Molecular Characteristics and Aqueous-Phase Formation Processes of Organics in Cloud Water Revealed by Fourier Transform Ion Cyclotron Resonance Mass Spectrometry

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Abstract

Cloud water serves as a medium for the aqueous-phase reactions of dissolved gases and aerosols, thereby contributing to the formation of secondary organic aerosols. However, little is known regarding the molecular-level composition and aqueous-phase reactions of organics in cloud water. In this study, cloud water and aerosol samples were collected at Mt. Tianjing in southern China in 2018 and 2020, and the concentrations of water-soluble ions and organic carbon (WSOC) were determined. The organic molecular composition was characterized by electrospray ionization in negative mode coupled with Fourier transform ion cyclotron resonance mass spectrometry. The strong correlation observed between water-soluble organic compounds (WSOM) and aqueous secondary matter (e.g., NO3- and oxalate), the positive matrix factorization results, and the elevated WSOM/K+ ratios in cloud water compared with aerosols suggest the formation of in-cloud aqueous-phase secondary organic matter (aqSOM). WSOM in cloud water are dominated by CHON (formulas containing carbon, hydrogen, oxygen, and nitrogen elements), with a fraction exceeding 40% in relative abundance. This fraction is higher than that of aerosols, indicating enhanced formation of nitrogen-containing compounds in cloud water. Thirty-seven characteristic molecules of in-cloud aqSOM were identified using linear discriminant analysis effect size (LefSe), mainly including dinitrophenols, dicarboxylic acids, and nitrophenols. Some of these compounds exhibit low or extremely low volatility and may contribute to secondary organic aerosol formation after droplet evaporation. The molecular composition of water-insoluble organic compounds (WIOM) was also detected. WIOM are characterized by lower oxidation states of carbon atoms, higher carbon number, and lower unsaturation relative to WSOM. Intermolecular relationship analysis showed that WIOM has the potential to transform into WSOM through oxidation reactions. These results reveal the modifying effects of in-cloud processes on aerosol organic composition at the molecular level and could improve our understanding of aerosol–cloud interactions.

Early Career Scientist

YES, I am an early career scientist.

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