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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*

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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

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.

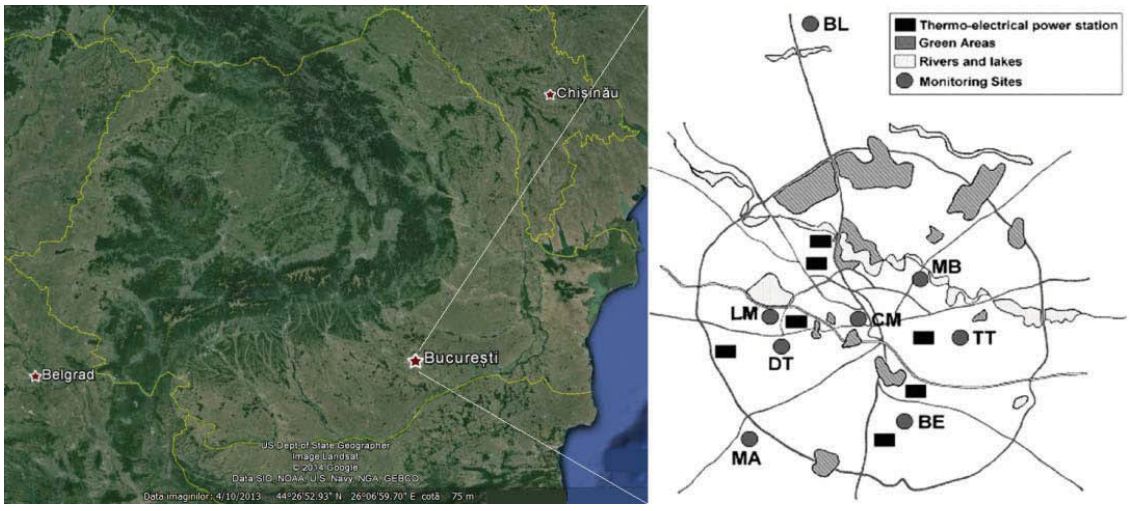

Figure 1. *Map showing Romania and the AQ network in Bucharest Greater Area (grey circles represent the monitoring stations; black rectangles show major thermo–electrical power plants).*