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NOx AND O_3 VARIABILITY AND ITS RELATION WITH WEATHER CONDITIONS IN IAȘI CITY

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Abstract. The paper presents the temporal variability of surface ozone (O_3) and its precursors (oxides of nitrogen - NO_x) from January 2012 to December 2015. In the same time, this study is aimed to explore the influence of the weather elements on these two major atmospheric pollutants in the area of Iași city. The maximum record of nitrogen oxides reached during the 4 analyzed years was 1200 μ/m^{-3} on October 27, 2015, a period of the year when atmospheric stability prevails. During the warm season (considered from May to September) the highest concentration of ozone was recorded at 155 μ/m^{-3}. The winds direction from SSE or NNW and the winds speed greater than 2 m/s can significantly increase the concentration of the O_3 and NO_x respectively. Also, the stable atmospheric conditions can increase especially the concentration of the nitrogen dioxides. The statistical results illustrate a strong Pearson's correlation of surface ozone with solar radiation/maximum air temperature (r >0.5). The correlations were strongest during the summer months. Using the composite method, between warm season and cold season a difference of the pollutants concentration greater than 30 units for NO_x was observed. Synoptic conditions associated with high pollution are also described.

Introduction

The topic of atmospheric pollution is currently intensely discussed as a result of the industrial development combined with the steep increase of the population living in and around the cities worldwide and the increase in the car traffic in the last decennia as a major contributor to atmospheric pollution, especially within the cities and mainly with nitrogen oxides. Beyond their origins, it is widely known that the concentration and the distribution of

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pollutants are in straight relation with the weather conditions (Bruno, 2008; Grivas et al, 2004).

NO_x and O_3 pollution are major public health concerns in metropolitan regions (Lippmann, 1989). The major sources of nitrogen oxides (NO_x) are represented by combustion processes (EEA Report, 2015). Despite some efforts to mitigate the problem, the emissions of nitrogen oxides (NO_x) from road transport have not sufficiently decreased to meet air quality standards in many urban areas.

In general, NO_x and VOCs are referred to as ozone precursors. Ozone is produced when the primary pollutants NO_x and VOCs (often called non-methane hydrocarbons, NMHC) interact under the action of sunlight (Logan, 1985). Additional mechanisms for the formation of tropospheric ozone include stratospheric injection and processes that influence the abundance of NO_x (Sun et al, 2010). Some studies, as Mcke (1997), showed that tropospheric ozone originates mainly from two sources: the intermittent engulfment of stratospheric ozone in the troposphere and the *in situ* formation from chemical processes among tropospheric trace gases, basically initiated by the incoming solar radiation and high temperatures. Also, the land use changes over the past century affect ozone production by altering vegetations patterns (Zenone et al., 2016; Zhou, 2016). Apart from that, small quantities are also formed from atmospheric discharges at storms, as well as from other anthropogenic sources (Solomon et al. 2007). Additionally, the surface heat forcing driven by urban land surface influences the flux dynamics of the atmospheric boundary layer and influence the pollutant transportation (Oke, 2002). Ozone pollution attracted a lot of concern since this element has a strong oxidant properties, which may cause damage to humans, animals, vegetation and materials under conditions of increasing surface ozone concentration because of smog photochemical reactions in the presence of growing atmospheric pollution. Many studies have prroven that the ozone exposure is especially associated with small increases in asthma morbidity and even mortality (Kalabokas et al., 2000; Posede, 2008).

The weather conditions - indicated by the air temperature, solar radiation, relative air, humidity, wind speed and direction - are playing a major influence on pollutants concentration. Such relationships between meteorological conditions, precursors (NO_x) and ozone concentrations have been examined in several studies (Peton et al., 2000, Abdul-Wahab et al., 2005; Kalbarczyk et al, 2007, Mihăilă et al., 2018). Relationships between meteorological conditions and ozone concentrations have been examined in several studies which have used a combination of statistical regression and graphical analysis. Also, local climate, due to the topography, may influence the distribution of pollutant elements. In the region of Iași the role of local climate in polluants dispersion or stagnation was analized by Ichim (2014) and Sfică et al. (2018).

In Iași city NO_x and O_3 pollution episodes became a major environmental problem in the last years. Our study describes the general features of NO_x and O_3 concentration and distribution in Iași, and their relation to weather conditions, representing a first mandatory step in the current attempt to mitigate this local environmental problem.

2. Data and methods

For this study we have used multiple types of data:

- hourly concentration of the nitrogen oxides, ground level ozone and weather elements for 4 years (2012-2015) were taken from the Environmental Protection Agency (EPA, 2018);
- daily data of the weather elements for the Iași airport station (solar radiation, air temperature, relative humidity, precipitation, atmospheric pressure, wind direction, wind speed) were taken from Climate Prediction Center (1987);
- vertical temperature gradient calculated at daily level on the basis of the observations from Dancu (70 m height) and Păun (380 m height) monitoring points within the experimental temperature monitoring points of the Faculty of Geography and Geology from "Alexandru Ioan Cuza" University of Iași;
- synoptic maps from the reanalysis dataset, provided by National Centers for Environmental Prediction (NCEP) and the National Center for Atmospheric Research (NCAR) Reanalysis (Kalnay et al., 1996). For this we have selected and analyzed the days with polutants concentration within the daily peak values (the first 35 days according to daily concentration for the entire period).

The analysis of this dataset was done in order to show the hourly evolution of the pollutants at daily, monthly, seasonly and semestrial level.

Library OpenAir from R Studio software was very useful to build graphics and visualisation in order to analyse air pollution data. A matrix of Pearson's correlation was use as a statistical method in order to explain the influence of the meteorological parameters on the pollutants concentration (nitrogen oxides and ground level ozone). Additionally, the correlation between pollutant, wind speed and wind direction was calculated by the Polar Plot graphic. This method is a useful diagnostic tool for quickly gaining an idea of potential sources of pollution. The correlation between the weather conditions and pollutants elements was calculated at semestrial level (warm semester, cold semester). Regarding the influence of the weather elements on the pollutants analyzed, we used a composite analysis for which the concentrations of pollutants were grouped according to the increasing values of the weather elements, then the difference between the upper and lower third of the daily pollutants was calculated. The difference obtained was tested for significance using a t-test and it can give a idea about the magnitude of changes in pollutant concentration determined by the synoptic conditions.

3. Results

Monitoring network results. In Iași, the NO_x normalized values have two peaks during the day: one at the 11:00 AM, and a second one at the 23:00 PM (Figure 1a). The first peak is directly in relation with the the accumulation in the maximum traffic hours (7-9,30), while the second one is probably related with the increasing atmospheric stability during the night. Also, this link with traffic conditions and urban climate conditions is sustained by the lowest values of the nitrogen dioxides during the weekend days when the traffic is reduced and the NO_x day maximum is lower than the night maximum.

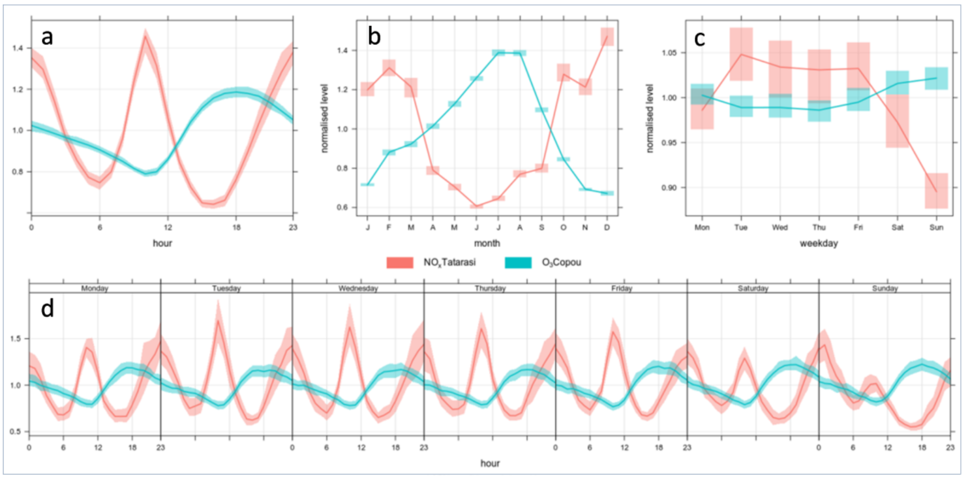

Fig. 1 - The hourly(a), monthly(b), weekly(c) and hourly by day (d) concentrations of the nitrogen oxides (red) and ozone ground level (blue) in Iași for 2012-2015(normalised values)

O_3 instead is more dependent on the diurnal weather conditions due its cycle of formation, the solar radiation and air temperature - two elements favoring the formation of this element - often having the highest value of the day during the noon or during the early afternoon hours (Figure1c, 1d). Rezultate similare pentru evoluția diurnă și săptămânală, Mihăilă et al., 2018 pentru Suceava.

Thus, meteorological conditions and ozone concentrations are far less day-of-week dependent than are changes in NO_x and, as a result, comparison of weekdays to weekends is an effectively and widely used tool to study the NO_x dependence on O_3 formation (Murphy et al., 2006; LaFranchi et al., 2011; Pollack et al., 2012). In Iași NO_x concentration is typically 40–50% lower during weekends than during the weekdays, a phenomenon caused by the reduced weekend car traffic, especially heavy Diesel traffic (Marr et al., 2002).

The dependence of NOx concentration on the atmospheric stability is to be seen also on its normalised annual evolution (Figura 1b) which inidcates a high concentration season from October to March, corresponding with the maximum atmospheric stability in the region. Therefore, the maximum hourly concentration of nitrogen dioxides (1200 μ/m^3) was reached on October 27, 2015. In this month of the year the thermal inversions are frequent in the region (Ichim et al., 2014) as a result of the dominance of high pressure centers (Sfîcă, 2015).

As concerning O_3, the annual maximum is reached during summer (Figure 1b) and as a result the maximum hourly of ground level ozone concentration (152.2 μ/m^3) was recorded on 8 July during the afternoon hours. Noteworthy is that no monitoring station recorded values greater 180 μ/m^3. However, the monitoring station Copou - Sadoveanu records values between 80-150 μ/m^3 for all the analyzed period, as a consequence of its periurban position in the city. The years 2012 and 2015 are noted because the values 80-100 $\mu g/m^3$ wich are recorded in Mars, respectively in April (Fig. 3).

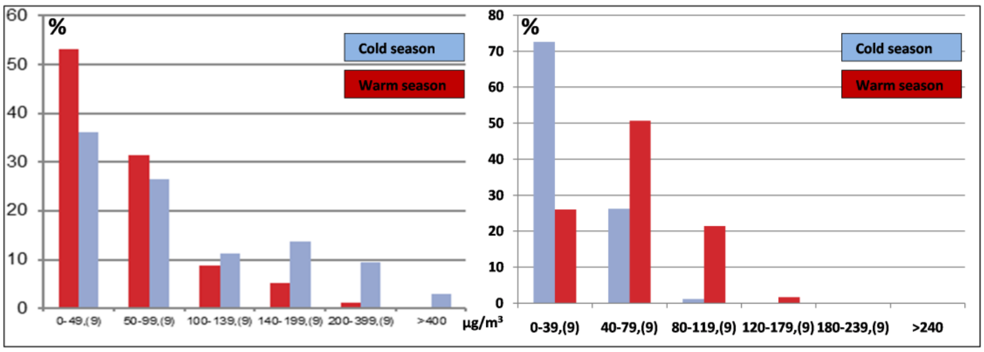

Fig. 2 - The semester concentrations of the nitrogen oxides (left) and ozone ground level (right) in Iași city (2012-2015)

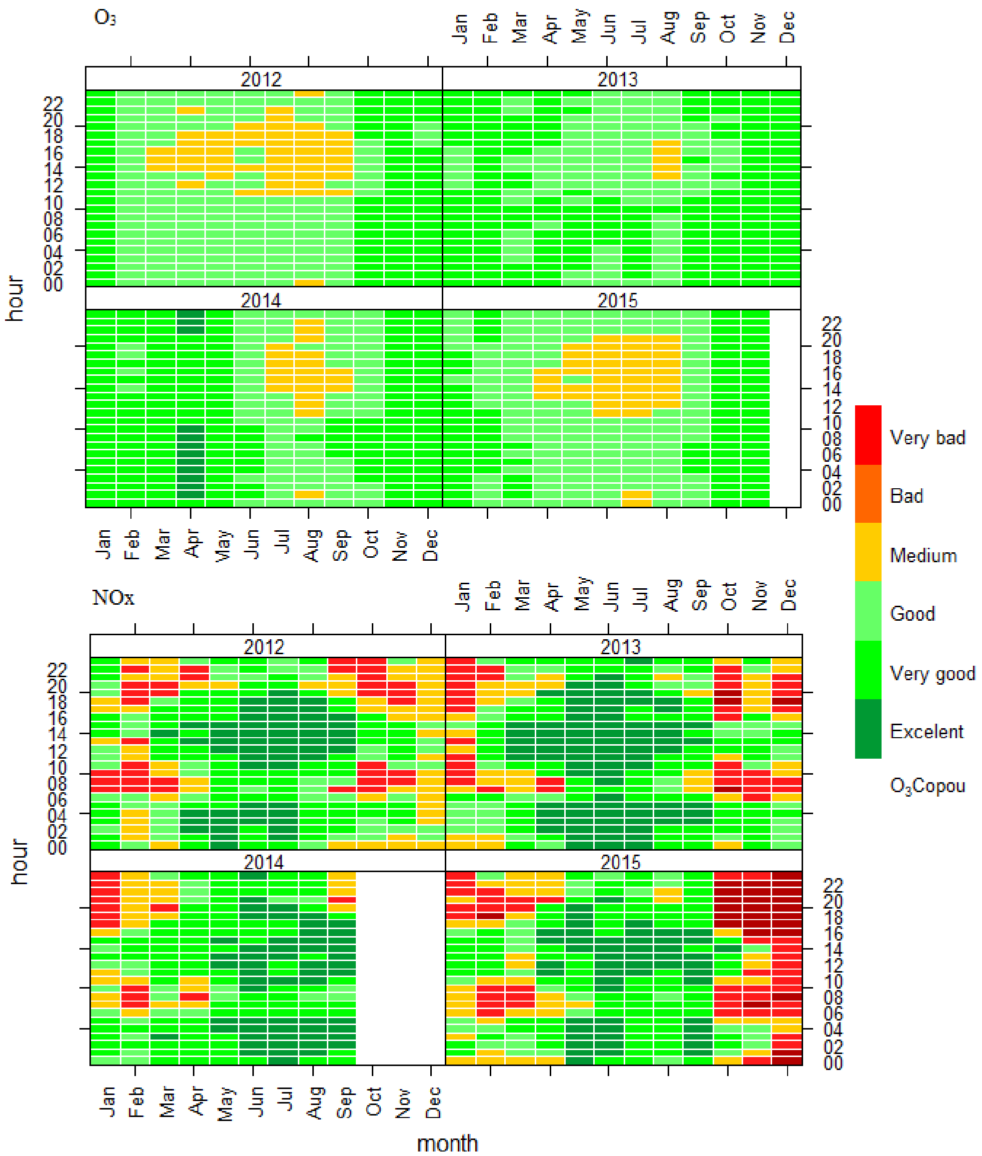

Fig. 3 - The monthly hourly index concentration of the nitrogen oxides (down) and ozone ground level (up) for the period 2012-2015 in Iaşi city

The maximum daily values for the NOx were recorded during the cold season of 2012 and during the autumn of 2015. In the last interval the daily values greater than 400 µg/m^3 in four consecutive days were also recorded. Also, the difference between semesters is showed by the values 120-179, (9)/ >400 µg/m^3 which are recorder only in the cold semester/warm semester for the nitrogen oxides/ ground level ozone. During the cold semester the values 0-39(9) µg/m3 of the ground ozone level has a high frequency while during the warm semester they decrease (Fig. 2).

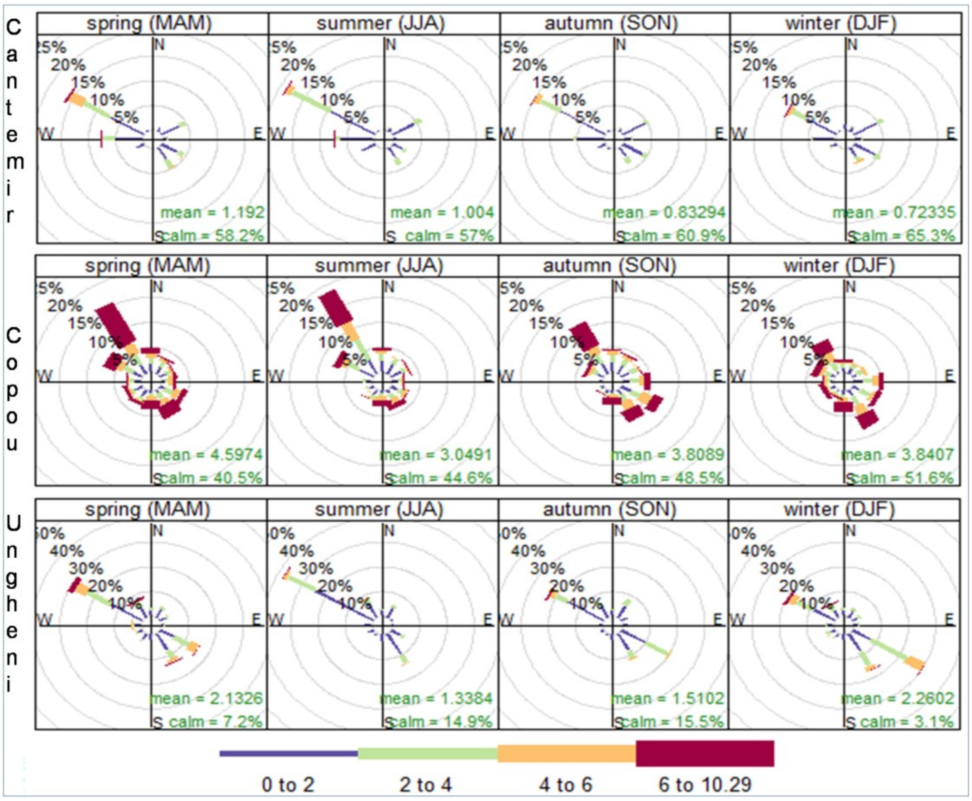

Fig. 4 - Frequency of wind direction (%) and the mean wind speed by direction in Cantemir (top), Copou (middle), Ungheni (bottom) monitoring points of EPA Iași (2012-2015)

Taking to account the importance of wind for the dispersion but also for the formation of NOx and O_3 (Gorai, 2017) we have analyzed in detalis the influence of wind on these two pollutants in Iași. The prevailing wind direction in

Iași is North-West (Figure 4), as effect of the prevailing westerly atmospheric circulation in the region. As well, in many winter weather situations, this is the region in Romania that heats up in the first place after very cold periods as a result of air advection of Atlantic origin on a north-western direction (Apostol and Sfîcă, 2011).

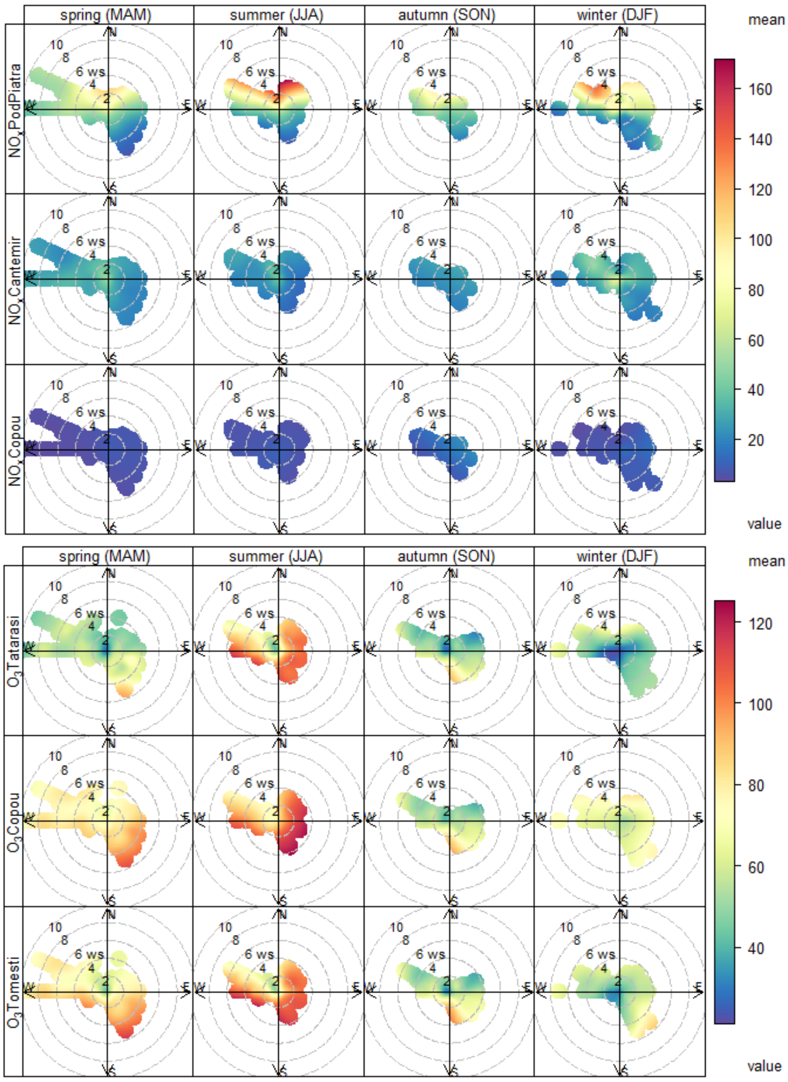

Fig. 5 - Nitrogen oxides (up) and ozone ground level (down) mean concentrations (ppb) in relation with the wind direction and speed in Iași (2012-2015)

The Copou monitoring point - situated in the northern hilly part of the city - records the highest wind speed values as a results of its higher altitude. In the Cantemir station the lowest wind speeds are recorded insted, presenting also 60 % frequency of atmospheric calm, according to its location in the riverbed of Bahlui river and in the same time within the city center.

The calm wind condition within the urban region was one of the contributing factors for the accumulation of O_3 precursors and O_3 exceedance. The wind direction at the Cantemir and Ungheni monitoring stations is canalized by the predominant orientation of the Bahlui Valley (WNW-ESE) and the NW wind direction has a 20 % frequency during the spring time. Moreover, the wind direction North- Western has the higher frequency in the summer (30 %). In winter time, the E , NE and E-SE wind direction has a 30 % frequency as an effect of anticyclonic activity centered in the eastern part of the Europe.

For better understanding the impact of the wind on pollutants concentrations, a wind rose showing the pollutant concetration as a function of wind direction and speed for each station and each pollutant is presented in Figure 5.

For nitrogen oxides, the North-NorthWest wind direction is a pollutant factor. The wind moves the NOx from the northern area of the station, bringing them into the central area, according with the shape of the Bahlui Valley. In Podul de Piatră station, the wind speed of 10 m/s NW-N direction can increase the concentration of nitrogen dioxides all the periods, while S-SE direction leads to a strong dispersion of the nitrogen oxides. In the Cantemir monitoring station, the low wind speeds of 5 m/s are the main contributor of high pollution. This is linked to frequent stability condition in the Bahlui Valley. According to Alexe (2008) and Ichim (2014) the frequency of the thermal inversions is upper to 35 % in the autumn. Also, wind coming from southeast to northwest with sustained wind magnitude (> 5 m/s) produces a peak of NO_x in a Copou–Sadoveanu monitoring station as a result of pollutant transportation from the city center toward the city periphery.

Regarding O_3, concentrations above 60 mg/m^3 are measured for the winds greater than 5 m/s from south and southwest direction. In summer, ozone concentrations are high regardless of the direction of the wind, but a peak of ozone concentration occurs during the lowest wind speed (<7 m/s). The south and south-west component wind is linked to intense tropical air mass advection. The high maximum air temperatures and low humidity is the characteristic of this air mass. During the winter months (DJF) the easterly winds and a wind speed exceeding 7 m/s leads to low concentrations of ozone. Often, these periods are characterized by the heavy fog, low radiation and minimum temperatures.

Statistical results. The statistical analysis was performed by a Pearson's correlation matrix applied for Copou-Sadoveanu monitoring point of EPA-Iași using NOx/O_3 and weather data. This was detailed through a composite analysis for all the available EPA monitoring points.

The obtained results are in line with previous findings on the field (Abdul-Wahab et al. 2005; Yerramsetti et al. 2013). Correlation coefficient between O_3 and NOx is significantly negative as expected: -0.60/-0.66. This relation between O_3 and NO_x concentrations has been previously explained in other studies (Modarres and Dehkordi 2005, Pudasainee et al. 2006). Abdul (2002) found that the O_3 concentration were negatively correlated with NO and NO_2 (Figure 8) because it is known that these pollutants are precursors of O_3.

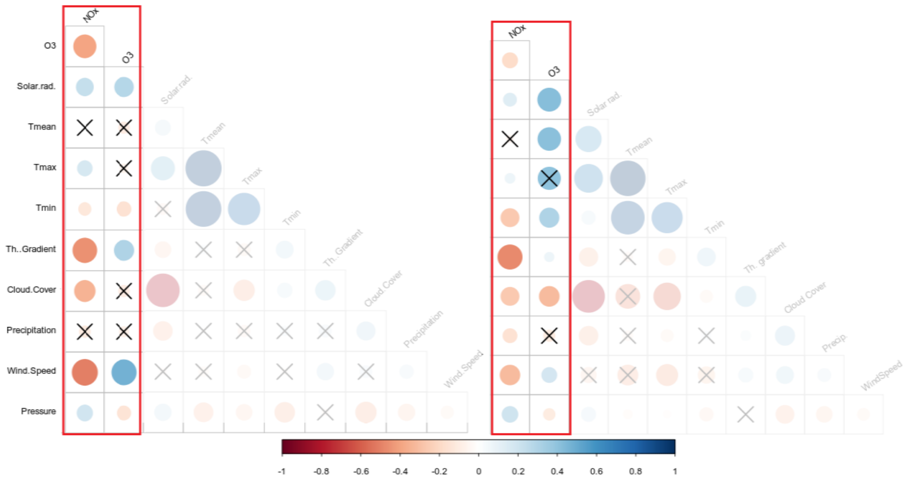

Fig. 6 - The Pearson's correlation matrix between NO_x/O_3 and weather elements during cold semester (left) and warm semester (right) in Iași - statistically significant ($p<0.01$) values with dots

The Pearson's correlation matrix shows that the thermal gradient, the minimum air temperatures and the wind speed have the highest impact on the annual run of nitrogen oxides concentration (r > +/-0.40). In brief, when the synoptic situation favors the occurrence of lowest minimum air temperatures - often on the anticyclone situation in the winter time - next to a steap thermal inversion, the nitrogen oxides concentrations reach their maximum values (Figure 8). Obviously, this is indicated by the positive correlation of NOx with atmospheric pressure. Interestingy, the role of the thermal gradient remains high even for the warm season, an indicator of the important role played by the radiative temperature inversion for the occurence of NOx pollution in this period. As well, high wind speed are more efficient in NOx dispersion during the cold season. Also, we should remark that high amount of precipitation play a dispersion role for NOx only for warm season, while during cold season the correlation is not significant.

Higher O_3 concentration values are mainly caused by high amount of solar radiation and high air temperature. In fact, air temperature acts as a proxy parameter, representing directly the diurnal variation of solar radiation (Figure 8). The relation between temperature and O_3 concentration has been mentioned as well by several authors (Mihăilă et al., 2018). During cold season instead, we remark that higher values of O_3 are induced by very low minimum temperature. This is a result of the stratospheric ozone intrusion in troposphere during extreme coldwaves associated with polar vortex disruption. Such an event occured in January-February 2012 when very high O_3 concentrations occured throughout Romania (Păunescu, 2012) at the end of a sever winter episode (Sofroni et al., 2013). Also, low pressure conditions and a high thermal gradient/atmospheric instability favor together the increase of O_3 concentration, especially during cold season.

Tab. 1 - Composite analysis values at annual, cold semester and warm semester level (year/cold semester/warm semester) for nitrogen oxides in Iași city (2012-2015); differences tested with z-test and not shown when statistically insignificant

	Podul de Piatră	Cantemir	Copou	Tomești
T mean	-9,5/ /+6,4	-2,3/+3,9/		
Pressure	+11,9/-12,2/	+2,7/-13,7/	+6,5/-21,0/	+12,5/-16,3/
Wind Speed	/-10,0/	/-13,4/	/-11,4/	/-10,4/
Thermal Gradient	-32,5/-45,7/-9,3	-28,6/-15,6/-14,3	-13,3/-22,8/-12,2	-12,1/-32,4/-8,6
Precipitation	-10,1/-7,7/	-6,6/-8,5/		-3,8/ /

These results are supported also by the composite analysis (Table 1, Table 2) which indicate the differences between the pollutants concentration associated to upper third and lower third of the most important weather elements sorted from their higher to lower values. This analysis reconfirms the role of the vertical thermal gradient and the maximum air temperature for the occurrence of high concentrations of NOx, respectively of O_3 in air quality. The thermal gradient, in combination with the high atmospheric stability has a strong influence on the concentration of pollutants. For instance, during cold semester, for the upper third of thermal gradient values in Iași (atmospheric instability), the NOx concentration is 45,7 μg/m^3 lower than for the days within the lower third of thermal gradient values (atmospheric stability). According to this analysis the high wind speed plays an important role in NOx dispersion only in cold season.

As regarding O_3 the most remarkable differences are imposed by the thermal gradient. Atmospheric instability determines O_3 concentrations 35,3/26,4 μg/m^3 higher in Tătărași/Tomești, monitoring points than during atmospheric stability conditions. In the upper area of the city instead, in Copou-Sadoveanu monitoring point the values of O_3 are 20,5 μg/m^3 lower in atmospheric instability than during atmospheric stability. As well, air temperature imposes great differences at annual level, with higher air temperature bringing higher values of O_3 concentrations (15-30 μg/m^3) at annual level. The above discussed contribution of O_3 stratospheric intrusion during cold season can be observed, leading to higher O_3 concentrations when the air mean temperature is very low. Not at last, the solar radiation determines higher concentrations during sunny days.

Tab. 2 - Composite analysis values at annual, cold semester and warm semester level (year/cold semester/warm semester) for nitrogen oxides in Iași city (2012-2015); differences tested with z-test and not shown when statistically insignificant

	Tătărași	Copou - Sadoveanu	Tomești
T mean	15,3/-12,6/13,1	30,5/-11,9/24,3	26,4/-11,8/14,7
Solar Rad.	21,5/10,9/20,6	25,7/11,4/20,5	22,9//21,4
Cloud Cover	-25,4/-21,4/	-20,4/-21,3/-22,4	-22,2/-11,4/-12,3
Wind Speed	-8,2/-10,3/-8,6	-20,2/-13,4/-15,4	-7,6/-21,4/-7,2
Thermal Gradient	35,3/-5,3/13,0	-20,5/-15,9/10,6	26,4/8,9/14,3
Precipitation	-3,9//-12,1	-8,3//-9,8	//-10,6

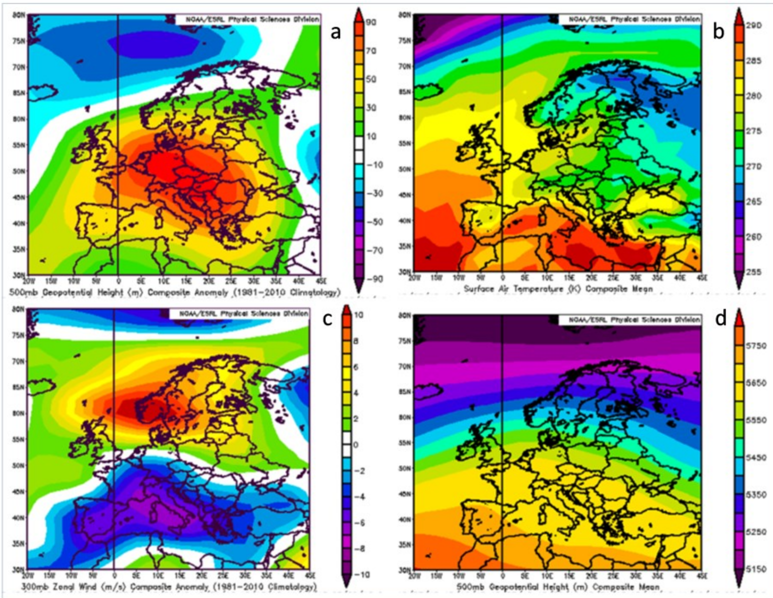

Fig. 7 - Daily mean composites of the 500 hPa geopotential height anomaly (a), surface air temperature (b), 850 hPa temperature anomaly (c), and 500 hPa geopotential height (d) - NCEP/NCAR

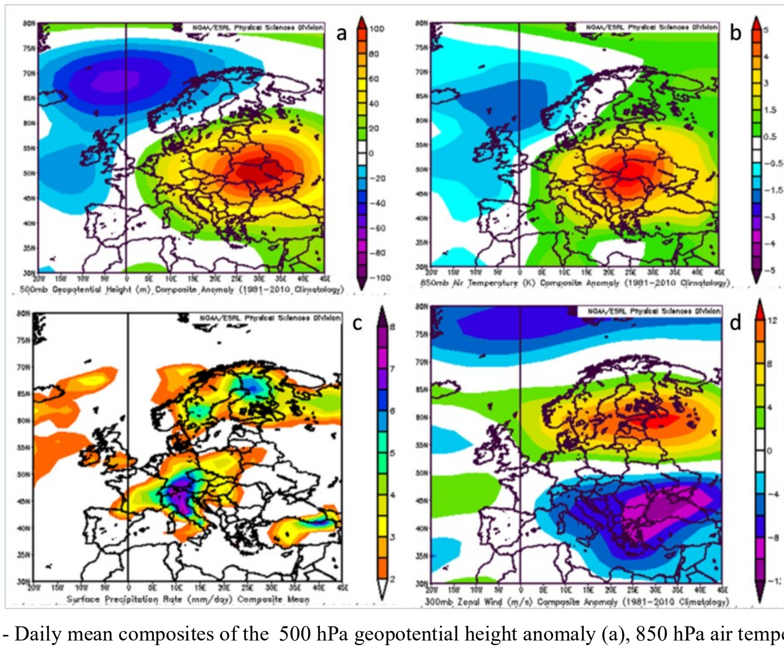

Fig. 8 - Daily mean composites of the 500 hPa geopotential height anomaly (a), 850 hPa air temperature anomaly (b), surface precipitation rate mean (c), 300 hPa zonal wind anomaly (d) - NCEP/NCAR

Synoptic results. The synoptic situation for the days within the top 35 daily values for nitrogen oxides concentrations are characterized by a high pressure anomaly over Central and Eastern Europe (Figure 7a). Concerning the air temperature the emergence of a cold air mass in the North-Eastern region of Romania (Figure 7b) is observed. According to 500 hPa geopotential aneomaly and 300 hPa wind anomaly such conditions are characterizing an intense blocking activity at continental level. These conditions involves a high atmospheric pressure field in the south and the center of the continent (Figure 7d) which is sustained by a high altitude ridge towards the north of the Scandinavian Peninsula (Trigo et al, 2007). These synoptic characteristics (dry and cold) supports the development of thermal inversions that favor the accumulation of NOx and their non-dispersion (Escourru, 1999). Being known that NOx high concentration are specific for the cold season, it is clearly that anticyclonic conditions over central Europe are the most prone for the occurrence of NO_x pollution in Iași city.

The synoptic situation associated with high O_3 concentration is is controlled by a deep low-pressure anomaly located in north-western Europe and a high pressure anomaly located over its eastern and southeast part (Figure 8a). Thus, this pressure pattern favors the warm air advection over the central and central eastern part of Europe, implicitly on the territory of Romania. This effect is seen in the 850 hPa air temperature anomaly that reach its maximum in the nort-east Romania (Figure 8b). It should be underline that eastern and southeastern part of Europe the precipitations are absent as a result of the advance of hot and dry air in North Africa (Figure 8c). The synoptic situation corresponds to heat waves in Romania, where, according to the recent study (Sfîcă et al. 2017) they have an increasing frequency, where 40% of the heat waves recorded in Romania for 55 years are concentrated in only 15 years. In this idea, we should espect an increase in O_3 concentration during summer in our region.

It is noteworthy that the continental pattern of 300 hPa wind anomaly is similar for O_3 and NO_x high concentrations (Figure 7c, 8d), indicating both blocking activity over the continent.

4. Conclusions

Depending on the pollutants, they can be influenced differently by weather elements. Generally, NO_x are increased during the highest minimum temperature and O_3 are increased when highest maximum temperature is recorded. Secondly, the low wind speed and the N-NV wind direction can increase the concentration of the NOx and the high wind speed (7 m/s) with South-South West direction can increase significantly the concentration of the tropospheric O_3.

This context above offers the possibility for local climate to express themselves in pollutants concentration, especially as an effect of thermal inversions and wind sheltering. According to the composite analysis, the rural monitoring points are more influenced by meteorological elements than the urban or industrial monitoring stations.

From a synoptic point of view, during the daily maximum of pollutants concentration, Central and Eastern Europe showed a high pressure or a positive pressure anomaly which is a marker of blocking activity over the continent.

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