Title:	In-cloud sulfate addition to single particles resolved with sulfur isotope analysis during HCCT-2010
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Abstract:	In-cloud production of sulfate modifies aerosol size distribution, with important implications for the magnitude of indirect and direct aerosol cooling and the impact of SO2 emissions on the environment. We investigate which sulfate sources dominate the in-cloud addition of sulfate to different particle classes as an air parcel passes through an orographic cloud. Sulfate aerosol, SO2 and H2SO4 were collected upwind, in-cloud and downwind of an orographic cloud for three cloud measurement events during the Hill Cap Cloud Thuringia campaign in autumn 2010 (HCCT-2010). Combined SEM and Nano SIMS analysis of single particles allowed the δ34S of particulate sulfate to be resolved for particle size and type. The most important in-cloud SO2 oxidation pathway at HCCT-2010 was aqueous oxidation catalysed by transition metal ions (TMI catalysis), which was shown with single particle isotope analyses to occur primarily in cloud droplets nucleated on coarse mineral dust. In contrast, direct uptake of H 2SO4 (g) and ultrafine particulate were the most important sources modifying fine mineral dust, increasing its hygroscopicity and facilitating activation. Sulfate addition to "mixed" particles (secondary organic and inorganic aerosol) and coated soot was dominated by in-cloud aqueous SO2 oxidation by H2O2 and direct uptake of H2SO4 (g) and ultrafine particle sulfate, depending on particle size mode and time of day. These results provide new insight into in-cloud sulfate production mechanisms, and show the importance of single particle measurements and models to accurately assess the environmental effects of cloud processing.
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