

Supporting Information

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

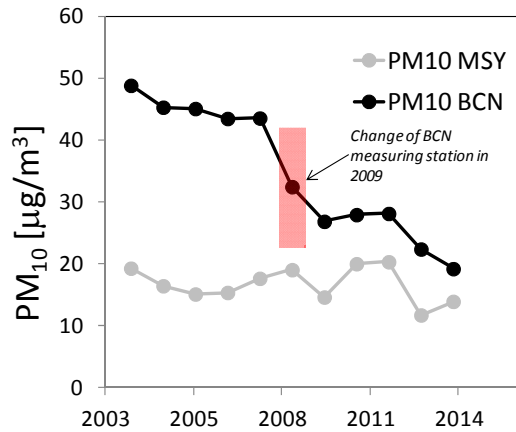


Figure SI-1: Trends of PM₁₀ concentrations from gravimetric measurements at BCN and MSY. Red rectangle highlights the decrease of PM₁₀ concentration at BCN due to the change of the location of the BCN measuring station in 2009.

2) Trend of Zn concentrations in PM_{2.5} at MSY

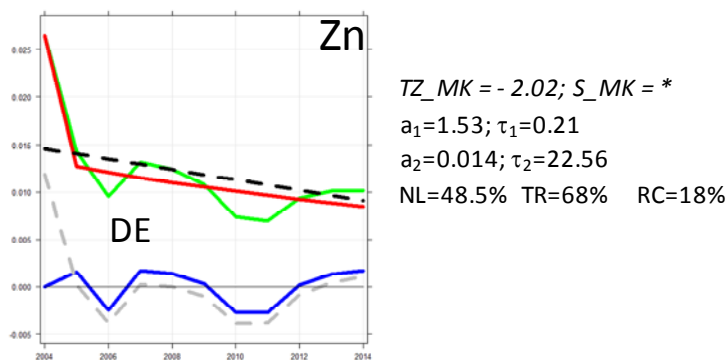


Figure SI-2: (a) Trend of Zn in PM_{2.5} at MSY;

3) Trend of Sn concentrations in PM_{2.5} at MSY

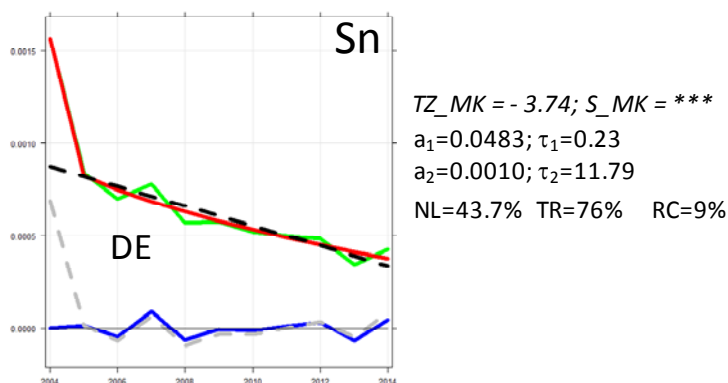


Figure SI-3: (b) Trend of Sn in PM_{2.5} at MSY;

4) PMF source profiles at BCN and MSY

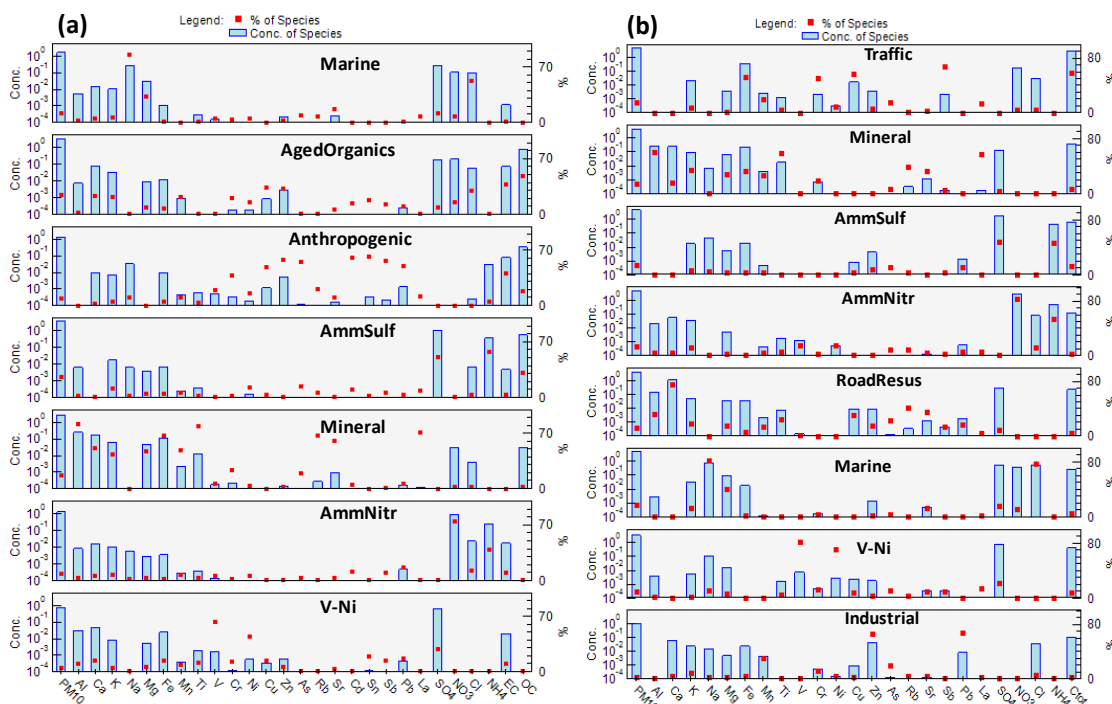


Figure SI-4: Chemical profiles of the PMF sources at MSY (a) and BCN (b)

Source profiles from PMF analysis at BCN and MSY. Common sources at both BCN and MSY were: *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 SO_4^{2-} and 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 $5.14 \mu\text{g}/\text{m}^3$ (15.1%)), *road resuspension* (traced by both crustal elements, mainly Ca, and traffic tracers such as Sb, Cu and Sn and contributing $4.25 \mu\text{g}/\text{m}^3$ (12.5%)) and *Industrial/metallurgy* (traced by Pb, Cd, As and Zn and contributing $0.96 \mu\text{g}/\text{m}^3$ (2.8%).

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

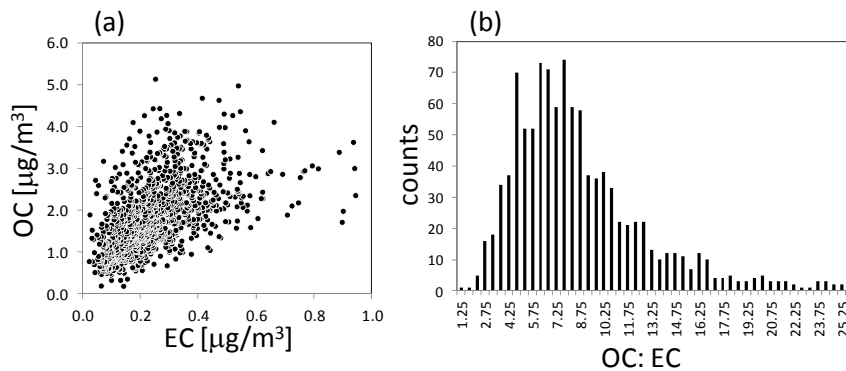


Figure SI-5: OC and EC scatterplot (a) and frequency distribution of the OC:EC ratio (b) at Montseny (MSY) station.

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)**

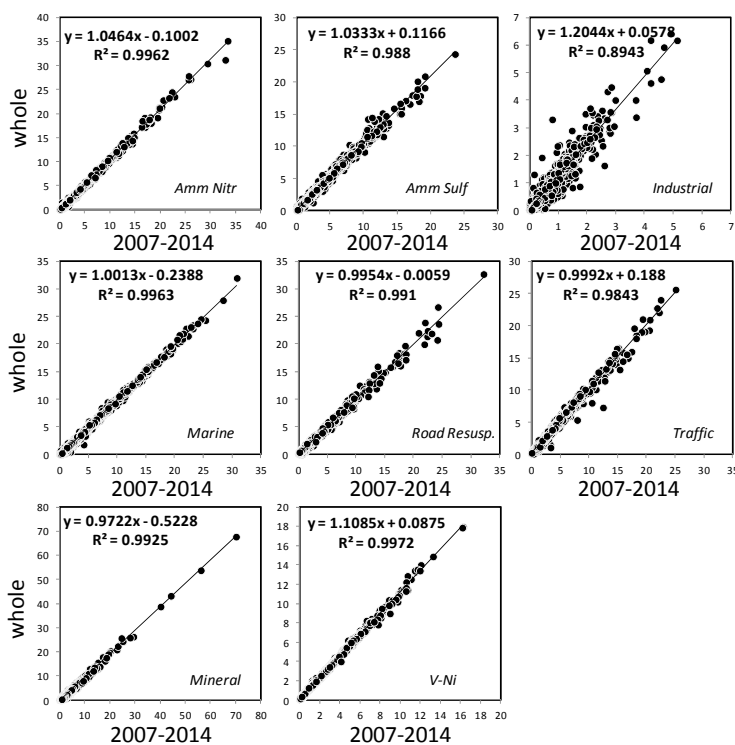


Figure SI-6: Comparison between PMF results at BCN obtained using the period 2007-2014 (separate OC and EC measurements available) and using the whole period 2004-2014 (Cnm was used in PMF).

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

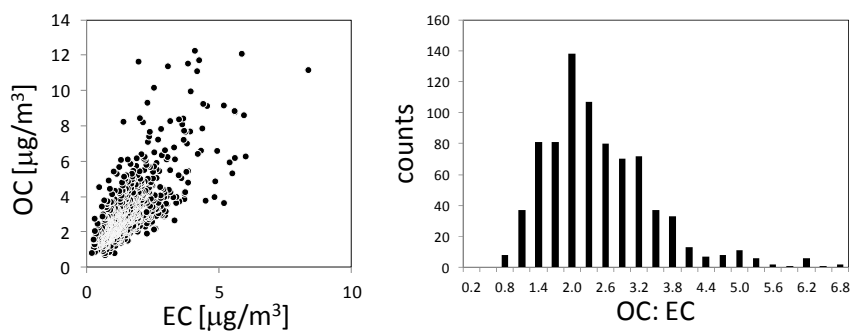


Figure SI-7: OC and EC scatterplot (a) and frequency distribution of the OC:EC ratio (b) at Barcelona (BCN) station.