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Cloud chemistry in eastern China: Observations from Mt. Tai

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Until recently, studies of fog and cloud chemistry in China have been rare - even though the fate of China's large sulfur dioxide emissions depends, in part, on the ability of regional clouds to support rapid aqueous oxidation to sulfate. Sulfur dioxide oxidized in regional clouds is more likely to be removed by wet deposition while sulfur dioxide that undergoes slower gas phase oxidation is expected to survive longer in the atmosphere and be transported over a much broader spatial scale. Two 2008 field campaigns conducted at Mt. Tai, an isolated peak on the NE China plain, provide insight into the chemical composition of regional clouds and the importance of various aqueous phase sulfur oxidation pathways. Single and two-stage Caltech Active Strand Cloudwater Collectors were used to collect bulk and drop size-resolved samples of cloudwater. Collected cloudwater was analyzed for key species that influence in-cloud sulfate production, including pH, S(IV), H2O2, Fe and Mn. Other major cloud solutes, including inorganic ions, total organic carbon (TOC), formaldehyde, and organic acids were also analyzed, as were gas phase concentrations of SO2, O3, and H2O2.

A wide range of cloud pH was observed, from below 3 to above 6. High concentrations of cloudwater sulfate were consistent with abundant sulfur dioxide emissions in the region. Sampled clouds were also found to contain high concentrations of ammonium, nitrate, and organic carbon. Peak TOC concentrations reached approximately 200 ppmC, among the highest concentrations ever measured in cloudwater. Hydrogen peroxide was found to be the dominant aqueous phase S(IV) oxidant when cloud pH was less than approximately 5.4. Despite its fast reaction with sulfur dioxide in cloud droplets, high concentrations of residual hydrogen peroxide were measured in some clouds implying a substantial additional capacity for sulfate production. Ozone was found to be an important S(IV) oxidant when cloud pH was high. Oxidation of S(IV) by oxygen, catalyzed by Fe (III) and Mn(II) was generally the second or third fastest pathway for sulfate production. Differences between the pH and trace metal concentrations of small and large cloud droplets were observed, giving rise to aqueous phase sulfate production rates that were drop size-dependent for the ozone and metal-catalyzed pathways.