Evaluation and development of Scanning Flow CCN Analysis

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Atmospheric aerosols influence the climate and hydrological cycle of Earth by acting as cloud condensation nuclei (CCN). The ability of aerosol particles, to act as CCN, depends on the particle size and chemical composition as well as the ambient water vapour supersaturation (SS). Köhler theory (Köhler 1936) is commonly used to predict and describe the ability of the particles to activate into cloud droplets. However, the activation of complex mixtures of particles in ambient air is not fully explained nor understood by theory. There is a demand for better temporal and supersaturation resolution for ambient measurements as well as in laboratory experiments. A more rapid and continuous technique would enable measurements of e.g. small samples, fast processes, or a better understanding of a rapidly ageing aerosol. For regional and global measurement networks a standardization of measurement procedure and techniques at different sampling sites are of great importance.

Here, the Continuous-Flow Streamwise Thermal-Gradient Cloud Condensation Nuclei Counter (CCNC, DMT-100), a commercial instrument, has been evaluated when running the instrument in an altered mode of operation. By scanning the flow in the instrument, while maintaining a constant pressure and temperature gradient through the instrument column, rapid and continuous measurements are made possible. Moore and Nenes (2009) introduced the Scanning Flow CCN Analysis (SFCA), with a detailed description of the measurement procedure accompanied by results from both laboratory and ambient measurements. They also present a comparison between the former, "conventional" stepwise-ΔT way of operation with the new mode with scanning flow. However, there are still uncertainties of how to operate the CCNC in SFCA mode for long-term measurements. This evaluation of the SFCA is part of the EU FP7 ACTRIS Infrastructure project (www.actris.net) and is the first step towards a standardization of CCN measurements with high temporal and supersaturation resolution within the European network.

To induce a supersaturation change in the "conventional" operation mode, the flow is kept constant, while the temperature gradient is varied in a stepwise way. Due to the slow temperature stabilization, measurement of whole supersaturation spectra is time consuming. By adding a small software change to the robust and well-established hardware of the CCNC, the flow is instead varied through the column in a controlled manner, while the streamwise temperature gradient (ΔT) and pressure is maintained constant. The flow rate in the chamber is varying in a scan cycle, where the flow increases/decreases linearly, as well as a shortly kept constant at maximum/minimum flow rates. When calibrating the instrument the flow and the corresponding activated fraction of the particles generate a supersaturation curve with a "critical flow rate", Q_{50}. From knowledge of the particle dry diameter and chemical composition Q_{50} is translated to a critical supersaturation (SS_c) using Köhler theory. Hence, every instantaneous flow rate corresponds to a critical supersaturation. However, the calibration curves are specific for the chosen ΔT, scan time and pressure.

SFCA enables measurements of many supersaturation spectra during a short time period, allowing a better temporal resolution (Fig.1) and also smaller samples are needed. The inlet temperature can be kept closer to ambient conditions and therefore minimizing biases from volatilization of semi-volatile compounds in the instrument.

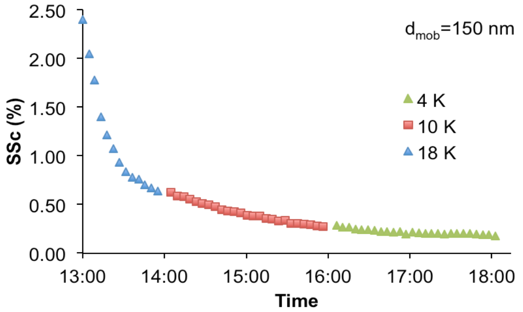

Figure 1. Results of measurements using SFCA for the ageing process of diesel soot agglomerates (mobility diameter 150 nm). SFCA entails high temporal resolution of the CCN spectra. The shift of ΔT makes it possible to cover the whole aging process.

To fully cover the supersaturation spectra of ambient air, a three-step change of the temperature gradient (ΔT = 4, 10, and 18 K) has been introduced (Fig.1). For better resolution and more reliable data of the activation step we recommend an optimal scan cycle of 5 min. To keep the aerosol sample flow rate at constant, a second flow has been introduced, mirroring the flow in the CCNC.

Laboratory experiments of calibration substances such as ammonium sulphate and sucrose, with good repeatability, has been performed. SFCA has also been used for measurements of soot agglomerates during ageing processes (Fig.1) and for other organic mixtures.

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