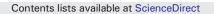
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Effect of atmospheric mixing layer depth variations on urban air quality and daily mortality during Saharan dust outbreaks



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HIGHLIGHTS

• The ML thinning makes more toxic the ambient air we breathe.

• The lower the MLH the higher the concentrations of local anthropogenic pollutants.

• The NAF episodes can cause a reduction of MLH thus worsening the air quality.

· Secondary particles (high MLH) are less toxic than primary particles (low MLH).

• Relevance of a synergic effect of atmospheric pollutants on health

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ABSTRACT

Several epidemiological studies have shown that the outbreaks of Saharan dust over southern European countries can cause negative health effects. The reasons for the increased toxicity of airborne particles during dust storms remain to be understood although the presence of biogenic factors carried by dust particles and the interaction between dust and man-made air pollution have been hypothesized as possible causes. Intriguingly, recent findings have also demonstrated that during Saharan dust outbreaks the local man-made particulates can have stronger effects on health than during days without outbreaks. We show that the thinning of the mixing layer (ML) during Saharan dust outbreaks, systematically described here for the first time, can trigger the observed higher toxicity of ambient local air. The mixing layer height (MLH) progressively reduced with increasing intensity of dust outbreaks thus causing a progressive accumulation of anthropogenic pollutants and favouring the formation of new fine particles or specific relevant species likely from condensation of accumulated gaseous precursors on dust particles surface. Overall, statistically significant associations of MLH with all-cause daily mortality were observed. Moreover, as the MLH reduced, the risk of mortality associated with the same concentration of particulate matter increased due to the observed pollutant accumulation. The association of MLH with daily mortality and the effect of ML thinning on particle toxicity exacerbated when Saharan dust outbreaks occurred suggesting a synergic effect of atmospheric pollutants on health which was amplified during dust outbreaks. Moreover, the results may reflect higher toxicity of primary particles which predominate on low MLH days.

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1. Introduction

Epidemiological studies consistently have shown an association between air pollution and respiratory illness and the number of deaths from cardiovascular and respiratory diseases. Associations have been found for particulate matter (PM) and individual species such as vapours, gases, and carbonaceous aerosols, among others (i.e. WHO, 2013; Kloog

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et al., 2013; Lepeule et al., 2012; Pope et al., 2004). Concentration and toxicity of these species increase in areas influenced by anthropogenic activities worsening the quality of the air and increasing the number of premature deaths. Recently in the Mediterranean Basin attention has been given to the effect on health of atmospheric dust originating from the North African desert which is the main source of natural dust in the atmosphere causing most of the daily exceedances of the PM_{10} (PM finer than 10 µm) limit value of the EU Directive induced by 'natural' processes in the Mediterranean (Pey et al., 2013). Increased associations with mortality of PM_{10} have been observed during Saharan dust (NAF from

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now on) outbreaks in several southern European cities (Perez et al., 2008, 2012a, 2012b; Mallone et al., 2011; Tobías et al., 2011; Diaz et al., 2012) and it has been hypothesized that the increased risk might be due in part to the biological materials contained within the dust. Intriguingly, a recent study conducted in Barcelona (Spain) during 2003–2007 has shown that the local man-made particulates during NAF outbreaks have stronger effects on health than on other days (Perez et al., 2012b). This local PM₁₀ mass under NAF outbreaks, which represents the mass that would be measured if there were no Saharan dust outbreaks (cf. Paragraph 2.4), is then more toxic compared with the PM₁₀ measured when no NAF episodes occur. The reasons for this increased toxicity of local PM during NAF days remain to be understood, although the condensation of secondary components from gaseous precursors onto the surface of dust particles has been proposed (Perez et al., 2012b).

This study seeks to identify the possible cause triggering the observed increased toxicity of local PM during NAF episodes. With this aim we calculated more than 2500 mixing layer heights (MLHs) from daily radiosoundings launched in Barcelona (Spain) during the period 2003–2010 and studied how the variations of MLH during NAF days affected the concentrations of PM and their toxicity. For the first time, we correlated the calculated MLH with all cause daily mortality stratifying the analysis by different intensities of NAF episodes and compared the calculated associations with those obtained for different PM fractions. The effect of the variations of MLH on particle toxicity and mortality during days with and without NAF episodes is presented and discussed.

2. Methods

2.1. Mixing layer height (MLH) determination

The MLH in Barcelona (NE Spain) was calculated by means of the simple parcel method (Holzworth, 1964) by using the vertical profiles of pressure (P) and temperature (T) from radiosondes launched every day at 12:00 UTC. Taking into account that the potential temperature (θ) tends to be constant in the mixing layer, the MLH is taken as the equilibrium level of an air parcel with θ calculated at ground level. The potential temperature of an air parcel at pressure P is the temperature that the parcel would acquire if it is adiabatically brought to a standard reference pressure P₀ (usually 1000 hPa) and is given by:

$\theta = T \times \left(1000/P\right)^{0.286}\!\!.$

The parcel method is highly effective for the determination of the MLH heights in the case of marked inversions, which are usually observed at midday when convective activity is high. Based on this method, a total of 2513 MLH were determined for the period under study. It should be noted that the calculated MLH at midday represents the highest top reached by the Planetary Boundary Layer (PBL) during the day in Barcelona (Pandolfi et al., 2013), being the PBL height lower at night as a consequence of the reduction of convective activity driven by the heating of the earth's surface by sunlight and the corresponding nocturnal radiative cooling of the ground.

2.2. Particulate matter, particle number and gaseous pollutant concentration measurements

Measurements of PM concentrations were conducted at an urban background monitoring site located in southwest Barcelona, influenced by vehicular emissions from one of the city's main traffic avenues located at approximately a distance of 300 m with a mean traffic density of 132,000 vehicles/day. Details of PM sampling and analytical methods have been reported elsewhere (i.e. Perez et al., 2009). Briefly, an optical counter (Grimm Labortechnik GmbH & Co. KG model 1107) was used for real time measurements of PM₁, PM_{2.5} and PM₁₀ (particulate matter finer than 1 μ m, 2.5 μ m and 10 μ m, respectively), and these data were continuously validated and corrected by gravimetric measurements

performed every 2/3 days using high volume samplers (DIGITEL and MCV) with PM₁, PM_{2.5} and PM₁₀ cut-off inlets (DIGITEL and MCV; $30 \text{ m}^3 \text{ h}^{-1}$). Real-time measurements of gaseous pollutant and particle number concentrations were acquired by conventional air pollution monitors (Thermo Scientific, Model 42i for NO_x and MCV S.A., model 48AUV for O₃) and condensation particle counter (CPC, Model 3787), respectively.

The main limitation of our study is that we used a single urban monitoring station to estimate individual exposure to PM levels. However, a study assessing the relationships between ultrafine particle number and PM_{2.5} mass concentrations measured at a central site and inside the study homes in four European cities, suggested that using a central site for exposure assessment may result in less accurate estimations of exposure to ultrafine particles than to larger size particles (Hoek et al., 2008). We assumed that the single monitoring site used in this work represented vehicle traffic exposure in the entire city. This assumption is probably valid because past observations have shown that fine and coarse PM levels at our monitoring station are strongly correlated with PM levels in other parts of the city. An additional analysis also concluded that, at least for dense traffic areas, one fixed monitoring site could represent a wider urban area, and this may not result in additional substantial measurement errors for ultrafine particles compared to fine particles (Puustinen et al., 2007).

2.3. Detection of NAF episodes

In order to identify the NAF episodes we used the same methodology proposed in previous studies (Pey et al., 2013; Querol et al., 2009; Escudero et al., 2005, 2007; Rodríguez et al., 2001). This procedure consists in the interpretation of: 1) air mass back-trajectories (HYSPLIT: http://ready.arl.noaa.gov/HYSPLITtraj.php), 2) aerosol maps (BSC-DREAM: http://www.bsc.es/projects/earthscience/DREAM/; NAAPSNRL: http://www.nrlmry.navy.mil/aerosol/; SKIRON: http://forecast.uoa.gr/ dustindx.php), 3) meteorological products (NCEP/NCAR: http://www. esrl.noaa.gov/psd/data/composites/hour/), and 4) satellite images (Sea-WiFS: http://oceancolor.gsfc.nasa.gov/SeaWiFS/; MODIS: http://modis. gsfc.nasa.gov/). It is worth to remark that in the Western Mediterranean Basin (WMB) the African air masses frequently travel at high altitudes and the African dust may affect PM levels at ground up to 2 days after the episode ends (Pey et al., 2013; European Commission, 2011). Thus, a final evaluation of PM levels was conducted incorporating these possible delays by using PM₁₀ levels measured at a regional background site located 40 km to the NNE of Barcelona (Montseny site, 41°46'N, 02°21′E, 720 m a.s.l.) as described in Pey et al. (2013).

2.4. Saharan dust contribution to PM₁₀ concentrations

The daily contributions of Saharan dust to PM₁₀ concentrations were determined by applying a statistical methodology based on the application of 30 days moving 40th percentile to the PM₁₀ data series collected at the Montseny regional background station after excluding those days impacted by NAF. For these days a percentile value was obtained which was assumed to be the theoretical background concentration of PM if African dust didn't occur. Then, the African dust contribution was obtained by difference between the experimental PM₁₀ concentration measured at the Montseny station and the calculated 40th percentile. The calculated African dust contribution was then subtracted to the PM₁₀ concentrations measured in Barcelona to estimate the local PM₁₀ mass under NAF outbreaks in Barcelona. Details on this methodology and validation procedure can be found in previous publications (Escudero et al., 2005; Escudero et al., 2007). Currently, this methodology is one of the official methods proposed by the European Commission to evaluate the occurrence of African dust outbreaks and to quantify their contributions to PM₁₀ and PM_{2.5} concentrations (European Commission, 2011).

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