Atmospheric Pollution Research 6 (2015) 824-834

Atmospheric Pollution Research

www.atmospolres.com

Annual air pollution level of major primary pollutants in Greater Area of Bucharest

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ABSTRACT

In the international context of incomplete information on air pollution in East Europe, we made a review–image of the air pollution problem in Bucharest metropolitan area, Romania, by assessing concentrations, variability, and compliance with the EU regulations of the primary pollutant levels (PM_{10}, $PM_{2.5}$, SO_2, CO, and NO_X) from eight sampling stations for six years of continuous sampling (2005–2010) and analyzing for factors affecting the seasonal and spatial variations of PM levels. Investigation of temporal and spatial variation of PM_{10} and $PM_{2.5}$ concentrations, as well as their relationships with the measured gaseous air pollutants and with meteorological variables includes correlation analysis, trend analysis, multiple linear regression analysis, and atmospheric back–trajectory analysis. Starting with systematic exceedances of the limit values in 2005 for PM_{10} and NO_X, we observed negative trends for all main pollutants at majority of sites. Exception appears in the background levels where no major improvement was seen. SO_2 and CO were found in compliance with the EU regulations in 2010, but PM_{10} and NO_X still remain a problem. Over the years, $PM_{2.5}$ represents a significant fraction (70–80%) of PM_{10}, irrespective the type of monitoring site. PM levels are higher than those registered for other cities over the Western, Central, and Northern Europe. Combustion–related PM_{10} fraction varies from 73% (cold season) to 59% (warm season) and is higher with 22–26% than in similar polluted area in Southeast Europe. The contributions are site dependent but the background sites experience comparative combustion–related PM_{10} contributions to that of sites within Bucharest ring. Wind speed and temperature are the main factors that influence the PM levels; atmospheric pressure and humidity play a secondary role. Pollution events in Bucharest area are mainly caused by local anthropogenic emissions and not by advections from long distances.

Keywords: *Air pollution, Eastern Europe, particulate matter, gaseous pollutants*

doi: 10.5094/APR.2015.091

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Article History:
Received: 13 August 2014
Revised: 01 March 2015
Accepted: 01 March 2015

1. Introduction

Over the last 50 years, many publications dealing with different issues of air pollution due to particulates and with their environmental effects including reviews of air pollution conditions and detailed long–term monitoring studies (e.g. Blanchard et al., 2013a; Blanchard et al., 2013b) have been published worldwide. With respect to Europe, most of reports cover Western, Northern, and Central Europe (e.g. Bellander et al., 1999; Lenschow et al., 2001; Vallius et al., 2003; Querol et al., 2004; Kukkonen et al., 2005; Salvador et al., 2007; Querol et al., 2008a; Querol et al., 2008b) and some of them analyzed Mediterranean area (e.g. Kocak et al., 2007; Cusack et al., 2012; Onat et al., 2013). For Eastern Europe, the publications are not numerous, even if an increasing number of studies for some urban areas were reported from the 2000s (e.g. Houthuijs et al., 2001; Rajsic et al., 2004; Chaloulakou et al., 2005; Arsene et al., 2011). Some comparison studies between geographically different sites in Europe were also performed (e.g. Vardoulakis and Kassomenos, 2008). Modeling studies at large–scale including East Europe in their simulation domain bring valuable information on regional pollution but they cannot capture the details at local scale, which can be viewed from in–situ measurements. Information on pollution due to particulate matter (PM) and main gaseous pollutants in Romania (e.g. Balaceanu and Stefan, 2004; Arsene et al., 2011; Grigoras and Mocioaca, 2012) is still very scarce in the mainstream of scientific journals despite of the annual reports of National Agency of Environmental Protection and other governmental institutions and despite the importance determined by, for example, PM effects on health (e.g. Pope and Dockery, 2006) or climate (e.g. Iorga and Stefan, 2007; Iorga et al., 2007; IPCC, 2007), which are multiple and complex. There are very few studies with in–situ measurements reported so far including Bucharest. During CESAR (Central European Study on Air Pollution and Respiratory Health) Project, PM_{10} (particles of diameter less than 10 µm) and $PM_{2.5}$ (particles of diameter less than 2.5 µm) concentrations were measured between November 1995 and October 1996, and their levels were between 73 (non–heating period) and 78 µg m^{-3} (heating period) for PM_{10} and between 35 and 57 µg m^{-3} for $PM_{2.5}$, respectively (Houthuijs et al., 2001). Citing a report from World Research Institute, Baldasano et al. (2003) give the following mean annual concentrations in 1995: 82 µg m^{-3} for total suspended particles, 10 µg m^{-3} for SO_2 and of 71 µg m^{-3} for NO_2 in Bucharest. Partial investigations following the specific interest at due moment using some of the measured concentrations of principal air pollutants from Air Quality (AQ) Monitoring Network in Bucharest were presented by (Raducan and Stefan, 2009; Raicu and Iorga, 2009; Balaceanu and Iorga, 2010; Stefan et al., 2014). In order to bring more information on PM in Bucharest, recent researches started to report additional data, including aerosol optical properties and micro–morphological aspects (e.g. Radu et al., 2008; Barladeanu et al., 2012; Olaru et al., 2012a; Olaru et al., 2012b), while Iorga and collaborators (unpublished data) determined the size–segregated mass concentrations of water–soluble ions and carbonaceous fractions in different size ranges between 0.06 and 16 µm in first intensive field campaign in summer of 2010. Nevertheless, such

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data can be matched with collected observations by the Bucharest AQ Network between 2005 and 2010 only for very short periods.

Although the few previously mentioned results were reported, to the best of our knowledge, no one has published, for Bucharest or for Romania, analyses of the large datasets of PM and gaseous pollutant concentrations in order to give an overview of the general air pollution covering a long time period and identify factors that influence the PM levels in Bucharest metropolitan area. Using different methods widely recognized, in the present study we aimed for a simple, practical feasible, sufficient accurate and computationally inexpensive approach in order to obtain an image of the air pollution in Bucharest area. The approach could be extended as far as the monitoring station network develops and the corresponding datasets become larger. The present work analyses primary pollutant (PM_{10}, $PM_{2.5}$, SO_2, CO, and NO_X) concentration data simultaneously collected by the Bucharest AQ–monitoring network over the city greater area for the entire year of 2005, as first year when the AQ network was fully operational, and their time evolution up to the end of 2010. We created a synthetic database based on common time periods in order to: (i) assess the annual average concentration levels at all monitoring stations in Bucharest Greater Area (BGA) checking upon the degree of compliance with the EU–legislated air quality standards (EC, 2008) for particulates and primary gaseous pollutants for the entire period 2005–2010, (ii) provide an overall statistical examination of spatial and temporal variation of PM_{10} and $PM_{2.5}$ concentrations, as well as their relationships with other measured gaseous air pollutants, and to investigate the possible factors affecting the seasonal and spatial variations of PM levels in BGA; (iii) compare PM levels recorded in BGA with measured concentrations reported for other European sites, with a focus on southern Europe.

2. Data and Methods

2.1 Experimental set–up, air quality, and meteorological data

The capital of Romania, Bucharest (approx. 44°26'N, 26°06'E), represents the most developed city of the country, and is located less than 70 km North from the Danube River. Detailed information about city can be found in the Supporting Material (SM). The monitoring sites of the Air Quality Network of Bucharest (Figure 1) are distributed at different spatial levels (inner core city, larger urban zone and sub–city area) covering the main types of anthropogenic activities:

- Two traffic sites in the core–city: Mihai Bravu (MB) and Cercul Militar (CM, in the very center of the Bucharest);
- Three industrial sites: Drumul Taberei (DT), Titan (TT) and Berceni (BE), located near industrial platforms of the city and in proximity of the largest thermo–electrical power plants and residencies;
- Three background stations: Lacul Morii (LM, urban background site), Magurele (MA, suburban background, located near of the surrounding ring of Bucharest that is used for heavy transport) and Balotesti (BL, regional background site, located north, outside the main area of the city at about 22 km).

Beginning with 2004, particulates PM_{10} and $PM_{2.5}$ and gaseous pollutants (NO_2, NO_X, CO, SO_2, O_3) were monitored at above stations on a daily and hourly basis, from midnight to midnight, respectively. The determination techniques were chemiluminescence (NO_X), UV fluorescence (SO_2), non–dispersive IR absorption (CO) and UV photometry (O_3). All analyzers have the detection limit below 1 ppb, precision 1 ppb, resolution 0.001 ppm (0.0001 ppm for O_3), and linearity ±1% over the temperature range 5–40 °C. PM_{10} and $PM_{2.5}$ mass concentrations were obtained by gravimetric method following the standards SR EN 12341:2002 (ASRO, 2002) and SR EN 14907:2006 (ASRO, 2006), the repeatability equals to 1 $\mu g\, m^{-3}$ for 24–h averages. Many differences in the monitoring periods during the year 2004 were found and we excluded these records from our analysis.

We focus here on PM_{10} and $PM_{2.5,}$ and NO_X, SO_2, CO (Table 1), as primary gaseous pollutants that accumulate in urban atmosphere and significantly contribute to the photochemical formation of ozone and other oxidants and to a fraction of the particulate matter (Monks, 2000). O_3 daily averages were added in Section 3.2 in order to seek if they could help to a better understanding of the correlations between particulates and primary gaseous pollutants. A synthetic database of daily averaged datasets of pollutants and local meteorology series (air temperatures, relative humidity, atmospheric pressure, wind speed and direction) was prepared in order to have completeness for all sites for the whole period 2005–2010 by merging data from National Environmental Protection Agency, European AQ Database Airbase (EEA, 2012), and National Meteorological Administration. Conversion to daily averages of hourly gaseous pollutants and local meteorology data was done by averaging over 24–h periods from midnight to midnight.

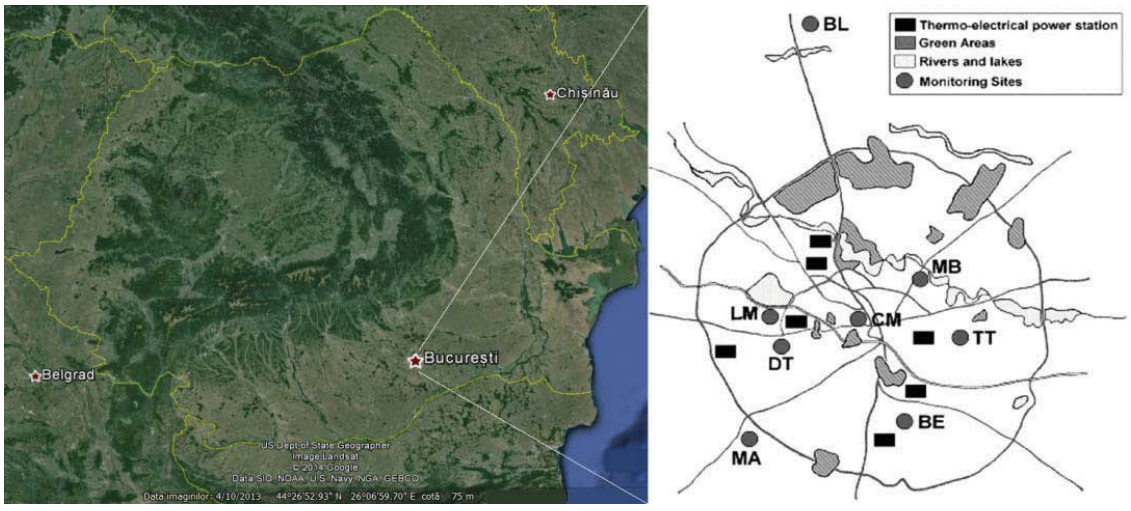

Figure 1. *Map showing Romania and the AQ network in Bucharest Greater Area (grey circles represent the monitoring stations; black rectangles show major thermo–electrical power plants).*

Table 1. *Monitoring stations in Bucharest Greater Area*

Station Name, Site Designation	Station Type	Latitude	Longitude	Altitude [a] (m a.s.l.)	Pollutants Monitored and Included in Analysis
Mihai Bravu, MB	Traffic	44°26'26" N	26°09'04" E	81	PM_{10}, NO_X, SO_2, CO
Cercul Militar, CM	Traffic	44°25'44" N	26°07'15" E	80	PM_{10}, $PM_{2.5}$, NO_X, SO_2, CO
Drumul Taberei, DT	Industrial	44°24'42" N	26°03'08" E	89	PM_{10}, $PM_{2.5}$, NO_X, SO_2, CO
Titan, TT	Industrial	44°24'40" N	26°11'05" E	71	PM_{10}, NO_X, SO_2, CO
Berceni, BE	Industrial	44°23'45" N	26°08'54" E	81	PM_{10}, NO_X, SO_2, CO
Lacul Morii, LM	Urban background	44°26'33" N	26°03'36" E	90	PM_{10}, $PM_{2.5}$ [b], NO_X, SO_2, CO
Magurele, MA	Suburban background	44°20'56" N	26°02'01" E	72	PM_{10}, NO_X, SO_2, CO
Balotesti, BL	Regional background	44°37'12" N [a]	26°05'11" E [a]	94 [a]	PM_{10}, $PM_{2.5}$ [c], NO_X, SO_2, CO

[a] *Estimated coordinates, BL station is located within a military unit, exact coordinates not available*
[b] *Starting with 2009*
[c] *Between 2005–2007*

2.2. Statistical analyses and the selection of pollution episodes

Statistical examination of temporal and spatial variation of PM_{10} and $PM_{2.5}$ concentrations, as well as their relationships with the measured gaseous air pollutants and meteorological variables includes:

- Correlation analysis, expressed by Pearson coefficients, statistically significant at 95% confidence interval;
- Linear regression and multiple linear regression analysis, between daily PM as the dependent variable and meteorological factors and gaseous pollutants as independent variables, respectively. Traditionally, the heating period in Bucharest is centrally imposed, starts on 15th October and stops on 15th April, and corresponds (within a few days) to cold (15th October – 14th April), and warm season (15th April – 14th October), respectively. Therefore, differences in PM concentrations between cold (with heating) and warm (without heating) season were explored, as we expect that higher levels of pollutant concentrations might occur due to the domestic heating.
- Temporal trend analysis for detecting and estimating a monotonic annual and seasonal trend of ambient pollutant concentrations was performed using the non–parametric Mann–Kendall's test and Sen's method using the MAKESENS software (Salmi et al., 2002). The calculated trends (as percent change in the concentration per unit time) were calculated for minimum data coverage of 70% of valid data per site and per year for at least 5 years out of 6 (exception was BL site and for $PM_{2.5}$ data were percentages slightly below 70% for 2005 were accepted) and results were considered statistically significant for level of significance 0.1 or lower. For linear regression and correlation analyses, the same minimum data coverage as in trend analysis was used.
- Atmospheric back–trajectory analysis for each day corresponding to a pollution episode was performed in order to provide additional arguments for the cause of a pollution episode (influence from local sources or atmospheric long–range transport).

Selection of pollution episodes proved to be a difficult task. The first criterion for selecting a particular episode was that 24–h PM_{10} concentration to exceed the limit value of 50 μg m^{-3} at each of the monitoring sites. Severity of the condition (very large number of exceedances at all sites, as in the SM is shown) determined us to use two new criteria: by each monitoring site, a particular episode was be attributed if 24–h PM_{10} concentration exceeded the PM_{10} annual average plus one or two times standard deviation (SD), which translates into 68% or 95% of the measured daily values of PM_{10} that fit within the interval of ±1 SD and ±2 SD, respectively. Episodes when PM_{10} levels are higher than PM_{10} annual average plus 1 SD but less than PM_{10} annual average plus 2 SD were named "pollution episodes (events)" (criterion 1), whilst the episodes when PM_{10} levels exceeded PM_{10} annual average plus 2 SD named "strong pollution episodes (events)" (criterion 2). Both of these criteria led to a corresponding analyzable number of pollution events. Analysis of pollution events in 2005 is presented in the SM.

3. Results and Discussion

3.1 Air pollution levels and trends in Bucharest Greater Area between 2005 and 2010

The overall picture of primary pollutants. Figure 2a and Figure 2b provide a box–plot comparison of the annual levels of daily averages of PM_{10}, and $PM_{2.5}$ mass concentrations by site for the period 2005–2010. Results for annual levels of gaseous pollutants are available in the SM, Figures S4–S6. Observed annual concentrations (mean, median, and 95% percentile) of all pollutants indicate that all sampling sites within the Bucharest inner area experience the relatively same pollution level. At traffic, industrial, urban background and even suburban background site, majority of 24–h PM_{10} ($PM_{2.5}$) concentrations are up to 150 μg m^{-3} (120 μg m^{-3}). Gaseous pollutants show a little variability between measurement sites: while NO_X reach highest levels at traffic sites MB, and respectively CM, of about 400–500 μg m^{-3}, NO_X concentrations at industrial sites may reach 300 μg m^{-3} (TT and BE) to 400 μg m^{-3} (DT). Urban (LM) and suburban background (MA) sites register NO_X values up to about 250–200 μg m^{-3}. SO_2 and CO concentrations are generally below 100 μg m^{-3} (6 mg m^{-3}, respectively) and only occasionally they go up to over 120 μg m^{-3} (8 mg m^{-3}) at traffic sites during 2005-2008. Average and median values show a minimum in 2007–2008, which we identify with the moment of a massive vehicle fleet change during a national program from the old technology gasoline cars to diesel cars. The dip in pollutant levels during 2007–2008, obvious in PM_{10} and $PM_{2.5}$, is not as clear in the annual gaseous pollutants concentrations. For some sites, as BE and LM, a drop in the 95th percentile SO_2 concentrations are observed whereas the mean levels increase.

It seems that only the regional background site BL has a clear lower pollution level comparing with the rest of the sites. However, the box–plots of CO concentrations measured at BL show increases in average, median and maximum values.

A good correlation (R^2>0.8) was found between 24–h average mass concentrations of $PM_{2.5}$ and PM_{10} during the entire period of 2005–2010. The average $PM_{2.5}/PM_{10}$ mass ratios in BGA fall in the range of 0.7–0.8 indicating the fine fraction contributed significantly to all samples. Lower $PM_{2.5}/PM_{10}$ mass ratios of about 0.5–0.6 indicating a contribution of coarse natural particles were observed only occasionally like in the period 11th–20th June 2010 when the air masses came from Sahara desert (see the SM). Our observations lie within the range of $PM_{2.5}/PM_{10}$ mass ratios (0.5–0.9) measured at most sites across the Europe (Putaud et al., 2010).

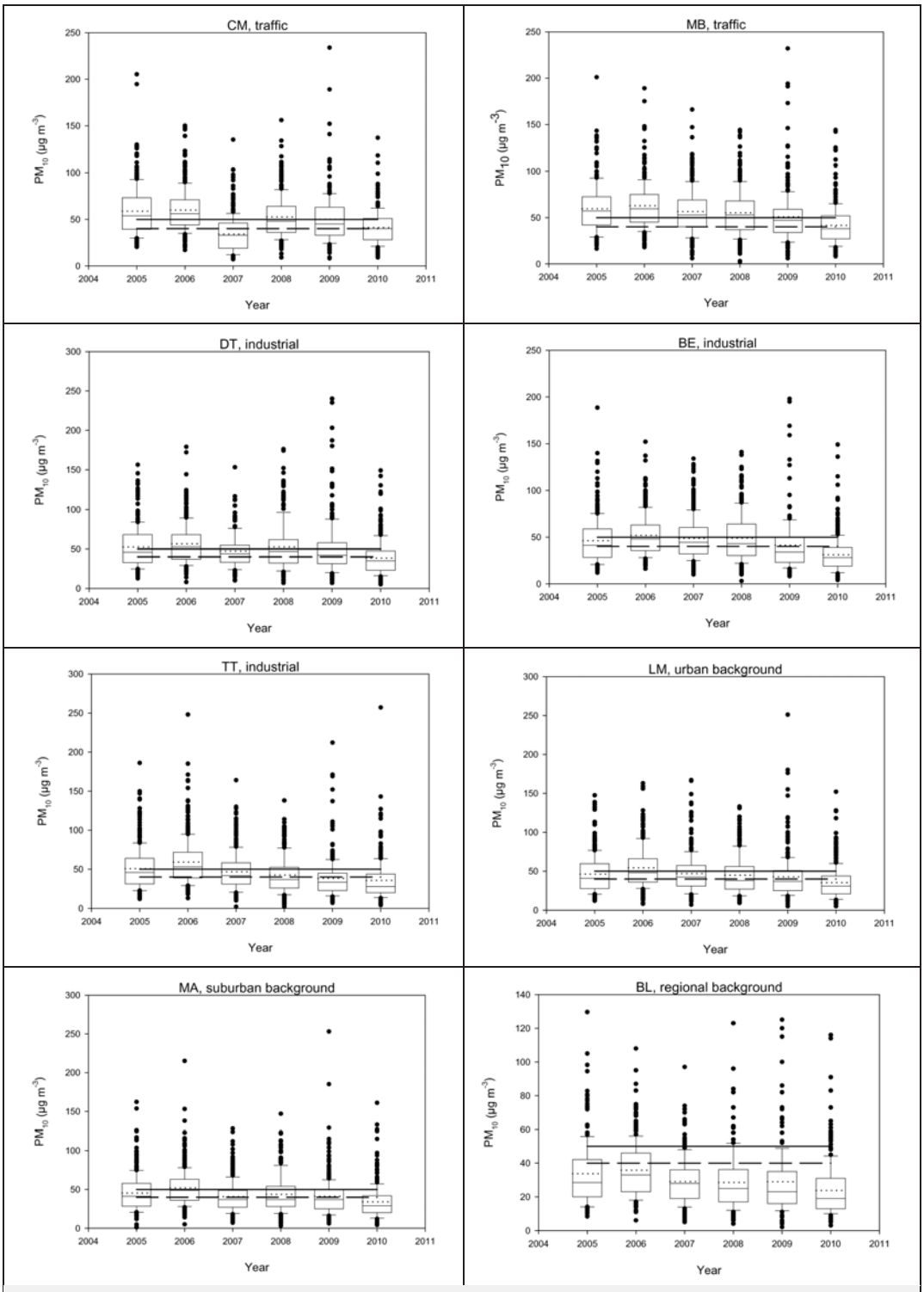

Figure 2a. *Levels of daily PM_{10} mass concentrations for the period 2005–2010 in BGA, including median, the 5th, 25th, 75th, and the 95th percentiles of their 24–h concentrations. Dotted line represents the annual average. The EU annual limit value of 40 µg m^{-3} (long dashed line) and EU 24–h limit of 50 µg m^{-3} (long continuous line) are included. Black circles represent first and last 5% of observed PM_{10}.*

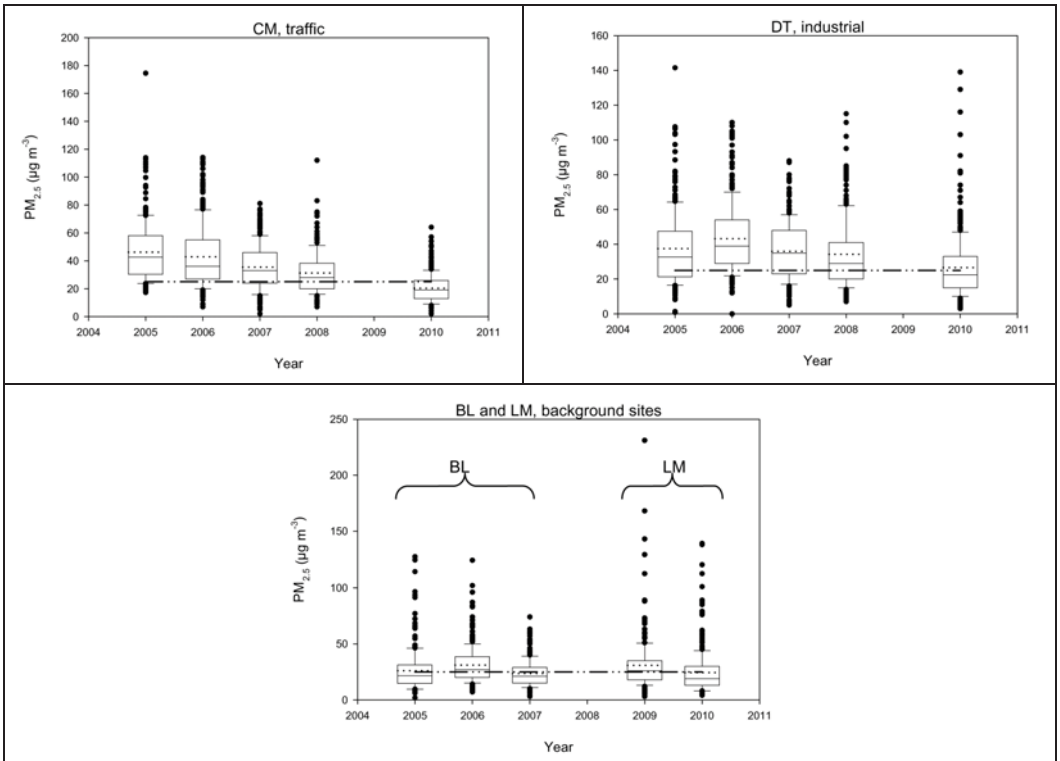

Figure 2b. *As in Figure 2a, but for $PM_{2.5}$ concentrations. Dash–dot–dot line represents the EU–2010 target value of 25 μg m^{-3}.*

The calculated differences between 2005 and 2010 in PM_{10} ($PM_{2.5}$) average seasonal levels show a site–dependent variation from 10 (7) μg m^{-3} to 25 (33) μg m^{-3} during cold season and from 8 (1) to 21 (18) μg m^{-3} during the warm season, with lowest differences in concentrations at background sites and the highest at the traffic site.

Pollutant annual and seasonal average concentrations at all sites were further investigated in order to determine if temporal trends could be revealed. Calculated annual and seasonal trends of PM_{10}, $PM_{2.5}$, NO_X, SO_2, CO by site, and examples of time series and trends are shown in Table S1 and Figure S3 (see the SM) at level of significance at least 0.1. All sites showed a significant annual decreasing trend for PM_{10}, with the most pronounced trend at the MB traffic site, closely followed by the city center (CM). Even if they are not always as strong as for particulates, gaseous pollutants also present negative trends. Some exceptions appear when the calculated Sen's slopes of the trend was found to be non–significant at the imposed level of significance or, as in the case of industrial site BE, a probably increasing trend in SO_2 and CO was found. Seasonal trends were more difficult to mark out; mostly seasonal trends in particulates were found significant. In general, the calculated trends for BGA are small. As an example, the SO_2 annual trends varied between –1.28% yr^{-1} and –3.73% yr^{-1} in Bucharest area. More pronounced reductions (from –6.6 to –14.9% yr^{-1}) were recently reported for different locations in western Europe, UK (Jones and Harrison, 2011) and between –5.1 and –9.7% yr^{-1} in southeastern United States (Blanchard et al., 2013a). Blanchard et al. (2013a) also reported decreases of annual mean CO and NO_X concentrations at rates ranging from 1.2 to 7.2% yr^{-1} (6.0 to 9.0% yr^{-1}) which are also higher than the corresponding decreasing rates determined for Bucharest area. At the background sites BL and MA, both located outside the Bucharest core, no major improvement has been attained in 2010 for the air quality despite of the regulations applied to the major pollutant sources within the city. It can be seen a general decrease from 2005 to 2010 only for SO_2: calculated annual trends are of –1.28% yr^{-1} at MA and –1.36% yr^{-1} at BL whereas seasonal trends were found to be significant only at MA site (–1.16% yr^{-1} in cold season and –1.33% yr^{-1} in warm season). CO trends were found non–significant for MA site whereas a probable positive trend for BL site is suggested in the context of the increase of annual CO national emissions (see the SM, Figure S3). All pollutant trends are less pronounced at BL; moreover, while negative trends for PM_{10} and SO_2 were detected, a slow positive trend was calculated for CO. A positive CO trend for MA site was found non–significant.

Decreasing trend in gaseous pollutants observations follows generally the reduction in emissions (e.g. Torseth et al., 2012) at EU scale and at the country scale (local or regional emissions were not available), as a result of implementations of reduction policies like using fuels with lower sulfur content at the power generation stations (see the SM, Figure S2). Availability of natural gas for domestic heating has also increased recently and this should translate into a reduction of both CO and SO_2. The period 2007–2010 also corresponds to the period of economic boom in Romania. Therefore, implementation of strategy environmental development programs, including modernization of power plants facilities, transport infrastructure, and the vehicle fleet change in 2007–2008 superposed in the context of a fast economic development in Bucharest Greater Area. Probably the emissions strengths in Figure S2 (if we assume that emission trends at the country scale could be applied to Bucharest area), the lack of experience in implementation of reduction policies, together with the shorter length of our data sets might be a part of the explanation and currently the direction of the trends represents the most important finding.

Relationship of primary pollutants to air quality objectives and EU limit values. The PM_{10} EU limits (EC, 2008) were widely exceeded in the early studied period. Despite of the negative annual trends of PM_{10} concentration at all sites (see also the SM, Table S1), and the fact that the annual limit of 40 μg m^{-3} in force during 2005–2009 is exceeded in 2010 only at the traffic sites by 2 μg m^{-3}, the new EU–2010 annual limit value of 20 μg m^{-3} is still exceeded. Only at the background site BL the annual level (24 μg m^{-3}) is close to

the limit value. At the other sites, annual concentrations increase by factors from 1.5 (industrial sites) to about 2 at traffic site in the very center of the city. Figure 2b shows that $PM_{2.5}$ annual concentrations are slightly below the EU–2010 target value of 25 μg m^{-3} at traffic site (20.4 μg m^{-3}) and urban background site (24.4 μg m^{-3}) and above it at the industrial site (26.58 μg m^{-3}).

Although in 2005 some exceedances were found, the compliance with the EU regulations are evident in 2010 for both SO and CO concentrations. By contrast, nitrogen oxide concentrations remain in 2010 above the annual limit for protection of ecosystems at all sites within the city and we identify hotspots as about 350 μg m^{-3} at both traffic sites, which are characterized by annual average concentrations of 109 μg m^{-3} (CM), and of 92 μg m^{-3} (MB). However, depending on the site, the maximum values are much more variable: 162 μg m^{-3} (LM and MA), 260 μg m^{-3} (BE) to 340 μg m^{-3} (DT), pointing to the local factors in Bucharest.

3.2 Factors affecting the particulate matter levels

In this Section we investigated some factors affecting the PM levels in Bucharest area in order to understand how local environmental characteristics concur to air pollution pattern.

Particulate matter inter–site correlations. Inter–site correlation analysis was performed for each year using daily PM_{10} values. Because no significant differences were found in the pattern of the correlation coefficients by each year, we present PM_{10} inter–site correlation coefficients in 2010 (Table 2). All sites in BGA show a positive correlation (coefficients vary from 0.66 to 0.93) suggesting they all suffer from probably the same sources affecting the whole area, including industrial and the relatively well–distributed traffic sources. In 2005, correlation coefficients were lower (from 0.55 to 0.88) but the small difference could be explained by the increasing sampling performance over the years. Correlation coefficients from 0.55 to 0.84 were also found in South–Eastern Europe, Athens Area (Grivas et al., 2008).

Highest correlation coefficients are between the urban background LM site and traffic site CM (0.84), followed by industrial sites BE (0.88) and DT (0.93). Followed by industrial sites BE (0.88) and DT (0.93). TT and DT sites are also quite well correlated. This suggests that LM site is exposed to the influence from the East and Southeastern part of the city, in favorable synoptic conditions, in addition to the high traffic on the nearby avenue (Figure 1). As the altitude slightly increases from the Southeast to the Western part of the city (Table 1), following more or less the Dambovita riverbed and being in the close proximity with LM, the site DT also receives air masses carrying the pollution from E–SE direction; the wind direction data (E–NE/W–SW is the prevailing wind direction) and linear analysis of PM_{10} and $PM_{2.5}$ versus each gaseous pollutant support this hypothesis. The lowest correlation coefficients were obtained for the background site BL (in north of the city, not in the main path of the air masses) which is characterized by very low traffic (average local $PM_{2.5}/PM_{10}$ mass ratio is 0.77). Probably the pollution from the inner city spreads over the larger area including this background site. Area's topography with slow altitude variations point also to this conclusion (Grigoras and Mocioaca, 2012). The inter–sites correlation based on CO data (as an alternative indicator) in 2005 (correlation based on 70% of daily averaged concentrations), 2010 (80%) and for the entire period 2005–2010 (76%) shows similar correlation pattern to that found using PM_{10}.

Relationships between PM and gaseous pollutants. Table 3 synthesizes the relationships between daily means of PM_{10} and $PM_{2.5}$ and gaseous pollutants over the entire period 2005–2010, separately for the cold (15th October–14th April) and warm (15th April–14th October) season. It shows reasonable correlations between both PM_{10} and $PM_{2.5}$ and NO_X (highest coefficients), suggesting a common road traffic origin. Correlation PM_{10}–CO and $PM_{2.5}$–CO follows in rank, followed by a less defined correlation with SO_2. Similar correlation coefficients (0.5–0.6 for $PM_{2.5}$–NO_X relationship, about 0.4 for $PM_{2.5}$–CO and about 0.3 for $PM_{2.5}$–CO) were reported by Harrison et al. (2012) at different sites in UK. However, the strength of these correlations varies among sites and between seasons. Some seasonality at some sites can be observed in PM_{10} and $PM_{2.5}$–CO and SO_2 relationships (with higher coefficients during cold season, better pronounced for MA and BL sites). Hardly any difference in coefficients is found for PM_{10}–NO_X relationship between seasons, although the correlation $PM_{2.5}$–NO_X points to seasonal variation both inside and outside the city area. Higher $PM_{2.5}$–SO_2 coefficient (0.44) was found at the traffic site during cold season, which is probably due to SO_2 associated with the residual sulfur in vehicle fleet (Arsene et al., 2011; Harrison et al., 2012). Daily averaged particulate matter concentrations correlated always negatively in cold season and mostly negatively with O_3 (with few exceptions during warm season), but they were less defined than the rest of the relationships and less than correlations found in UK. The negative PM_{10} and $PM_{2.5}$–O_3 correlations indicate thus lower O_3 concentrations being associated with higher particulates concentrations (associated with increased NO_X, which leads to lower O_3 concentrations). As in some situations in the UK atmosphere (Harrison et al., 2012), short periods with positive correlation particulate matter–O_3 during photochemical episodes were reported in Bucharest Greater Area during 2005–2007 by Stefan et al. (2014). They identified a primary maximum in April and a secondary maximum in July, with a difference between springtime and summertime maxima higher at BL site compared to CM which. These results fit into the European mean seasonal cycle of O_3 containing a springtime maximum of background origin and a summer peak due to ozone precursors' emissions (Monks, 2000) and a diurnal amplitude of O_3 variability is getting higher as we move from northern to southern latitudes. Our positive correlation coefficients might indicate such situations when both particulate matter and O_3 are generated by photochemical activity for some sites in warm season but the calculated coefficients are very low and maybe these episodes are swamped by the 6–year analysis. In this respect and due to the lack of detailed information on ozone precursors' (abundances, reactivity) during our study period in the Bucharest area, we conclude that there are insufficient results to generate a robust picture at the moment.

Table 2. *Pearson inter–sites correlation coefficients using daily PM_{10} concentrations in 2010*

Site	MB	CM	DT	TT	BE	LM	MA	BL
MB	1.00							
CM	0.86	1.00						
DT	0.81	0.86	1.00					
TT	0.77	0.78	0.85	1.00				
BE	0.86	0.89	0.83	0.90	1.00			
LM	0.78	0.84	0.93	0.81	0.88	1.00		
MA	0.79	0.76	0.80	0.71	0.82	0.85	1.00	
BL	0.72	0.74	0.74	0.66	0.74	0.74	0.71	1.00

Table 3. *Pearson correlation coefficients between daily PM_{10} and $PM_{2.5}$ and daily averaged gaseous pollutant concentrations in cold (15th October–14th April) and warm (15th April–14th October) seasons during the period 2005–2010*

Site/Season	CO		NO_X		O_3		SO_2	
	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm
				PM_{10}				
MB	0.18	0.37	0.69	0.60	–0.12	–0.34	0.19	0.18
CM	0.34	0.35	0.45	0.52	–0.04	–0.31	0.33	0.17
DT	0.16	0.18	0.64	0.59	–0.11	–0.12	0.10	0.08
TT	0.55	0.53	0.60	0.70	–0.13	–0.04	0.35	0.06
BE	0.26	0.07	0.56	0.58	–0.03	0.08	0.17	–0.10
LM	0.19	0.29	0.63	0.43	–0.14	0.05	0.25	–0.02
MA	0.34	–0.04	0.58	0.58	–0.02	0.08	0.16	0.11
BL	0.22	–0.04	0.59	0.38	–0.10	0.24	0.40	0.17
				$PM_{2.5}$				
CM	0.46	0.04	0.45	0.50	0.01	–0.06	0.44	0.01
DT	0.23	0.04	0.47	0.56	–0.02	0.03	0.13	0.11
LM [a]	0.61	0.51	0.73	0.38	–0.01	–0.17	0.18	0.18
BL [b]	0.49	0.14	0.58	0.30	0.10	0.12	0.24	0.10

[a] *Based on samples between 2009 and 2010*
[b] *Based on samples between 2005 and 2007*

The associations between PM_{10} and primary gaseous pollutant levels in Bucharest's specific situation were investigated further by multiple linear regressions performed using daily mean PM_{10} values and daily averaged gaseous pollutants NO_X, SO_2, CO for the same seasonal periods. Linear regression analysis proposed by Harrison et al. (1997) who describe it in detail, is frequently used for urban areas (Deacon et al., 1997; Fuller et al., 2002; Thorpe et al., 2007) and has been applied earlier in Athens, South–Eastern Europe, by e.g. Vardoulakis and Kassomenos (2008). Various studies in urban areas (or paired urban/sub–urban sites) indicate NO_X can be used as a tracer for traffic combustion, SO_2 for large–scale combustion and CO for small–scale combustion (see the SM, Text S2). Based on the information from Romanian Emission Inventory Reports (Text S2), we assumed NO_X, SO_2 and CO as tracers for combustion, a major anthropogenic activity in BGA, and we estimated (modeled) the seasonal combustion–related processes' associations (slopes) due to concurrent presence of primary pollutants to PM_{10} in Bucharest. The intercepts we assume to represent the associations of PM_{10} with non–combustion processes. Figure 3 shows the seasonal mean non– and combustion–related PM_{10} in Bucharest Greater Area in 2005–2010.

Results show for Bucharest Greater Area that an average of 66% of PM_{10} is related to combustion processes and 34% of PM_{10} represents the other sources. The overall combustion processes contributed to 73% in cold season, and to 59% in warm season. During the warm season the non–combustion–related PM_{10} levels are increased, the heating processes can therefore lead to an about 14% difference in PM_{10} concentrations. This important finding is emphasized by the comparison of our data with Athens' situation (Vardoulakis and Kassomenos, 2008) and we indicate a higher contribution of the combustion processes in BGA with about 22% (cold season) –26% (warm season) with respect to Athens.

The contributions are site dependent, with a maximum difference encountered at the traffic site MB. The industrial site DT encounters the same contribution from combustion in cold season (72%) and the warm season (75%). The same situation appears for the industrial site BE (69% cold versus 73% warm season). This suggests that thermo-electrical power stations located in these areas constantly operate in similar parameters over the year. Some activities during the warm season on the industrial platform close to BE site possibly contribute to the higher percentage in warm season (see also the trends in Table S1 in the SM). The suburban background site MA shows a different situation: combustion–related PM_{10} is higher during warm period than during cold period with about 16%. Figure 3 also indicates that the background sites receive comparative combustion–related PM_{10} contributions to that of sites within Bucharest ring. This suggests once again that the pollution in the inner city easily disperses over large areas around, making more difficult to separate between local pollution sources and pollution coming from longer distances.

Relationships between PM and meteorological factors. Bucharest's meteorological conditions are presented in the SM, Text S1. Meteorological data at the city scale were available for all years but local meteorology dataset by each sampling site was available only for 2005. To check for existence of different pollution behaviors associated to PM–meteorology relationships within BGA, we made correlations between PM and daily averaged local meteorological variables (Table 4). Correlation analysis between particulates and temperature revealed a different seasonal pattern for PM_{10} and $PM_{2.5}$. Positive correlation with temperature with higher coefficients appeared in the warm season for both PM_{10} and $PM_{2.5}$, which is associated with stronger convection and instable atmospheric conditions (atmospheric pressure). The moderate positive correlation with temperature could also reflect a positive correlation with solar radiation, increased oxidation during summer months for example, and a negative correlation with temperature could be due to increased emissions from space heating combustion appliances. The negative correlation between $PM_{2.5}$ and PM_{10} at some sites in cold season suggests an inverse relation with the temperature and goes to the hypothesis of a reduced dispersion and stable atmospheric conditions, but the correlations are low.

Daily PM_{10} and $PM_{2.5}$ concentrations correlated negatively with relative humidity at all sites whatever the season but with relative higher coefficients during warm season. In hot humid days, pollutants may be scavenged by fog or cloud droplets and deposited onto surfaces leading to lower ambient concentrations. Highest correlation coefficients were found for CM site; therefore city center seems to be more influenced by the relative humidity than all the other sites.

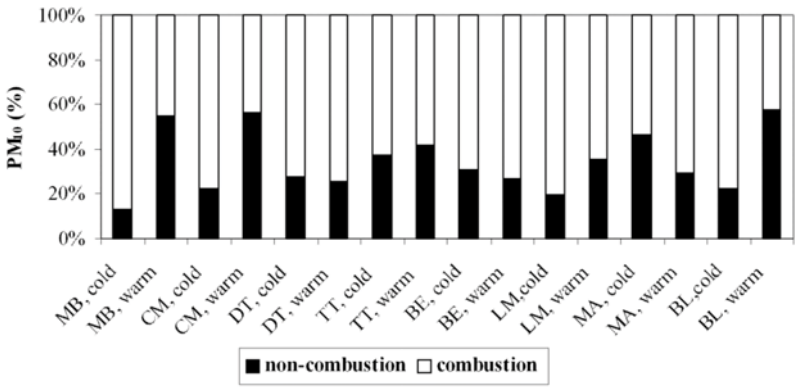

Figure 3. *Seasonal mean non– and combustion–related PM_{10} in BGA.*

Table 4. *Pearson correlation coefficients between daily PM_{10} and $PM_{2.5}$ concentrations and daily averaged local meteorological factors in cold (15th October–14th April) and warm (15th April–14th October) seasons in 2005*

Site/Season	Temperature		Relative Humidity		Wind Speed		Atmospheric Pressure	
	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm
				PM_{10}				
CM	0.02	0.36	–0.11	–0.35	–0.10	–0.39	0.36	0.27
DT	0.06	0.34	–0.09	–0.23	–0.36	–0.50	0.26	0.17
TT	0.09	0.28	–0.03	–0.17	–0.46	–0.33	0.29	0.20
BE	0.05	0.42	–0.03	–0.29	–0.42	–0.55	0.31	0.21
LM	0.08	0.34	–0.03	–0.35	–0.44	–0.38	0.19	0.13
MA	–0.02	0.31	–0.02	–0.17	–0.26	–0.26	0.25	0.27
BL	–0.20	0.38	–0.02	–0.12	–0.25	–0.42	0.14	0.27
				$PM_{2.5}$				
CM	–0.02	0.34	–0.16	–0.45	–0.10	–0.32	0.35	0.26
DT	–0.03	0.35	–0.04	–0.31	–0.35	–0.47	0.24	0.21
BL	–0.11	0.61	–0.02	–0.07	–0.33	–0.46	0.15	0.14

Finally, PM and wind speed are also negatively correlated at all sites, in both seasons: $-0.10 \leq R \leq -0.46$ (cold season) and $-0.26 \leq R \leq -0.55$ (warm season). This is consistent with the fact that PM concentrations decrease as wind speed and atmospheric dilution increase. The result is a cleaner atmosphere of both fractions of atmospheric particulates as pollutant dispersion became stronger due to stronger winds and as particulate matter is more scavenged in the water vapors. For background MA and BL sites, in cold season, low speed wind conditions and lower temperature could result in a low boundary layer that traps pollution to the ground. In warm season, more intense winds and higher temperature (that could reflect positive correlations with solar radiation) and higher boundary layer could result in pollution transport.

In a recent study, Chu et al. (2010) found that temperature, relative humidity, their squared terms, and their interactions explain much of the variation in airborne concentrations of $PM_{2.5}$ in Pittsburgh, other factors being of less importance. Similar studies on $PM_{2.5}$ in other cities suggest that the relative importance of different factors can vary greatly. Different results were obtained by Vardoulakis and Kassomenos (2008) for Athens and Birmingham. They found a better–defined correlation PM_{10}–wind speed during cold season ($-0.55 \leq R \leq -0.23$) than in warm season ($-0.45 \leq R \leq -0.15$) in both cities. In contrast with the results of Chu et al. (2010) and those for Athens and in similar case with Birmingham, for Bucharest, after the wind speed, the temperature represents a second factor that does influence the concentration of particulates and can explain about 30–40% of this variation. Atmospheric pressure and humidity play the third and fourth role, respectively.

3.3 Comparison with other European sites

Table S6 (see the SM) provides a comparison of PM concentrations by site type for some European sites, with a focus on Southern Europe. Few other sites in Western, Central and extreme Southern part were added. The Vienna urban area was chosen in order to compare with a location where the traffic is the only major contributor to the air pollution. Heating in Vienna is provided by natural gas (32.2%) and by district heating plants (43.5%) and is not expected to have much influence [energy data for 2003 in Hitzenberger et al. (2006)]. Moreover, strict regulations for abatement pollution were in force before 2005. Selection of monitoring sites in Vienna area was done in order to match the location characteristics (wide or canyon streets) of the sites in Bucharest area, and pollution data were derived mostly from the European Air Quality Database (EEA, 2012).

Our study generally shows that air pollution in Bucharest area was higher than in urban atmospheres of Europe. All sites in BGA are also characterized by clearly higher concentrations comparing to Vienna similar sites, the highest difference being for traffic sites by a factor of two, for both PM_{10} and $PM_{2.5}$ fractions. Both traffic sites in Bucharest encountered slightly lower PM_{10} annual averages (59 μg m^{-3} in 2005 to 41 μg m^{-3} in 2010) and similar $PM_{2.5}$ (about 46 μg m^{-3}) to Athens' center (PM_{10}=77 or 60 μg m^{-3} and $PM_{2.5}$=

40 μg m^{-3}) but higher levels than in greater Athens' area (PM_{10}= 54.23 μg m^{-3}) or at kerbside locations in Vienna or Budapest.

Bucharest industrial sites prove to be comparable with Barcelona, while at the MA suburban site PM_{10} concentrations (about 45 μg m^{-3}) are even generally higher than urban background values in Western Europe, and fit within the urban background values in Southern Europe. PM atmospheric concentrations at the regional background site BL indicates this site behaves like a suburban background site in Europe. Bucharest urban background site LM is characterized by concentrations that fits within the concentration range of similar sites and is very close to PM_{10} levels in Graz, despite of the very different topography of these cities (Graz is located in a semi–alpine basin at South of Alps with weak natural ventilation that could lead to high pollution levels, and no heavy industry).

Comparing with the European annual averages by region, calculated from data in European aerosol climatology (Putaud et al., 2010) we find that the PM concentrations at urban background LM site (about 46 μg m^{-3} for PM_{10} and about 27 μg m^{-3} for $PM_{2.5}$, average value 2009–2010) are very close to the urban Southern Europe PM levels, while PM concentrations at regional background BL site (about 34 μg m^{-3} for PM_{10} and of about 26 μg m^{-3} for $PM_{2.5}$, average value 2009–2010) are higher than those corresponding to the urban Southern Europe PM levels.

With respect to the gaseous pollutants, the annual averages of daily values from Airbase database in similar sites in Vienna for the year of 2005 were calculated and compared with the levels registered in Bucharest Greater Area. In the case of nitrogen oxides, we found comparable values between industrial sites and urban background sites (53.86 μg m^{-3} at Vienna, Kaiserebersdorf station and 54.48 μg m^{-3} at Vienna, Stephansplatz), but at traffic sites, both wide and canyon streets, the registered values in Bucharest are higher by a factor of 2.5 than concentrations in Vienna, Wahringer Gurtel and Taborstrasse, respectively. SO_2 is greater in similar sites in Bucharest by a factor ranging between 2 (Kaiserebersdorf: SO_2=6.18 μg m^{-3}) and 3 (Wiener Neustadt: SO_2= 3.96 μg m^{-3}), and CO by about 4 times (canyon street Taborstrasse: CO=0.48 mg m^{-3}). Despite of a clear decrease of all concentration levels of all gaseous pollutants during the 2005–2010 period, it results that ratios of gaseous pollutant concentrations in 2010 versus the corresponding pollutant concentrations in Vienna, range between 1.06 and 4.33, with the exception of traffic sites where CO levels decreased in 2010 to about 0.60 mg m^{-3}, which is comparable with Vienna, Taborstrasse values of 0.48 mg m^{-3} in 2005.

When we compare with the sites in Europe, it could also be of importance the classification of the monitoring sites also. The classification we use is that made by the National Institute in charge with the AQ monitoring at the time of 2005, while the Airbase database mention the BL site as rural background. The newest classification of the monitoring sites in Europe is that of Joly and Peuch (2012). They identified for Bucharest Greater Area: urban, suburban and rural monitoring sites for O_3, NO_2 and SO_2 and no specification was made yet for PM_{10}. Their proposed pollutant–specific classification could help in future, as longer pollutant time series will be available in order to better classify BL station. Analyzing our data over the period 2005–2010, considering the distance to the core city, topography of the region and the measurements, we indicate that BL site behaves as suburban background site and not as regional natural background as it was initially classified in 2004 at the beginning of the monitoring. Our finding completes the pollutant – specific classification of Joly and Peuch (2012) and fits into the classification of Van Dingenen et al. (2004). We consider that a better denomination/classification of a monitoring site should combine pollutant specificity with topography of the surrounding area, in order to get better data compatibility with similar classified sites. Nevertheless, we believe this may affect only the local representativeness of data and will not change significantly the overall picture of the pollution of Bucharest Greater Area when we compare with Europe.

4. Conclusions

In the international context of incomplete information on air pollution in East Europe, six–year PM_{10}, $PM_{2.5}$, NO_X, SO_2, CO concentrations (from January 2005 to December 2010) at eight monitoring sites (traffic, industrial and background) in Bucharest Greater Area were analyzed. This study, first of this type for Bucharest, Romania, reveals the following:

- All pollutants are characterized by a spatial gradient in concentrations across the city area from the highest averages associated with the hotspot traffic sites to the background sites. All sites in BGA show a PM positive correlation suggesting they all suffer from industrial sources affecting the whole area and from the relatively well–distributed traffic sources. Pollution in the inner city seems to have a certain impact over the larger area around, as topography and meteorological factors favor this situation. PM_{10}, $PM_{2.5}$ and NO_X levels at several industrial and traffic hotspots exceeded systematically the recommended EU pollution limits but CO and SO_2 do not put serious problems relative to their limits. Over the years, we observed significant negative trends for all pollutants at majority of sites and a general compliance with EU regulations indicating that control measures started to produce the desired effects; exception appears in the background levels where no major improvement was seen. At traffic sites PM_{10} ($PM_{2.5}$) levels decreased from about 59 (46) μg m^{-3} in 2005 to 41 (21) μg m^{-3} in 2010. At the background site BL, located outside Bucharest, PM_{10} levels diminished from about 34 μg m^{-3} in 2005 to 24 μg m^{-3} in 2010, whereas no clear improvement could be observed for $PM_{2.5}$ which decreased from about 26 μg m^{-3} in 2005 to only 24 μg m^{-3} in 2007, when the monitoring of PM fine fraction stopped here. Nitrogen oxides still remain an issue of concern even in 2010.
- PM levels are general higher than those registered at many other cities over the Western, Central and Northern Europe. Similar levels were observed in Southern Europe (Barcelona), and in South–East Europe (Athens). The combustion processes' correlation to PM_{10} is higher in Bucharest with about 22–26% than in relatively similar polluted urban area (Athens) in South–East Europe.
- $PM_{2.5}/PM_{10}$ ratios generally show a large contribution (70–80%) of fine particles to the particulate mass, but within the range of those reported for Europe.
- An average of 66% of PM_{10} is related to combustion processes and 34% of PM_{10} represents the other sources. The overall combustion processes contributed to 73% in cold season, and to 59% in warm season. During the warm season the non–combustion related PM_{10} levels are increased, as we expected, the heating processes can therefore lead to an about 14% difference in PM_{10} concentrations. The contributions are site dependent, with a maximum difference encountered at the traffic site MB. The background sites experience comparative combustion–related PM_{10} contributions to that of sites within Bucharest ring.
- After the wind speed, the temperature represents an important factor that does influence the concentration of particulates and can explain about 30–40% of this variation. Atmospheric pressure and humidity play secondary roles.
- Identified pollution episodes can reach high concentration levels ranging from 135 μg m^{-3} outside Bucharest to hotspot values of 205 μg m^{-3} in the very center of Bucharest. Pollution events are mainly caused by local anthropogenic emissions and not by advections coming from long distances (based on 2005 data).

Acknowledgements

G. Iorga thanks to Prof. Dr. R. Hitzenberger for her valuable critical feedback on the manuscript and to Dr. J. Burkart for preparing a first form of Table S6. The financial supports provided by UEFISCSU, Project PN–II–ID–PCE–2011–3–0471, Contract: 200/05.10.2011, from ÖAD Austria, Program WTZ, Project RO 02/2009 and from ANCS Romania, Program PN II, Contract: 304/27.04.2009 are gratefully acknowledged. Bucharest pollution data during 2006–2010 were extracted from European AQ database (accessed in January 2012) and some from public ANPM data (accessed in October 2010). Dr. Ing. D. Ciocorva, Leader of the AQ Control Group–ICIM Bucharest, is acknowledged for providing the raw 2005 data and National Meteorology Administration for meteorology data. The authors acknowledge the NOAA Air Resources Laboratory for HYSPLIT transport and dispersion model and READY website (www.arl.noaa.gov/ready.php) used in this publication.

Supporting Material Available

Detailed description of Bucharest Greater Area (Text S1). Daily mean temperature and frequency of wind direction in 2005 in Bucharest (Figure S1). Supplementary information on anthropogenic emission sources of CO, SO_2, NO_X (Text S2). Annual emissions of PM_{10}, $PM_{2.5}$, $PM_{10-2.5}$, CO, SO_2, NO_X at country scale between 2005 and 2010 (Figure S2). Calculated annual and seasonal trends of PM_{10}, $PM_{2.5}$, NO_X, SO_2, CO by site (Table S1). Examples of time series and trends of annual mean mass concentrations of PM_{10}, NO_X, SO_2, CO (Figure S3). Levels of daily averages of gaseous pollutant mass concentrations for the period 2005–2010 at all sites: NO_X (Figure S4), SO_2 (Figure S5), CO (Figure S6). Annual air pollution pattern in BGA (Text S3) including: Daily range and annual mean particulate (Table S2) and gas–phase (Table S3) concentrations and monitoring data recovery; Compliance with the EU regulations for all sites in BGA in 2005 and 2010 (Table S4), Seasonal behavior of daily averages of observed PM_{10}, NO_X, SO_2 and CO concentrations at urban background site (Figure S7), Example of diurnal variation of pollutants, traffic counts and meteorological parameters (Figure S8). Pollution episodes in Bucharest Greater Area in 2005 (Text S4). Identified pollution episodes in BGA in 2005 (Table S5). Air mass back–trajectories and wind rose during the episode in 8th–14th February 2005 (Figure S9). PM_{10} and $PM_{2.5}$ concentrations for some European sites (Table S6). This information is available free of charge via the Internet at http://atmospolres.com.

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