Atmospheric Environment 45 (2011) 6557–6566

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Determination and analysis of PM_{10} source apportionment during episodes of air pollution in Central Eastern European urban areas: The case of wintertime 2006

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ARTICLE INFO

Article history:
Received 11 April 2011
Received in revised form
2 August 2011
Accepted 8 August 2011

Keywords:
Urban particulate air pollution
PM_{10}
Trace metals
Source apportionment
Backward trajectories
Poland

ABSTRACT

Source apportionment of air pollution due to particulate matter with an aerodynamic diameter <10 μm (PM_{10}) was investigated in Central Eastern European urban areas. A combination of four methods was developed to distinguish long-range transport (LRT) and regional transport (RT) from local pollution (LP) sources as well as to discern the involvement of traffic or residential sources in LP. Sources of PM_{10} events of pollution were determined in January 2006 in representative Polish cities using monitored air quality and meteorological data, backward air mass trajectories, correlation and principal component analysis (PCA). Daily patterns of PM_{10} levels show that several peak episodes were registered in Poland; January 21–30th being the most polluted days. Air mass back-trajectory analysis shows that all cities were under the influence of LRT from North-eastern origins (Russia–Belarus–Ukraine), most were also under LRT from Southern origin (Slovakia, Czech Republic), and northern cities were under national RT influence. PCA analysis shows that ion-sums of secondary inorganic aerosols account for LRT pollution while arsenic and chromium represents markers of RT (industrial) and LP (residential) sources of PM_{10}, respectively. Determination of several ratios (REG/UB, REG/TRAF, TRAF/UB) calculated between PM_{10} levels measured at regional background (REG); urban background (UB) and traffic (TRAF) monitoring sites shows that, with ratios REG/UB ≥ 0.57, PM_{10} episodes in both Szczecin and Warsaw bore a marked RT origin. The lower REG/UB ≤ 0.35 in the Southern cities of Cracow and Zabrze indicates that LP was the main contributor to the observed episodes. Only PM_{10} episodes in Southern-western Poland (Jelenia Góra) were clearly of LP origin as characterized, by the lowest REG/UB ratio (<0.2). The high TRAF/UB ratios obtained for all cities (close to 1) indicate that there was a great uniformity of PM levels on an urban scale owing to the meteorologically stagnant conditions. A high correlation between PM_{10}, NO_2 and CO confirms that traffic emission represented a common and an important LP source of urban pollution in most Polish cities during January 2006. On the other hand PM_{10} which is also highly correlated with SO_2 in 4 cities out of 6, indicates that coal combustion through domestic heating or industrial activities was also an important LP source of PM_{10}. Finally, extremely unfavourable meteorological conditions caused by the influence of a Siberian high-pressure system were found to be associated with the occurrence of severe PM_{10} episodes of pollution.

1. Introduction

According to current knowledge, particulate matter (PM) consists of a complex mixture of solid and liquid particles of organic matter, mineral dust, secondary inorganic aerosols (SIA including sulphates, nitrates, ammonia) and trace metals (TMs), as well as water and unspecified compounds. PM categories that are identified according to their aerodynamic diameter, as either ultrafine $PM_{0.1}$ (particles with an aerodynamic diameter, $d_a < 100$ nm), fine $PM_{2.5}$ ($d_a < 2.5$ μm), coarse $PM_{10-2.5}$ ($d_a \geq 2.5$ μm and $d_a < 10$ μm) or PM_{10} ($d_a < 10$ μm) arise from variety of natural and anthropogenic sources and of both primary and secondary origins. Primary PM, generated mainly from fossil fuel combustion processes in power and heat generation industries, in the residential sector and in diesel vehicles as well as from industrial processes are mostly found in the fine $PM_{2.5}$ mode (Chow et al., 1996; Harrison et al., 1999; Nelson, 2007). Fine particles formed in the combustion processes of fossil fuels also contain TMs such as, As, Be, Cd, Cr, Co, Pb, Mg, Ni, Zn and Se whereas traffic-related PM includes a variety of particles generated by road surface wear, tyre and brake wears. Particles created by brake abrasion are in general

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doi:10.1016/j.atmosenv.2011.08.020

found in the fine $PM_{2.5}$ mode, while those generated by the road/tyres wear were found in the coarse $PM_{10-2.5}$ mode (Manoli et al., 2002; Wåhlin et al., 2006). Moreover, tyre wear was found to be an important source of Zn, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, and Pb (Legret and Pagotto, 1999; Li et al., 2001; Sörme et al., 2001; Davis et al., 2001; Hjortenkrans et al., 2006), while brake abrasion proved more the source of Cd, Cr, Cu, Ni, Pb, Sb, and Zn (Weckwerth, 2001; Monaci et al., 2000; Hjortenkrans et al., 2006). In contrast, although less is known about particles emitted from small-scale combustion utilities (small residential or domestic boilers), it is thought that these emissions affect mostly the particulate matter load in the coarse $PM_{10-2.5}$ mode. Wind-blown soil and re-suspended street dust are also present largely in the coarse particle fraction (Harrison et al., 1999; Putaud et al., 2004) and this latter source can make a significant contribution to particle mass. Secondary particles, generated in the atmosphere by photochemical gas-to-particle conversion processes are in general belonged to the submicron $PM_{1.0}$ mode ($d_a < 1.0\ \mu m$) (Seinfeld and Pandis, 2006). A number of European aerosol phenomenology studies performed by several authors (Van Dingenen et al., 2004; Putaud et al., 2004) have shown that the annual background PM_{10} and $PM_{2.5}$ concentrations for continental Europe are strongly affected by the regional aerosol background (Van Dingenen et al., 2004). As for chemical composition, the European study of Putaud et al. (2004) demonstrated that organic matter appears more often as the major component of both PM_{10} and $PM_{2.5}$, with the exception of natural and rural background sites, where sulphate contribution appear to be much greater. Furthermore, the source apportionment study performed in seven selected European urban sites (Querol et al., 2004) revealed that, carbonaceous aerosols (organic matter and elemental carbon) and SIA accounted for a major fraction of PM_{10} and particularly for the $PM_{2.5}$ (Querol et al., 2004). Concerning sources of emission and their origins, the long-range transport (LRT) and regional transport (RT) from local pollution (LP) could be distinguished. Most of reported winter episodes in Europe were caused by LP or RT sources of PM, such as coal combustion (e.g. Cracow, Juda-Rezler, 2006; Houthuijs et al., 2001) or wood combustion for heating (e.g. Oslo, Kukkonen et al., 2005), as well as by increased traffic emissions due to unfavourable winter driving conditions (Milan and London, Kukkonen et al., 2005). In Poland, combustion of coal (the main energy carrier) for electricity and heat production contributes to the emission of both primary particles, from the inorganic material in the coal, and secondary aerosols, from atmospheric oxidation of sulphur dioxide (SO_2) and nitrogen oxides (NO_x) species to sulphate and nitrate. Whereas emissions from large power and cogeneration plants, as well as from other industrial facilities, are becoming efficiently controlled by various types of air-pollution control devices (APCD), emissions from small residential units remain largely unregulated and represent a significant problem as the poorest and least expensive types of fuel and even wastes are burned (Pacyna et al., 2007). During adverse meteorological conditions which occurred in urbanized, industrialized and remote areas of Poland in winter (January–February) 2006, extremely high PM_{10} concentrations (up to hourly values of 1000 $\mu g\ m^{-3}$) were recorded (Juda-Rezler, 2006). However, to our knowledge very little data on PM episodes were reported from Poland with the exception of the aforementioned articles and the studies performed by Hoek et al. (1997) and Houthuijs et al. (2001). Recently, the comparison of TM concentrations (Cd, Cr, Cu, Fe, Mn, Ni, and Pb) associated with PM at both the intersection and the background sites of the heavily polluted Upper Silesian city of Zabrze, has shown that the ten fold increase of all analysed TM concentrations at the intersection site was mainly associated with PM_{10}, while in contrast, it was increased by as little as two or three fold in $PM_{2.5}$ (Pastuszka et al., 2010). In this latter particulate fraction, only an increase of Fe by five times and no increase of Cr were observed (Pastuszka et al., 2010). Moreover, as car engines emit mostly fine and ultrafine particles, these authors have concluded that emissions from vehicular non-exhaust sources, including tyre wear and brake wear and the resuspension of dust from the road surface, represent one of the most important processes involved in the pollution at intersection sites. It should be pointed out that if both LP and RT pollution sources are involved to a different degree in PM_{10} concentrations, the transboundary LRT particulates also represent a main source of episodic pollution, as in the case of the Helsinki Metropolitan Area where frequent PM episodes due to LRT of pollutants originate from Eastern Europe (Kukkonen et al., 2005; Aarnio et al., 2008; Niemi et al., 2009). LRT of aerosol particles was shown to contribute mainly to both $PM_{2.5}$ and PM_{10} elevated urban levels, although to a lesser extent for this latter pollutant (Aarnio et al., 2008). The strongest and longest LRT episodes of $PM_{2.5}$ described by Niemi et al. (2009) were caused by emissions not only from open biomass burning, but rather from ordinary Eastern European anthropogenic sources (e.g. from energy production).

In Poland, domestic coal (hard and brown) is the main energy carrier, still amounting to approximately 60% in the structure of primary energy consumption and about 95% in the structure of electricity consumption. Such structure of energy production is unfavourable to maintain a high air quality as it causes relatively high emissions of air pollutants, especially SO_2 and PM. As high as 99% of SO_2 is emitted from combustion processes (in energy and transformation industries: 72%, in residential sources: 20%, and in manufacturing industry: 7%). Sources of PM_{10} are more diverse, but combustion processes are also responsible for the majority (79%) of total PM_{10} emission (residential sector: 50%, transport: 11%, manufacturing industry: 11%, and energy and transformation industries: 7%). A better understanding of the contribution of either RT and LRT sources or of LP combustion or other processes in the generation of elevated PM levels in urban areas is of great importance. As such, the objectives of the present paper are to distinguish between all these sources those that are involved preferentially or in association with the occurrence of such episodes of pollution. The case study of severe PM episodes that occurred during winter 2006 in Poland has been selected for investigation for this purpose. Four methods were developed and combined in an original way, in order to distinguish LRT, RT and LP sources of pollution in the occurrence of polluted events in representative cities of Northern, Central and Southern parts of Poland. The contribution of regional background, traffic and residential sources to the overall urban pollution was also assessed.

2. Materials and methods

2.1. Investigated cities and main emission sources

Extremely severe PM_{10} episodes during January 2006 that were predominantly caused by the Poland's high coal consumption were selected and can be referred to as 'coal-burning episodes' following the classification of air quality episodes described by Kukkonen et al. (2005). The air quality was considered in six cities situated in Northern (Szczecin, Diabla Góra), Central (Warsaw) and Southern (Cracow, Zabrze, Jelenia Góra) Poland (see Fig. 1).

In Warsaw, located in Central-eastern Poland, the majority of the city's households are connected to a central heating supply system which greatly limits the pollution that originates from private use of fossil fuels for domestic heating. The most important LP sources of PM remain vehicular traffic and cogeneration plants whereas the contribution of small residential units to PM_{10} concentrations appear to be significant in only a few districts in the city. Warsaw, which does not have yet a peripheral ring road, is one of the most congested cities in Poland. The PM_{10} concentrations in street level air are consequently dominated by the combustion,