

Name of research institute or organization:

Institute for Atmospheric and Environmental Sciences
Goethe University Frankfurt am Main

Title of project:

Study of new particle formation in the free troposphere (NUCLACE-2013)

Project leader and team:

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Project description:

The aim of this project is to study the formation of new particles (nucleation) and the underlying mechanisms in the free troposphere. It is well known that aerosol particles affect the climate both through direct absorption and scattering of light as well as indirectly through their ability to act as cloud condensation nuclei (CCN). A recent study estimates that about 50% of the CCN are formed by a gas to particle conversion mechanism. However, information about the relevant processes in the free troposphere is scarce although sulfuric acid is considered to be the main precursor gas for new particle formation. Therefore, it is an important question which other vapors are involved in the very first steps of nucleation when molecules collide and form small clusters. The identification of the elemental composition of the clusters during a nucleation event is of utmost importance if one wants to understand new particle formation.

Thus, a Chemical Ionization-Atmospheric Pressure interface Time Of Flight (CI-APi-TOF) mass spectrometer was deployed to measure the elemental composition of neutral (uncharged) clusters with a maximum mass of up to about 2000 atomic mass units. Due to the high mass resolving power and the high mass accuracy of the time of flight instrument, it is in principle possible to determine the elemental composition of the clusters. CI-APi-TOF utilizes a charging unit to charge the neutral clusters; therefore, quantitative measurement of cluster concentrations is feasible if the charging and detection efficiencies are known.

Important information that can be derived from the CI-APi-TOF data is the sulfuric acid monomer concentration ($[H_2SO_4]$). Figure 1 shows an example of the diurnal cycle of $[H_2SO_4]$ during the measurement period (January and February 2013). Sulfuric acid is photochemically produced; therefore, a clear diurnal cycle can be observed under favorable conditions (generally when there is low cloud cover). Although, CI-APi-TOF can in principle measure the sulfuric acid concentration with high accuracy (typically the error is within $\pm 50\%$), the conditions at JFJ are substantially different than under normal operating conditions at ground level. The low temperature and the low pressure at JFJ very likely influence the detection efficiency of sulfuric acid and lead to a much larger uncertainty in the derived concentrations. For this reason only relative changes in the concentration are shown in Figure 1. Further data evaluation and comparison to the results obtained from a second CI-APi-TOF (operated by the University of Helsinki, Finland) are required. This is also the case regarding the measurement of neutral clusters. Elevated signals could be observed at higher masses (beyond the charged sulfuric acid monomer HSO_4^- , which is observed at m/z 97) but it is not clear yet if these are significantly above their background values and if so, to which neutral species they belong. Despite the challenges encountered at JFJ, promising results were obtained during this first deployment of a CI-APi-TOF at a high-altitude measurement station and further studies will be conducted in the near future.

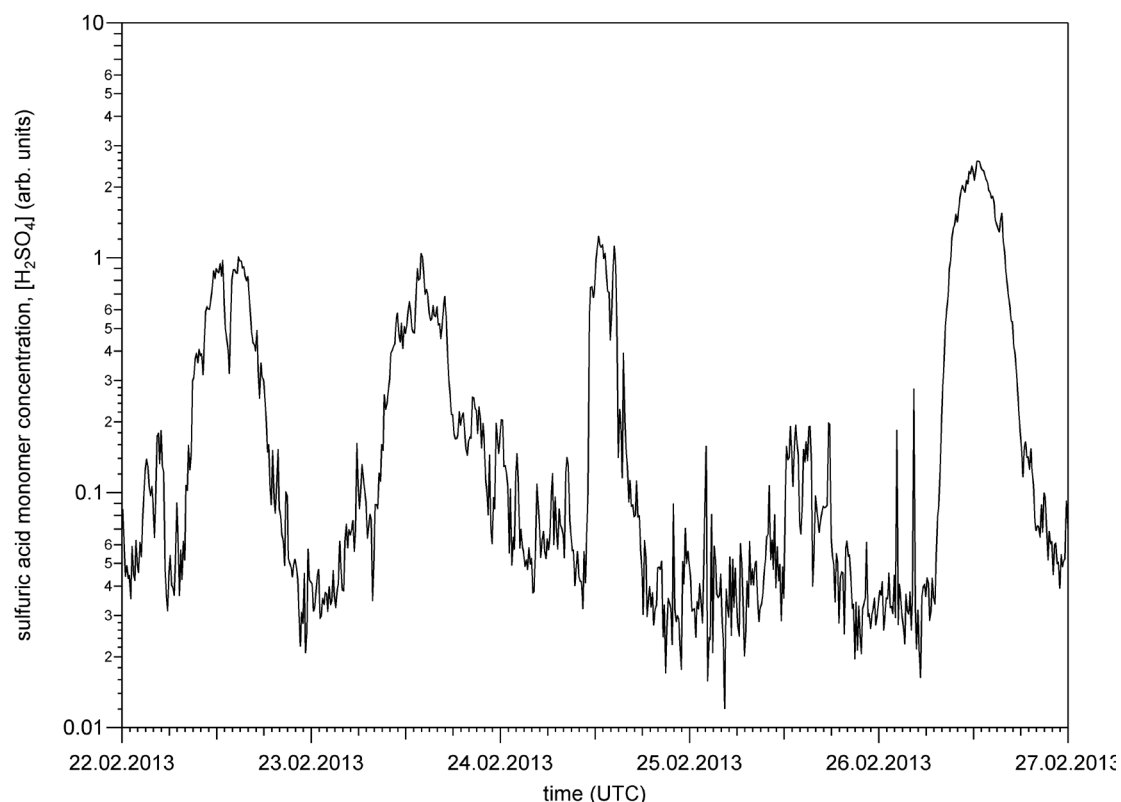


Figure 1. Observed diurnal variation of the sulfuric acid concentration measured by the CI-API-TOF from Frankfurt.

Key words:

Nucleation, Chemical Ionization-Atmospheric Pressure interface-Time Of Flight (CI-API-TOF) mass spectrometer, sulfuric acid, clusters

Collaborating partners/networks:

Paul Scherrer Institute, University of Helsinki

Scientific publications and public outreach 2013:

Conference papers

Bianchi, F., H. Junninen, J. Troestl, J. Duplissy, L. Rondo, M. Simon, A. Kuerten, A. Adamov, J. Curtius, J. Dommen, E. Weingartner, D. Worsnop, M. Kulmala, and U. Baltensperger, Particle nucleation events at the high alpine station Jungfraujoch, 19th International Conference on Nucleation and Atmospheric Aerosols, Fort Collins, Colorado, USA, June 23-28, 2013.

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