



A detailed analysis of the organic aerosol budget over Paris during the MEGAPOLI campaign

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We performed a case study of the organic aerosol (OA) budget on the MEGAPOLI summer campaign data to gain new insights into the sources and diurnal production of OA over Paris. We combined Aerosol Mass Spectrometer (AMS) datasets and observations of gas-phase chemistry and boundary layer dynamics with a 1D model, including a detailed description of OA formation and photochemical evolution in the boundary layer. Semi-volatile organic components are distributed into volatility bins based on their saturation concentration and can partition into the aerosol phase. Furthermore, the semi-volatile organics in the gas phase can react with OH radicals leading to compounds with lower volatility and hence continued OA formation. The model explicitly represents the gas-phase chemistry leading to the formation of semi-volatile organics and the diurnal dynamics of the atmospheric boundary layer.

An evaluation of the model against observations shows that it reproduces boundary layer dynamics and gas-phase chemistry satisfactorily. A base case simulation of the evolution of several OA components forms the basis for a series of detailed numerical experiments. We find that during daytime hydrocarbon-like OA (HOA) concentrations over Paris are mainly driven by dilution due to boundary layer dynamics, and semi-volatile oxidized OA (SV-OOA) concentrations by local photochemistry. We analyze the formation of SOA from biogenic and anthropogenic VOC, and from fuel combustion semi- and intermediate-volatile organic compounds (S/IVOCs). Budget calculations are performed to show the contribution of the various processes (formation of semi-volatile organics, gas/particle partitioning, dilution of the background OA, etc.) to the calculated OA mass.

Finally, the influence of uncertainties in several processes that determine the OA budget is systematically analyzed through sensitivity analyses. These include 1) the number and range of the volatility bins, 2) the number of gas-phase aging steps of the S/IVOCs and 3) the volatility and concentration of the background aerosol. We evaluate the ability of the model to reproduce observed OA mass factors and O:C ratios for the combinations of these parameters. We conclude with recommendations for large-scale OA modeling.