Impact of mixing layer height variations on air pollutant concentrations and health in a European urban area: Madrid (Spain), a case study

Pedro Salvador, Marco Pandolfi, Aurelio Tobías, Francisco Javier Gómez-Moreno, Francisco Molero, Marcos Barreiro, Noemí Pérez, María Aránzazu Revuelta, Isabel Martínez Marco, Xavier Querol & Begoña Artíñano

Abstract

The occurrence of local high-pollution episodes in densely populated urban areas, which have huge fleets of vehicles, is currently one of the most worrying problems associated with air pollution worldwide. Such episodes are produced under specific meteorological conditions, which favour the sudden increase of levels of air pollutants. This study has investigated the influence of the mixing layer height (MLH) on the concentration levels of atmospheric pollutants and daily mortality in Madrid, Spain, during the period 2011–2014. It may help to understand the causes and impact of local high-pollution episodes. MLH at midday over Madrid was daily estimated from meteorological radio soundings. Then, days with different MLH over this urban area were characterized by meteorological parameters registered at different levels of an instrumented tower and by composite sea level pressure maps, representing the associated synoptic meteorological scenarios. Next, statistically significant associations between MLH and levels of PM10, PM2.5, NO, NO2, CO and ultra-fine particles number concentrations registered at representative monitoring stations were evaluated. Finally, associations between all-natural cause daily mortality in Madrid, MLH, and air pollutants were estimated using conditional Poisson regression models. The reduction of MLH to values below 482 m above-ground level under strong atmospheric stagnation conditions was accompanied by a statistically significant increase in levels of NO, NO2, CO, PM2.5 and ultra-fine particle number concentrations at urban-traffic and suburban monitoring sites. The decrease of the MLH was also associated to a linear increase of the daily number of exceedances of the UE NO2 hourly limit value (200 µg/m3) and levels of air pollutants at hotspot urban-traffic monitoring stations. Also, a statistically significant association of the MLH with all-natural cause daily mortality was obtained. When the MLH increased by 830 m, the risk of mortality decreased by 2.5% the same day and by 3.3% the next day, when African dust episodic days were excluded. They were also higher in absolute terms than the increases in risk of mortality that were determined for the exposition to any other air pollutant. Our results suggest that when the prediction models foresee values of MLH below 482 m above-ground level in Madrid, the evolution of high-contamination episodes will be very favourable. Therefore, short-term policy measures will have to be implemented to reduce NO, NO2, CO, PM2.5 and ultra-fine particle emissions from anthropogenic sources in this southern European urban location.

Introduction

Many plans and measures have been implemented in the last decades to reduce air pollution and improve air quality (AQ) in Europe. In spite of these efforts, concentrations of some pollutants still exceed the European Union AQ standards for the protection of health in many European densely populated regions (EEA 2018).

For most air pollutants, the AQ standards are established as an annual concentration limit value, which is linked to the effects due to the long-term exposition. Additionally, a concentration limit value is established for some pollutants for shorter time periods (1 h, 1 day, maximum daily 8-h mean,…) to protect the population from acute short-term exposure to air pollutants (WHO 2013a). These episodes typically happen when air pollution levels reach relatively high values during brief time periods, due to the combination of two factors, namely, a suddenly increase of air pollution emissions from their baseline levels and adverse weather conditions favouring the build-up of pollution within the air masses (EEA 2014).

In 2016 (EEA 2018), 19% of all the AQ monitoring stations reporting data to the European Environmental Agency-EEA exceeded the PM10 (the mass of particulate matter which passes through a size-selective impactor inlet with a 50% efficiency cut-off at 10 µm aerodynamic diameter) daily limit value (50 µg/m3 not to be exceeded more than 35 days per year, according to the 2008/50/EC European Directive). Moreover, 1.3% of them also reported exceedances of the NO2 hourly limit value (NO2-HLV). The NO2-HLV was established as 200 µg/m3 in the 2008/50/EC European Directive and cannot be exceeded more than 18 h per year. Approximately 97–98% of these monitoring stations were located in urban or suburban areas all across Europe (EEA 2018). Exceedances of the NO2-HLV were mostly registered at urban-traffic stations in Turkey, Spain, France, the UK, Germany, Norway and Italy.

In Spain, Madrid with more than 6 million inhabitants is the most densely populated metropolitan area. It includes Madrid City (more than 3 million inhabitants) and the surrounding towns (Fig. 1). In the city of Madrid, the NO2-HLV, as well as the annual limit value (40 µg/m3, according to the above directive) have been exceeded in a systematic way at many AQ urban-traffic monitoring stations over the last decade (MITECO 2018). Moreover, a recently published study has shown a statistically significant association between NO2 exposure and daily mortality due to different causes in this city (Linares et al. 2018). Other regulated pollutants that presented relatively moderate long-term average concentrations in this area, such as PM10 and PM2.5 (the mass of particulate matter which passes through a size-selective impactor inlet with a 50% efficiency cut-off at 2.5 µm aerodynamic diameter), may still reach fairly high concentrations during the occurrence of urban high-pollution episodes (Borge et al. 2016). Moreover, it has been documented that other pollutants that are not commonly measured at AQ monitoring stations such as PM1, i.e. particles smaller than 1 µm (Borge et al.

2016); ultra-fine particles (UFP), i.e. particles smaller than 0.1 µm (Borge et al. 2018); and NH_3 (Artíñano et al. 2018) can reach high concentration values during these events in Madrid.

Fig. 1

figure1

Geographical location of the Madrid air basin, the Madrid metropolitan area and the air quality and meteorological monitoring sites used in this study

In addition to the intensive air pollutant emissions from Madrid urban sources, meteorological parameters such as the mixing layer height (MLH) might highly influence levels of air pollutants at the local and/or regional scale. The MLH is considered as the height of the layer adjacent to the ground over which pollutants or any constituents emitted within this layer or entrained into it become vertically dispersed by convection or mechanical turbulence within a time scale of about an hour (Seibert et al., 2000). Thus, it is a critical parameter that determines the atmospheric volume available for the dispersion of ground-level emitted pollutants by local sources.

In the recent work of Borge et al. (2018), an urban high-pollution episode produced in the Madrid metropolitan area in December 2016 was analysed in detail. These authors underlined the strong relationship between the MLH and the occurrence of this episode. They suggested that the MLH could be used for forecasting the development of these episodes. The main aim of this study was addressing this topic by studying a heterogeneous group of long time series of levels of air pollutants and meteorological parameters instead of a single case study. It is also important to stress that PM_{10}, $PM_{2.5}$, NO_2 and O_3 are the air pollutants whose health effects have been most closely studied in the last decade worldwide (WHO 2013b) and specifically in the Madrid region (Díaz et al. 2017, 2018; Linares et al. 2015, 2018; Ortiz et al. 2017). However, evidences on the effects of health induced by meteorological variables, such as MLH, are scarce (Pandolfi et al. 2014) and still under study.

In brief, this study was designed to estimate the influence of the variations of the MLH obtained at midday on daily mean air pollutant concentrations and on daily mortality in Madrid, during the period 2011–2014. The relationship between this meteorological index and the occurrence of air pollution episodes and exceedances of the NO_2-HLV in this urban area was specifically addressed.

Methodology

Area of study

The area of study is the Madrid metropolitan area, which is located in the Madrid air basin (Fig. 1). Road traffic and to a lesser extent commercial, institutional and household activities are the main sectors contributing to the levels of NOx, CO and primary PM10 and PM2.5 in this area. Heavy industry does not generate relevant contributions to air pollutant levels (Gómez-Moreno et al. 2011; Salvador et al. 2012, 2015; AM 2016; López et al., 2019). It should be noted that road traffic presently represents the main source of air pollution in many Mediterranean urban areas (Llop et al. 2017).

Estimation of the mixing layer height over Madrid

The MLH is not commonly provided by standard meteorological services, but can be estimated from profile measurements of atmospheric variables derived from in situ measurements, such as radiosondes, tethered balloons or masts and remote sounding systems, such as lidars, sodars or ceilometers (Seibert et al. 2000). Radio soundings are performed twice daily at 00 UTC and 12 UTC at many meteorological stations located all across the globe for a long time. These stations provide data to the World Meteorological Organization and many other institutions. Such data files are continuously quality controlled as they are used as inputs in weather forecast models and other applications. On one hand, they have the limitation that only provides a “snapshot” view of the vertical structure of the atmosphere at specific times of the day. But on the other hand, the availability of long time series of radio sounding data files can be used to obtain long time series of MLH estimations at specific sites. The Spanish Meteorological Agency (AEMET) carries out radio soundings every day at the Madrid airport (40.47° N, 3.56° W, 610 m above sea level, asl; Fig. 1) at 00 and 12 UTC. In this study, the MLH was calculated for each day of the study period by means of the simple parcel method (Holzworth, 1964) and the vertical profiles of pressure (P) and temperature (T) from these radiosondes. 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. This method is highly effective for the determination of the MLH during daytime, when convective activity usually prevails over mechanical turbulence (e.g. Pandolfi et al. 2014). For this reason, only daytime data, when convection is present, were employed in this study. Datasets for the midday radio soundings were available for 94% of the days of the period 2011–2014. Finally, 1375 MLH values were determined with this procedure for the period 2011–2014.

Then, this set of MLH daily values were classified by their magnitude and assigned to three different categories for analysis: high, medium and low MLH days were defined as days with MLH higher than or equal to the 75th percentile of the whole data set, between the 25th and 75th percentiles and lower than or equal to the 25th percentile, respectively (MLH-H, MLH-M and MLH-L). Additionally, the ventilation coefficient (VC) was computed from radio sounding data to evaluate the vertical and horizontal dilution capability of the atmosphere (Kleinman et al. 1976). The VC (m2/s) was obtained as the product between the average wind speed within the mixing layer and the MLH. Higher values of this parameter imply a stronger atmospheric dilution capability and consequently a reduction of the levels of air pollutants (Liu et al. 2019).

Next, this classification of days with different MLH over Madrid was assessed by means of different meteorological parameters and synoptic meteorological scenarios. This validation procedure pretended checking the physical meaning of such classification. Meteorological information registered at the instrumented tower installed at the CIEMAT urban-background research site (40.45° N, 3.72° W, 672 m asl, Fig. 1) was thus obtained. At this site, temperature and wind direction and speed were measured at 54 m above-ground level (agl), global horizontal irradiance at 35 m agl, temperature and humidity at 4 m agl and pressure at ground level. Data are recorded every 10 min. The vertical temperature variation (ΔT) is computed from the difference of the temperature records at 54 m and 4 m agl. Global horizontal irradiance is the total amount of shortwave radiation received from above by a horizontal surface. For the sake of simplicity, this parameter will be denoted as solar irradiance from now on. The evolution of these meteorological parameters registered during days with different MLH was analysed and interpreted. Besides, a characterization of the synoptic meteorological scenarios associated to the days with different MLH over Madrid was carried out. To this end, composite synoptic maps were obtained by averaging the sea level pressure, using the data fields corresponding to MLH-H, MLH-M and MLH-L days. NCEP/NCAR global reanalysis dataset fields of sea level pressure were obtained from the NOAA/ESRL Physical Sciences Division, Boulder, CO, USA (Kalnay et al. 1996).

Air pollutant data from air quality monitoring stations

AQ in the Madrid air basin is regularly monitored by the Madrid City and Regional Air Quality Networks (AQNs), which are managed by the Madrid municipality and the Madrid regional government, respectively. The Madrid City AQN contributed with 24 stations distributed throughout the city. The Madrid Regional AQN consists of 23 stations located in different sites of the basin. In the period 2011–2014, the Madrid City and Regional AQNs contained 9 and 6 urban-traffic AQ monitoring stations, respectively, (Fig. 1). The rest of the stations were located at suburban and rural sites to determine the urban-background and regional-background levels of air pollutants in this region.

Time series of daily mean concentrations of PM10, PM2.5, NO, NO2 and CO measured at selected stations of the Madrid City AQN were obtained and analysed in this work with the aim to evaluate associations with the MLH. One urban-traffic monitoring station situated in the Madrid downtown (“Escuelas Aguirre”, 40.42° N, 3.68° W, 692 m asl, Fig. 1) and a suburban monitoring station placed inside the largest park of the city (“Casa de Campo”, 40.41° N, 3.74° W, 645 m asl, Fig. 1) were chosen. They were the stations of the Madrid City AQN that registered the highest number of air quality and meteorological parameters. Since the year 2009, when deep changes were performed in this network, not all the stations register the levels of the same pollutants. They also represented different but representative environmental characteristics for air quality assessment in the city of Madrid.

Besides, UFP number concentrations were measured by a scanning mobility particle sizer (TSI-SMPS model 3936) at the CIEMAT research site. This instrument provides in situ continuous measurements of aerosols from 15 to 660 nm (Wang and Flagan 1990) and is included in the European network ACTRIS (Aerosols, Clouds, and Trace Gases Research Infrastructure Network) (Wiedensohler et al., 2012). It should be noted that UFP concentrations are mostly referred to their number because they represent a very small fraction of the PM mass. Previous studies have demonstrated that the main source of primary UFP in Madrid is road traffic (Gómez-Moreno et al. 2011; Brines et al. 2015). Hence, this parameter is regarded as a good tracer of road transport emissions, in spite of the fact that UFP can also be created from nucleation processes of anthropogenic and biogenic gases, during the warmer months of the year (Gómez-Moreno et al. 2011; Carnerero et al. 2018).

The Kruskal-Wallis non-parametric test was applied for the statistical comparison of the daily mean levels of PM10, PM2.5, NO2, NO, CO and UFP number concentration and meteorological parameters registered at the different monitoring sites ("Escuelas Aguirre", "Casa de Campo" and CIEMAT) between days with MLH of different intensities (MLH-H, MLH-M and MLH-L). This non-parametric method is frequently used to perform multiple sample comparisons (Dinno 2015). When the test led to the rejection of the null hypothesis (all the samples were drawn from the same population), it indicates that levels of the AQ parameter under study were significantly influenced by the values of the MLH over Madrid. Then, a post hoc test (the Bonferroni procedure, Dunn 1961) was applied to find out specifically which groups were significantly different.

Finally, the time series of hourly data of NO2 from all the urban-traffic monitoring stations of both Madrid City and Regional AQNs were obtained and analysed, with the aim to evaluate the incidence of the MLH on the exceedances of the NO2 limit values registered at these stations. Daily and hourly mean variations of the other air pollutants when NO2-HLV exceedances happened were also analysed and related with the associated mean MLH values.

Short-term effects on daily mortality

The association between all-natural cause daily mortality (International Classification of Diseases, ICD9: 001-799 ICD10 A00-R99) and daily values of MLH at midday in Madrid was investigated using a sound methodology that has been employed elsewhere (Pérez et al. 2012; Pandolfi et al. 2014; Salvador et al. 2019 among others). Data were analysed using conditional Poisson regression models (Armstrong et al. 2014). 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) was adopted (Jaakkola 2003). Such 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). Regression models were adjusted for temperature, by using one temperature average to control for the immediate effects dominated by heat (average of the exposure day and the 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), and for public holidays (Stafoggia et al. 2016). Estimates were reported as the percentage increase in risk of mortality (IRR), defined as (relative risk − 1) × 100% and its 95% confidence intervals (CIs) for an interquartile range (IQR) increase in the environmental exposure. Analyses were done using the Stata statistical software (StataCorp, College Station, TX, USA, version 14). The short-term effects were examined at different lags, up to 4 days. This type of analysis was also performed with the aim to find associations between daily mortality by all-natural causes and daily measurements of the main air pollutants (daily mean PM10, PM2.5, NO, NO2 mass concentrations registered at "Escuelas Aguirre" and UFP number concentrations registered at CIEMAT). Daily mortality data were obtained from the Spanish Statistical Institute (INE) for the study period between 1st January 2011 and 31st December 2014 in Madrid City.

Otherwise, some recently published papers have associated the occurrence of African dust outbreaks with increases in the mortality rates in many regions of Spain (Díaz et al. 2017 and references therein) and other countries located in the Mediterranean basin (Stafoggia et al. 2016 and references therein). For this reason, the influence of these events was also taken into account in this study as in Salvador et al. (2019). To detect the African dust outbreaks in the central region of Spain during the study period, a procedure based on the daily interpretation of HYSPLIT air mass back trajectories, several desert dust models forecast images and meteorological maps were applied. It is described in detail in Escudero et al. (2007) and Querol et al. (2013).

Results and discussion

Seasonal evolution and meteorological characterization of mixing layer height

An average value of 941 ± 553 m agl (mean ± standard deviation) was obtained for the MLH in Madrid during the period of study (Table 1). This parameter followed a clear seasonal evolution with low mean winter heights (475 ± 397 m agl), intermediate autumn ones (794 ± 423 m agl) and much higher spring (1073 ± 504 m agl) and summer (1365 ± 452 m agl) values. The strong relationship between the incoming solar radiation and wind speed and the vertical development of the mixing layer was evidenced by the fact that the daily mean levels of solar irradiance registered at the CIEMAT monitoring site followed the same seasonal evolution (88 ± 43, 139 ± 68, 201 ± 78 and 283 ± 44 W/m2 in winter, autumn, spring and summer, respectively). Solar irradiance at 12 UTC also followed the same trend but with a lower difference between winter and summer mean values (368 ± 174, 507 ± 222, 637 ± 245 and 841 ± 144 W/m2 in winter, autumn, spring and summer, respectively). Besides, lower mean values of wind speed were obtained in winter and autumn (3.3 ± 1.9 and 3.2 ± 1.3 m/s, respectively) than in spring and summer (3.7 ± 1.5 and 3.6 ± 1.0 m/s, respectively).

Table 1 Mean values (and standard deviation) of mixing layer height (MLH) and ventilation coefficient (VC) at midday registered at the Madrid Airport and daily mean values of meteorological variables and concentrations of atmospheric pollutants registered at the monitoring stations of CIEMAT, “Escuelas Aguirre” and “Casa de Campo”, during all the days of the period 2011–2014 (ALL) and the days with low, medium and high MLH (MLH-L, MLH-M and MLH-H, respectively)

Full size table

Next, MLH days were classified as MLH-H (≥ 1309 m agl), MLH-M (481–1309 m agl) and MLH-L (≤ 481 m agl) days. Most MLH-H days (84%) occurred in spring and summer. During the warmest months of the year, turbulence was mainly driven by buoyancy in the mainland of the Iberian Peninsula (Crespí et al. 1995). It usually forms a thermal low during the central hours of the day (Millán et al. 1996). The thermal low was clearly detected in the composite synoptic map of SLP during MLH-H days at 18 UTC, when it usually gets its highest intensity (Fig. 2a). During thermal low conditions, the intense ground heating usually favours the mixing layer development up to higher altitudes than usual due to the associated convective processes. Adding to this, it should be noted that significantly higher daily mean radiation was registered at ground level in MLH-H than in MLH-M and MLH-L days (Table 1). In fact, a low number of MLH-H days were identified in the winter months (12 days), under unusual conditions of relatively high mean values of daily mean solar irradiance and wind speed for this season (50% and 48% higher than the mean values registered in winter, respectively).

Fig. 2

figure2

Composite synoptic maps of sea level pressure for days with high (MLH-H), medium (MLH-M) and low (MLH-L) values of mixing layer height at midday over Madrid, during the period 2011–2014 at 18 UTC (a) and 12 UTC (b and c)

The development of MLH-M days was evenly distributed among the spring and autumn months (30–31% in each season) and to a lesser extent in the summer and winter seasons (25% and 14% of the days, respectively). MLH-M days occurred under different synoptic meteorological situations. The resulting averaged synoptic situation shows moderate sea level pressure values over the Iberian Peninsula and prevailing northwestern wind flows driven by the action of the Azores high (Fig. 2b). Finally, most MLH-L days occurred in autumn (27%) and winter (60%) but rarely in summer (1%). The majority of MLH-L days occurred under the presence of stationary high-pressure systems over the Iberian Peninsula (Fig. 2c). Daily mean radiation and wind speed at ground level in Madrid during MLH-L days were significantly lower than in the other days (Table 1 and Fig. 3). Besides, the mean daily values of ΔT were significantly higher in MLH-L than in MLH-M and MLH-H days (Table 1 and Fig. 3). These results confirmed that convective and dynamic processes that drive the vertical development of the mixing layer over Madrid were inhibited. However, Crespí et al. (1995) demonstrated that the prevailing evolution of the mixing layer in this type of meteorological synoptic situation was also convective, despite the fact that its growth was lower than during summer. Under this scenario, the persistence of the anticyclone over the Iberian Peninsula favoured the large-scale downward motion of the air masses. These are compressed and heated by the resulting increase in atmospheric pressure, and as a result, the lapse rate of temperature is reduced. If the air mass sinks low enough, the air at higher altitudes becomes warmer than at lower altitudes, producing a subsidence temperature inversion. In fact, the mean daily evolution of the temperature vertical variation at the CIEMAT monitoring station (Fig. 4) clearly depicts the development of deeper nocturnal surface thermal inversions in MLH-L than in MLH-M and MLH-H days. Moreover, mean values of the VC in MLH-L days were 62% lower than in MLH-M days and 44% lower in MLH-M than in MLH-H days (Fig. 3). It points to a poor atmospheric dilution capability during MLH-L days in Madrid. Such stagnant conditions often lead to high-pollution episodes in Madrid and other regions which can affect large urban areas and persist for several days (EEA 2018).

Fig. 3

figure3

Percentage variation of daily mean levels of atmospheric pollutants and meteorological variables at Madrid monitoring stations for days with different values of midday mixing layer height (MLH) over Madrid, during the period 2011–2014 (low: MLH-L, medium: MLH-M and high: MLH-H). UFP, ultra-fine particle number concentration; RAD, global horizontal irradiance; WS, wind speed at 54 m agl; T, surface temperature; ΔT, temperature vertical variation = temperature at 54 m agl − temperature at 4 m agl; VC, ventilation coefficient. a, Statistically significant differences between MLH-H and MLH-M days at the 95% confidence level. b, Statistically significant differences between MLH-M and MLH-L days at the 95% confidence level

Fig. 4

figure4

Daily mean evolution of the temperature vertical variation (ΔT = temperature at 54 m agl − temperature at 4 m agl) at CIEMAT monitoring site during days with different values of mixing layer height at midday (MLH) over Madrid, in the period 2011–2014 (low: MLH-L, medium: MLH-M and high: MLH-H)

Associations between the daily concentrations of gaseous pollutants and PM (mass and number) and the mixing layer height

Daily mean UFP number concentration measured at the CIEMAT urban-background monitoring station increased from MLH-H to MLH-M (18%) and from MLH-M to MLH-L days (62%) (Table 1 and Fig. 3). The same result was obtained for daily mean concentrations of PM2.5, NO, NO2 and CO at the urban and suburban monitoring sites (Table 1 and Fig. 3). It can be seen that the highest increase was obtained for NO at both stations from MLH-H to MLH-M (52% and 85% at “Escuelas Aguirre” and “Casa de Campo”, respectively) and from MLH-M to MLH-L days (153% and 402% at “Escuelas Aguirre” and “Casa de Campo”, respectively). Overall, the Bonferroni test results have showed that daily mean UFP number concentration, PM2.5, NO, NO2 and CO levels registered during MLH-L days were significantly higher than those obtained during MLH-M and MLH-H days (Fig. 3). It suggests that emissions of NOx, CO and PM2.5 and UFP from local and regional sources accumulated as a consequence of a thinner mixing layer and a lower VC. As a consequence, the typical urban high-pollution episode is produced over the Madrid metropolitan area during MLH-L days, when highly stable atmospheric conditions are combined with higher primary PM and NO2 emissions from road traffic and residential combustion in the winter period (Pujadas et al. 2000; Artíñano et al. 2004) and their subsequent accumulation in the area.

However, differences in mean PM10 levels during days with different MLH were not statistically significant at any of the analysed monitoring sites (Table 1). Aside from the contribution of primary PM10 from road traffic and commercial, institutional and household activities, significant inputs of coarse particles from other types of sources are currently produced in the summer period at the regional and long-range scales. In this period, when the MLH reached the highest values in Madrid, wind-blown and road dust resuspension, biological material and desert dust transport events during specific synoptic meteorological situations (Querol et al. 2004, 2008; Salvador et al. 2008, 2015; López et al., 2019) generally contribute to increase the PM10 levels in this region. It should be noted that downward trends in levels of many air pollutants have been recorded since the last years of the 1990s decade in Spain (Querol et al. 2014) and specifically in Madrid (Salvador et al. 2012, 2015). The diminution of the emissions of air pollutants from road traffic and residential heating at the urban agglomeration explained most of the decreasing rates. Such reduction of emissions was produced due to the implementation of different air pollution management and abatement actions. Besides, the economic recession produced a strong decrease in fuel consumption and a reduction of construction, demolition and roadwork activities in Madrid since 2008. These facts also contributed to the observed decreasing trends in this metropolitan area (Saiz-López et al. 2017). However, no statistically significant trend was detected for the regional-background levels of PM10 (Salvador et al. 2015). For these reasons, PM10 levels probably did not show significant increases from MLH-H to MLH-M to MLH-L days, like the other pollutants that have been analysed (Fig. 3). In good accordance with these results, Amato et al. (2014) pointed to the low reduction of urban traffic flows (and associated road dust and vehicle wear emissions) and the influence of African dust outbreaks as the major causes of the lack of decreasing trends in PM2.5–10 levels at rural, urban and industrial sites in southern Spain in the period 2003–2010.

Associations between the mixing layer height and the exceedances of the NO_2 air quality standards

AQ assessment reports are annually published by the Madrid City Council (AM 2019) and the Madrid Regional Government (CAM 2019). These reports stated that in the period 2011–2014, the NO_2 annual limit value was exceeded at a high percentage of the total number of urban-traffic stations of the Madrid metropolitan area (Fig. 1). This limit was exceeded at all the Madrid City AQN urban-traffic stations during at least 2 years of the period of study (Table 2). It was also exceeded at 3 out of 6 Madrid Regional AQN urban-traffic stations which were located in the main surrounding towns, “Coslada”, “Getafe” and “Leganés” (Fig. 1), but only in 1 or 2 years of the study period (Table 2).

Table 2 Number of years and hours exceeding the limit values of the air quality standards for NO_2, at urban-traffic monitoring stations of Madrid City and Madrid Regional Air Quality Networks (AQN) in 2011–2014

In the case of the NO_2-HLV, it was exceeded the maximum acceptable (18 h per year) at a lower percentage of the Madrid City and Regional AQN urban-traffic stations than the NO_2 annual limit value. It was exceeded during all the years of the period 2011–2014 at 3 stations (“Ramón y Cajal”, “Fernández Ladreda” and “Barrio del Pilar”), during 2 years at the “Escuelas Aguirre” and “Getafe” stations and only in 1 year at the “Cuatro Caminos” and “Coslada” stations (Fig. 1 and Table 2).

The main difference between these heavily traffic-influenced (hotspot) stations and the other urban-traffic stations was the intensity of the traffic flow in the closest road lanes. It was higher than 30,000 vehicles per day at the hotspot stations and was in the range 5000–30,000 vehicles per day at the rest of the stations (AM 2019; CAM 2019).

Our results showed that the NO_2-HLV could be exceeded from 1 h up to a maximum of 5 h the same day at the hotspot urban-traffic stations, when the MLH decreased to values close to 742, 614, 550, 467 and 336 agl, respectively (Fig. 5a). A clear linear relationship ($R^2 \geq 0.90$) was also obtained between the daily mean values of CO, NO, NO_2 and $PM_{2.5}$ registered at these urban-traffic stations and the mean values of MLH, when the NO_2-HLV exceedances were reached (Fig. 5b–e). PM_{10} was the air pollutant that showed the worst correlation ($R^2 = 0.77$) (Fig. 5f). Correlations between the maximum hourly value of air pollutant concentrations, each day that NO_2-HLV exceedances were registered, and the associated mean values of the MLH were quite similar than those obtained using the daily mean values. Namely, higher determination coefficients for CO, NO, NO_2 and $PM_{2.5}$ ($R^2 = 0.94$–0.97) and a lower value of this coefficient for PM_{10} ($R^2 = 0.73$) (Fig. Supplementary 1).

Fig. 5

figure5

Cross-correlation between the daily number of exceedances of the NO_2 hourly limit value (200 μg/m3) and the corresponding mean mixing layer height at midday over Madrid, in the period 2011–2014 (a). Data were averaged for those stations that exceeded the NO_2 hourly limit value more than 18 h per year, during at least one year of the period of study. Ninety-five percent confidence intervals for the mean mixing layer height values are shown. Daily mean levels of CO, NO, NO_2, PM2.5 and PM10 at Madrid urban-traffic air quality monitoring stations during days with NO_2 hourly limit value exceedances at urban-traffic stations were also correlated with the corresponding mean mixing layer height at midday over Madrid (b–f)

It means that the lower the midday MLH, the more intense the local air pollution episode associated to stationary high-pressure systems (Fig. 2c) and, consequently, the levels of air pollutants and the number of NO2-HLV exceedances per day registered at the Madrid urban-traffic AQ monitoring stations. These results suggest that during MLH-L days, when this parameter was lower than 482 m agl and local air pollution episodes happen, the NO2-HLV could be exceeded 4 times per day on average at the urban-traffic stations. Hence, the NO2-HLV could be exceeded the maximum acceptable per year at any of these stations during local air pollution episodes, which lasted more than 4 days.

Associations between mixing layer height and mortality

Statistically significant associations of MLH with all-natural cause daily mortality were obtained in this study (Table 3). The strongest effects were observed at lags 0 and 1. The IRR decreased (in absolute value) with the increasing of the MLH. That is, for an IQR increase of the MLH, 830 m, the risk of mortality decreased by 1.8% the same day and by 2.7% the next day. This result implies that as the MLH increased over Madrid, the associated IRR decreased, because there was available a higher volume of air for dilution of the air pollutants within the mixing layer. As a consequence, the air became less toxic. The opposite effect can thus be expected. The thinning of the mixing layer during the occurrence of urban high-pollution episodes made more toxic the ambient in Madrid, probably due to the continuous accumulation of gaseous pollutants and PM2.5 and UFP. Hence, the lower the MLH in Madrid, the higher the daily mortality.

Table 3 Percentage increase in risk of mortality (IRR) and its 95% confidence interval (95% CI) for an interquartile range (IQR) increase of mixing layer height (MLH) for all days of the period 2011–2014 and for MLH, PM2.5, PM10, NO_2, NO and ultra-fine particle (UFP) number concentrations for all days excluding African dust episodic days and for African dust episodic days identified in this period

The IRR values obtained for the exposition to air pollutants in Madrid, using the time series of PM10, PM2.5, NO2 and NO registered at “Escuelas Aguirre” and of UFP number concentrations obtained at “CIEMAT”, are shown in Fig. 6 and Table 3. The analysis was performed for all days of the period 2011–2014 excluding African dust episodic days and specifically only for African dust episodic days.

Fig. 6

figure6

Percentage increases in risk of mortality (IRR) for an interquartile range increase of mixing layer height at midday (MLH), PM2.5, PM10, NO2, NO and of ultra-fine particle (UFP) number concentrations and their 95% confidence interval, for the association with daily mortality by all-natural causes during all days of the period 2011–2014, excluding African dust episodic days (a and b) and during African dust episodic days (c and d)

Results indicated that, in the first case, statistically significant associations between daily mortality and the exposition of air pollutants were only obtained for PM2.5 at lag 0 (IRR = 0.9%). At lag 1, significant increases in the risk of mortality were obtained for all pollutants (Fig. 6a, b). The IRR ranged from 0.8% to 1.2%, 1.4%, 1.6% and 1.8% for IQR increments of NO, PM2.5, UFP number concentrations, PM10 and NO2, respectively. However, higher IRRs in absolute terms were obtained for IQR increases of MLH for lags 0 and 1 (− 2.5% and − 3.3%, respectively). It is evident that the effects on health were mainly produced one day after the exposure of air pollutants in Madrid during days with low MLH values. This is probably due to the fact that low MLH values are usually accompanied by the creation of a mixture of gaseous air pollutants, PM2.5 and UFP from local sources.

Lower decreases in the risk of daily mortality (1.1% for the whole period and 0.99% for African dust episodic days at lag 1) for an IQR increase in the value of the MLH were obtained in a similar study carried out in Barcelona in 2003–2010 (Pandolfi et al., 2014). This could be attributed to the fact that different periods of time were analysed in both studies. Besides, the evolution of the mixing layer at a Mediterranean coastal site such as Barcelona is highly controlled by the strong geographical gradients of height and the development of sea breezes (Sicard et al., 2006). These factors hampered the formation of highly stable atmospheric conditions associated to the presence of high-pressure systems. This kind of situation (Fig. 2c) generated a reduced MLH in the centre of the Iberian Peninsula (Table 1) and frequently the occurrence of high-pollution episodes. A slightly lower relative risk (1.009, 95% CI = [1.006; 1.013]) for NO2 due to natural cause mortality in Madrid was calculated by Linares et al. (2018) in 2000–2009. These differences could be attributed to the fact that it was obtained for increases of 10 μg/m3 in NO2 levels, instead for IQR increases as in the present study (IQR = 22 μg/m3), and for a different time period.

For the African dust episodic days, statistically significant associations with all-natural cause daily mortality were only obtained for PM10 at lag 1 (Fig. 6c, d). In this case, the IRR for IQR increments of PM10 was higher (2.2%) than when all days excepting African dust episodic days were considered (1.6%). It reveals the high impact of the external contributions in the levels of PM10 registered in the Madrid metropolitan area and partially explains the different behaviour of this air quality parameter when compared with the NOx, PM2.5 and UFP. For these air pollutants, the highest values were registered during days with low MLH. For this reason, the reduction of the MLH over Madrid produced an increase in the levels of air pollutants and also in the risk of mortality. It should be taken into account that the mean MLH during all African dust episodic days was slightly higher (993 ± 523 m agl) than during the other days (932 ± 558 m agl) of the period 2011–2014. Besides most African dust episodic days (83%) were classified as MLH-H or MLH-M days. Recent studies have revealed that intense African dust events with large natural dust contributions caused a reduction of the MLH over Barcelona and Madrid (Pandolfi et al. 2014; Salvador et al. 2019). However, this effect did not happen during all African dust episodic days. In the Madrid case, it was determined that it only happened when the net dust load contribution to PM10 levels exceeded a threshold value. It was obtained as the 50th percentile of the time series of value of dust contribution to PM10 in the central region of Spain (8 µg/m3) for the period 2011–2014 (Salvador et al., 2019). Probably, this is the reason why the IRR for an IQR increase in MLH was not statistically significant during all African dust episodic days of the study period. Díaz et al. (2017) estimated a lower relative risk for PM10 in African dust episodic days (1.007, 95% CI = [0.999; 1.015]) due to natural cause mortality in Madrid in 2004–2009. This value was also obtained for increases of 10 µg/m3 in PM10 levels and for a different time period.

Weakness of the study

A limitation to our work is that MLH changes along the day according to different turbulent processes. Remote sounding systems, such as lidars, sodars or ceilometers, have demonstrated to be very useful for determining the diurnal evolution of the MLH during short time periods in observational studies (Liu et al. 2019). However, these systems have been applied to MLH determination since recent time, reducing the temporal database available everywhere. Adding to these, the high operational costs and difficulty of management of these techniques have not favoured their use in continuous mode for obtaining long time series of vertical profiles of meteorological variables. In the case of Madrid, there are no available long-term data sets for MLH estimations from remote sounding systems. Many numerical models can also provide estimations of the MLH for a given geographical location. However, a careful study must be previously performed for validating model results. That is, for assessing the range of deviation of MLH estimations calculated by the model against MLH estimations obtained from real-world measurements. For these reasons, long-term data sets provided by radio soundings carried out at the Madrid airport were analysed in this study, despite the fact that they only provided MLH estimations at midday. Moreover, recently published studies have demonstrated that variations of levels of air pollutants registered in the Madrid metropolitan area during different high-pollution events were highly related with the MLH determined at the Madrid airport (Borge et al. 2018; Salvador et al. 2019).

Another limitation of our study is that we used a single urban-traffic monitoring station to estimate individual exposure to air pollutant levels in the city of Madrid. It could be argued that the average of the levels from all the stations of the Madrid City AQN should be used. The main problem with this approach is related to the fact that not all the stations register the levels of the same pollutants. Since 2009, NO2 is registered at the 24 stations (9 urban-traffic, 12 urban-background and 3 suburban), PM10 at 12 stations (5 urban-traffic, 6 urban-background and 1 suburban), CO at 10 stations (5 urban-traffic, 4 urban-background and 1 suburban) and PM2.5 is only measured at 6 stations (4 urban-traffic, 1 urban-background and 1 suburban). Thus, the network average cannot be considered homogeneous and comparable between all the air pollutants.

We assumed that the urban-traffic monitoring station used in this work was the best option for representing air pollutant exposure in the entire city, taking into account that road traffic is the main emission source in this metropolitan area. We also obtained similar variations of the daily mean levels for all air pollutants taking into account our classification of days with different MLH at midday at “Escuelas Aguirre” and “Casa de Campo” stations (Fig. 3) and at other stations (Fig. Supplementary 2).

Conclusions

In this study, a 4-year-long time series of heterogeneous data of meteorological parameters, air quality variables and daily mortality in the city of Madrid has been analysed. It could be demonstrated that the reduction of the mixing layer height (MLH) over Madrid to values below 482 m agl (MLH-L days) is one of the main factors that give rise to the development of acute urban pollution episodes in this metropolitan area. Such reduction of the MLH was associated to the occurrence of specific synoptic meteorological situations, which generated strong atmospheric stagnation conditions. The reduction of the MLH was accompanied by a statistically significant increase in levels of NO, NO2, CO, PM2.5 and ultra-fine particle number concentrations at urban-traffic and suburban monitoring sites. PM10 was the only air pollutant that did not show a clear association between its levels at these monitoring sites and variations of the MLH. It was a consequence of the different local, regional and long-distant sources that contributed to its levels in this area.

The MLH reduction was also closely related to the occurrence of exceedances of the NO2 hourly limit value in urban-traffic air quality monitoring stations located in Madrid. This is the main air quality problem that currently Madrid City faces. On average, this hourly limit value would be exceeded at least once per day at any of these stations, when the MLH reach values lower than 742 m agl and even 4 times per day during MLH-L days.

A statistically significant decrease in the risk of daily mortality by all-natural causes, was determined for an interquartile increase in the value of the MLH over Madrid. When the MLH increased by 830 m, the risk of mortality decreased by 1.8% the same day and by 2.7% the next day. These values were even higher, by 2.5% the same day and by 3.3% the next day, when African dust episodic days were excluded. They were also higher in absolute terms than the increases in risk of mortality that were determined for all the other air pollutants. Hence, it can be interpreted that the narrowing of the mixing layer during episodes of high atmospheric stability increased daily mortality due to the accumulation of PM2.5, ultra-fine particles and gaseous pollutants from anthropogenic sources. These results suggested that MLH were able to influence all-natural cause daily mortality more than any single air pollutant. Hence, future studies dedicated to explore the associations between air pollutant exposure and adverse health effects in urban areas should include MLH as one of the potential factors that could contribute to increase mortality.

Finally, it should be noted that the MLH is one of the basic parameters that are forecasted by the main numerical weather prediction models that are presently in use, such as HIRLAM, MM5 and WRF among others. After a careful validation process of the MLH estimated by any of these numerical models, this information should also be taken into account when it comes to analysing and forecasting the occurrence of local pollution episodes. Specific control strategies could consequently be triggered for this and other urban sites seriously affected by air pollution.