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Trends and sources vs air mass origins in a major city in South-western Europe: Implications for air quality management

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HIGHLIGHTS

- We analysed a 17-year of PM10 levels, gaseous pollutants and inter-annual variations.
- A PCA identifies the sources contributing to traffic city affected by Saharan dust.
- An increase in potentially toxic elements during Saharan dust outbreaks is obtained.
- The process of discounting the number of exceedances should be reviewed.

GRAPHICAL ABSTRACT

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ABSTRACT

This study presents a 17-years air quality database comprised of different parameters corresponding to the largest city in the south of Spain (Seville) where atmospheric pollution is frequently attributed to traffic emissions and is directly affected by Saharan dust outbreaks. We identify the PM10 contributions from both natural and anthropogenic sources in this area associated to different air mass origins. Hourly, daily and seasonal variation of PM10 and gaseous pollutant concentrations (CO, NO_2 and SO_2), all of them showing negative trends during the study period, point to the traffic as one of the main sources of air pollution in Seville. Mineral dust, secondary inorganic compounds (SIC) and trace elements showed higher concentrations under North African (NAF) air mass origins than under Atlantic. We observe a decreasing trend in all chemical components of PM10 under both types of air masses, NAF and Atlantic. Principal component analysis using more frequent air masses in the area allows the identification of five PM10 sources: crustal, regional, marine, traffic and industrial. Natural sources play a more relevant role during NAF events (20.6 $\mu g \cdot m^{-3}$) than in Atlantic episodes (13.8 $\mu g \cdot m^{-3}$). The contribution of the anthropogenic sources under NAF doubles the one under Atlantic conditions (33.6 $\mu g \cdot m^{-3}$ and 15.8 $\mu g \cdot m^{-3}$, respectively). During Saharan dust outbreaks the frequent accumulation of local anthropogenic pollutants in the lower atmosphere results in poor air quality and an increased risk of

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mortality. The results are relevant when analysing the impact of anthropogenic emissions on the exposed population in large cities. The increase in potentially toxic elements during Saharan dust outbreaks should also be taken into account when discounting the number of exceedances attributable to non-anthropogenic or natural origins.

1. Introduction

Air pollution has been declared the biggest health risk in the world by The World Health Organization (WHO, 2000). A large number of epidemiological studies have reported evidence of adverse health effects of airborne particulate matter (Pope and Dockery, 2006) leading to increases in mortality as a result of cardiovascular, cerebrovascular or respiratory diseases (Brook et al., 2010; Samoli et al., 2013; Stafoggia et al., 2013). Recent investigations have also examined the relationship between particulate matter (PM) chemical composition and mortality or hospital admissions (Basagaña et al., 2015).

In urban environments, traffic related air pollution is a significant source of atmospheric PM (Su et al., 2015; Amato et al., 2009). Research undertaken in large cities such as Madrid (Tobías et al., 2011) and Barcelona (Perez et al., 2009, 2012) indicate that exposure to PM during Saharan dust events accelerates adverse effects on health. PM originated from Saharan dust and vehicle traffic degrades air quality in urban areas, increasing mortality from cardiovascular and respiratory diseases and from cancer (Garrison et al., 2014). However, natural sources, like North African (NAF) dust, are not taken into account when determining the number of exceedances of the maximum permitted threshold (2008/50/EC), even though many studies have shown that desert dust is very frequently mixed with particulate pollutants (Rodríguez et al., 2011). In addition, when the intensity of dust outbreaks increases the planetary boundary layer (PBL) height is progressively reduced causing anthropogenic pollutants to accumulate and increase the toxicity of local ambient air (Pandolfi et al., 2014).

Source apportionment (SA) models are commonly applied to the field of atmospheric sciences in order to assess pollution sources and quantify the contributions to ambient air pollution levels. This reliable and quantitative information is essential for the implementation of the Air Quality Directives (2008/50/EC and 2004/107/EC). Pollution source information is essential for identifying whether exceedances are due to natural or non-natural sources, such as road salting and sanding, for preparing air quality plans or for quantifying transboundary and long range transport pollution. There is a wide range of statistical models and modelling approaches currently available in the literature (Viana et al., 2008; Belis et al., 2013). Many authors have implemented additional tools to source contribution analysis in order to refine their results (Kim et al., 2003; Zhou et al., 2009; Zhao and Hopke, 2006, Amato and Hopke, 2012; Masiol et al., 2010). Nevertheless, despite the large number of SA studies undertaken, Belis et al. (2013), in a review of the current state of the field, note the lack of long-term speciated PM datasets, especially in urban areas, which would facilitate the interpretation and comparison of results and their application to the design of abatement measures.

This study focuses on identifying the sources that contribute to PM10 concentrations using air mass origins to quantify the impact that natural and anthropogenic sources have on a major city in southern Spain affected by traffic emissions and Saharan dust outbreaks. Toward that end, we analysed a 17-year record of PM10 levels, gaseous pollutants influenced by air masses originated from the Atlantic and North African (NAF). We also determined the chemical characterization of PM10 for both Atlantic and NAF air masses between 2007 and 2013, estimating the long-term trend of gaseous pollutants and PM10 and quantifying the impact of the different sources.

2. Methodology

2.1. Study area

Seville, the largest city in the south of Spain with approximately 1 M inhabitants, is situated in the lower Guadalquivir valley (in the south of the Iberian Peninsula), approximately 80 km from the Gulf of Cadiz, 90 km from the industrialized town of Huelva and 180 km from the Straits of Gibraltar and North Africa.

Seville has a Mediterranean climate, with warm dry summers and mild winters. The annual mean temperature is 19.2 °C, the highest in Europe, with about 3000 h of daylight and an average of 540 mm per year of rain concentrated between October and April. The wind regime shows two main directions, SW-NE, coinciding with the Guadalquivir Valley axis, and breeze regimes are frequent (Castell et al., 2010). The city experiences frequent photochemical pollution events, mainly in the warm season (Notario et al., 2012). A study by Adame et al. (2008) showed that the highest ozone concentrations occur during the spring and summer months. High ozone concentrations are generally influenced by local conditions such as the development of a breeze originated in the coast of the Gulf of Cadiz and channelled through the Guadalquivir valley (Castell et al., 2010).

Levels of PM10 and gaseous pollutants were measured hourly and daily particle samples were collected at the Príncipes station (37.375° N, 6.006° W, 8 m a.s.l.), which is part of the air quality monitoring network of the Regional Government of Andalusia located in a park in the southwest of the city of Seville (Fig. 1).

Of significance to the issue of air quality is the harbour of Seville, with an annual throughput of 4–5 Mt of cargo, and located to the south south-east of the monitoring station. Also relevant is the PERSAN chemistry industry, located in the east of the city, some 8 km from the monitoring site.

According to the annual emission inventory for 2005, NOx emissions in Seville were 30,631 t per year, with 53% attributable to traffic and 12% to agricultural activities (CMAJA, 2008). Interestingly, traffic has decreased about 14% in recent years, as the economic crisis has produced an increase in the use of public transport, boosted by the creation of a new tram network and the development of a cycling infrastructure. Vehicle use fell from 873,487 vehicles per day in 2009, to 756,545 vehicles per day in 2013, a decrease of around 117,000.

The classification of daily atmospheric episodes affecting the Príncipes monitoring site was determined through the use satellite images, meteorological parameters, mean pollutant levels and back-trajectory analysis (Querol et al., 2002). Back-trajectory analysis was carried out using NOAA Air Resources Laboratory's (ARL) HYSPLIT model (Stein et al., 2015), based on FNL (1996–2005) and GDAS (2006–2013) archives as the meteorological database. For the study period (1996–2013), North-West Atlantic (NWA) was the most frequent origin in western Andalusia (27%), followed by North-African (NAF, 25%), West Atlantic (WA, 23%), North Atlantic (NA, 12%), Mediterranean (4%) and other (3%). During the period, air masses originated from Atlantic accounted for 64% of the total, while NAF represented 26%. NAF air masses are more frequent between March and September, with summer being the most significant season. During these warmer months, Atlantic air masses have a lower incidence (principally NA and WA). The rest of the air mass origins show no defined seasonal pattern.

Various studies have previously been conducted in Seville. For example, Fernández Espinosa et al. (2001); Fernández-Espinosa and