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Insight into the aqueous formation of particulate oxalate

Xinhui Bi (1), Guohua Zhang (1), Qinhao Lin (1), and Duohong Chen (2)

(1) Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China (bixh@gig.ac.cn), (2) State Environmental Protection Key Laboratory of Regional Air Quality Monitoring, Guangdong Environmental Monitoring Center, Guangzhou 510308, PR China

Oxalic acid is globally the most abundant dicarboxylic acid. While ground-based works suggest the significance of in-cloud production (or aqueous formation) to oxalate, direct evidence is rare. With the in situ measurements performed at a remote mountain site (1690 m a.s.l.) in southern China, we first reported the size-resolved mixing state of oxalate in the cloud droplet residual (cloud RES), the cloud interstitial (cloud INT), and ambient (cloud-free) particles by single particle mass spectrometry. The results support the growing evidence that in-cloud aqueous reactions promote the formation of oxalate. Furthermore, oxalate was predominantly internally mixed with the aged biomass burning particles, highlighting the impact of biomass burning on the formation of oxalate. It can be interpreted by the individual particle mixing state that the aged biomass burning particles contained an abundance of organic components serving as precursors for oxalate. The results further show that in-cloud aqueous reaction dramatically improved the conversion of organic acids to oxalate. The abundance of glyoxylate associated with the aged biomass burning particles is the controlling factor for the in-cloud production of oxalate. Since only limited information on oxalate is available in the free troposphere, the results also provide an important reference for future understanding of the abundance, evolution and climate impacts of oxalate. We also provided the first direct field observational evidence for the enhanced aqueous formation of oxalate associated with Fe-containing particles. It is likely that reactive oxidant species (ROS) via Fenton reactions enhanced the formation of these organic compounds and their oxidation product oxalate. Gas-particle partitioning of oxalic acid followed by coordination with Fe might also partly contribute to the enhanced oxalate. Aerosol water content likely played an important role in the enhanced oxalate formation when the relative humidity is > 60%. The study could provide a reference for model simulation to improve understanding on the formation and fate of oxalate, and the evolution and climate impacts of particulate Fe.