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Diurnal Evolution of Organic Aerosols over Paris: Insights from the Combination of Measurements during the MEGAPOLI campaign with a 1D Model

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In spite of rapid developments in our understanding of organic aerosol (OA) physicochemical properties, representing the OA composition and evolution over urban areas remains a challenge. This study addresses the diurnal evolution of OA over Paris during the MEGAPOLI campaign. We analyze the observations with a model that aims at a balanced representation of the various processes that contribute to the diurnal variation of the organic aerosol budget. It is a 1D Eulerian model of the atmospheric boundary layer that contains advanced modules for gas-phase chemistry, gas/particle partitioning, and dry deposition. The model represents a computationally efficient framework for the accurate 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 are allowed to partition into the aerosol phase. Furthermore, the semi-volatile organics in the gas phase continue to react with OH radical leading to compounds with lower volatility and hence continued OA formation.

Model results are evaluated against available observations of OA, gas-phase chemistry and boundary layer dynamics. The model results are used along with the Aerosol Mass Spectrometer (AMS) dataset from the MEGAPOLI campaign to give new insights into the sources and diurnal production of OA over Paris. Furthermore, budget calculations are performed to show the contribution of the various processes (i.e. photochemistry, aerosol thermodynamics, boundary layer dynamics, etc.) to the calculated OA mass.

Finally, the influence of uncertainties in several processes that determine the OA budget on the calculated OA properties is systematically analyzed through a series of sensitivity analyses. These include emission fractions of semivolatile and intermediate volatile compounds (SVOC/IVOC), secondary OA yields for the various gas-phase precursors, gas-phase aging of SVOC and IVOC during several generations of oxidation, dry deposition of OA and its gas-phase precursors, the temperature dependence of gas/particle partitioning, and assumptions on the volatility and entrainment of the background OA concentration.