Intercomparison of sulphuric acid measurements and neutral cluster composition in the lower free troposphere

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Aerosol nucleation from trace vapours is a ubiquitous process in the Earth's atmosphere and has been observed at many different places (Kulmala et al., 2004). It is thought that sulphuric acid is a key species in the formation process of small clusters and particles. A correlation between the particle formation rate and the concentration of sulphuric has been frequently observed during ground-level field campaigns. However, data coverage on the sulphuric acid concentrations at higher altitudes is relatively scarce, although, these regions are very important on a global scale. In addition, it is not clear if and what other compounds (besides water) are participating in the formation of aerosol particles in these regions.

During January and February 2013 the **Nu**cleation, **Cl**oud and **A**erosol **C**haracterization **E**xperiment (NUCLACE) took place at the high alpine research station Jungfraujoch (JFJ) in Switzerland. The JJF station is located at 3580 meters altitude and mainly influenced by free tropospheric air during this time of the year. One of the goals of the NUCLACE campaign was to investigate the role of ions on the formation of new particles. However, in this study we focus on the measurement of the sulfuric acid concentration as well as on the detection of neutral clusters and their composition with state-of-the-art instruments.

For this purpose, two Chemical Ionization Atmospheric Pressure interface Time-Of-Flight Mass Spectrometers (CI-APi-TOFs) were used (Jokinen et al., 2012). Within the inlets of both instruments nitrate primary ions are generated which can ionize neutral sulphuric acid and clusters through a proton transfer reaction or by clustering. The primary and the product ions are detected with a high resolution time of flight mass spectrometer. The combination of these two techniques allows low detection limits (at ppq levels) and the identification of ion species through their exact mass. Although both CI-APi-TOF instruments have the same principle they differ in the design of their ion source. The CI-APi-TOF from the University of Frankfurt utilizes a corona discharge needle (Kürten et al., 2011) while the CI-APi-TOF from the University of Helsinki (Jokinen et al., 2012) utilizes a soft x-ray ion source. In addition, the geometry of the ion drift regions and the inlet geometries differ slightly. Additionally, the operation of the CI-APi-TOFs at JFJ turned out to be challenging due to the low pressure and strong temperature variations inside the building where the instruments were located. Therefore, an intercomparison of the two instruments is important (see figure 1). Furthermore, it turned out that an on-site calibration needed to be performed to take into account the conditions at the measurement site. For this, a calibration unit similar to the one described by Kürten et al. (2012) has been brought to JFJ. With this set-up a known concentration of H_2SO_4 can be generated and measured by one CI-APi-TOF at a time.

We will report on the overall comparison of the two CI-APi-TOFs with respect to the sulfuric acid monomer concentrations. In addition, the instruments will be compared regarding the detection of different neutral clusters. Details about the calibration will also be presented.

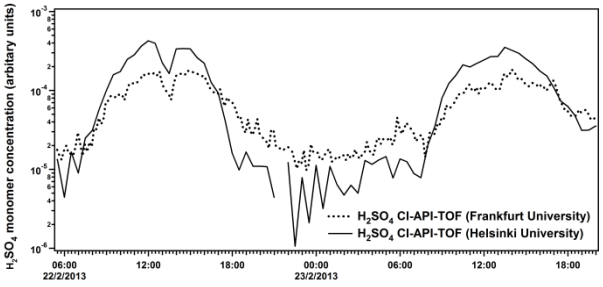

Figure 1. Observed diurnal variation of the sulphuric acid concentration measured by the two CI-APi-TOFs from Frankfurt and Helsinki.

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