



Evidence a major source of water insoluble secondary organic aerosols in the region of Paris (France) during wintertime

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Near real time measurements of carbonaceous aerosols were performed in the fine mode (aerodynamic diameter, A.D. < 2.5 μ m) during the winter 2009 at a suburban site of Paris (France). Elemental carbon (EC) and organic carbon (OC) were determined semi-continuously every hour using a Sunset Field instrument; absorption measurements were performed every 5 min using a seven wavelength aethalometer; and water soluble organic carbon (WSOC) was determined every 4 min using a particle-into-liquid-sampler (PILS) connected with a total organic carbon (TOC) analyzer. Filter sampling was performed in parallel and showed very good agreements for EC, OC, and WSOC with on-line analyzers.

The aethalometer model (Sandradewi et al., 2008) applied to semi-continuous measurements of OC was used to derive 3 sources of organic aerosols (OA): fossil fuel, wood burning, and secondary. This source apportionment was tested for primary OA (fossil fuel, wood burning) against time-resolved measurements of tracers commonly used for these combustion sources (levoglucosan, watersoluble potassium, methanol for wood burning; nitrogen oxide and xylene for fossil fuel). A good agreement was obtained from these comparisons, giving further confidence on the results of the Aethalometer model to properly determine primary OA. Based on this model, wood burning source appeared to contribute as much as 70% of the total mass of combustion aerosols (40% to the total mass of carbonaceous matter). Contribution of wood burning aerosols to PM_{2.5} was ranging from 4 to 56% (average of 17 \pm 11%). Residual Organic matter (i.e. secondary organic aerosols, SOA) showed significant concentration levels during our study, ranging from 1.5 to 10.0 μ g/m³ (average 4.88 \pm 1.56 μ g/m³). These SOA levels are among the highest reported in literature for Europe during wintertime.

In order to better characterize these SOA, a source apportionment of hourly concentrations of WSOC and WIOC (calculated as OC-WSOC) was performed using the 3 OA sources obtained with the Aethalometer model. A very large fraction (82 \pm 5%) of organic carbon originating from wood burning could be assigned as watersoluble in agreement with literature data. An independent estimate of the OC-OM conversion factor of 1.86 \pm 0.07 was calculated here for wood burning, which is in the range of those commonly reported in literature. Negligible (<1%) of fossil fuel was identified as watersoluble, which is also consistent with the idea that primary OA from fossil fuel are poorly oxidised and mainly water insoluble. About 20% of WSOC was found to be of secondary origin, with a clear diurnal pattern showing a maximum during daytime linked to local photochemical production.

Less expected is the large fraction of secondary organic carbon (~85%) identified as water insoluble. This fraction did not show an important diurnal variation suggesting a weak local photochemical production, and pointing to a more regional (long range transport) origin. The weak biogenic emissions during wintertime suggest that our water insoluble SOA could originate from anthropogenic VOC which is consistent with recent studies which have shown that semi-volatile primary anthropogenic OA could lead to significant amount of SOA (Robinson et al., 2007). These results bring new light on the commonly accepted idea that SOA is mainly water soluble. They have been obtained in a typical suburban area of France and then may be representative of a larger European area. They have also important implication for the SOA lifetime which is partly controlled by removal processes in which the water soluble properties play an important role.