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Levels, sources and seasonality of coarse particles (PM_{10}–$PM_{2.5}$) in three European capitals – Implications for particulate pollution control

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ABSTRACT

Coarse particles of aerodynamic diameter between 2.5 and 10 μm (PM_c) are produced by a range of natural (windblown dust and sea sprays) and anthropogenic processes (non-exhaust vehicle emissions, industrial, agriculture, construction and quarrying activities). Although current ambient air quality regulations focus on $PM_{2.5}$ and PM_{10}, coarse particles are of interest from a public health point of view as they have been associated with certain mortality and morbidity outcomes.

In this paper, an analysis of coarse particle levels in three European capitals (London, Madrid and Athens) is presented and discussed. For all three cities we analysed data from both traffic and urban background monitoring sites. The results showed that the levels of coarse particles present significant seasonal, weekly and daily variability. Their wind driven and non-wind driven resuspension as well as their roadside increment due to traffic were estimated. Both the local meteorological conditions and the air mass history indicating long-range atmospheric transport of particles of natural origin are significant parameters that influence the levels of coarse particles in the three cities especially during episodic events.

1. Introduction

Coarse particles are commonly defined as airborne particles with aerodynamic diameter between 2.5 and 10 μm (PM_c). These particles are produced by a wide range of processes such as mechanical grinding, windblown dust, sea spray, non-exhaust vehicle emissions, agriculture, construction and quarrying activities (Charron and Harrison, 2005). Moreover, primary emissions from industry and the traffic, although they mainly produce fine ($PM_{2.5}$) particles, are also potential contributors of coarse particles. A further contribution arises from surface soils and dusts on paved areas becoming resuspended due to the wind or to the turbulence induced by passing road traffic (Solazzo et al., 2007). Long-range transport of coarse dust particles is also an important process. Specifically the African dust outbreaks have been studied by Querol et al. (2009) and Pederzoli et al. (2010), who tried to isolate the African dust contributions to mean ambient PM10 mass levels across Mediterranean Basin and Italy respectively.

In urban environments road transport is one of the predominant sources of PM_{10} emissions. Coarse particles, belonging to the non-exhaust traffic-related fraction of PM_{10}, are mainly generated from the abrasion of brake and tyre components of motor vehicles and the abrasion of the road surface itself (Thorpe et al., 2007). Using coarse particles as a crude measure of non-exhaust particles, Harrison et al. (2001) found that non-exhaust sources accounted for an almost equal concentration of PM_{10} to that from engine exhaust on a heavily trafficked London road. Similarly, using data from a number of European cities, Querol et al. (2005) found that exhaust and non-exhaust sources contribute almost equally to total traffic-related emissions.

Coagulation is a process, affecting particle mass and size, that is often overlooked by the majority of field studies dealing with coarse particles. While the coagulation theory is very advanced (Seinfeld and Pandis, 1998), theoretical and modelling results are rather difficult to extrapolate to actual field measurement conditions. Therefore, the contribution of the coagulation processes to the recorded PM_c levels is largely unknown and difficult to estimate. Several studies have addressed the issue of increased fine particle removal by coagulation with coarse particles, under

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favourable conditions (Jung et al., 2002; Kim et al., 2007). However more research is needed on this topic.

For the purposes of air quality monitoring and health-based analysis, PM_{10} and $PM_{2.5}$ fractions are being used in the majority of research studies (Ostro et al., 2000; Furusjö et al., 2007; Boogaard et al., 2010). However, coarse particles are of particular interest from both public health and regulatory perspectives, as they may play an important role in certain aspects of morbidity and possibly contribute to mortality. A number of research studies conducted, mainly, in the US have demonstrated that particles in the coarse size range could pose a risk of adverse effects to the most sensitive regions of the respiratory tract above certain exposure levels (Ostro et al., 2000). A systematic review by Brunekreef and Fosberg (2005) showed that there is evidence to suggest that both the fine and coarse fractions of PM_{10} are able to cause adverse health effects, and concluded that, in cases, coarse particles have an independent effect on respiratory morbidity. However, the evidence of an independent effect of coarse particles on total mortality was less strong. Following studies, appear to verify these conclusions (Host et al., 2008; Halonen et al., 2009; Strickland et al., 2010).

Atkinson et al. (2010) investigated the associated between PM fractions and respiratory/cardiovascular mortality and morbidity in London. Results were largely dependant on the day lag examined and the type of health outcome The most significant effect were recorded for 1-day lagged respiratory hospital admissions, which was comparable to the corresponding effect of $PM_{2.5}$.

In the Southern European cities of Madrid and Athens, focus has been recently shifted towards the effects of long-range transported dust, as these study areas are frequently impacted by Saharan dust outbreaks. In Madrid, Tobias et al. (2011) investigated the effects of coarse and fine $PM_{2.5}$ on total mortality during Saharan dust and non-dust days. They found that during Saharan dust days, a 10 $\mu g\ m^{-3}$ increase of PM_c significantly raised total mortality, in comparison to non-dust days. In Athens, only recently, Samoli et al. (2011) addressed the issue of PM_{10} health effect modification by long-range transported dust.

The EU PM_{10} limit values for the protection of human health already in force are 40 $\mu g\ m^{-3}$ as an annual mean and 50 $\mu g\ m^{-3}$ as a daily mean (not to be exceeded more than 35 times in a calendar year). According to the Directive on ambient air quality and cleaner air for Europe (2008/50/EC), an annual mean limit value of 25 $\mu g\ m^{-3}$ should be achieved for $PM_{2.5}$ in the EU by 2015. Currently, there is no separate EU limit value for coarse fraction of PM_{10}. However, in order to enhance PM_{10} control strategies in the EU, it is important to characterise the sources and factors affecting coarse particle concentrations in different European cities.

In this paper, we chose to examine the levels, sources and seasonality of coarse particle concentrations in three European capitals (London, Madrid and Athens). Pollution characteristics in these cities are well studied and, moreover, they have been included in various European research projects, such as the Auto-Oil I&II programmes (Skouloudis and Suppan, 2000), which were salient in supporting E.U. regulatory decisions. We attempt to estimate the wind driven and non-wind driven resuspension contributions to coarse particles, their meteorological dependence, the local and long-range transport, and discuss implications for air quality management.

2. Methodology

2.1. Monitoring sites and datasets

We used air quality data from one traffic oriented and one urban background monitoring station in each of the three cities. Specifically, we analysed PM_{10}, $PM_{2.5}$ and NO_x concentrations from six monitoring stations in order to estimate the vehicle exhaust and non-exhaust contributions to the total PM_{10} and $PM_{2.5}$ background, based on the assumption that NO_x concentrations in cities are mainly due to emissions from motor vehicles (Vardoulakis and Kassomenos, 2008). For London, UK, hourly and daily mean PM_{10} and $PM_{2.5}$ concentrations were calculated using TEOM instruments, while corresponding NO_x concentrations were calculated from data obtained with standard chemiluminescence gas analysers located in Marylebone Road (LMR-traffic station) and London Bloomsbury (LBL-urban background) in 2005. Hourly meteorological data were retrieved from the Heathrow Airport archives of the British Atmospheric Database Centre.

For Madrid, Spain, both hourly and daily mean PM_{10} and $PM_{2.5}$ concentrations were calculated using TEOM oscillating microbalance monitors (Artíñano et al., 2004), while NO_x concentrations were calculated using standard chemiluminescence gas analyser data for 2005 from Paseo de Recoletos (MPR-traffic station) and Casa de Campo (MCC-urban background). Hourly meteorological data were also obtained from Casa de Campo, a background residential station located in the west of Madrid (Artíñano et al., 2004). The same London and Madrid sites were also used during the Auto-Oil II study (Skouloudis and Suppan, 2000).

From the Greater Area of Athens, one urban traffic and one urban background site were included in this study. The urban traffic site (API) is located south of the city centre in the area of Piraeus. The urban background site (AAP) is located in the suburb of Agia Paraskevi in the NE of the city centre. Daily average PM_{10} and $PM_{2.5}$ concentrations for the calendar year of 2007 were determined by continuous monitoring, using beta attenuation instruments (ESM Andersen, FH 62 I-R monitors). Concentrations of NO_x, determined by chemiluminescence methods, were also available for the same period. Meteorological data for the station of API were obtained from Psyttalia in close vicinity to the Piraeus coast, and for the station of AAP from measurements in the campus of the National Technical University of Athens located in the area of Zografos, about 4 km to the south of Agia Paraskevi.

All the data used in this study are yearlong (1st January–31st December). The completeness of the PM datasets as more than

Table 1
Coordinates, Type, site description and instrumentation of the stations used in the study. All the data are for one year (1st January–31st December) for the year 2005 for UK and spanish stations and for 2007 for Greek stations.

Station	Longtitude/Latitude (degrees)	Type	Local characteristics/Emission sources	Instrumentation PMs/NO_x
LMR (UK)	0.122814/51.518867	Urban traffic	Near the road/Heavy traffic conditions	TEOM/Chemiluminence gas analyser
LBL (UK)	0.156511/51.522486	Urban background	Inside a park/low traffic, residential area	TEOM/Chemiluminence gas analyser
MPR (Spain)	3.691908/40.422197	Urban traffic	Near the road/Heavy traffic conditions	TEOM/Chemiluminence gas analyser
MCC (Spain)	3.758522/40.421967	Urban background	Inside a park/No other sources low traffic	TEOM/Chemiluminence gas analyser
API (Greece)	23.647514/37.943333	Urban traffic	Near the harbor/Heavy traffic conditions	Beta attenuation ESM Andersen FH 62 I-R/Chemiluminence gas analyser
AAP (Greece)	23.819448/37.993332	Urban background	Inside a park/no significant sources in the vicinity, low traffic, residential area	Beta attenuation ESM Andersen FH 62 I-R/Chemiluminence gas analyser

90% for London and Madrid and 75–85% for Athens. Table 1 summarises the characteristics of the monitoring stations included.

The urban background sites included in this study are not directly affected by localized industrial or other emission sources, while the three traffic locations are characterised by heavy traffic throughout the year. Sources affecting atmospheric pollution in the three study areas have been extensively presented elsewhere (Kassomenos et al., 1998; Artinano et al., 2003; Bigi and Harrison, 2010).

Regarding the equivalence of PM10, PM2.5 measurements, obtained with TEOM and beta monitors, we are presenting TEOM measurements (PM_{10} and $PM_{2.5}$) from London and Madrid without any empirical corrections.

It must be noted that datasets for the year 2007 were used for Athens (instead of 2005 as for the other two cities) because this year was more climatologically representative and did not present exceptional sand storm episodes in comparison with 2005 or 2006. In all cases, coarse particle concentrations (PM_c) were calculated by subtracting $PM_{2.5}$ from PM_{10}.

2.2. Statistical methods

Harrison et al. (2001) introduced a methodology to estimate the relation between the two components of resuspension of coarse particles: one caused by the action of the wind ("wind resuspended") and the "non-wind driven" component which is caused by the action of moving vehicles that also re-suspend coarse particles. This methodology is based on the assumption that coarse particles could be resuspended either by the wind or mechanically, which is expressed by the subtraction of two curves, $PM_{2.5}$ and PM_c vs. wind speed, taking a low wind speed threshold below which there should be no wind induced resuspension of particles (Harrison et al., 2001). The resulting hyperbolic curve has the form of $C_r = au^b$ where C_r is the wind resuspended coarse particle concentration and u is the wind speed. While resuspension of particles has been observed to occur at any wind speed, it's rate for coarse particles is likely to rapidly increase above a threshold wind velocity, as processes of particle saltation and wind erosion become more effective (Nicholson, 1988).

In order to find an optimum threshold wind velocity, we varied the wind speed between 1 and 4 m s^{-1} and searched for the best fit in terms of the coefficient of determination. We assumed that this range of values is sufficiently high to induce resuspension of dust from the ground (Harrison et al., 2001). This method was used to derive the "wind resuspended" and "non-wind" contributions to the total coarse particle concentrations observed at all monitoring sites.

2.3. Atmospheric back trajectories

Long-range transport of coarse particles from remote sources to the monitoring stations has been examined using back trajectories of air masses arriving in the area of the air quality measurements. This is just an indication of the possible long-range transport patterns of particles e.g. from the sea (sea salt particles) or the Sahara desert (dust affecting the south of Europe). This method has been used in the past for possible contribution of natural sources to particle levels measured in an area (Charron et al., 2007; Borge et al., 2007).

In order to examine possible relationships between ground level PM concentrations and long-range transport of polluted air masses over the cities, we identified the days with daily $PM_c/PM_{2.5}$ ratio higher than two and plotted the three-day long kinematic back trajectories (with arrival time 12.00 UTC) for the 10% of these days with the highest $PM_c/PM_{2.5}$ ratio for each city.

The kinematic back trajectories were modelled with the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT, version 4) developed by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (Rolph, 2003).

Table 2

a. Summary of PM_{10}, $PM_{2.5}$, PM_c and NO_x annual and seasonal concentrations at all sites. LMR and LBL are London (UK stations). MPR and MCC are Madrid (Spain) Stations and API and AAP Athens, Greece Stations. LMR, MPR and API are traffic stations (UT) and LBL, MCC and AAP are Urban background (UB) stations. b. Summary of PM_{10}, $PM_{2.5}$, PM_c and NO_x concentrations at all sites on weekly and daily bases. LMR and LBL are London (UK stations). MPR and MCC are Madrid (Spain) Stations and API and AAP Athens, Greece Stations. LMR, MPR and API are traffic stations (UT) and LL, MCC and AAP are Urban background (UB) stations.

	PM_{10} ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)	PM_c ($\mu g\ m^{-3}$)	PM_c/PM_{10}	NO_x (ppb)
a					
Annual mean					
LMR	33.4	19.2	14.2	0.43	153.7
LBL	20.4	12.8	7.4	0.36	54.0
MPR	38.2	20.4	17.8	0.47	52.3
MCC	31.8	13.9	17.9	0.56	14.7
API	48.3	35.9	12.4	0.26	66.9
AAP	28.1	19.1	9.0	0.32	12.3
Winter					
LMR	30.7	17.2	13.5	0.44	152.3
LBL	19.3	12.2	7.1	0.37	65.2
MPR	39.5	21.6	17.9	0.45	67.3
MCC	28.9	15.0	13.9	0.48	22.0
API	43.3	32.9	10.4	0.24	80.7
AAP	21.9	13.9	8	0.37	13.6
Spring					
LMR	33.8	19.0	14.8	0.44	150.0
LBL	21.3	12.7	8.6	0.40	50.4
MPR	34.8	19.8	15.0	0.43	36.1
MCC	30.8	12.6	18.2	0.59	10.6
API	45.5	34.7	10.9	0.24	61.3
AAP	28.6	19.9	8.7	0.30	12.9
Summer					
LMR	32.3	18.3	14.0	0.43	130.6
LBL	19.5	12.0	7.5	0.38	41.8
MPR	42.1	21.6	20.5	0.49	46.1
MCC	37.2	15.1	22.1	0.59	8.6
API	58.6	44.5	14.1	0.23	71.0
AAP	35.8	25.9	9.9	0.28	11.1
Autumn					
LMR	36.8	22.3	14.5	0.39	181.8
LBL	21.6	14.1	7.5	0.35	57.0
MPR	36.4	18.6	17.8	0.49	59.6
MCC	30.0	12.9	17.1	0.57	17.9
API	45.6	32.5	13.1	0.29	63.0
AAP	25.7	16.5	9.2	0.36	12.0
b					
Weekdays					
LMR	35.1	20.2	14.9	0.42	168.2
LBL	21.0	12.9	8.1	0.36	56.0
MPR	39.8	21.0	18.9	0.47	54.3
MCC	31.9	14.0	17.9	0.56	15.7
API	49.0	36.0	12.9	0.26	69.6
AAP	29.0	19.3	9.6	0.33	13.0
Weekends					
LMR	29.0	16.9	12.1	0.42	117.8
LBL	19.2	12.4	6.8	0.35	49.1
MPR	34.3	19.0	15.3	0.45	46.1
MCC	31.3	13.6	17.7	0.57	12.3
API	46.7	35.8	10.9	0.23	59.0
AAP	25.9	18.6	7.3	0.28	10.5
Day					
LMR	37.5	20.8	16.7	0.45	188.4
LBL	21.9	13.0	8.9	0.41	61.2
MPR	39.8	22.4	17.5	0.44	58.5
MCC	32.8	14.1	18.7	0.57	15.1
Night					
LMR	29.2	17.6	11.6	0.40	119.5
LBL	19.0	12.6	6.4	0.34	46.9
MPR	36.6	18.3	18.3	0.50	45.5
MCC	30.6	13.7	17.0	0.56	14.2

HYSPLIT has been widely used in air pollution modelling applications (Artíñano et al., 2001; Borge et al., 2007; Kassomenos et al., 2010) and can be run either online or offline on a PC (further information on the model can be found at: http://www.arl.noaa.gov/ready/hysplit4.html). The meteorological data used for the computation of the trajectories in the present study were obtained from the NOAA reanalysis database (http://www.arl.noaa.gov/archives.php), and the trajectory arrival height was 750 m above ground level in all cases. It must be noted that the accuracy of an individual trajectory is limited by uncertainties as meteorological inputs and analysis errors.

3. Results and discussion

3.1. Temporal and seasonal variability

The levels of PM_{10}, $PM_{2.5}$ and PM_c appeared to be lower by 6–16% at LBL and by 11–21% at LMR than those reported by Harrison et al. (2001) for the same monitoring stations in London but for the period 1997–1998. In agreement with Harrison et al. (2001), a noticeable seasonal variability of PM_c was observed showing lower seasonal mean values during winter compared to summer, autumn and spring (Table 2a). This pattern is observed for both traffic and urban background stations not only in London but also in Athens and Madrid where the highest PM_c levels are measured during summer. This can be attributed to the drying up of surfaces due to the very low humidity levels during the warm season, which exacerbates processes of particle resuspension (Table 2a).

In Athens and London, annual mean PM_c concentrations were higher at the roadside locations compared to the corresponding urban background sites. By contrast, annual mean PM_c concentrations were very similar at the two sites in Madrid, but during spring and summer PM_c levels were higher at the MCC (urban background) station compared to MPR. The latter could be attributed to the dry conditions prevailing during these periods of the year that support the resuspension of heavier particles as well as the higher frequency of African dust outbreaks (Querol et al., 2009).

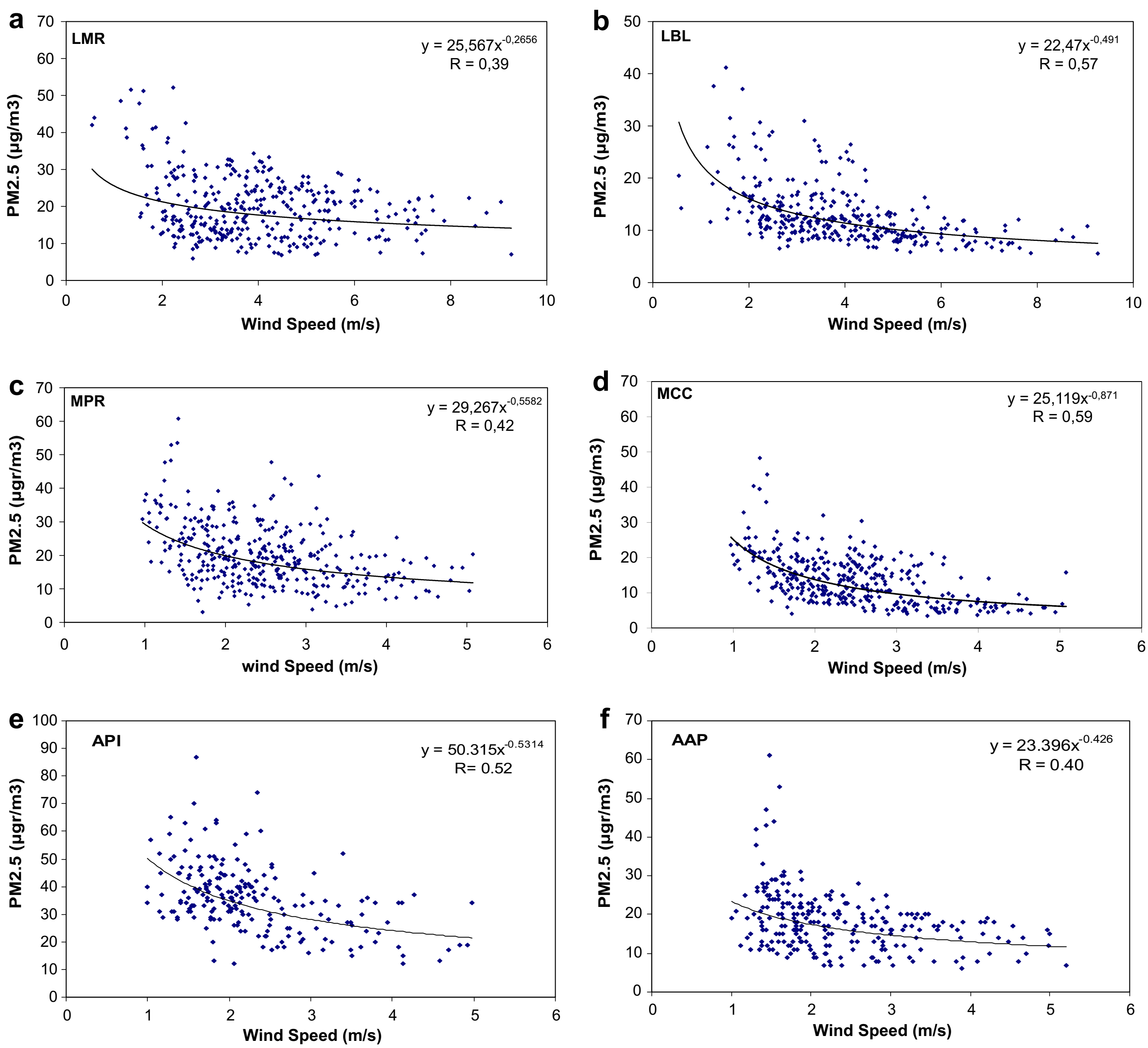

Fig. 1. The concentration of $PM_{2.5}$ vs. wind speed in (a) LMR, (b) LBL, (c) MPR, (d) MCC, (e) API, (f) AAP and best fit line.

The effect of road traffic is very clearly seen in the substantial elevations of both PM_{10} and $PM_{2.5}$ at LMR compared to LBL. In average, the roadside PM_{10} and $PM_{2.5}$ increments were 13 μg m^{-3} and 6.4 μg m^{-3} respectively for the 12-month period. These values are significantly higher than the PM_{10} and $PM_{2.5}$ increment values of 10.5 and 5.4 μg m^{-3} respectively reported by Harrison et al. (2001). London stations recorded the highest levels of nitrogen oxides, especially at the traffic station of LMR, due to the proximity to road (5 m) and the very high traffic flows which exceed 80,000 vehicles per day (Jones et al., 2008). Annual mean concentrations at LBL were comparable to those recorded at the traffic sites of Madrid and Athens. Increased LBL concentrations as opposed to the other two urban background sites are probably related to the site being located in the centre of London, while MCC and API sites are situated at some distance from the city centre. The two Southern European study areas displayed a similar traffic-background pattern for NO_x, with very low levels measured at MCC and AAP.

Annual mean PM_c levels were nearly twice as high at LMR compared to LBL. This can be attributed to more particle resuspension due to traffic induced turbulence and tyre, brake and road wear at the roadside location (Thorpe et al., 2007; Gietl et al., 2010). It must be noted that road and building works are considerable contributors to the high values of PMc recorded in London, (Fuller and Green, 2004; Charron and Harrison, 2005). The extent of the impact these sources have in Athens and Madrid is unknown up to now. On the other hand the roadside PM_{10} and $PM_{2.5}$ increments in Madrid are almost the same for both fractions (6.5 μg m^{-3}) for a 12-month period.

In Athens the ratio of coarse particles to PM_{10} is significantly higher (by almost 30%) in AAP compared to API. This is due to the clearly different characteristics of the two sites including the land cover of the areas as well as the intensity of traffic sources.

During working days the values of PM_{10} were about 19% higher than those found during weekends at LMR but the differences between weekday and weekend concentrations were significantly smaller at LBL (around 9%) confirming the work of Jones et al. (2008). An even smaller difference (4%) was observed for $PM_{2.5}$ between weekdays and weekends at LBL indicating that this station is not substantially affected by road traffic. A similar pattern of smaller reductions of NO_x levels between weekdays and weekends

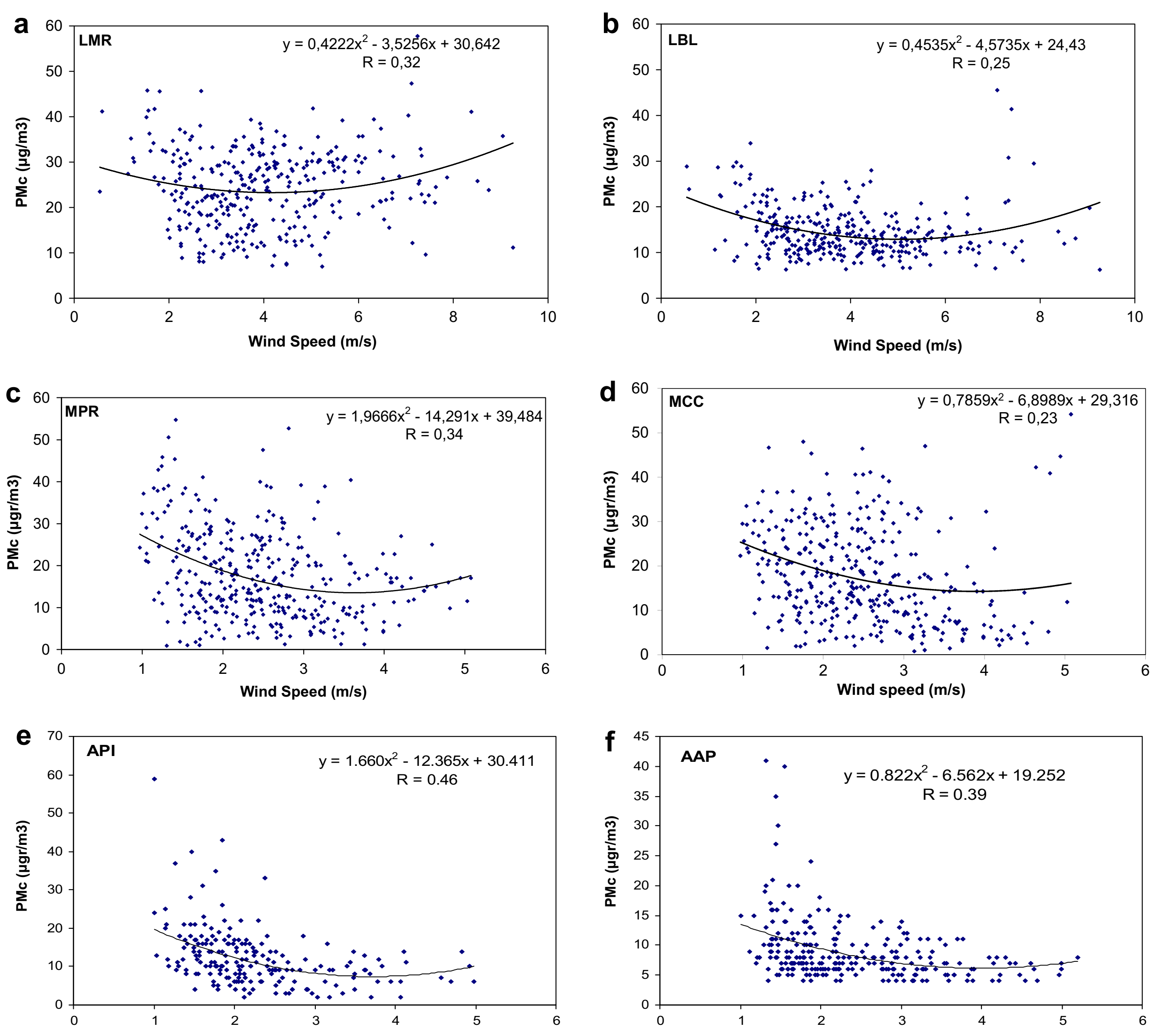

Fig. 2. The concentration of PM_c vs. wind speed in (a) LMR, (b) LBL, (c) MPR, (d) MCC, (e) API, (f) AAP and best fit line.

at LBL (13%) compared to LMR (35%) was also observed in Vardoulakis and Kassomenos (2008). Similarly in Madrid the weekday PM concentrations were higher by 10–15% compared to the weekend levels at MPR but practically remained unchanged at MCC indicating that the urban background site was not significantly affected by local traffic (Table 2b).

In Athens, however, the PM values over weekdays were higher (by around 10–12%) at both monitoring stations compared to weekends. This increment is less pronounced in API which experiences significant human and commercial activity even on weekend days.

PM_{10} and PM_c mean values at the London sites during nighttime were substantially lower (14–25% and 33–36%, respectively) compared to daytime concentrations at both monitoring stations. However, $PM_{2.5}$ values during nighttime were only 3% lower compared to daytime at the urban background station (LBL), while this difference was around 17% for the roadside station (LMR). The above analysis, demonstrates the influence of local traffic intensities endorsing the different behaviour found between daytime and nighttime (Charron and Harrison, 2005).

During nighttime, the values of PM_{10} and $PM_{2.5}$ at both Madrid sites were 7–9% and 5–19% lower respectively due to the lower traffic volumes as also indicated by the values of NO_x. It should be noted that in MCC-urban background site the PM_c concentrations were higher during daytime, which is opposite to what was observed at the MPR site (Table 2b).

Pearson correlation coefficients between the two fractions of PM_{10} were lower at the background sites (0.64–0.73) compared to the traffic ones (0.72–0.78) at the three cities indicating common PM_c and $PM_{2.5}$ sources around the traffic sites.

3.2. Analysis of the wind driven resuspension of coarse particles

We have plotted $PM_{2.5}$ concentrations against local wind speed at all monitoring sites (Fig. 1). The best fit curves are all of the hyperbolic form and indicative of the wind driven dilution process,

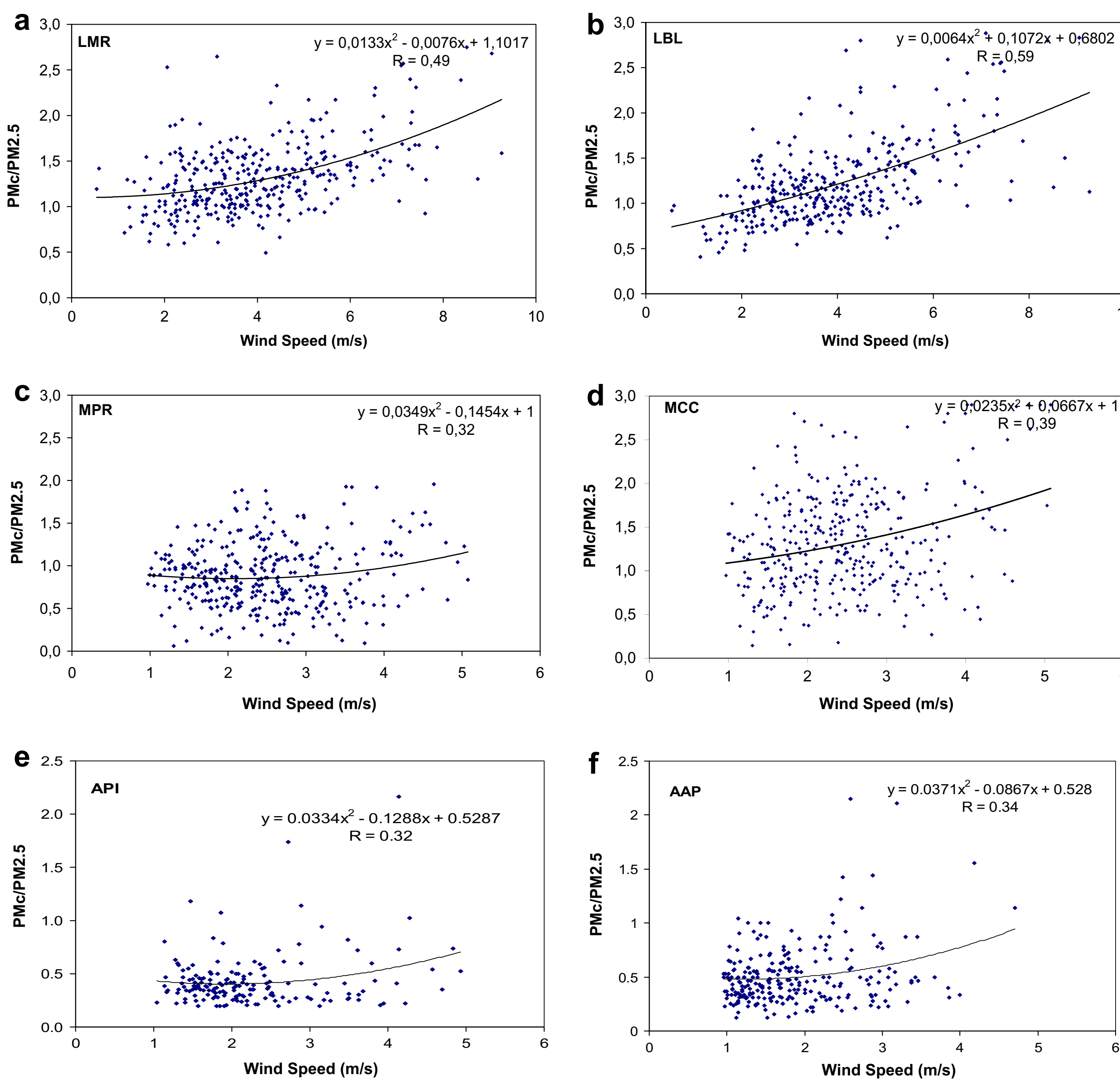

Fig. 3. The ratio $PM_c/PM_{2.5}$ vs. wind speed in (a) LMR, (b) LBL, (c) MPR, (d) MCC, (e) API, (f) AAP and best fit line.

as $PM_{2.5}$ values decrease with wind speed increases at all sites. However, there are some minor differences as the correlation coefficients at urban background stations are higher than those at traffic stations in London and Madrid while the opposite is observed in Athens.

We have also plotted the PM_c concentrations against the wind speed (Fig. 2). In this case, the curves indicate a U-shape relationship at all stations and cities. Low to moderate correlations were calculated (R: 0.23–0.46), however, these values are generally comparable to those reported by Harrison et al. (2001). All correlations were statistically significant at the 0.99 confidence level. The shape of the curves may be interpreted as two simultaneously existing components of coarse particles. The first one is diluted with increasing wind speed (similar to $PM_{2.5}$, as displayed on Fig. 2). The second component indicates increased resuspension of coarse particles when wind speed exceeds a lower threshold (Charron and Harrison, 2005; Liu and Harrison, 2011). Thorpe et al. (2007) verified the presence of the second component after investigating the association of the resuspension emission factor with wind speed.(Fig. 3)

The $PM_c/PM_{2.5}$ ratio shows an increase with wind speed (Fig. 4), with this trend being clearer in London (in particular at LBL). This reflects the increased particle resuspension at higher wind speeds in an area, as the $PM_c/PM_{2.5}$ vs. wind speed relationship effectively eliminates the impact of dilution processes (Harrison et al., 2001).

Differences of the resuspended fraction of coarse particles were found for the weekdays/weekends and daytime/nighttime (Table 3). If the resuspension was only due to the wind blowing, then the resuspended concentration of PM_c would be similar on weekdays and weekends, but greater during daytime compared to

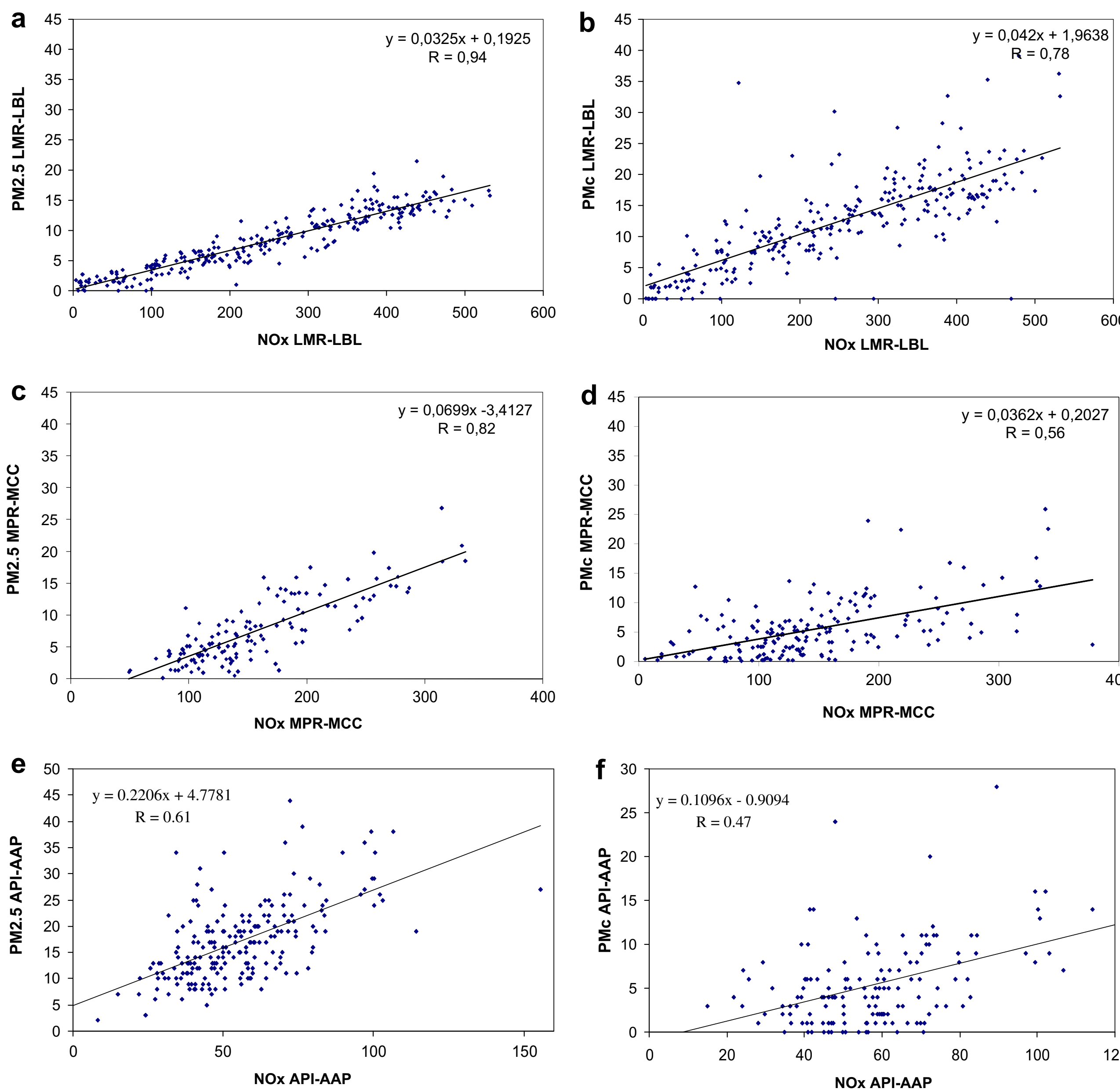

Fig. 4. Relationships between daily mean $PM_{2.5}$ and NO_x and mean PMc and NO_x expressed as concentration difference between roadside and background site in (a, b) London, (c, d) Madrid, (e, f) Athens.

nighttime. In fact, there is a substantial weekday-to-weekend difference, and there is also a significant day-to-night difference indicating a small resuspended particle mass component during nighttime when road traffic is light, and wind speed and atmospheric turbulence lower.

The non-wind driven component of coarse particles is also higher on weekdays than weekends and daytime compared to nighttime for the two London sites but this is more unclear for the Madrid stations. In Athens, the weekend/weekday variation of the non-wind driven component is similar to London. It is possible that these non-wind driven coarse particles are not only of natural origin, but perhaps are related to anthropogenic sources such as tyre and break wear and abrasion of road surfaces, and can be resuspended by traffic induced turbulence (Almeida et al., 2006).

Table 3

a. Summary of seasonally averaged estimates of "wind resuspended" and "non-wind driven" components of coarse particles, in yearly and seasonal basis, "*a*" and "*b*" are the coefficient and the exponent of the equation $C = au^b$. The optimum wind speed and correlation coefficient (R) were also presented. b. Summary of seasonally averaged estimates of "wind-resuspended" and "non-wind driven" components of coarse particles, in weekly and daily basis, "*a*" and "*b*" are the coefficient and the exponent of the equation $C = au^b$. The optimum wind speed and correlation coefficient (R) were also presented.

	Wind speed (ms^{-1})	R	a	b	C_{total} ($\mu g\ m^{-3}$)	$C_{wind\text{-}resuspended}$ ($\mu g\ m^{-3}$)	$C_{non\text{-}wind\ driven}$ ($\mu g\ m^{-3}$)	Wind-resuspended rate (C_r/C_t)
a								
Whole period								
LMR	1.5	0.62	0.66	1.80	9.5	2.4	7.1	0.25
LBL	2.0	0.51	0.43	3.00	20.0	4.1	15.9	0.21
MPR	2.5	0.43	0.82	1.25	16.4	2.6	13.8	0.16
MCC	2.0	0.46	0.19	2.31	15.8	2.7	13.1	0.17
API	2.5	0.48	0.68	1.14	12.4	1.8	10.6	0.14
AAP	3.0	0.42	0.89	1.82	9.0	1.8	7.2	0.20
Winter								
LMR	2.0	0.72	0.90	0.99	9.5	2.7	6.8	0.28
LBL	1.5	0.64	0.19	3.72	18.3	4.6	13.7	0.25
MPR	2.0	0.54	0.26	1.79	16.1	2.1	14.0	0.13
MCC	1.5	0.55	0.38	1.80	12.8	2.3	10.5	0.18
API	3.0	0.47	0.39	1.62	10.4	1.6	8.8	0.15
AAP	3.0	0.52	0.73	0.90	8.0	1.6	6.4	0.20
Spring								
LMR	1.5	0.67	0.83	1.42	13.5	3.9	8.9	0.29
LBL	1.5	0.70	0.64	1.96	20.3	4.8	7.1	0.24
MPR	2.0	0.52	0.15	1.58	13.8	1.8	12.0	0.13
MCC	1.5	0.55	0.27	2.31	15.5	3.0	12.5	0.19
API	2.0	0.72	0.38	1.65	10.9	1.7	9.2	0.16
AAP	2.0	0.59	0.58	1.07	8.7	1.4	7.3	0.16
Summer								
LMR	2.5	0.48	0.21	2.09	20.6	2.8	17.8	0.14
LBL	3.5	0.56	0.75	1.01	10.5	2.7	7.8	0.26
MPR	3.0	0.62	0.29	2.15	17.9	2.9	15.0	0.16
MCC	2.5	0.59	0.35	1.34	19.1	2.9	16.2	0.15
API	2.0	0.58	0.23	1.74	14.1	1.5	12.6	0.11
AAP	2.0	0.50	0.65	0.90	9.9	1.4	8.5	0.14
Autumn								
LMR	2.5	0.76	0.23	1.47	11.7	2.5	9.2	0.21
LBL	2.5	0.72	0.72	1.02	20.8	3.3	17.5	0.16
MPR	1.5	0.58	1.38	1.01	15.4	2.9	12.5	0.19
MCC	2.0	0.53	0.92	0.78	16.5	2.8	13.7	0.17
API	3.0	0.44	0.34	1.36	9.9	1.4	8.5	0.14
AAP	2.5	0.62	0.73	1.04	9.2	1.9	7.3	0.28
b								
Weekdays								
LMR	1.5	0.44	0.64	1.95	19.0	2.5	17.5	0.14
LBL	2.0	0.56	0.09	2.84	17.4	3.9	13.5	0.22
MPR	3.0	0.57	0.23	1.68	17.2	2.8	14.4	0.16
MCC	3.0	0.62	0.48	0.97	17.3	3.2	14.1	0.18
API	2.5	0.41	0.81	1.09	12.9	1.9	11.0	0.19
AAP	3.0	0.56	1.13	0.81	9.6	1.7	7.9	0.23
Weekends								
LMR	3.5	0.43	0.31	2.33	18.0	1.8	16.2	0.12
LBL	3.5	0.51	0.86	2.20	11.9	1.8	10.1	0.15
MPR	2.0	0.40	0.27	1.14	16.2	2.0	14.2	0.12
MCC	2.0	0.39	0.84	1.36	16.1	2.1	14.0	0.13
API	2.0	0.47	0.24	1.73	10.9	1.3	9.6	0.12
AAP	3.0	0.51	0.85	0.89	7.3	1.7	5.6	0.23
Day								
LMR	2.0	0.45	0.27	2.14	16.7	3.1	13.6	0.19
LBL	2.5	0.55	0.46	1.19	8.9	2.3	6.6	0.26
MPR	2.0	0.57	0.42	1.05	17.5	3.1	14.4	0.18
MCC	2.0	0.41	0.31	1.39	18.7	3.2	15.5	0.17
Night								
LMR	3.0	0.67	0.34	1.84	11.6	2.2	9.4	0.19
LBL	2.5	0.69	0.46	0.84	6.4	1.5	4.9	0.23
MPR	3.0	0.51	0.02	2.01	18.3	2.2	16.1	0.12
MCC	3.0	0.57	0.16	2.04	17.0	2.3	14.7	0.14

3.3. Roadside enhancement of different PM fractions

We subtracted hourly mean concentrations of PM_c, $PM_{2.5}$ and NO_x measured at urban background stations from those recorded in traffic stations, in order to estimate the daily "roadside increment" of the three pollutants.

Fig. 4 show scatter plots of the daily mean enhancement of particle concentrations ($PM_{2.5}$ and PM_c) against the NO_x roadside enhancement. For London a strong correlation is found for both PM_c and $PM_{2.5}$ with NO_x concentrations ($R = 0.78$ and $R = 0.94$ respectively) but this relationship is stronger for $PM_{2.5}$ (Fig. 4a and b). This reflects the common sources of $PM_{2.5}$ and NO_x, i.e. vehicle exhaust emissions. The relationship between daily increment of PM_c vs. NO_x presents lower correlation coefficients ($R = 0.78–0.47$) than $PM_{2.5}$ vs. NO_x increments ($R = 0.94–0.61$) in all the cities studied, indicating that PM_c is associated with a wider range of sources included non-exhaust vehicle emissions and natural sources. A similar pattern was observed in Madrid (Fig. 4c and d) and Athens (Fig. 4e and f), although the correlation coefficients were lower than in London probably because of the larger contribution of windblown dust events to coarse particle levels observed in these two cities.

This analysis indicates that vehicle related sources have an impact on observed coarse particle levels measured at roadside locations, but this impact is generally smaller for PM_c compared to $PM_{2.5}$ and varies substantially between cities.

The net roadside increment is 6.8 $\mu g\ m^{-3}$ for London and 3.4 $\mu g\ m^{-3}$ for Athens. If the difference between roadside and background mean concentrations is wholly attributed to traffic differences, then the roadside traffic contribution to the mean annual concentrations observed at LMR and API would be 48% and 27%, respectively. For the background site, a lower bounds estimate on the impact of traffic can be provided by the weekday-weekend difference of concentrations (Jones et al., 2008). The weekday increments at LBL and AAP are 1.3 $\mu g\ m^{-3}$ and 2.3 $\mu g\ m^{-3}$ corresponding to 19% and 26% of the annual average, respectively.

3.4. Coarse particles and local meteorology

One of the main questions related to coarse particles is whether there is a significant correlation between their concentrations and meteorological parameters other than wind speed. Table 4 presents the correlation coefficients between temperature, relative humidity, precipitation, atmospheric pressure and wind speed with coarse particle concentrations. With the exception of LMR, the coarse particles concentrations are correlated with temperature, relative humidity and atmospheric pressure at 95% confidence level. The stronger correlations were found at MCC for temperature (positive correlation) and relative humidity (negative correlation). It is expected that relative humidity and precipitation anti-correlate with PM_c since wetter conditions are associated with lower PM_c concentrations due to the hygroscopic nature of coarse particles. A significant positive correlation was found between PM_c concentrations and atmospheric pressure indicating an increase of PM_c with anticyclonic conditions, due to the associated stable conditions that favours resuspension of PMcs, in London (LBL only) and Madrid (MPR and MCC) but not in Athens.

Table 4
Correlation coefficients of PM_c concentrations vs. meteorological variables.

	Temperature (°C)	Relative humidity (%)	Atmospheric pressure (hPa)	Precipitation (mm)	Wind speed ($m\ s^{-1}$)
LMR	0.22*	0.012	−0.031	0.019	0.32*
LBL	0.12*	−0.22*	0.25*	−0.21*	0.25*
MPR	0.18*	−0.12*	0.24*	−0.20*	0.34*
MCC	0.38*	−0.37*	0.19*	−0.22*	0.23*
API	0.14*	−0.25*	−0.11*	−0.08	0.46
AAP	0.30*	−0.12*	−0.21*	−0.08	0.39*

*Values significant at 95% confidence level.

3.5. Long-range transport

The analysis of the back trajectories arriving in London shows that the air masses associated with high values of the $PM_c/PM_{2.5}$ ratio can be mainly split into two categories, those whose origin is over the Atlantic Ocean (77%) and those with origin over continental Europe (23%). The air masses having their origin over continental Europe are associated with higher concentrations of all fractions of particulates, probably due to the substantial emissions of natural and anthropogenic particles over that region. It must be noted however that all these trajectories have relatively short pathways, which can also result in higher coarse particle loads. Oceanic trajectories could be further divided in two almost equal sub-classes (in terms of number of members) consisting of trajectories with long and short pathways all coming from the west or southwest of the Ocean (Table 5) and not directly affected by African dust transportation. It must be noted that, as Ryall et al. (2000) found, African dust events contrarily to the other two capitals are quite rare in London. Pathways with distance of less than 1000 km between their origin and the point of arrival are defined as short (Markou and Kassomenos, 2010).

For Madrid, the analysis of back trajectories revealed four different classes. The first consisted of back trajectories having their origin over the Atlantic Ocean (further separated in trajectories having short (9.4%) or long (40.6%) pathways). The second group consisted of short (3.1%) or long (18.8%) trajectories over France or western Europe in general. The third category comprised trajectories originating from Sahara desert with only short pathways (9.4%), and finally the last category contained back trajectories having their origin over the Iberian peninsula (having all short pathways) (18.7%). The third class (of Sahara desert origin) is associated with the highest concentrations of PM_c, $PM_{2.5}$ and PM_{10} at the two Madrid stations (Table 5). This is consistent with finding from previous studies indicating that long-range atmospheric transport from North Africa can have a significant impact on PM_{10} levels in Madrid (Borge et al., 2007).

For Athens, the analysis revealed four patterns, the first consisting of 44.8% of the high $PM_c/PM_{2.5}$ days is associated with trajectories with origin either over North Sahara desert (mainly over Libya), or the Gulf of Sidra and the maritime area between Crete and the North African Coast, having all short pathways, the second (27.6%) consists of air masses coming from the west and having their origin over the western Mediterranean Sea (long pathways), the third (13.8%) is similar to the second but with short pathways, while the fourth (13.8%) is associated with air masses coming from the north having short pathways. The second category is associated with the highest concentrations of both PM_{10} and PM_c following by the first category. The latter is associated with lower $PM_{2.5}$ indicating that long-range transport of coarse particles from Sahara desert may have a significant contribution to PM_{10} episodes in Athens (Grivas et al., 2008; Vardoulakis and Kassomenos, 2008).

The third category (Western Mediterranean origin, short pathways) may be associated with the transport of mainly sea salt particles, due to its pathway over the sea, and the observed particle ratio is shifted towards $PM_{2.5}$, having a smaller influence on coarse particles, as compared to African dust outbreaks (Table 5). These results are consistent with the findings of Grivas et al. (2008).

Table 5
Origin of the air masses associated with high $PM_c/PM_{2.5}$ values, percentage of occurrence, mean $PM_c/PM_{2.5}$ ratio, PM_c, $PM_{2.5}$ and PM_c values.

	%	$PM_c/PM_{2.5}$	PM_c ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)	PM_{10} ($\mu g\ m^{-3}$)	Mean length (m)
LONDON						
1. Atlantic Ocean (Short)	30.8	1.36	11.8	9.3	21	632
2. Atlantic Ocean (Long)	46.2	1.46	14.0	9.6	23.6	2525
3. Continent	23	1.36	14.5	10.4	24	545
MADRID						
1. Atlantic Ocean (Short)	9.4	2.6	25	9.3	34.2	832
2. Atlantic Ocean (Long)	40.6	2.8	27	11	37.9	2101
3. Continent (Short)	18.8	2.7	29.8	12.1	42.4	472
4. Continental (Long)	3.1	2.1	19.3	9.4	28.7	1897
5. Local	18.8	1.8	29.4	10.8	37.3	393
6. Sahara	9.4	2.1	46.3	22.3	68.7	565
ATHENS						
1. North Africa	44.8	1.2	16.2	13.3	35.0	674
2. West Mediterranean (Long)	27.6	1.3	13.0	9.4	22.4	1874
3. West Mediterranean (Short)	13.8	1.1	19.0	16.0	35.0	875
4. North Balkans	13.8	1.0	15.5	16.0	31.5	713

4. Conclusions

In this study, an analysis of the levels, seasonality, origin and meteorological factors affecting coarse particle concentrations in three European capitals was carried out. The levels of PM_c range between 25% and 48% of PM_{10} at the selected traffic stations in all cities while the relevant percentages were about 33%–52% in the urban background sites. PM_c levels appeared generally higher during the drier periods of the year in all three cities. As expected, PM_c concentrations were higher (by 19–35%) during working days especially at the traffic stations compared to weekends, while these differences were less marked at the urban background stations. Similarly, daytime PM_c concentrations were higher (from 10 to 36%) than the nighttime levels at all stations.

The wind resuspended component of the coarse particles was around 10–26% of the total coarse particle mass. On the other hand, the non-wind driven component of the coarse particles was around 74–90% of the total coarse particle mass in the three examined cities.

This has significant implications for air quality management, as the wind resuspended component of total coarse particle matter is very difficult to control. PM_c is therefore expected to play an increasingly significant role in the control of PM_{10} in urban areas, as direct vehicle exhaust emissions of the finer fraction of particulate matter ($PM_{2.5}$) are likely to decline in Europe due to the introduction of improved vehicle technologies.

The sources of the coarse particles were also investigated using atmospheric back trajectory modelling. The modelling results indicate that there are significant long-range transport sources of coarse particles in the three urban areas, in addition to local sources. The examination of air masses arriving in London during episodic conditions (high concentrations of coarse particles) revealed associations with trajectories originating from continental Europe. In Athens high PM_c levels were mainly associated with air masses originating either from north Africa or the western Mediterranean, possibly carrying desert dust and/or sea salt particles. In Madrid, in addition to long-range transport from North Africa, a significant contributor to PM_c is the transport of coarse particles from the north of Spain and France. These long-range influences in the three cities are likely to reduce the effectiveness of local PM_{10} control policies. Finally, local meteorology plays a marked role in the dilution of coarse particles in the majority of the cases examined. Coarse particle concentrations are correlated positively with temperature and anti-correlated with relative humidity and precipitation indicating that wetter and rainy conditions reduce resuspension of coarse particles while drier periods enhance resuspension.

Overall, this paper supports the notion that local control of particulate emissions can only have a limited effect in reducing PM_c and PM_{10} in urban streets and background locations (Harrison et al., 2008) and this must be taken in mind in the design of future pollution reduction strategies. This issue has been addressed by several research studies across Europe, evaluating the effectiveness of active local abatement measures, mainly for road dust (e.g. street sweeping and washing). Results however don't display a uniform pattern and appear to depend on local weather conditions (Norman and Johansson, 2006; Amato et al., 2009; Keuken et al., 2010; Karanasiou et al., 2011).

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