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Oxidation of organic aerosol over the Po Valley basin observed at Mt. Cimone (2165 m asl), Italy

Matteo Rinaldi (1), Stefania Gilardoni (1), Stefano Decesari (1), Marco Paglione (1), Sandro Fuzzi (1), Paolo Cristofanelli (1), Paolo Bonasoni (1), Vanes Poluzzi (2), Silvia Ferrari (2), and Maria Cristina Facchini (1)
(1) CNR, ISAC, Bologna, Italy (m.rinaldi@isac.cnr.it), (2) Arpa Emilia-Romagna, Bologna, Italy

High resolution time of flight aerosol mass spectrometer (HR-ToF-AMS) measurements have been performed, for the first time, at Mt. Cimone GAW station (44°12' N, 10°42' E, 2165 m asl) between June and July 2012, under the framework of the EU project PEGASOS and the Emilia-Romagna Region project SUPERSITO.

The main aerosol components (organics, sulphate, ammonium and nitrate) show a clear concentration trend, at the sampling site, as a result of the planetary boundary layer (PBL) daily evolution. The highest concentrations are observed during the day, when the site is within the PBL and is affected by the aerosol sources located within the Po Valley basin. Conversely, the concentrations reach their minimum at night, when the top of Mt. Cimone is above the shallow nocturnal layer, in the residual layer (LR), disconnected to the underlying aerosol sources.

Thanks to the peculiar location of the site, it is possible to use the dataset to investigate the ageing of organic aerosol (OA) occurring at regional scale over the Po Valley basin. In fact, particles sampled at Mt. Cimone during the day (PBL samples) are fresher and are representative of the early stages of aerosol atmospheric oxidation. During the night, the aerosol sampled at the site (RL samples) is more aged, as the residual layer contains particles with an age from several hours to days.

Elemental analysis performed with high resolution mass spectra (Aiken et al., 2007), revealed decreasing average H/C and increasing O/C ratio from PBL to RL samples. As a consequence, the average OM/OC ratio passes from 1.83 ± 0.05 in PBL, to 1.94 ± 0.08 in RL samples. These results evidence the progressive oxidation of OA over the Po Valley basin, from few hours after their emission/formation to one or more days of atmospheric processing. On a Van Krevelen space, the data produce a slope of ~ -1, suggesting that the observed regional scale oxidation processes occur mainly through the addition of carboxylic functional groups (Heald et al., 2010). This is further confirmed by the analysis of the HR mass fragments, showing an enhanced contribution of fragments containing carbon, hydrogen and more than one oxygen atom, in RL samples with respect to PBL ones.

Analysis of the AMS size distributions shows no appreciable aerosol growth with ageing, suggesting that the observed oxidation process occurs without significant production of new particulate mass. This points to heterogeneous reactions with gaseous oxidants as a significant ageing process.

Positive Matrix Factorization (PMF) was performed on the high resolution organic mass spectra collected through the whole campaign. Preliminary results support the above findings on aerosol ageing.

Aiken et al. (2007), Anal. Chem. 79, 8350-8358.

Heald et al. (2010), Geophys. Res. Lett. 37, L08803.