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Traffic and nucleation events as main sources of ultrafine particles in high-insolation developed world cities

M. Brines et al.

Correspondence to: M. Brines (mariola.brines@idaea.csic.es)

Cluster number reduction methodology

The number of clusters was conservatively chosen using the Dunn Index and the Silhouette Width. The larger the Dunn Index and Silhouette Width, the more compact and well separated were the clusters and the more similar were the elements within each cluster (Beddows et al., 2009). Preference was given to a solution with a higher cluster number to reduce the likelihood that any one of the clusters grouped together spectra reflects more than one source. Although we reduce the possibility of losing information by 'over-clustering', it is likely that when comparing the average size distributions - together with the corresponding gaseous pollutants, meteorological parameters, and various temporal trends (daily, weekday-weekend, monthly) - that more than one size distribution may (or even may not) originate from a similar process/source. More often than not, when considering the average size distributions and auxiliary measurements from over-clustered data (e.g. similarly low NO concentrations among the clusters, similar daily trends…), one or more clusters are combined together thus reducing the number of clusters in the final solution.

Uncertainty limits calculation for the cluster size distributions

The uncertainty limits for all the clusters size distributions have been calculated at each city using the confidence limits μ:

$$\mu = \text{mean}(x) \pm t\frac{\sigma}{\sqrt{n}}$$

where x are the size bin values dN/dlogDp, n is the number of values used in the average, σ is the standard deviation, t is the Student t-value. We approximated the degrees of freedom to ∞, due to the high number of hours contributing to each cluster - in the range of hundreds to thousands. We considered 99.99% of the confidence level, obtaining a Student t-value of 3.291 according to http://www.webassign.net/harrischem/4-02tab.gif (last entry Dec 2014).

Table S1: Exact locations of the selected SMPS sampling sites, their elevation and site type.

City	Site	Latitude	Longitude	Elevation (m.a.s.l.)	Site type
Barcelona, Spain	Palau Reial	41°23'14''N	2°6'56''E	78 m	Urban background
Madrid, Spain	CIEMAT	40°27'30''N	3°43'30''W	655 m	Suburban background
Brisbane, Australia	QUT	27°28'43''S	153°1'44''E	10 m	Urban background
Rome, Italy	Montelibretti	42°06'38''N	12°38'05''E	47 m	Regional background
Los Angeles, USA	USC	34°1'9''N	118°16'39''W	61 m	Urban background

Table S2: Sampling period of the SMPS instruments and their characteristics and size range at each selected city.

City	Sampling period	SMPS model	SMPS size range	Size bins
Barcelona (BCN)	30/07/2012-04/08/2013 (7295 h)	TSI (DMA 3081, CPC 3772)	11.3-358.7 nm	97
Madrid (MAD)	10/01/2007-12/12/2008 (12482 h)	TSI (DMA 3071, CPC 3022)	17.5-572.9 nm	34
Brisbane (BNE)	01/01/2009-31/12/2009 (6227 h)	TSI (EC 3080, CPC 3781)	10.2-101.8 nm	65
Rome (ROM)	26/09/2007-07/05/2009 (3373 h)	TSI (DMA 3081, CPC 3775)	15.1-224.7 nm (10.2-615.3 nm)	76 (87, 93, 104)
Los Angeles (LA)	04/09/2009-10/12/2009 (2184 h)	TSI (DMA 3081, CPC 3022A)	15.7-371.8 nm	89

Table S3: Summary of the measurements and sampling period at the selected cities. V^+ indicates the measurement site is different to the SMPS site.

City	Ancillary data site	Meteorological data					Gaseous Pollutants					Particulate Matter				Other	
		T	RH	ws/ wd	Rain	SR	NO	NO_2	O_3	CO	SO_2	PM_{10}	$PM_{2.5}$	PM_1	NO_3^-	N	BC
Barcelona (BCN)	Palau Reial, Fac. of Physics+	V^+	V^+	V^+	V^+	V^+	V	V	V	V	V	V	V	V		V	V
Madrid (MAD)	CIEMAT, Casa de Campo+	V	V	V	V	V	V	V	V	V^+	V	V^+	V^+		V		
Brisbane (BNE)	Rocklea+	V^+	V^+	V^+	V^+	V^+	V^+	V^+	V^+			V^+	V^+			V^+	
Rome (ROM)	Buf+	V^+	V^+	V^+	V^+	V^+	V^+	V^+	V^+		V^+						
Los Angeles (LA)	South Coast AQMD+	V^+	V^+	V^+	V^+		V^+	V^+	V^+	V^+	V^+		V^+			V	

Table S4: Overall occurrence (%) of each cluster at each site, classified into different categories (Traffic, Nucleation and Background pollution and Specific cases).

Category	Subcategory	Barcelona	Madrid	Brisbane	Rome	Los Angeles
Traffic	Traffic 1 (T1)	27%	25%	24%	7%	36%
	Traffic 2 (T2)	24%	22%	-	27%	-
	Traffic 3 (T3)	12%	11%	20%	7%	25%
Nucleation	Nucleation (NU)	15%	19%	14%	6%	33%
Background pollution and Specific cases	Urban Background (UB)	15%	6%	22%	25%	6%
	Summer Background (SB)	-	7%	-	-	-
	Regional Background (RB)	-	-	-	28%	-
	Nitrate (NIT)	7%	10%	-	-	-
	Growth 1 (G1)	-	-	10%	-	-
	Growth 2 (G2)	-	-	10%	-	-
Total		100%	100%	100%	100%	100%

Table S5: Log-Normal fitting peaks for each cluster category *k*-Means size distribution at the supplementary sites and the corresponding peak area percentage.

Category	Subcategory	Rome	Los Angeles
Traffic	Traffic 1 (T1)	37±1 nm (65%), 130±7 nm (35%)	21±1 nm (100%)
	Traffic 2 (T2)	59±2 nm (91%), 102±8 nm (9%)	-
	Traffic 3 (T3)	19±1 nm (20%), 75±1 nm (80%)	<15 nm (73%), 66±1 nm (27%)
Nucleation	Nucleation (NU)	23±1 nm (43%), 102±2 nm (57%)	<15 nm (62%), 67±3 nm (38%)
Background pollution and Specific cases	Urban Background (UB)	27±2 nm (46%), 105±1 nm (54%)	45±1 nm (100%)
	Summer Background (SB)	-	-
	Regional Background (RB)	89±1 nm (100%)	-
	Nitrate (NIT)	-	-
	Growth 1 (G1)	-	-
	Growth 2 (G2)	-	-

Table S6: Cluster categories (Traffic, Nucleation and Background pollution and Specific cases) and their occurrence at the supplementary sites.

Category	Rome	Los Angeles
Traffic	41%	61%
Nucleation	6%	33%
Background	53%	6%
	100%	100%

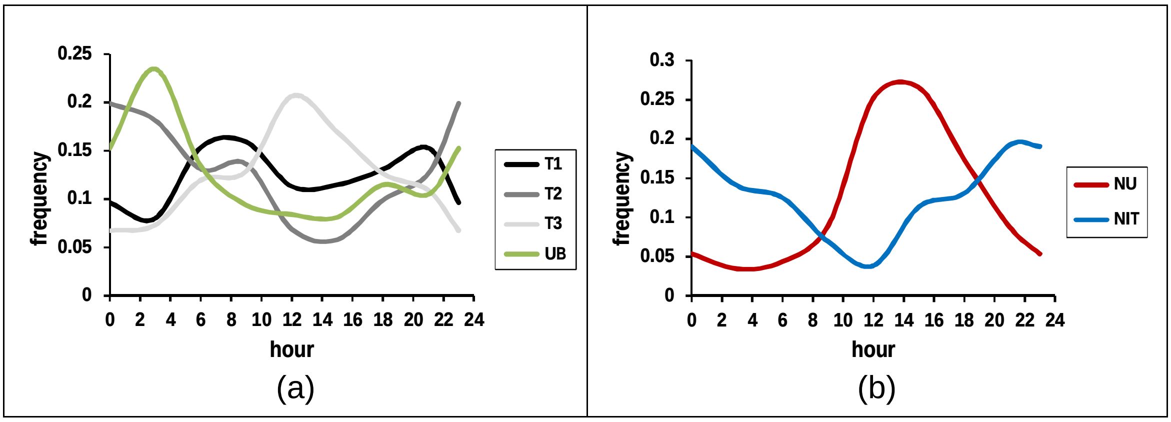
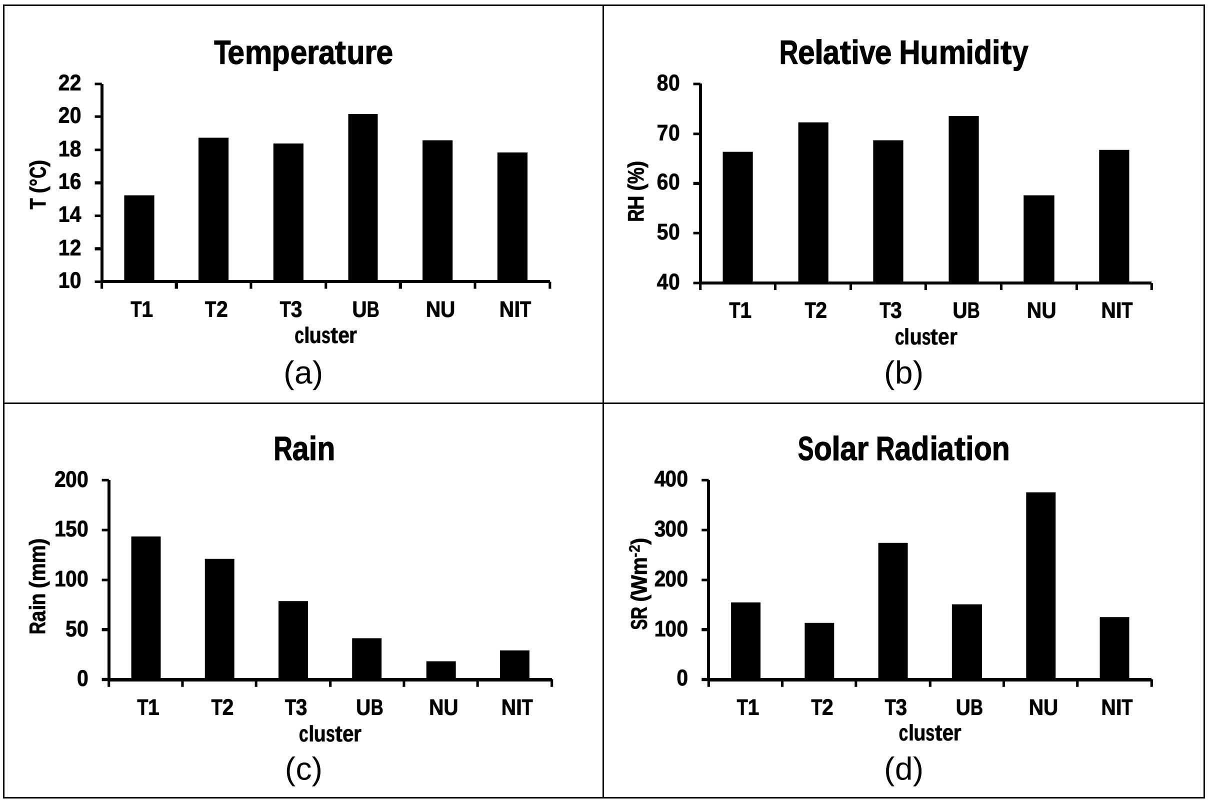
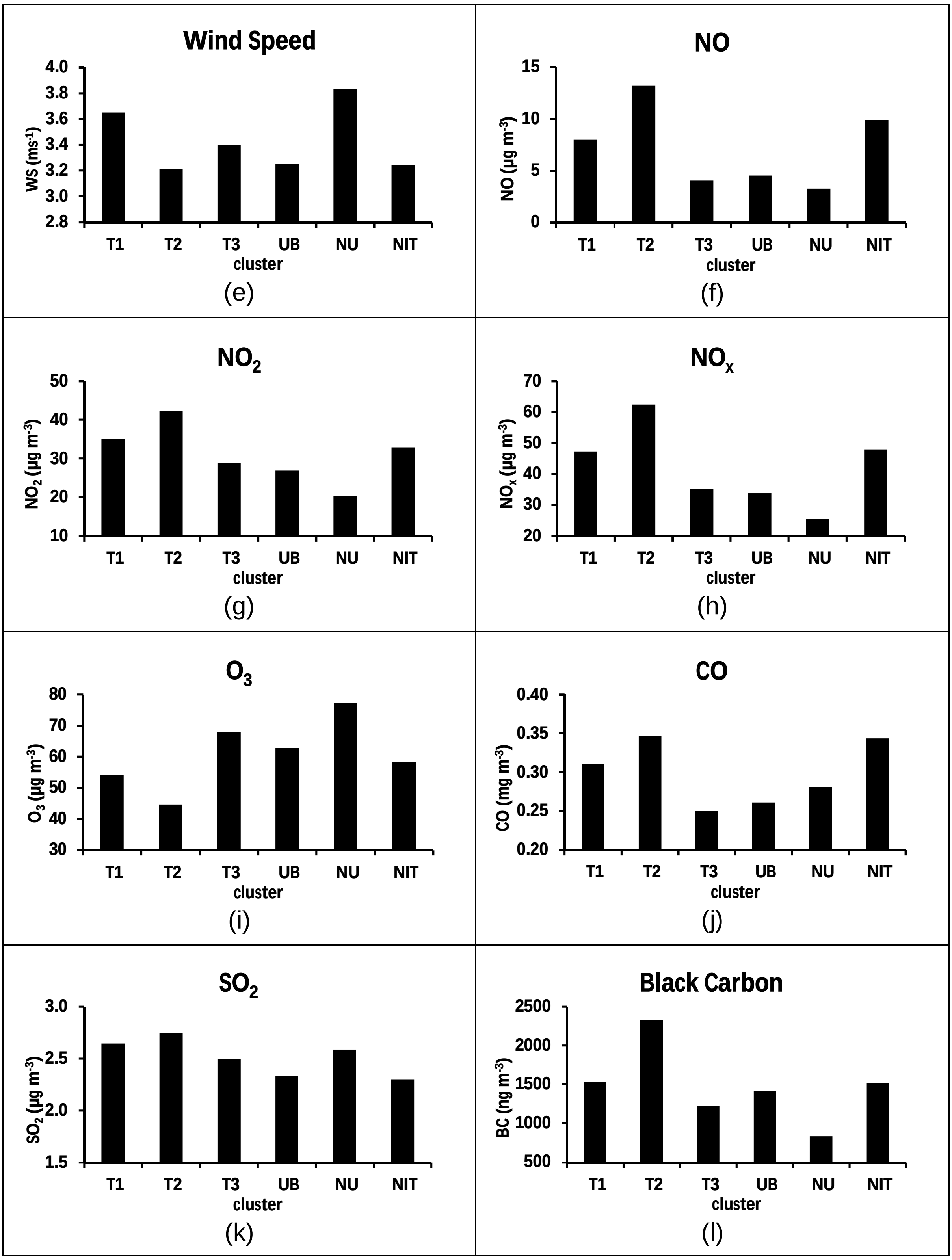


Figure S1: Diurnal trends for the main clusters: a) Traffic1 (T1), Traffic2 (T2), Traffic3 (T3) and Urban Background (UB); b) Nucleation (NU) and Nitrate (NIT). Although they are extracted from the results for Barcelona, they are representative of the rest of the cities.




Wind Speed
WS (ms-1)
4.0
3.8
3.6
3.4
3.2
3.0
2.8
T1 T2 T3 UB NU NIT
cluster
(e)
NO
NO (µg m-3)
15
10
5
0
T1 T2 T3 UB NU NIT
cluster
(f)
NO2
NO2 (µg m-3)
50
40
30
20
10
T1 T2 T3 UB NU NIT
cluster
(g)
NOx
NOx (µg m-3)
70
60
50
40
30
20
T1 T2 T3 UB NU NIT
cluster
(h)
O3
O3 (µg m-3)
80
70
60
50
40
30
T1 T2 T3 UB NU NIT
cluster
(i)
CO
CO (mg m-3)
0.40
0.35
0.30
0.25
0.20
T1 T2 T3 UB NU NIT
cluster
(j)
SO2
SO2 (µg m-3)
3.0
2.5
2.0
1.5
T1 T2 T3 UB NU NIT
cluster
(k)
Black Carbon
BC (ng m-3)
2500
2000
1500
1000
500
T1 T2 T3 UB NU NIT
cluster
(l)

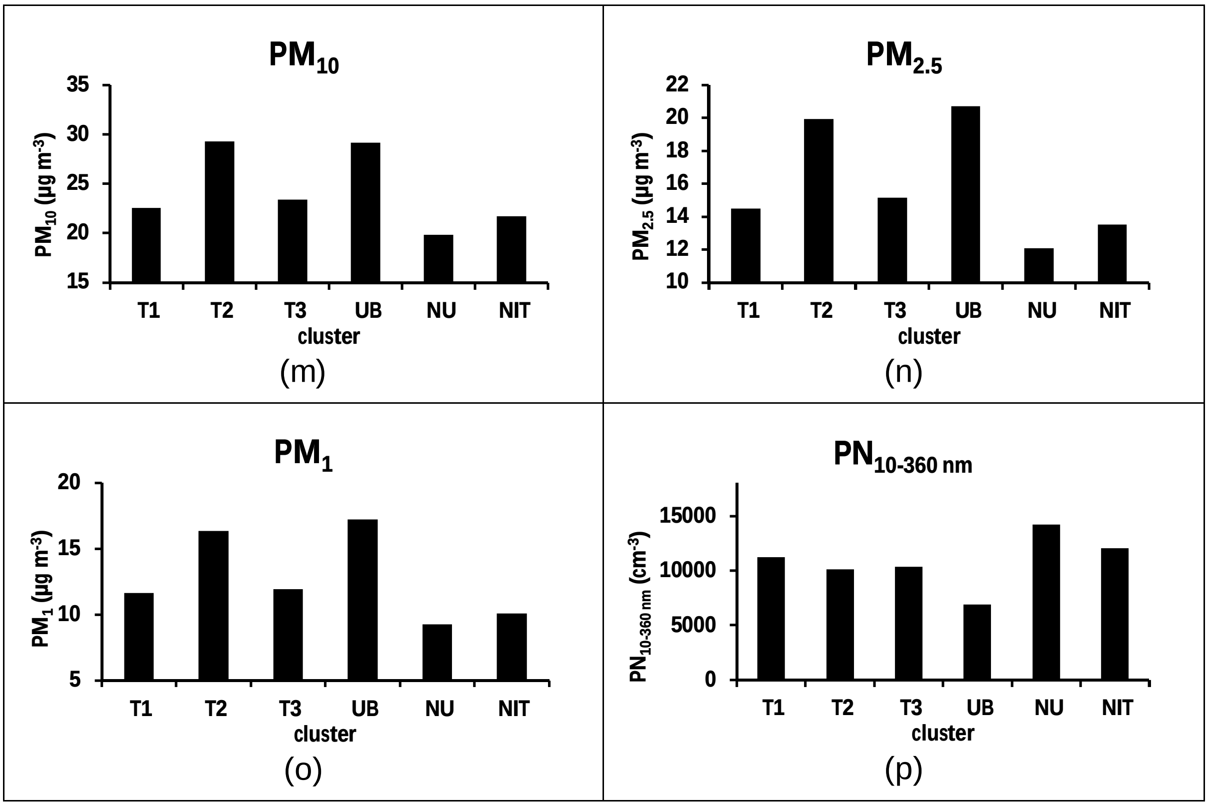


Figure S2: Meteorological parameters and gaseous pollutants for the main clusters: a) Temperature, b) Relative Humidity, c) Rain, d) Solar Radiation, e) Wind Speed, f) NO, g) NO_2, h) NO_x, i) O_3, j) CO, k) SO_2, l) Black carbon, m) PM_{10}, n) $PM_{2.5}$, o) PM_1, p) PN_{10-360}. Although they are extracted from the results for Barcelona, the trend followed by clusters is representative of the rest of the cities.

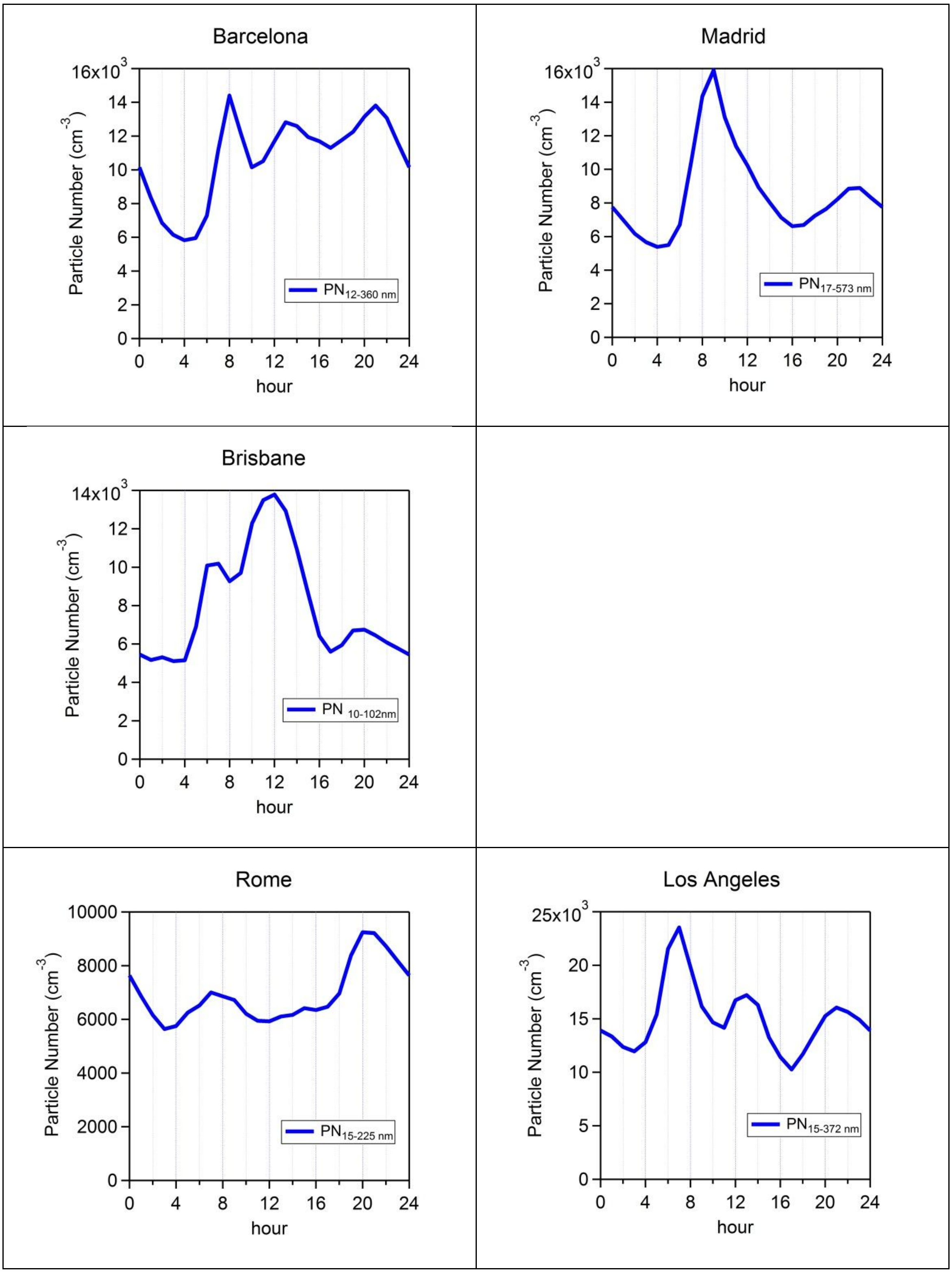


Figure S3: Average daily particle number concentration for the study periods at each city.