Experimental and model investigations of the multiphase chemical processing of glyoxal at the CESAM cloud chamber

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Chemical processes in clouds have been suggested to contribute substantially to organic aerosol particle mass (OM), however, considerable uncertainties still exist, e.g. with regards to the nature of the resulting aerosol particles. Therefore, the project PARAMOUNT aimed at investigations of chemical cloud processing of organic constituents such as glyoxal in the CESAM chamber. To study the multiphase processing, a large set of online instruments was used, e.g. Aerosol Mass Spectrometers (AMS), PTR-MS, CHARON-PTR-MS, and FT-IR. The chamber results demonstrated the feasibility of such adiabatic experiments for the first time and, even if the cloud lifetime remains short, an aqueous-phase processing can be studied. Under the deliquesced aerosol conditions, the OM formation was observed by online mass spectrometers including various higher molecular weight compounds. The glyoxal-related experiments showed that the processing is not only restricted to the in-cloud period but can interestingly occur after the droplet evaporation. There, e.g., a rapid formation of formic acid was observed. Finally, the performed CESAM experiments were modeled with a multiphase chemistry model using MCM/CAPRAM investigating chemical process under both deliquesced aerosol and cloud conditions. The applied model was able to reproduce the microphysical cloud formation and evaporation cycle for adiabatically formed clouds. The measured AMS mass was reproduced by the model, however, sensitivity studies also demonstrated that the modeled OM concentration is very sensitive to the applied kinetic forward and backward rate coefficients of the glyoxal hydration process. Excitingly, the simulations reveal that the water evaporation proceeds on a faster timescale than the adjunct repartitioning of dissolved glyoxal back to the gas phase. As a consequence, the kinetically delayed evaporation of glyoxal leads to higher aqueous-phase concentrations after the cloud evaporation fostering the formation of dimers and formic acid during the post-evaporation period. So, the formation and subsequent release of the less volatile organic compound, formic acid, by a non-radical oxidation process in evaporating droplets/particles was modeled. So, the chamber observations were successfully supported by the accompanied CESAM modeling. Overall, combined CESAM-experiment-model studies emphasized that cloud and cloud evaporation periods both provide conditions for chemical cycling of aqSOA.

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