Ab initio nanoparticle sizing during new particle formation in precisely controlled chamber experiments

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The contribution of new particle formation to the number of cloud condensation nuclei (CCN) crucially depends on the initial growth of freshly nucleated particles. Only if the newly formed particles grow sufficiently fast will a substantial fraction of the particles survive to become CCN (Weber *et al.*, 1997). Huge efforts have been put into the quantification of early nanoparticle growth in the past, however, fundamental limitations have prevented monitoring of the size evolution at reasonable time resolution or counting statistics (Winkler *et al.*, 2012).

In this study we present the first results from a newly designed differential mobility analyser (DMA) train, which was used to track particle formation events in the CLOUD chamber. The CLOUD experiment at the European Centre for Nuclear Research (CERN) in Geneva, Switzerland, is used to study new particle formation under atmospheric conditions (Kirkby *et al.*, 2011). The CLOUD chamber consists of a 26 m³ stainless steel tank in which conditions can be precisely controlled, and contamination levels are extremely low. A comprehensive set of instruments is attached to the chamber simultaneously monitoring the gas-phase composition, as well as the cluster and particle composition, concentration, and size evolution during new particle formation.

For the CLOUD10 campaign in fall 2015 we assembled an unprecedented array of instruments to measure neutral particle growth from 1.5 nm to 60 nm in the presence of highly oxidised biogenic vapours and sulphuric acid. We operated our DMA train during the CLOUD10 measurement campaign to monitor nanoparticle growth primarily in the critical size range below 5 nm. The DMA train consists of six DMAs (Grimm SDMA) in parallel for high time resolution measurements of nanoparticle growth starting from 1.5 nm. Each DMA channel is operated at a fixed voltage allowing precise measurement of the evolution of six individual particle sizes. For the detection of particles we use five butanol based condensation particle counters (CPC) (TSI3776) and one water based CPC (TSI3788). For two sub-2 nm channels two Airmodus A10 particle size magnifiers (PSM) (Vanhanen *et al*., 2011) are used.

The performance of the DMA train was evaluated by performing a comparison with two other instruments that monitored the particle size evolution in parallel. A PSM was operated in scanning mode to obtain size information between approximately 1 and 3 nm. In addition, a fast-scanning nanoSMPS (TSI 3938) was used for particle sizing up to 60 nm. The principle is illustrated in Figure 1 showing the instruments used for nanoparticle sizing. The PSM and nanoSMPS have been used regularly during the past CLOUD campaigns. While the PSM covers the very initial growth, the nanoSMPS starts monitoring particles from 5 nm upwards leaving a gap in the critical part of the size distribution. Intercomparison of the three instruments shows that the DMA train perfectly closes this gap. We will present the most detailed and precise particle growth rate measurements so far obtained by CLOUD, with particular emphasis on the critical 1.5-10 nm size range where particle losses to pre-existing aerosol particles are most important. We are therefore now at a stage where we can perform quantitative growth law analysis (Rao and McMurry, 1989) to analyse nanoparticle evolution from the very beginning.

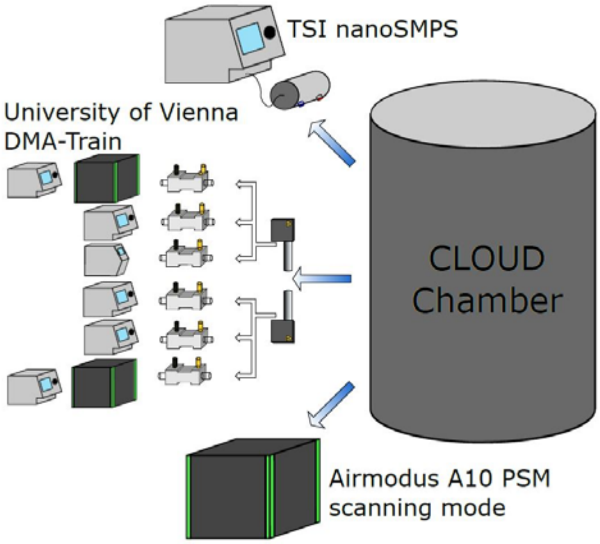

Figure 1. Set-up of nanoparticle sizing instruments at the CLOUD chamber.

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