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Advanced modelling of the multiphase chemistry of methylamines with CAPRAM

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Amines are important but poorly studied organic constituents in the marine atmosphere. There is strong evidence that within the marine boundary layer, the formation of new aerosol particles and the increase in particle mass is influenced by amines. However, very high uncertainties still exist with respect to: the sources, the further chemical reactions within the multiphase chemical system of the marine atmosphere, and the contribution to the marine aerosol mass. A deeper understanding of the amine-initialized formation of organic nitrogen in marine aerosol particles requires fundamental mechanistic modeling studies of the multiphase oxidation of amines.

Therefore, a detailed multiphase chemistry mechanism, the CAPRAM Amine module, has been developed to describe the oxidation of ammonia (NH_3), monomethylamine (MMA), dimethylamine (DMA) and trimethylamine (TMA) in the atmosphere. This mechanism is the first that considers the formation of the auto-oxidation products from DMA and TMA and their further oxidative fate in both the gas and aqueous-phase. Overall, the mechanism currently contains 541 reactions, thereof 232 gas-phase reactions 52 phase transfer reactions, and 257 aqueous-phase reactions.

In the present study, first simulations of an idealized marine environment were performed investigating the chemical processing of amines under cloud and non-cloud conditions. The simulations indicate that uptake is a main loss term for DMA whereas for TMA it is oxidation in the gas phase. This shows that models considering amine driven new particle formation have to consider both uptake and gas-phase oxidation. Interestingly, the chemical rates analyses revealed an unexpected important chemical conversion of amines during cloud conditions, where TMA, DMA and MMA are degraded into DMA, MMA and NH_3 , respectively. This unexpected fate of amines can have implications for the current unknown high DMA concentrations in the marine boundary layer that cannot be explained by oceanic emission alone. Because of the importance of DMA and TMA for new particle formation, the uncovered processes have to be thus analyzed by laboratory studies in more detail.