Supporting Information

1) **Effect of the change of the location of the measuring station in BCN in 2009**

![Figure SI-1](image): Trends of PM$_{10}$ concentrations from gravimetric measurements at BCN and MSY. Red rectangle highlights the decrease of PM$_{10}$ concentration at BCN due to the change of the location of the BCN measuring station in 2009.

2) **Trend of Zn concentrations in PM$_{2.5}$ at MSY**

![Figure SI-2](image): (a) Trend of Zn in PM$_{2.5}$ at MSY;
3) Trend of Sn concentrations in PM$_{2.5}$ at MSY

![Figure SI-3: (b) Trend of Sn in PM$_{2.5}$ at MSY;](image)

TZ$_{MK} = -3.74$; $S_{MK} = ***$

$a_1=0.0483$; $r_1=0.23$

$a_2=0.0010$; $r_2=11.79$

NL=43.7% TR=76% RC=9%

4) PMF source profiles at BCN and MSY

![Figure SI-4: Chemical profiles of the PMF sources at MSY (a) and BCN (b)](image)

Source profiles from PMF analysis at BCN and MSY. Common sources at both BCN and MSY were: Ammonium Sulfate (secondary inorganic source traced by SO$_4^{2-}$, NH$_4^+$ with contribution from OC), Ammonium nitrate (secondary inorganic source traced by NO$_3^-$ and NH$_4^+$), V-Ni bearing source (traced mainly by V, Ni and SO$_4^{2-}$ it represents the direct emissions from heavy
oil combustion), mineral (traced by typical crustal elements such as Al, Ca, Ti, Rb, Sr), aged marine (traced by Na and Cl mainly with contributions from \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \)). Non common sources at MSY were: Anthropogenic source (traced by EC, OC, Cr, Cu, Zn, As, Cd, Sn, Sb and Pb includes contributions from anthropogenic sources such as traffic and metallurgic) and aged organics (traced by OC and EC mainly with maxima in summer indicating a biogenic origin). Non common sources at BCN were: traffic (traced by Cnm, Cr, Cu, Sb and Fe mainly and contributing 5.14 µg/m³ (15.1%)), road resuspension (traced by both crustal elements, mainly Ca, and traffic tracers such as Sb, Cu and Sn and contributing 4.25 µg/m³ (12.5%)) and Industrial/metallurgy (traced by Pb, Cd, As and Zn and contributing 0.96 µg/m³ (2.8%).

5) **OC:EC ratio statistic at Montseny (MSY)**

![OC:EC ratio statistic at Montseny (MSY)](image)

**Figure SI-5**: OC and EC scatterplot (a) and frequency distribution of the OC:EC ratio (b) at Montseny (MSY) station.

Mean and median values of the OC:EC ratio at MSY were 9.1 and 7.8, respectively.
6) **PMF Barcelona: 2007-2014 (with OC and EC) vs. 2004 – 2014 (with Cnm)**

![Graphs showing PMF results comparison](image)

**Figure SI-6**: Comparison between PMF results at BCN obtained using the period 2007-2014 (separate OC and EC measurements available) and using the whole period 2004-2014 (Cnm was used in PMF).

7) **OC:EC ratio statistic at Barcelona (BCN)**

![Graphs showing OC:EC ratio statistics](image)

**Figure SI-7**: OC and EC scatterplot (a) and frequency distribution of the OC:EC ratio (b) at Barcelona (BCN) station.