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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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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 or 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 pollutants 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

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was amplified during dust outbreaks. Moreover, the results may reflect higher toxicity of primary particles which predominate on low MLH days.

Keywords

Saharan dust outbreaks; health effect; mixing layer depth

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 vapors, gases, carbonaceous aerosols, among others (i.e. WHO, 2013; Kloog 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 now on) outbreaks in several southern European cities (Perez et al., 2008; Perez et al., 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 PM_{10} 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 PM_{10} 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 (MLH) 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 are presented and discussed.

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2. Methods

2.1 Mixing Layer Height (MLH) determination

The MLH in Barcelona (NE Spain) were 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 P_0 (usually 1000 hPa) and is given by:

$$\theta = T \times (1000/P)^{\hat{}0.286}$$

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 Sun light and the corresponding nocturnal radiative cooling of the ground.

2.2 Particulate matter, particle number and gaseous pollutants 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 132000 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 PM_1, $PM_{2.5}$ and PM_{10} (particulate matter finer than 1 μm, 2.5 μm and 10 μm, 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 PM_1, $PM_{2.5}$ and PM_{10} cut-off inlets (DIGITEL and MCV; 30 $m^3\ h^{-1}$). Real-time measurements of gaseous pollutant and particle number concentrations were acquired by conventional air pollution monitors (Thermo Scientific, Model 42i for NOx and MCV S.A., model 48AUV for O_3) 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

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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/HYSPLIT traj.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 PM_{10} 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 PM_{10} concentrations

The daily contributions of Saharan dust to PM_{10} concentrations were determined by applying a statistical methodology based on the application of 30 days moving 40th percentile to the PM_{10} 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 PM_{10} concentration measured at the Montseny station and the calculated 40th percentile. The calculated African dust contribution was then subtracted to the PM_{10} concentrations measured in Barcelona to estimate the local PM_{10} mass under NAF outbreaks in Barcelona. Details on this methodology and validation procedure can be found in previous publications (Pey 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 PM_{10} and $PM_{2.5}$ concentrations (European Commission, 2011).

For the specific aims of this study, the above described methodology was applied also to the PM_1 data series. It is worth mentioning that during intense NAF episodes this technique may lead to an overestimation of PM_{10} concentrations of African origin calculated at Barcelona due to the different altitudes of Barcelona (80m a.s.l.) and Montseny (720m a.s.l.) stations. Given that African air masses often travel at high altitudes in the WMB it is possible that the

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concentrations of African dust at Montseny station are higher than in Barcelona during intense NAF episodes.

2.5 All-cause mortality data

Daily mortality by all-external causes (International Classification of Disease - ICD9: 001-799, ICD10 A00-R99) for people older than 65 years was obtained from the Catalan Mortality Registry. For the period under study (2003-2010) a total of 105733 deaths were considered, corresponding to an average of around 35 deaths per day.

2.6 Statistical analysis

The association between all-cause mortality and daily measurements of MLH and the above mentioned air quality parameters was investigated using a case-crossover design that compares exposure at case days (i.e. death) with exposure at days in which the event did not happen (control days) (Jaakkola, 2003). Control days were selected using a time-stratified approach from the same day of the week, month and year as case days (Levy et al., 2001). Data were analyzed using conditional Poisson regression models (Armstrong et al., 2011) with adjustment for temperature using one temperature average to control for the immediate effects dominated by heat (average of the exposure day and day before exposure) and a second temperature average to control for effects of lower temperatures at longer lags (average of the second to fourth days before exposure). Models were also adjusted for dummy variables for bank holidays and flu epidemic weeks, as used in previous studies (Perez et al., 2009, 2012a; Ostro et al., 2011). The effect modification by NAF days was examined by creating a dummy variable for the presence or absence of Saharan dust at exposure days. The potential for effect modification was examined by including an interaction for the previously defined dummy variables and MLH determinations and the anthropogenic parameters in the conditional Poisson regression models. The effects were examined at maximum lags previously reported (Perez et al., 2009), i.e. at lag1. Estimates from conditional Poisson regression models are reported as the percentage increase in risk of mortality, defined as (relative risk − 1) × 100% (%IRR) and its 95% confidence intervals (CIs) for an interquartile range (IQR) increase in the environmental exposure. Moreover, all these estimates are also reported as relative risks in the Supporting Information (Tables S2,S3). All analyses were done using Stata, version 12, statistical software (StataCorp, College Station, TX, USA).

3. Results and Discussion

3.1 Relationships between MLH, pollutant concentrations and different NAF episodes intensity

The MLH is the height of the top of planetary boundary layer (PBL) at midday when solar radiation and convective activity are at their maximum. At any other time of the day the height of the PBL will be lower (or at maximum equal) than the MLH due to a progressive reduction of convective activity until its cessation at night. Pollutants emitted at ground cannot easily escape from this lower part of the atmosphere, thus the MLH roughly represents the maximum volume of air at disposal for vertical dilution of pollutants during the 24h.

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During the period under study the MLH in Barcelona significantly decreased with increasing intensity of NAF episodes (Figure 1). Thus, for the NAF>p90 (NAF days with natural dust contribution to PM_{10} higher than the 90th percentile) the MLH was reduced by 34% with respect the non-NAF days (days without Saharan dust outbreaks). Figure 1 shows that the mean MLH for non-NAF days (1987 samples) was 889±359 m, slightly higher compared with the mean MLH (2513 samples) during the period 2003-2010 (859±376 m; cf. yellow rectangle in Fig.1). The mean MLH decreased with increasing intensity of NAF episodes: from around 735±267 m (233 samples) for NAF days with natural contribution to PM_{10} lower than the 75th percentile ($<15\ \mu gm^{-3}$; NAF<p75), to 590±179 m (33 samples) for NAF days with dust contribution above the 90th percentile ($>28\ \mu gm^{-3}$; NAF>p90). These differences were statistically significant.

The ML thinning under NAF days was likely due to a concomitance of factors. The presence of high amounts of dust in the free troposphere can reduce the solar radiation reaching the surface, thus limiting the convective growth of ML at midday. The daily mean and median values of solar radiation reaching the surface during intense (NAF>p90) NAF episodes were around 30% and 40%, respectively, lower compared with the mean and median values during days with NAF<p75 (cf. Supporting Information, Fig. S1). Moreover, the warm African air masses usually reach the WMB travelling at high altitudes (Pey et al., 2013). This transport pattern could form a new and lower than usual thermal inversion at the altitudes where the warm African air mass ends above the colder local air over Barcelona (Pandolfi et al., 2013), thus also contributing to the observed MLH reduction during NAF days.

The decrease of the MLH during NAF days led to the progressive accumulation of PM (Figure 1), inorganic gaseous pollutants (NO, NO_2, CO and SO_2; Figure 2a,b) and particle number concentration (Figure 2c).

The natural contribution of African dust to PM_x (PM_1, $PM_{2.5}$, PM_{10}) concentrations contributed to the observed PM_x increases: aerosols transported from African regions reached the surface increasing the levels of ambient PM under NAF days. However, it is expected that African dust contributes more to the mass concentrations of PM_{10} rather than to the fine mode (PM_1) (Querol et al., 2004). The mean PM_{10} and PM_1 concentrations during NAF>p90 were around 66 and 30 $\mu g/m^3$, respectively (Fig.1). The methodology described in Method Section for the separation between the local and natural (African) contributions to PM, yielded to a mean African dust contribution to PM_{10} of around 42 $\mu g/m^3$ (66%) during these days with NAF>p90. Thus, around 24 $\mu g/m^3$ of PM_{10} (34%) measured during NAF>p90 were of local origin. The same methodology applied to the PM_1 concentrations produced a natural (African) dust contribution to PM_1 of around 9 $\mu g/m^3$ (30%) during NAF>p90 and a local/anthropogenic contribution of around 21 $\mu g/m^3$ (70%). Following these numbers, the resulting PM_{1-10} concentration during NAF>p90 was around 36 $\mu g/m^3$ (~33 $\mu g/m^3$ of African origin and ~3 $\mu g/m^3$ of local origin). The mean PM_{10}, PM_1 and PM_{1-10} during non-NAF days for the period 2003-2010 were around 35 $\mu g/m^3$, 16 $\mu g/m^3$ and 19 $\mu g/m^3$, respectively. The low local PM_{1-10} calculated during NAF>p90 (~3 $\mu g/m^3$) was likely due to an overestimation of the African dust contribution to PM_{10} (and to a lesser extent to PM_1) measured in Barcelona during these very intense NAF>p90 episodes

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(cf. Paragraph 2.4). However, even in the case of an overestimation of African dust contribution to PM_{10} and PM_1, it is possible to state that the concentration of locally generated/formed PM_1 particles in Barcelona markedly increased during NAF>p90. Thus, the observed MLH thinning during NAF days favored the accumulation of local fine particles likely mainly primary.

Moreover, during the period 2003-2010 the mean concentrations of CO, NO, NO_2 and SO_2 increased by around 10%, 15%, 21% and 12%, respectively, during NAF>p90 compared to non-NAF days (Fig. 2a,b). Conversely, the O_3 concentrations reduced by around 12% due to the titration by increasing NO concentrations. The differences of the median concentrations between NAF>p90 and non-NAF days were higher and around 15%, 90%, 21%, 37% and −31% for CO, NO, NO_2, SO_2 and O_3, respectively.

The mean and median particle number concentrations increased by around 10% and 20%, respectively, during NAF>p90 compared with non-NAF days (Fig. 2c). The mean temperature, relative humidity, wind speed and solar radiation during 2003-2010 averaged for different NAF episode intensities are reported in Figure S1 in Supporting Information, whereas the occurrence of seasons in each considered NAF category is reported in Figure S2.

3.2 Associations between MLH and mortality

3.2.1 Case I: Associations calculated by using all available days—

In this Paragraph the daily estimated MLH were correlated with the number of daily deaths in Barcelona during the period 2003-2010. All available days (NAF and non-NAF days) were used for this analysis. An association of MLH with all cause daily mortality was observed with a reduction of the risk of mortality (IRR) of −1.05% (p=0.055) at lag 1 for an interquartile range (IQR) increase of MLH. Thus, as the MLH increased the IRR reduced because the pollutants confined within the ML had a larger volume of air at disposal for dilution and, consequently, the air was less toxic. The IRR for MLH decreased up to −1.62% (p=0.011) during weekdays, whereas no effect of MLH on mortality was observed during weekends when anthropogenic emissions were lower. The interactions were not statistically significant. The MLH adjusted by levels of PM showed lower and not statistically significant (0.165<p<0.210) IRR ranging from −0.74% for MLH adjusted by $PM_{2.5}$ to −0.82% for MLH adjusted by PM_1. During the period 2003-2010 increases in the risk of mortality of 0.61%, 0.91%, 1.27% and 0.69% for PM_1, $PM_{2.5}$, PM_{10} and $PM_{2.5\text{-}10}$, respectively were estimated for IQR increments of 11.2 $\mu g/m^3$ (PM_1), 12.9 $\mu g/m^3$ ($PM_{2.5}$), 20.4 $\mu g/m^3$ (PM_{10}) and 10.9 $\mu g/m^3$ ($PM_{2.5\text{-}10}$). The increase in the risk of mortality was statistically significant for PM_{10} (p=0.017) and marginally significant for $PM_{2.5}$ (p=0.058).

3.2.2 Case II: Associations calculated for different NAF episodes intensity—

For non-NAF days (days without Saharan dust outbreaks) during the period 2003-2010 a marginally statistically significant (p=0.091) reduction in the risk of mortality of −1.0% for an IQR increase of MLH was observed (circles in Figure 3).

For non-NAF days, a marginally significant increase in the risk of mortality was observed only for the PM_{10} fraction (p=0.081) with an IRR of 1.1% for an IQR increase in PM_{10}

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concentrations (Fig. 3). If only those NAF days with large natural dust contribution (NAF>p90) were included in the analysis, the IRR for an IQR increase in MLH during the period 2003-2010 decreased to −11.0% even if it was not statistically significant (p=0.150; Fig.3). However, a clear tendency of MLH IRR to increase (in absolute value) with increasing intensity of NAF episodes was observed, as with PM_x fractions (Fig.1), despite the low statistical significance. At NAF>p90 increases in the IRR were observed for all PM_x concentrations. In absolute values the IRR calculated for PM_x concentrations during 2003-2010 were lower compared with the MLH IRR and ranged from 3.0% for PM_1 to 5.8% for PM_{10}. Regarding PM under NAF>p90 the association with mortality was marginally significant for $PM_{2.5}$ (p=0.070) and PM_{10} (p=0.082) (Fig. 3). The interactions were not statistically significant (p-inter>0.15).

A previous study (Perez et al., 2012b) reported an increase in the risk of mortality of 2.8% per 10 $\mu g/m^3$ at lag 1 due to PM_{10} in Barcelona during the period 2003-2007 for non-NAF days. The lower IRR reported in this work for PM_{10} for non-NAF days during 2003-2010 (1.1%; circles in Fig.1) was likely due to the weighting down effect of the period 2008-2010 on particle toxicity when PM_x concentrations were considerably lower compared with the period 2003-2007. Recent studies (Cusack et al., 2012; Barmpadimos et al., 2012) have shown that the implementation of pollution abatement strategies at local/regional/European level, together with specific meteorological cycles linked with the North Atlantic Oscillation (NAO) led to a reduction of the continental background concentrations of PM_{10} and $PM_{2.5}$ over the last decade. A minimum in the concentrations of PM_x in Barcelona was observed in 2010 (cf. Supplementary Information; Fig. S3). Moreover, such negative NAO phase (especially during the years 2009 and 2010) affected the transport of Saharan dust towards the region under study (Pey et al., 2013) causing a reduction in the mean annual contribution of Saharan dust to PM_{10} (cf. Fig. S3).

This decrease in PM concentrations had a clear effect on health, indicating a reduction in particles toxicity. In fact, when all data during the period 2003-2007 were considered, the reduction in the risk of mortality for an IQR increase of MLH increased up to −1.65% (p<0.02). For PM_x the increases in the risk of mortality were 1.01%, 1.58%, 2.37% and 1.36% for PM_1, $PM_{2.5}$, PM_{10} and $PM_{2.5\text{-}10}$, respectively, and all fractions were statistically significant [$0.0007(pm_{10})<p<0.081(PM_1)$] and higher compared with the period 2003-2010. For weekdays during 2003-2007 the IRR for MLH reached −2.37% (p=0.003), around 45% higher compared with 2003-2010, and no effect on daily mortality was observed during weekends (IRR=0.31; p=0.81). The interaction was now statistically significant (p=0.07). Finally, the MLH adjusted by levels of PM_x showed IRR ranging from −1.09% for MLH adjusted by $PM_{2.5}$ to −1.32% for MLH adjusted by $^{PM}2.5\text{-}10$.

For non-NAF days during 2003-2007 the reduction in the risk of mortality for an IQR increase of MLH reached −1.4% (p=0.073) and statistically significant IRRs, ranging from 1.4% (p=0.055) for $PM_{2.5\text{-}10}$ to 2.9% (p=0.001) for PM_{10}, were observed for PM_x fractions (triangles in Figure 3). As expected, during the period 2003-2007 the IRR calculated for PM_{10} was comparable with the IRR reported in the previous study for the same period (Perez et al., 2012b). During the episodes with NAF>p90 (2003-2007) the IRR for an IQR increase in MLH reached −15.5% which was statistically significant (p=0.075). The IRR for

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PM_x during 2003-2007 ranged from 5.7% for PM_1 to 11.1% for PM_{10} and in all cases the reductions in the risk of mortality were statistically significant ($0.006<p<0.068$). For the period 2003-2007 the interactions were statistically significant for PM_{10} ($p=0.04$) and $PM_{2.5}$ ($p=0.095$) and no significant for MLH, PM_1 and $PM_{2.5-10}$, although in all cases clear clinically relevant differences were observed among the 4 considered categories (i.e. non-NAF, NAF<p75, p75<NAF<p90 and NAF>p90).

3.2.3 Case III: Associations calculated for different MLH during non-NAF days

—An exploratory analysis was performed in order to study the relative importance of MLH variations during NAF and non-NAF days on daily mortality. For this analysis only the non-NAF days during the period 2003-2010 were used and the analysis was stratified by different heights of the mixing layer (<300m, <450m, <530m, <600m and <1000 m). The IRR for MLH and PM_{10} for each different height were then calculated. The results are reported in Figure 4.

As observed in Figure 4 the IRR of MLH decreased (in absolute value) with increasing MLH, from −3.5% when MLH was <300m to −1.3% for MLH<1000m. The IRR of PM_{10} followed a very similar pattern ranging from 3.1% (MLH<300m) to 1.2% (MLH<1000m). Thus the same concentration of PM_{10} was less toxic on days with higher mixing heights. This may reflect lower toxicity of transported secondary particles than primary particles which predominate on low mixing height days. The reason for the increasing toxicity of PM with decreasing MLH during non-NAF days was the accumulation of anthropogenic pollutants which accompanied the ML thinning. In fact, as shown in Supplementary Information (Figure S4 and S5) the mean gaseous pollutant (CO, NO, NO_2 and SO_2), PM_x (PM_1, $PM_{2.5}$, PM_{10} and $PM_{2.5-10}$) and particle number concentrations progressively increased with decreasing MLH during non-NAF days.

For the period 2003-2010 the IRR of MLH obtained for intense NAF days (NAF>p90), when the mean MLH was around 590 m (Fig. 1), was −11% (Fig.3). Thus, under similar MLH the MLH effect on daily mortality was higher during NAF compared with non-NAF days.

4. Conclusions

This work shows that the MLH is a key parameter to take into account since it is able to influence all-cause daily mortality more than PM. The results put forward that other atmospheric components may have adverse health effects in urban environments and reflect the relevance of a synergic effect of atmospheric pollutants on health. In Barcelona the depth of ML controlled the degree of accumulation of local man-made pollutants and the lower the MLH the higher the concentration of pollutants. Thus, the thinning of ML made more toxic the ambient air through a progressive accumulation of fine particulate matter, gaseous compounds and particle number of anthropogenic origin. The outbreaks of Saharan dust (NAF) caused a reduction of MLH thus likely explaining the increased harmful effects on health of particulate matter during NAF days observed in past studies. Overall, the MLH in Barcelona had statistically significant associations with all cause daily mortality and the variations of MLH determined the degree of toxicity of ambient air. As the MLH reduced

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the risk of mortality associated with the same concentration of PM increased due to a progressive accumulation of anthropogenic pollutants. The observed high reduction of risk of mortality for IQR increase of MLH under intense NAF episodes suggested that in addition to a direct health effect of African dust, the observed increasing concentrations of local pollution with decreasing MLH may have favored the formation of specific relevant PM species likely from condensation of accumulated gaseous precursors on the surface of dust particles and/or formation of new fine particles. Moreover, the evidence that the same concentration of PM_{10} was less toxic on days with higher mixing layer heights reflected lower toxicity of transported secondary particles than primary particles which predominated on low MLH days. Finally, the reduced particle concentration and toxicity observed during 2003-2010 compared to 2003-2007 demonstrated the effectiveness and the clear benefit on health of the implementation of the local/regional/European pollution abatement strategies. Further studies would be useful to explore the associations between MLH and mortality in other cities where data similar to those collected in Barcelona are available.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Bibliography

Armstrong BG, Chalabi Z, Fenn B, Hajat S, Kovats S, Milojevic A, Wilkinson P. Association of mortality with high temperatures in a temperate climate: England and Wales. J. Epidemiol. Community Health. 2011; 65:340–345. [PubMed: 20439353]

Barmpadimos I, Keller J, Oderbolz D, Hueglin C, Prévôt ASH. One decade of parallel fine (PM2.5) and coarse (PM10–PM2.5) particulate matter measurements in Europe: trends and variability. Atmos. Chem. Phys. 2012; 12:3189–3203. doi:10.5194/acp-12-3189-2012.

Carslaw, DC. Manual for version 0.7-0. King's College; London: 2012. The openair manual – open-source tools for analysing air pollution data.

Carslaw DC, Ropkins K. Openair – an R package for air quality data analysis. Environ. Modell. Softw. 2012:27–28. 52–61.

Cusack M, Alastuey A, Pérez N, Pey J, Querol X. Trends of particulate matter (PM2.5) and chemical composition at a regional background site in the Western Mediterranean over the last nine years (2002-2010). Atmos. Chem. Phys. 2012; 12:8341–8357.

Diaz J, Tobias A, Linares C. Saharan dust and association between particulate matter and case-specific mortality: a case–crossover analysis in Madrid (Spain). Environ. Health. 2012; 11(1):11–21. [PubMed: 22401495]

Escudero M, Castillo S, Querol X, Avila A, Alarcón M, Viana MM, Alastuey A, Cuevas E, Rodríguez S. Wet and dry African dust episodes over Eastern. Spain. J. Geophys. Res. 2005; 110:D18S08. doi: 10.1029/2004JD004731.

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Escudero M, Querol X, Pey J, Alastuey A, Pérez N, Ferreira F, Alonso S, Rodríguez S, Cuevas E. A methodology for the quantification of the net African dust load in air quality monitoring networks. Atmos. Environ. 2007; 41:5516–5524.

European Commission. SEC(2011) 208 final: Commission staff working paper establishing guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe. 2011

Hoek G, Kos G, Harrison R, de Hartog J, Meliefste K, ten Brink H, Katsouyanni K, Karakatsani A, Lianou M, Kotronarou A, Kavouras I, Pekkanen J, Vallius M, Kulmala M, Puustinen A, Thomas S, Meddings C, Ayresi J, Van Wijnen J, Hamerih K. Indoor-outdoor relationships of particle number and mass in four European cities. Atmos. Environ. 2008; 42:156–169.

Holzworth CG. Estimates of mean maximum mixing depths in the contiguous United States. Mon. Weather. Rev. 1964; 92:235–242.

Jaakkola JJ. Case–crossover design in air pollution epidemiology. Eur. Respir. J. Suppl. 2003; 40:81–85.

Kloog I, Ridgway B, Koutrakis P, Coull BA, Schwartz JD. Long- and short-term exposure to PM2.5 and mortality: Using novel exposure models. Epidemiology. 2013; 24(4):555–561. [PubMed: 23676266]

Lepeule J, Laden F, Dockery D, Schwartz J. Chronic Exposure to Fine Particles and Mortality: An Extended Follow-up of the Harvard Six Cities Study from 1974 to 2009. Environ. Health. Perspect. 2012; 120(7):965–970. DOI:10.1289/ehp.1104660. [PubMed: 22456598]

Levy D, Lumley T, Sheppard L, Kaufman J, Checkoway H. Referent selection in case-crossover analyses of acute health effects of air pollution. Epidemiology. 2001; 12:186–192. [PubMed: 11246579]

Mallone S, Stafoggia M, Faustini A, Gobbi GP, Marconi A, Forastiere F. Saharan Dust and Association between Particulate Matter and Daily Mortality in Rome, Italy. Environ. Health. Perspect. 2011; 119:1409–1414. [PubMed: 21970945]

Ostro B, Tobias A, Querol X, Alastuey A, Amato F, Pey J, Pérez N, Sunyer J. The effects of particulate matter sources on daily mortality: A case-crossover study of Barcelona. Spain, Environ. Health Perspectives. 2011; 119(12):1781–1787.

Pandolfi M, Martucci G, Querol X, Alastuey A, Wilsenack F, Frey S, O'Dowd CD, Dall'Osto M. Continuous atmospheric boundary layer observations in the coastal urban area of Barcelona during SAPUSS. Atmos. Chem. Phys. 2013; 13:4983–4996. doi:10.5194/acp-13-4983-2013.

Perez L, Tobias A, Querol X, Künzli N, Pey J, Alastuey A, Viana M, Valero N, González-Cabré M, Sunyer J. Coarse particles from Saharan dust and daily mortality. Epidemiology. 2008; 19(6):800–807. [PubMed: 18938653]

Perez L, Medina-Ramón M, Künzli N, Alastuey A, Pey J, Pérez N, Garcia R, Tobias A, Querol X, Sunyer J. Size Fractionate Particulate Matter, Vehicle Traffic, and Case-Specific Daily Mortality in Barcelona, Spain. Environ. Sci. Technol. 2009; 43(13):4707–4714. DOI: 10.1021/es8031488. [PubMed: 19673255]

Perez L, Tobías A, Querol X, Pey J, Alastuey A, Díaz J, Sunyer J. Saharan dust, particulate matter and cause-specific mortality: A case-crossover study in Barcelona (Spain). Environ. International. 2012a; 48:150–155.

Perez L, Tobías A, Pey J, Pérez N, Alastuey A, Sunyer J, Querol X. Effects of Local and Saharan Particles on Cardiovascular Disease Mortality. Epidemiology. 2012b; 23(5):768–769. [PubMed: 22872119]

Pey J, Querol X, Alastuey A, Forastiere F, Stafoggia M. African dust outbreaks over the Mediterranean Basin during 2001–2011: PM10 concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology. Atmos. Chem. Phys. 2013; 13:1395–1410. doi: 10.5194/acp-13-1395-2013.

Pope CA, Burnett RT, Thurston GD, Thun MJ, Calle EE, Krewski D, Godleski JJ. Cardiovascular Mortality and Long-Term Exposure to Particulate Air Pollution: Epidemiological Evidence of General Pathophysiological Pathways of Disease. Circulation. 2004; 109:71–77. [PubMed: 14676145]

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Puustinen A, Hameri K, Pekkanen J, Kulmala M, de Hartog J, Meliefste K, ten Brink H, Kos G, Katsouyanni K, Karakatsani A, Kotronarou A. Spatial variation of particle number and mass over four European cities. Atmos. Environ. 2007; 41:6622–6636.

Querol X, Alastuey A, Rodíguez S, Viana MM, Artĩñano B, Salvador P, Mantilla E, do Santos SG, Patier RF, de La Rosa J, de la Campa AS, Meńendez M, Gil JJ. Levels of particulate matter in rural, urban and industrial sites in Spain. Sci. Tot. Environ. 2004:334–335. doi:10.1016/j.scitotenv. 2004.04.036. 359–376.

Querol X, Pey J, Pandolfi M, Alastuey A, Cusack M, Moreno T, Viana M, Mihalopoulos N, Kallos G, Kleanthous S. African dust contributions to mean ambient PM10 levels across the Mediterranean Basin. Atmos. Environ. 2009; 43:4266–4277.

Rodríguez S, Querol X, Alastuey A, Kallos G, Kakaliagou O. Saharan dust contributions to PM10 and TSP levels in Southern and Eastern Spain. Atmos. Environ. 2001; 35:2433–2447.

Tobías A, Perez L, Díaz J, Linares C, Pey J, Alastruey A, Querol X. Short-term effects of particulate matter on total mortality during Saharan dust outbreaks: a case– crossover analysis in Madrid (Spain). Sci. Tot. Environ. 2011:386–9. 412–413.

WHO. Review of evidence on health aspects of air pollution – REVIHAAP project: final technical report. 2013

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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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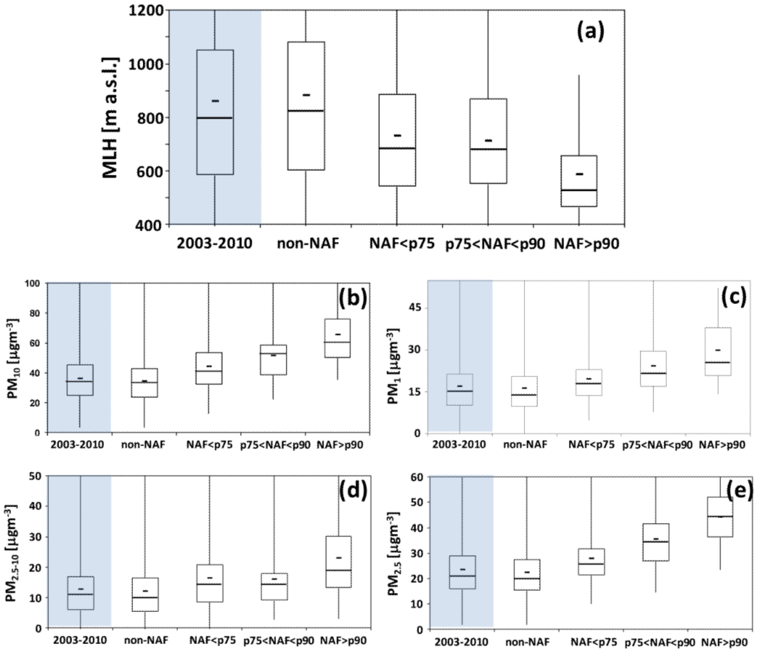

Figure 1.

Period 2003-2010. **(a)** Mean MLH and **(b-e)** PMx concentrations (**(b)** PM_{10}; **(c)** PM_1; **(d)** $PM_{2.5-10}$; **(e)** $PM_{2.5}$) calculated for non-NAF days and for different intensities of NAF episodes estimated as contribution of natural dust to PM_{10} concentrations lower than the percentile 75 (NAF<p75; 15 μgm^{-3}), between the 75th percentiles and the 90th percentile (p75<NAF<p90; 15-28 μgm^{-3}) and higher than the 90th percentile (NAF>p90; >28 μgm^{-3}). Mean MLH and PMx concentrations during NAF days and non-NAF days are compared with the mean values highlighted by the grey rectangles and calculated by using all samples during 2003-2010. Horizontal lines within the box-and-whiskers plots are the medians (50th percentile), the bottom and top of each box are 25th and 75th percentiles, respectively, and whiskers are 5th and 95th percentiles. Dots represent the mean values.

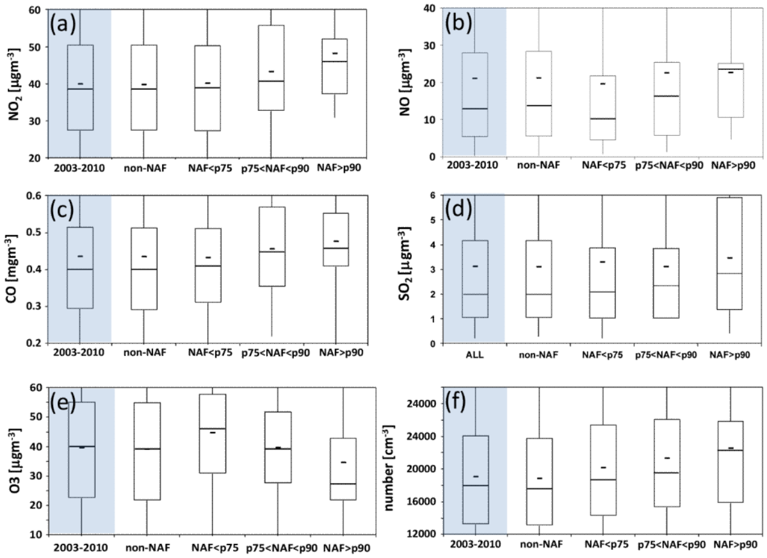

Figure 2.

Period 2003-2010. **(a-e)** Mean gaseous pollutants and (**f**) number of particles (N; D50>5nm) concentrations calculated for non-NAF days and for different intensities of NAF episodes estimated as contribution of natural dust to PM_{10} concentrations lower than the percentile 75 (NAF<p75; 15 µgm^{-3}), between the 75th percentiles and the 90th percentile (p75<NAF>p90; 15-28 µgm^{-3}) and higher than the 90th percentile (NAF>p90; >28 µgm^{-3}). Mean gaseous pollutant and particle number concentrations during NAF and non-NAF days are compared with the mean values highlighted by the grey rectangles and calculated by using all samples during 2003-2010. Horizontal lines within the box-and-whiskers plots are the medians (50th percentile), the bottom and top of each box are 25th and 75th percentiles, respectively, and whiskers are 5th and 95th percentiles. Dots represent the mean values.

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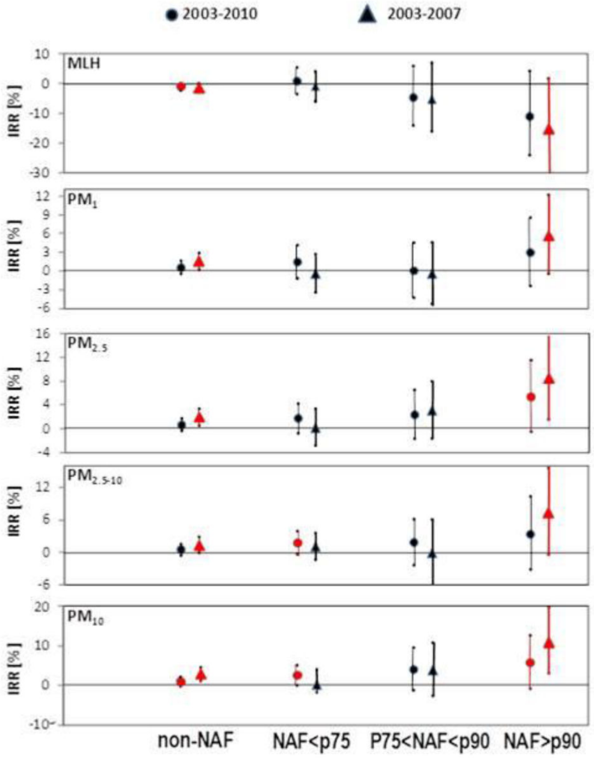

Figure 3.

Periods 2003-2010 (circles on the left) and 2003-2007 (triangles on the right): IRR (%) for increases by interquartile ranges (IQR) of MLH and particulate matter (PM) fractions, and its 95% confidence interval (95% CI), for the association with all cause daily mortality during non-NAF days and during NAF days. These latter estimated as contribution of natural dust to PM_{10} concentrations lower than the percentile 75 (NAF<p75; 15 μgm^{-3}), between the 75th percentiles and the 90th percentile (p75<NAF<p90; 15-28 μgm^{-3}) and higher than the 90th percentile (NAF>p90; >28 μgm^{-3}). Red colour highlights statistically significant associations.

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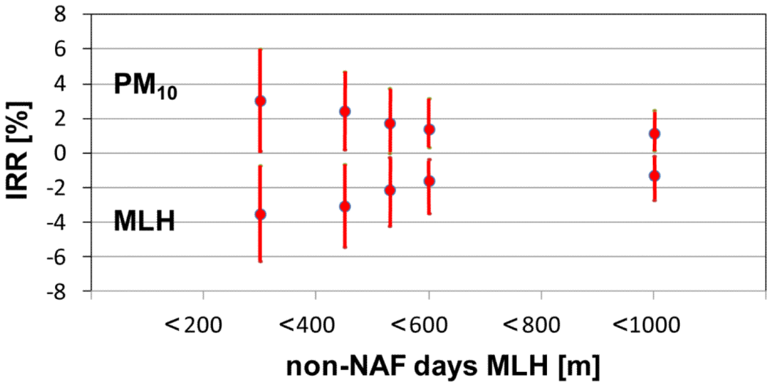

Figure 4.

Period 2003-2010. IRR (%) for increases by interquartile ranges (IQR) of MLH (below) and PM_{10} (above), and its 95% confidence interval (95% CI), for the association with all cause daily mortality during different MLHs (<300m, <450m, <530m, <600m and <1000 m) during non-NAF days. Red colour highlights statistically significant associations.