Supplement of


Marco Pandolfi et al.

Correspondence to: Marco Pandolfi (marco.pandolfi@idaea.csic.es)

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Supporting Information

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

Figure SI-1: 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) PMF source profiles at BCN and MSY

![Chemical profiles of the PMF sources at MSY (a) and BCN (b)](image)

Figure SI-2: Chemical profiles of the PMF sources at MSY (a) and BCN (b)

Source profiles from PMF analysis at BCN and MSY. Common sources at both BCN and MSY were: **Secondary Sulfate** (secondary inorganic source traced by SO$_4^{2-}$, NH$_4^+$ with contribution from OC), **Secondary nitrate** (secondary inorganic source traced by NO$_3^-$ and NH$_4^+$), **V-Ni bearing** (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 SO$_4^{2-}$ and NO$_3^-$). Non common sources at MSY were: **Industrial/Traffic source** (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/m$^3$ (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/m$^3$ (12.5%)) and **Industrial** (traced by Pb, Cd, As and Zn and contributing 0.96 µg/m$^3$ (2.8%).
3) *OC:EC ratio statistic at Montseny (MSY)*

**Figure SI-3:** 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.

4) *PMF Barcelona: 2007-2014 (with OC and EC) vs. 2004 – 2014 (with Cnm)*

**Figure SI-4:** 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).
5) **OC:EC ratio statistic at Barcelona (BCN)**

![Figure SI-5: OC and EC scatterplot (a) and frequency distribution of the OC:EC ratio (b) at Barcelona (BCN) station.](image)