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Changes in hygroscopicity and cloud-activation of diesel exhaust aerosols upon ageing

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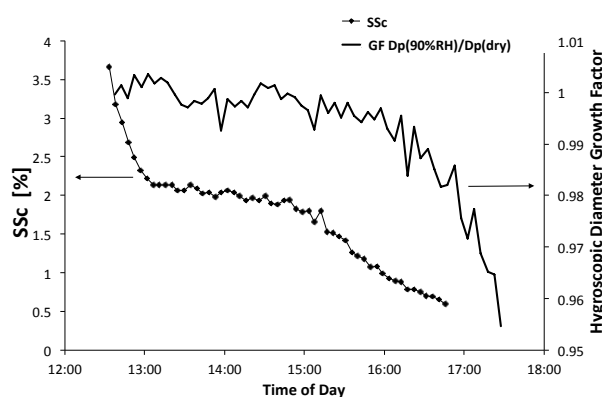
Aerosol particles play an important role in cloud formation processes. There is a direct effect from primary particles as well as an indirect effect from secondary aerosol particles formed in the atmosphere via complex gas-particle conversion processes. Secondary organic aerosols (SOA) can be formed when volatile organic compounds (VOCs) are present. Then part of the oxidation products from the VOCs condenses on pre-existing particles and produce SOA. This alters the particle properties, via condensation of the oxidised VOCs, which may lead to a lowered critical supersaturation (SS_c). Though, organic compounds will typically not suppress the water activity as much as inorganic salts. In general, the understanding of the properties of the SOA is scanty relative to the understanding of the major inorganics and is thereby of great interest for investigation.

This study is a continuation of a previous campaign, where exhaust from light duty vehicles were transferred to a smog chamber and photochemically aged (Nordin *et al.*, 2012). The aerosol properties were monitored using an extensive instrumental set-up, obtaining information on morphology, density, volatility, particulate and gas composition as well as hygroscopic growth and cloud droplet nucleation. The present study focus on experiments using soot (from a flame soot generator and diesel car exhaust) and different SOA precursors (Toluene and m-Xylene). During the ageing procedure, the transformation of the hygroscopic behaviour and its link to the effect on cloud droplet activation were related to the organic fraction in the particle as well as particle size and morphology.

Hygroscopic properties were analysed using a Hygroscopic Tandem Differential Mobility Analyser (H-TDMA; Nilsson *et al.*, 2009), the cloud-activation properties were measured using a Cloud Condensation Nucleus Counter (CCNC; DMT 100), and a soot particle aerosol mass spectrometer (SP-AMS, Aerodyne research) to determine the composition of the cores and the coatings of the particles. The particle mass-mobility relationship was characterized using a Differential Mobility Analyser-Aerosol Particle Mass Analyzer (DMA-APM; McMurry *et al.*, 2002; Kanomax Japan 3600). A thermodenuder was introduced in series between the DMA and the APM, used for quantification of the size dependent mass fraction condensed onto the non-volatile soot cores. The particles were first size selected by the DMAs, according to their mobility diameter. This was followed by measurements in the APM, the H-TDMA and CCNC. Furthermore, it was investigated whether the diameter growth factor (GF;

Fig.1), derived from H-TDMA data, could be used to predict SS_c for particles of a certain mobility diameter. During the latter campaign, the CCNC was operating using a modified measurement procedure, i.e. Scanning Flow CCN Analysis (SFCA) (Moore & Nenes, 2009), enabling rapid measurements with high time resolution of the supersaturation spectra, revealing more detailed, accurate and continuous results.

Fresh diesel soot particles show no hygroscopic growth and require a higher SS_c than predicted when using a simple model, based on the Köhler theory. The model only take the Kelvin effect into account, using the mobility diameter and assuming wettable particles (Rissler *et al.*, 2010). This is probably due to the agglomerated structure of the soot particles (e.g. Khalizov *et al.* 2009). At 90% relative humidity (RH), the soot particles collapse when coating exceeds 15-40% condensed organic material. Moreover, the cloud droplet activation is improved long before the particles show any hygroscopic growth at all, at 90% RH (Fig.1).



Figur 1. Preliminary SS_c decline for diesel soot particles ($d_{dry}=150$ nm) (data points), as well as the corresponding GF at 90% RH (solid black line) over time.

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