2.051 Insight into the in-cloud formation of oxalate based on in situ measurement by single particle mass spectrometry.

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Abstract:

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, with ~15% of the cloud RES and cloud INT particles containing oxalate, in contrast to only ~5% of the cloud-free particles. Furthermore, individual particle analysis provides unique insight into the formation and evolution of oxalate during in-cloud processing. Oxalate was predominantly (>70% in number) internally mixed with the aged biomass burning particles, highlighting the impact of biomass burning on the formation of oxalate. In contrast, oxalate was underrepresented in aged elemental carbon particles, although they represented the largest fraction of the detected particles. 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. Through the analysis of the relationship between oxalate and organic acids (-45[HCO_2]$^-$, -59[CH_3CO_2]$^-$, -71[$C_2H_3CO_2$]$^-$, -73[C_2HO_3]$^-$), the results 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.