Molecular characterization of organic compounds in aerosol particles and in cloud water – a high-resolution mass spectrometry study at the Schmücke mountain site

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Cloud droplets play a pivotal role in atmospheric multiphase systems, providing a ubiquitous medium for formation and chemical transformations of organic aerosol (OA) constituents in the aqueous phase. Despite the large relevance of clouds for hydrological and climatic systems, knowledge on the chemical composition of dissolved organic compounds in cloud water and in-cloud chemistry of OA is still limited. Moreover, possible effects of surface-active organic compounds on cloud formation lack a complete understanding and are associated to large uncertainties.

Here, we investigated molecular connections between the chemical composition of OA particles and cloud water at a Central European mountain site (Schmücke, Germany). During summer 2016, aerosol particles and cloud water were collected at the mountain's summit during cloud-free and cloudy periods, respectively. The samples were analyzed by high-resolution Orbitrap mass spectrometry in combination with liquid chromatography (LC-HRMS). In contrast to previous studies, the application of LC allowed us to separate isomers, and furthermore, to reduce possible artifacts from matrix components and ion suppression – a severe drawback that is commonly observed for direct-infusion HRMS analysis. Subsequently, we identified significant signals and determined unambiguous molecular formulas of organic compounds present in OA and cloud water, using a customized non-target analysis approach. Remarkably, from the large number of detected compounds in aerosol particle and in cloud water samples only a rather small percentage was found in both sample types, indicating aqueous-phase chemistry and gas-phase uptake to determine largely the composition of organic matter in cloud water droplets. Our results expand current knowledge on understanding of aqueous secondary OA formation and provide unique insights into the factors influencing the chemical composition of dissolved organic matter in cloud water.