

Formation of Brown Carbon by Carbonyl and Reduced Nitrogen Species: Reaction Mechanisms, Production Distribution and Light-Absorbing Capacity

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Abstract

Reactions between carbonyl and reduced nitrogen species (RNS) serve as key sources of secondary brown carbon (BrC), which significantly impacts global radiative balance through absorption in the near-UV and visible regions of solar spectrum. However, their chemically intricate formation pathways hinder a comprehensive understanding of their molecular composition, posing challenges for evaluating their optical properties. In this study, we employed high-resolution mass spectrometry (HRMS) techniques, including HPLC-PDA-MS and FT-ICR-MS to investigate the profiles of product distribution in the dark reactions of carbonyls (i.e., glyoxal (G) and methylglyoxal (MG)) with RNS (i.e., ammonium salts and amino acids). Amine precursors generate more diverse and abundant products with higher light-absorbing capacities compared to ammonium. Carbonyl precursors result in the diversity of molecular distribution. Specifically, when using MG as a carbonyl precursor, BrC formation reactions generated more diverse, conjugated, and less nitrogenous products, compared to using G. Products from ammonia + MG reactions exhibited enhanced aromaticity compared to those from ammonia + G reactions, contrasting with similar aromaticity observed when using organic amine precursors. In addition, in ammonia reactions, the use of ammonium sulfate as a precursor resulted in the production of a wider range of light-absorbing products compared to ammonium chloride and ammonium nitrate. This highlights the significance of considering the impact of anions when modeling the global radiative effects of BrC. Additionally, formic acid was formed in both G and MG-involved reactions, whereas acetic acid is only observed in MG system, which serves as a significant precursor of secondary organic aerosols, and potentially contributes to atmospheric acidity.

Early Career Scientist

YES, I am an early career scientist.

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