NO, NO_2, O_3, PM_{10}, air pollution, meteorological variables.

Introduction

Air quality is a key element of the people wellbeing and quality of life. It is well-known air pollution harms human health and environment. Although emissions of many air pollutants decreased, air quality problems persist, especially in cities where particulate matter, nitrogen dioxide, and ground-level ozone are recognized as the pollutants that most significantly affect human health. 80% of cases of premature death from heart disease and strokes are due to air pollution, lung disease and lung cancer according to (WHO, 2014). Polluted air increases the incidence of a wide range of diseases like respiratory and cardiovascular diseases and cancer with both long- and short-term health effects (EEA, 2015).

Many settlements around the world are located in valleys, and air quality problems often occur when pollutant sources are located within or near the valleys. The high concentration of industrial and power plants and the dense road network are responsible for a considerable amount of gases and aerosols released in the atmosphere over Sofia (Kolev *et al.*, 2000).

Photo-oxidants and particulate matters are at present a major issue. PM_{10} and NO_2 effect air quality in Sofia, where 79% of

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inhabitants live in areas with exceedance of daily limit value for PM_{10} and 1% residents are exposed to high NO_2 concentrations. The health-related threshold of the O_3 target value was exceeded more than 25 times in 2013 in 18 of the 28 EU members including Bulgaria, in particular, Sofia (EEA, 2015).

Due to the adverse effects of air pollution, there is great interest in improving knowledge (1) on the spatiotemporal evolution of air pollutant concentrations in urban and rural areas and (2) on meteorological influences on the spatiotemporal pattern of air pollutant concentrations. Extended knowledge will help us to develop management options with the aim to reduce air pollution, to prevent adverse impact of air pollution on human health and ecosystems, and to mitigate climate change.

Materials and Methods

The study was carried out in the territory of the city of Sofia. The capital has an area of 492 km^2. It is the largest city in Bulgaria and the 15-th in the European Union with more than 1.3 million inhabitants. Sofia is located in the central part of western Bulgaria, in the Sofia Valley that is surrounded by Vitosha Mountain in South, Lulin Mountain in the west and Balkan Mountain in the north. The average altitude of the valley is 550 a.s.l. Because of its location, Sofia faces air pollution problems due to reduced potential of self-cleaning caused by the mountains that surround the valley. The pool thus formed has most often a stable stratification along its depth and is called a valley inversion (WHITEMAN, 1982). The city belongs to the near-mountain and low-mountain climatic region of Western Middle Bulgaria. The orography of the region is characterized by the predominance of sloped and bulging terrain. The winter is cold and dry, with average air temperature in January being -2.4 °C while the summer is relatively cool, with average July temperature of 20.2 °C (KOLEV *et al.*, 2000).

Meteorological and air pollution data was obtained from six Automatic Measurement Stations, part of the Bulgarian Air Quality National Monitoring Network. Three of them are urban background stations and one rural background station. They are situated away from air pollution sources. The rural background station is at considerably high altitude: 1250 m. The other two posts are transport measurement stations located on key roads where the traffic is intensive, and the concentrations of the air pollutants from vehicles are high. The data was provided by Executive Environment Agency**.**

In Table 1 is given a brief characteristic of the measurement stations such as year of establishment, territory coverage of the posts, measured variables, and station's type. The main differences between the two station types are their location and their scope. The background stations are located in neighborhoods in different parts in Sofia. The scope of the background posts is 100 – 2000 m while the transport monitoring stations cover a very localized area 10 – 15 m. They are established at crossroads with intensive traffic. All stations record data for common pollutants such as NO, NO_2, O_3, and PM_{10}, and meteorological data as air temperature (AT), global solar radiation (GSR), relative humidity (RH), and wind speed (WS). Only transport station Orlov most does not record ozone data. The above listed variables were used for the spatiotemporal analysis of the air pollutant concentrations in Sofia. The data is recorded as one-hour averaged value for seven years period 2009 – 2015. For the goals of the current study, it was essential all monitoring stations obtain an equal amount of data. By the above statement, the survey period was set from 2009 to 2015.

In Fig.1 are shown the locations of the monitoring stations.

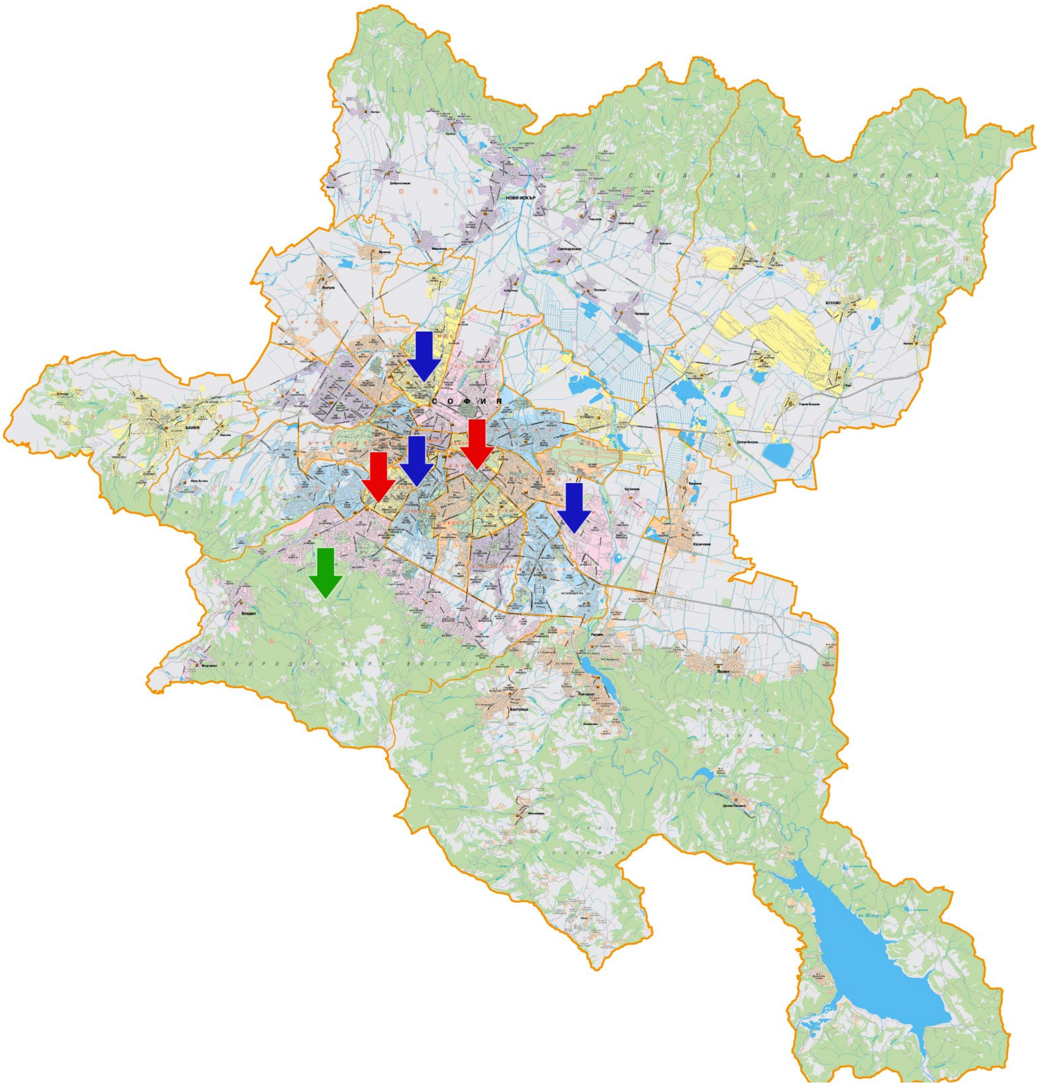

Fig. 1. Location of rural background (green arrow), urban background (blue arrow), and transport (red arrow) air quality monitoring sites.

Table 1. Characteristic of the automatic measurement stations.

Station ID	Station name	Station type	Measured variables	Scope	Establishment
1	Druzhba	Urban background	NO; NO_2; O_3; PM_{10}; AT; GSR; RH; WS	100 – 2000 m	2005 –
2	Hypodrum	Urban background	NO; NO_2; O_3; PM_{10}; AT; GSR; RH; WS	100 – 2000 m	2005 –
3	Nadezhda	Urban background	NO; NO_2; O_3; PM_{10}; AT; GSR; RH; WS	100 – 2000 m	2005 –
4	Kopito	Rural background	NO; NO_2; O_3; PM_{10}; AT; GSR; RH; WS	2 – 10 km	2008 –
5	Orlov most	Transport	NO; NO_2; PM_{10}; AT; GSR; RH; WS	10 – 15 m	2005 –
6	Pavlovo	Transport	NO; NO_2; O_3; PM_{10}; AT; GSR; RH; WS	10 – 15 m	2009 –

As first step was performed data quality check of meteorological and air pollution data. The data series were checked for abnormal values, gaps, and breakpoints. All unusual values were removed from the datasets. Data availability was calculated for each station by components. As a final stage from this step was performed a gap filling procedure by Matlab software. The aim of the gap filling is to provide complete data series for the further processing of the data.

To define spatial similarity in measurements of the air pollutants and the meteorological variables was used Biorthogonal decomposition (BOD). By BOD the local influence of meteorological variables on air pollution concentrations can be separated from regional. For this purpose, all data series were grouped by components in matrix forms and a sole variance of each component from the general variance of the dataset was determined (AUBRY *et al.*, 1991; HÉMON & SANTI, 2003; VENTURI, 2010). For every component (NO, NO_2, O_3, PM_{10}, air temperature, solar radiation, relative humidity and wind speed) was made a matrix L for example L_{NO}, L_{NO2}, L_{O3} L_{PM10}, L_{AT}, L_{GSR}, L_{RH}, L_{WS}.

$$L_{NO}=\sum_{k=1}^{\infty}\sqrt{\beta_k}.\,\mu k.\gamma k \qquad (1)$$

Where:

k is an index which defines the BOD components through decomposition

μk and γk describe component connected parts of inherent temporal and spatial variance of L_{NO}

$\sqrt{\beta_k}$ is a real weight factor which interprets the variance of the BOD-components and sorts down the spatial variance of the BOD-components in descending order.

Decomposition of the matrix L_{NO} results in six spatial components. The number of BOD-components corresponds to the number of the stations. Not all BOD-components consist significant information about the influence of meteorological variables on the air pollutants. Significant BOD-components define the regional contribution to the spatial variance in the studied area. The significance of the BOD-components was tested by Kaiser criterium KAISER (1960). Then they were used for reconstruction of the regional components of the dataset. The reconstruction of the dataset results in matrix L_{reNO}, L_{reNO2}, L_{reO3}, L_{rePM10}…

By the Wavelet coefficient air pollution data was sorted out into 12-time scales. The time-scales are as follow (Table 2):

Table 2. Time-scales, where: Si – time-scale number; h – hours.

S1	2h	S4	12h	S7	168h	S10	2160h
S2	4h	S5	24h	S8	336h	S11	4380h
S3	6h	S6	48h	S9	730h	S12	8760h

Data decomposition was performed by flexible Morlex-Wavelet Function. The data fitting to the different time scales was achieved through compressing and stretching of the data series. Time-scale variance was defined from calculated wavelet coefficients PERCIVAL & WALDEN (2010).

Scaling function:

$$\omega_{n,s}(t)=\omega\left(\frac{t-n}{s}\right) \qquad (2)$$

Where:

S – dilation parameter

n – displacement parameter

t – time-interval

The Wavelet transformation of data series f(t) represents a folding between f(t) and $\omega_{n,s}$(t) TORRENCE & COMPO (1998):

$$c_n(s)=\frac{1}{s}\int_{-\infty}^{+\infty} f(t)\,\omega_{n,s}\,dt \qquad (3)$$

Where:

$c_n(s)$ - calculated Wavelet coefficient. By this coefficient data series variance is calculated.

$$W(s) = \int_{-\infty}^{+\infty} \left|c_n(s)\right|^2 dn \qquad (4)$$

W (s) – interpret the explained variance.

Results

Table 3 represent the number of significant BOD components that describe the variance of the data series. Each variable is described by only one significant BOD component. The percentage of the explained variance (EV) differed for the different variables. The explained variance for the air pollutants fluctuates between 84% for L_{O3} and 94% for L_{PM10}. These results mean that significant BOD components could explain a large part of the matrix L variance.

Table 3. An explained variance of the different data series by some significant BOD components.

Data series	Matrix	Some significant components	Explained variance (%)	Reconstructed matrix
NO	L_{NO}	1	87	$L_{re,NO}$
NO_2	L_{NO2}	1	90	$L_{re,NO2}$
O_3	L_{O3}	1	84	$L_{re,O3}$
PM_{10}	L_{PM10}	1	94	$L_{re,PM10}$

Fig. 2 represents he explained variance of NO-, NO_2-, O_3- and PM_{10} data series for each station. Where correlation coefficient (R) measures the deviation of the station-specific air pollution data from the whole air pollutant data, which means that the study area has a local influence on air pollutants if the R value is close to null. At the areas of Orlov most and Hipodrum NO concentrations have the most prominent regional influence R-value (0.95 and 0.92), while for Nadezhda, Druzhba, and Pavlovo R-value is 0.82 – 0.83. NO concentrations at the Kopitoto locality demonstrates strong local influence and do not depend on transportation of NO from the other observed areas. NO_2 levels have very strong regional character: R-value (0.90 - 0.96) in Hipodrum, Nadezhda, Pavlovo and Orlov most, while 81 % from NO_2 measured in Druzhba depends on the regional factors for its occuarrence. Kopito station stands out with only 12 % of regional influence. Ozone concentrations have similar regional influence for Hipodrum, Nadezhda, and Pavlovo R-value (0.85 - 0.89). Druzhba differ a little from the other areas with regional influence (R-value: 0.76). Ozone concentrations presented a strong local character at Kopito locality. Regarding PM_{10} concentrations, Druzhba and Kopito show a strong local character which is more apparent at Kopito locality R-value (0.04) rather at Druzhba R-value (-0.36). For the rest observed areas, the character of the PM_{10} concentrations was regional.

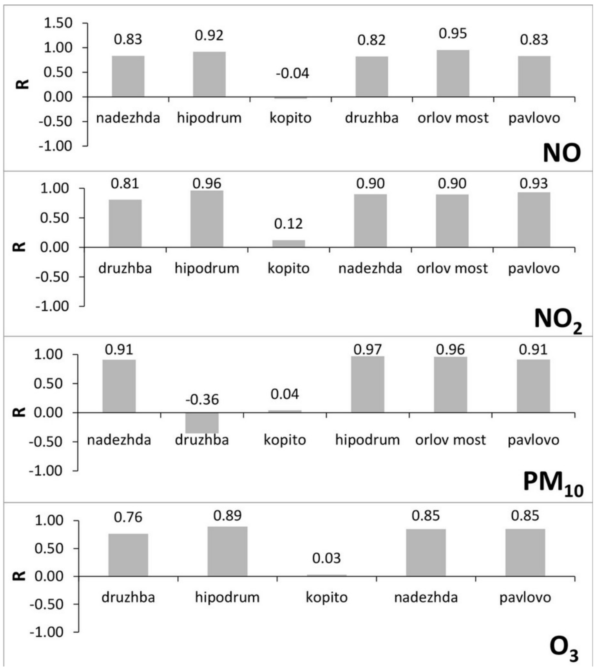

Fig. 2. Correlation coefficient (R). Air pollutants explained variance by significant BOD component.

In Fig. 3 are shown the changes in air pollutant concentrations depending on different time scales. Obtained results are for the whole investigation period.

NO concentrations are dominated by (46.1 %) from S3 (12.4 %), S4 (18.3 %), and S5

(15.4 %), which appeared to be the most important time scales for emerging and developing of NO gasses in the atmosphere. Regarding NO_2 concentrations, they had very similar behavior, where 57.2 % from NO_2 concentrations depend on S2 (10.7 %), S3 (13.6 %), S4 (16.8 %), and S5 (16.1 %). Ozone concentrations demonstrate very prominent dependence on S4 (18.8 %) and S5 (25.8 %) which together contain (44.6 %) of explained variance for the whole time scale. About particulate matter, there is no clear dependence on some definite time scale, which is the case with the previous three air pollutants. The explained variance is evenly distributed at the different time scales; it varies from (3.8 %) to (13.4 %).

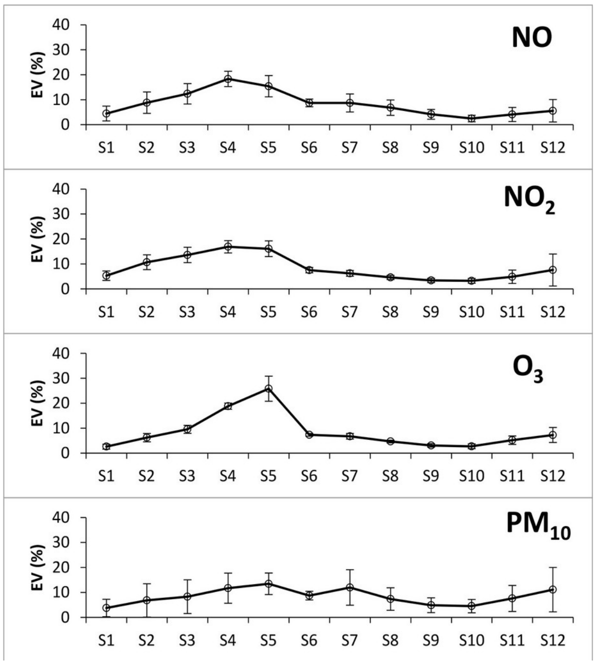

Fig. 3. Explanied variance of air pollutants by Morlex – Wavelet Function in time scale from S1-S12. Error bars represent the standard deviation.

Results from performed partial least-squares regression (PLSR) at five different time scales are represented in Table 4.

The values in the cells expressed the dependence of air pollutant concentration on meteorological variables. Meteorological variables have the highest influence on NO concentrations at S7 (41.3 %), S9 (47.5 %), and S12 (40.4 %). Meteorological variables have a little bit more importance for NO_2 concentrations and are reason for the emergence of NO_2 at S5 (48.4 %), at S6 (46.4 %), and at S9 (54.2 %). Ozone expressed the highest dependence on meteorological variables at each time scale among the air pollutants. It is the most influenced by meteorological variables at S5 (68.3 %) and S12 (64.0 %). Dependence on meteorological variables is less prominent for PM10, where S9 (49.1 %) has the highest explained variance.

Table 4. Partial least-squares regression. Explained variance of air pollutants for the most important time scales by meteorological variables (%).

Air pollutant	S5	S7	S9	S11	S12
NO	36.8	41.3	47.5	36.9	40.4
NO_2	48.4	46.4	54.2	32.8	38.2
O_3	68.3	55.0	55.4	53.8	64.0
PM_{10}	36.5	35.9	49.1	35.1	38.8

Discussion

Regardless air pollutants emergence at different locations, they are mixed and transported by movement of the air masses above Sofia. The process is deeply explained by (KOLEV *et al.*, 2000). After them, due to the influence of the mountain wind, a gradual destruction of the urban heat island and formation of surface inversion starts, which results in an anti-mountain wind which carrying warm air and aerosol from the central parts of the urban island and thus give rise to the so-called urban plume. Such air quality problems experience many population centers located within or near the valleys, where the specific local relief has a key role in the spatial distribution of air pollutants. The same phenomenon is observed by (GIRI *et al.*, 2008). The topography of the Sofia valley in combination with local meteorology predetermines the regional character of the air pollutants in Sofia. An exception from this assertion is Kopitoto locality, where the results prove the local origin of the pollutants. The main reason for this effect is the valley inversion which traps air pollutants above the city, and high altitude of the area that together act as a barrier for transportation of air pollutants from Sofia. These results are quite intriguing because the same factors which define the regional origin of the air pollutants in Sofia determine the local character of pollutants in Kopitoto locality. The outcomes of the current research stressed out the importance of the topography in spatio-temporal evolution and distribution of the air pollutants. Concerning PM_{10} pollution, it was interesting to observe Druzhba district is a local source of PM_{10} emissions, whereas it has a regional character for the other study's areas. Moreover, a small scale PM_{10} transportation was observed from other parts of the city. The predominant local character of PM_{10} pollution in Druzhba could be addressed to the negative relief, in some cases, denivelation between Druzhba and the surrounded areas reaches more than 80 m and the weight of the particulate matters. This prevents the spread of PM_{10}, originated in Druzhba, to the other parts of the city. Apart from the spatial pattern of air pollution, the current study improves the knowledge about the temporal evolution of the air pollutants in Sofia. Where most important periods for emerging of air pollutants such as NO, NO_2, and O_3 are 12 and 24-hours periods, especially for O_3 24-hours period is most critical for its accumulation in the lower atmosphere. PM_{10} pollution does not express a strong dependency on any given period, probably because of its nature of origin. As regards the influence of meteorological variables on concentrations of air pollutants such as NO, NO_2, and PM_{10}, they have the strongest impact on their monthly concentrations. The levels of ozone in ambient air mostly depend on meteorological variables at diurnal scale. Likewise, ozone exhibits the strongest dependence on meteorological variables form the other pollutants because of its nature as secondary photochemical pollutant. The advanced knowledge, as an overall result from the current research, on the spatiotemporal evolution of air pollutant concentrations in urban areas and on meteorological influences on the spatiotemporal pattern of air pollutant concentrations, could be in use of developing management options with the aim to decrease air pollution and to prevent adverse impacts of air pollution on human health and ecosystems.

Conclusions

For most of the observed areas, air pollution concentrations have a regional influence. Exception from this rule is Kopito locality, where air pollutants have a local character. Concentrations of NO, NO_2 and O_3 have clearly marked regional origin in Sofia. It is important to notice that concentration of PM_{10} has a very strong regional influence in the capital, whereas PM_{10} could be considered as a local pollutant in "Druzhba" district. The meteorological variables have the strongest impact on the monthly concentrations of NO, NO_2, and PM_{10}, respectively (47.5%), (54.2%) and (49.1%), whereas O_3 concentrations expressed the strongest dependence on meteorological variables at diurnal scale (68.3%).

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