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Sulphuric acid and neutral cluster measerements with CI-APi-TOF

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Sulphuric acid is a key compound in atmospheric nucleation (Sipilä et al., 2010) and in the atmosphere it is mainly produced photochemically via reaction of OH with SO_2. The concentration of sulphuric acid is typically very low and rarely exceeds 108 molecules cm-3. The low concentrations set requirements for the detector used for quantitative measurements. A sensitive method measuring gas phase sulphuric acid was developed by Eisele and Tanner (1993). They used chemical ionization with nitrate ion, NO_3-, and its clusters with HNO_3, to selectively ionize sulphuric acid and detected it with a quadrupole mass spectrometer.

We present a technique in which the atmospheric pressure chemical ionization (CI) inlet, with geometry similar to Eisele and Tanner (1993), is coupled with a high resolution atmospheric pressure interface time-of-flight mass spectrometer (APi-TOF, Tofwerk Ag., Junninen et al., 2010). In the present setup the high resolution and mass range of the TOF allows separation and summing of different clusters. The advantage of this method is that it allows one to find neutral sulphuric acid containing clusters formed by nucleation in the atmosphere (Kuang et al., 2008) or in laboratory systems (Sipilä et al., 2010).

The first ambient measurements using the CI-APi-TOF for sulphuric acid and neutral cluster detection are also presented. We found the CI-APi-TOF a highly stable and sensitive tool for molecular sulphuric acid detection. The limit of detection for sulphuric acid was 3•104 molecules cm-3 for two hour averaging. Signals from sulphuric acid clusters up to the tetramer were obtained but were found to result from naturally charged clusters formed by ion induced clustering in the atmosphere. Opposite to earlier studies with cluster mass spectrometers, we had no indication of neutral clusters. The reason is either less efficient charging of clusters in comparison to molecular sulphuric acid, or in low concentration of neutral clusters at our measurement site during these particular nucleation events. We showed that utilizing high resolution mass spectrometry is crucial in separating the weak sulphuric acid cluster signal from the other compounds.

The further aim is to use CI-APi-TOF with an ion filter to remove natural ion interference and to be able to analyze neutral sulphuric acid and its clusters exclusively.

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