



Modelling concentrations and properties of secondary aerosols in the Western Mediterranean

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In the framework of the Chemistry-Aerosol Mediterranean Experiment, measurements of both aerosol concentrations and properties are performed at a remote site (Ersa) on Corsica Island in the northwestern Mediterranean sea in the summers 2012, 2013, 2014 and the winter 2014, and above the western Mediterranean Sea in the summer 2014. This work aims at understanding the origins and processes leading to the formation of organic and inorganic aerosols, using the air-quality model Polyphemus. The contribution of marine emissions to inorganic aerosols (IA) is large, and a parameterization with an adequate wind-speed power law is chosen. Marine organic aerosols (OA), which are added to the model with a parameterization that uses the chlorophyll-a concentration as a proxy parameter to model the marine chemistry, contribute to OA by only 2% at the maximum. The ground-based and airborne model-to-measurements comparisons show the importance of an accurate description of shipping emissions to model sulfate and OA concentrations. However, this is not true for nitrate, ammonium and chloride concentrations, which are very dependent on the hypotheses used in the model for condensation/evaporation. During the summers 2012 and 2013, measurements show important concentrations of highly oxidized and oxygenated OA. For the model to reproduce not only the concentrations but also the oxidation and hydrophilicity properties of OA, three processes are added to the model: the autoxidation process leading to the formation of extremely low volatility organic compounds, the organic nitrate formation mechanism and the second generational ageing of monoterpenes. In agreement with the measurements, the model also shows the preponderant biogenic origin of OA during summer. OA concentrations are not very sensitive to the anthropogenic intermediate/semi-volatile organic compound (ISVOC) emissions missing from the inventories during summer. Nonetheless, the role and the contribution of ISVOC are more significant during the winter, with a large contribution from residential heating. Different parameterizations to represent the emissions and the ageing of ISVOC are implemented in the model, namely the volatility distribution of emissions, single-step vs multi-step oxidation scheme and non-traditional volatile organic compounds (NTVOC) chemistry. Sensitivity studies show that the volatility distribution at the emission is a key parameter to improve the modelling of OA concentrations. The model reproduces well the observed concentrations, but the observed organic oxidation and oxygenation states are strongly under-estimated, stressing the potential role of organic nitrate from anthropogenic precursors and highly oxygenated organic molecules.