Evidence of high acidity in individual aerosol particles in the marine atmosphere

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Aqueous aerosol thermodynamic modelling suggests that acids formed from anthropogenic and natural sources can result in high acidity in aerosols in the marine atmosphere. However, actual measurements of acidity or pH of aerosols is high challenging, particularly in individual aerosol particles. For this reason, observational evidence is lacking to confirm the model calculated high acidity in aerosols.

In this study, we carried out microscopic and nanoscale secondary ion spectroscopic analysis to show evidence of high acidity in individual atmospheric aerosol particles. We collected single particles from a research cruise over the East China Sea. Chemical analysis of hundreds of individual atmospheric particles show that iron-rich particles from coal combustion and steel industries were coated with thick layers of sulfate after 1-2 days of atmospheric residence. We detected highly acidic ammonium bisulfate in individual aerosol particles by selected area electron diffraction analysis. Furthermore, the iron in aged particles was present as “hotspots” of (insoluble) iron oxides and throughout the acidic sulfate coating in the form of (soluble) iron-sulfate, which increases with degree of ageing (thickness of coating). Since iron sulfate was not detected in the freshly emitted particles, and, there is no other source or mechanism of iron sulfate formation in the atmosphere, we conclude that insoluble iron in the primary particles were dissolved by acids. These results provide observational evidence of highly acidic individual aerosol particles in the marine atmosphere.