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Natural sources of atmospheric aerosols influencing air quality across Europe

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HIGHLIGHTS

- Natural sources of atmospheric aerosols across Europe were identified.
- Origin, frequency, magnitude, and spatio-temporal variability were assessed.
- The impact of natural episodes on air quality was significant in S and W Europe.
- Contributions from natural sources to annual PM_{10} means range between 1 and 8 μg/m^3.

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ABSTRACT

Atmospheric aerosols are emitted by natural and anthropogenic sources. Contributions from natural sources to ambient aerosols vary widely with time (inter-annual and seasonal variability) and as a function of the distance to source regions. This work aims to identify the main natural sources of atmospheric aerosols affecting air quality across Europe. The origin, frequency, magnitude, and spatial and temporal variability of natural events were assessed for the years 2008 and 2009. The main natural sources of atmospheric aerosols identified were African dust, sea spray and wildfires. Primary biological particles were not included in the present work. Volcanic eruptions did not affect air quality significantly in Europe during the study period. The impact of natural episodes on air quality was significant in Southern and Western Europe (Cyprus, Spain, France, UK, Greece, Malta, Italy and Portugal), where they contributed to surpass the PM_{10} daily and annual limit values. In Central and Northern Europe (Germany, Austria and Latvia) the impact of these events was lower, as it resulted in the exceedance of PM daily but not annual limit values. Contributions from natural sources to mean annual PM_{10} levels in 2008 and 2009 ranged between 1 and 2 μg/m^3 in Italy, France and Portugal, between 1 and 4 μg/m^3 in Spain (10 μg/m^3 when including the Canary Islands), 5 μg/m^3 in UK, between 3 and 8 μg/m^3 in Greece, and reached up to 13 μg/m^3 in Cyprus. The evaluation of the number of monitoring stations per country reporting natural exceedances of the daily limit value (DLV) is suggested as a potential tool for air quality monitoring networks to detect outliers in the assessment of natural contributions. It is strongly suggested that a reference methodology for the identification and quantification of African dust contributions should be adopted across Europe.

1. Introduction

Atmospheric aerosols may be emitted by natural and anthropogenic sources. Natural emission sources of primary and secondary airborne particulates include arid or semiarid regions, oceans, vegetation and volcanoes, among others. Anthropogenic emissions, on the other hand, mainly stem from industrial and combustion processes. On the global-scale, natural sources are responsible for ~98% of primary and secondary particle emissions with on average almost 12,000 Tg/year (Fig. 1; 11,990 Tg/year, based on Andreae and Rosenfeld, 2008; Durant et al., 2010; Guieré and Querol, 2010). The major natural emissions in terms of mass are sea spray (84%), and mineral dust (13%), with other sources being biological primary organic aerosols (POA), volcanic emissions, biogenic secondary organic aerosols (SOA), and volcanic and biogenic sulphate particles. Anthropogenic aerosols contribute with only 2% to global emissions, mainly in the form of anthropogenic sulphate (49%) and industrial dust (40%), with additional emissions of anthropogenic nitrate and SOA, and fossil fuel-derived POA. On the global-scale, primary aerosols are clearly dominant over secondary species (98% vs. 2%, Fig. 1).

This scenario is reversed on the local-scale. Even though the quantification of source contributions on the local-scale is more complex and it

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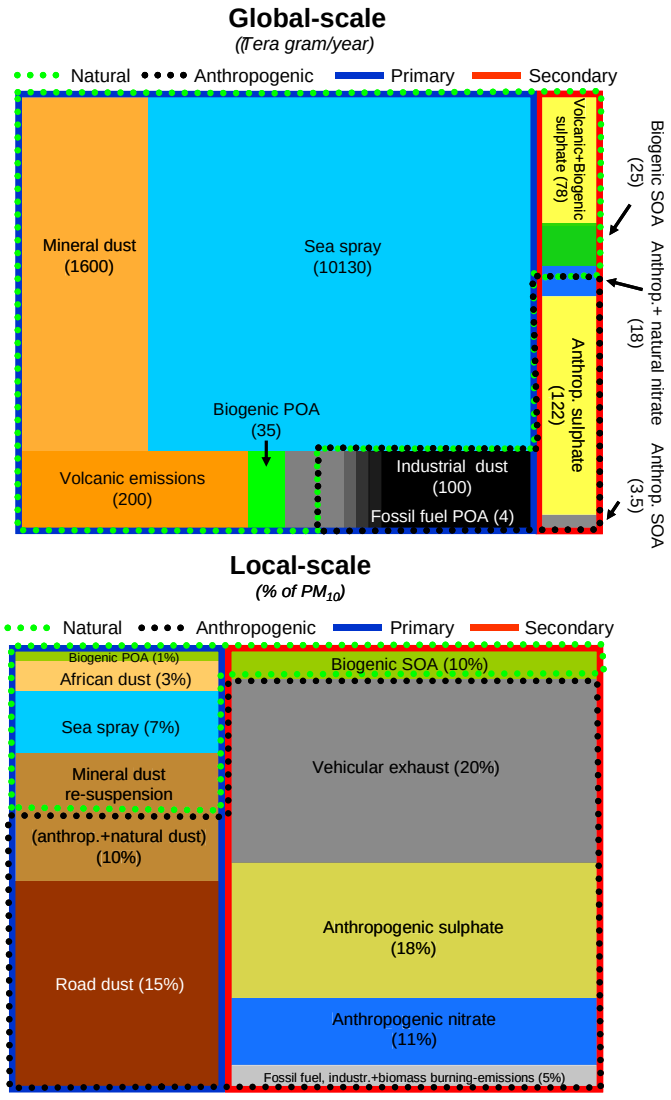

Fig. 1. Atmospheric aerosol sources on the global- and local-scales (modified from Andreae and Rosenfeld, 2008; Durant et al., 2010 and Guieré and Querol, 2010). Ratios for the local-scale in Barcelona based on Viana et al. (2008), Amato et al. (2009), and Minguillón et al. (2011).

may differ from one city/region to another given that it is strongly affected by a broad spatial variability, Fig. 1 evidences the change in the distribution between natural and anthropogenic sources for an example of a typical Southern European urban environment (Amato et al., 2009). Based on these data it may be estimated that anthropogenic sources account for 80% of particle emissions on the local-scale, the main contributors being vehicular exhaust, anthropogenic sulphate and nitrate, road dust and anthropogenically emitted mineral dust. Natural sources with an impact on urban air quality are mainly sea spray, biogenic emissions and mineral dust, with a contribution of 20% in terms of mass. In more Northern European regions the contribution from anthropogenic sources is similar or higher than in Southern Europe (Viana et al., 2008). The ratio between primary and secondary particles is also reversed on the local—with respect to the global-scale, with a prevalence of secondary aerosols (64%) on the local-scale (similar data for other EU regions not available at the time of writing).

Epidemiological studies have demonstrated strong associations between exposure to airborne particles (PM) and adverse health effects (Schwartz et al., 1996; Anderson et al., 2001; Klemm and Mason, 2000; von Klot et al., 2002; Pope and Dockery, 2006; Heal et al., 2012). Most of these studies conclude that fine particles may be more toxic because a large proportion of these particles are derived from traffic-related, industrial, and domestic emissions which contain abundant transition metals. However, coarse aerosols, with an aerodynamic diameter >2.5 μm and characteristic of natural sources, have also been linked to increased mortality and cardiovascular effects (Brunekreef and Forsberg, 2005; Mar et al., 2005; Pérez et al., 2008, 2012).

The main natural sources of atmospheric aerosols with an impact on urban air quality may be defined as:

- Windblown desert dust: this term refers to the transport of natural particles from dry regions, i.e. wind-blown desert dust. Most of the mineral dust is released to the atmosphere from arid or semiarid areas. In the case of Europe, arid zones in North Africa are the major source. In Europe, African dust may greatly increase ambient PM levels, especially in southern European countries (Bergametti et al., 1989), where it is a known source causing exceedances of the PM thresholds (Querol et al., 1998, 2009; Rodríguez et al., 2001; Viana et al., 2002; Escudero et al., 2005, 2007; Gerasopoulos et al., 2006; Kallos et al., 2006; Koçak et al., 2007; Mitsakou et al., 2008; Pey et al., 2009, 2010).
- Sea spray: sea spray is finely dispersed PM emitted from the sea surface. Sea spray is produced via the bubble-bursting processes typically resulting from whitecap generation, leading to the production of film and jet drops, resulting in sea-spray particles in the range of sub-micrometre size up to a few micrometres (O'Dowd & de Leeuw, 2007).
- Volcanic ash: emissions from volcanic eruptions and seismic activities. The main compounds emitted include water vapour, ash, CO_2, SO_2 and HCl (von Glasow et al., 2009). The impact of volcanic ash emissions may have a global impact due to the fact that emissions may be injected into the stratosphere. These may be of both primary and secondary origin.
- Carbonaceous aerosols from wildfires: wildfires are caused by the burning of non-managed and managed forests and other vegetation, strictly through natural processes. The emissions from these fires are of special relevance within Mediterranean countries where summers are drier and hotter, and fire outbreaks are commonly fanned by strong winds (Barbosa et al., 2009).
- Biogenic aerosols: attention on biogenic aerosols, primary and secondary particles emitted by vegetation, has increased in recent years (Claeys et al., 2004; Kourtchev et al., 2008). Previous studies have already highlighted biogenic aerosol influences on a global scale, as biogenic volatile organic compound (VOC) emissions were estimated to be 10 times higher than those of anthropogenic VOCs (Tsigaridis and Kanakidou, 2003).

The identification of natural events affecting air quality is a complex task which requires the combination of tools such as data on PM levels and chemical composition, aerosol maps, back-trajectory analysis, and dispersion and receptor modelling, among others. Natural contributions to PM levels and speciation in Europe have been characterised using different tools and approaches by numerous works in the literature (Artíñano et al., 2003; Escudero et al., 2007; Gobbi et al., 2007; Koçak et al., 2007; Korcz et al., 2009; Manders et al., 2009; Querol et al., 2001, 2009; Masson et al., 2010; Pederzoli et al., 2010; Beuck et al., 2011). All of these works focus on the local-, regional- or meso-scale, while to the best of our knowledge no work has so far taken an EU-wide approach to this issue. This is the aim of the present work. Thus, the present work aims to characterise the contributions from natural sources to atmospheric aerosols in urban environments, to establish their spatial distribution and to assess their impact on air quality in Europe. One additional aim is to present the state of the art regarding methodological approaches applied by the EU states to identify and quantify the impact of natural events on air quality. Natural events discussed in this work are limited to those caused by strictly natural causes, meaning those cannot be prevented or significantly reduced by human actions. As an example: wildfires may be considered as natural events only if they are started by natural causes (e.g., lightning).