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Changes in Hygroscopicity and Cloud-Activation of Diesel Soot upon Ageing

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Introduction

The contribution from fresh diesel exhaust particles to the cloud condensation nuclei (CCN) population is negligible (e.g. Tritscher *et al.*, 2011). However, complex gas-to-particle conversion processes in the atmosphere form secondary organic aerosol (SOA) from emitted exhaust gases and particles, which significantly may influence the cloud drop formation process. Results from two Lund University smog chamber campaigns (#I & #II), are presented. The CCN and hygroscopic properties of diesel soot particles and the accompanying organic coating were investigated during ageing.

Methods

Exhaust from a light-duty diesel vehicle at warm idling were transferred to a smog chamber and photochemically aged (Nordin *et al.*, 2012). VOCs and IVOCs (Intermediate Volatile Organic Compounds) in the diesel exhaust were used as SOA precursors. Selected amounts of toluene and m-xylene were added to allow investigations of the full particle transformation from agglomerates to spheres.

Hygroscopic properties were analysed using a Hygroscopic Tandem Differential Mobility Analyzer (H-TDMA; Nilsson *et al.*, 2009), and the cloud-activation properties were measured using a Cloud Condensation Nucleus Counter (CCNC; DMT 100). A soot particle aerosol mass spectrometer (SP-AMS, Aerodyne Research) determined the composition of the soot cores and the particle coatings. The particle mass-mobility relationship was characterized using a Differential Mobility Analyzer-Aerosol Particle Mass Analyzer (DMA-APM; Kanomax Japan 3600). During Campaign II, the CCNC measurement procedure was changed from the traditional Stepping-ΔT to Scanning Flow CCN Analysis (SFCA) (Moore & Nenes, 2009), enabling rapid measurements of the supersaturation spectra with high time resolution, revealing more detailed, accurate and continuous results.

Conclusions

During the ageing process, 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. The properties of the organic material strongly influence the activation of the coated soot particles, both regarding the change of chemical composition, and also the change of critical supersaturation (SS_c, Fig.1) upon UV exposure.

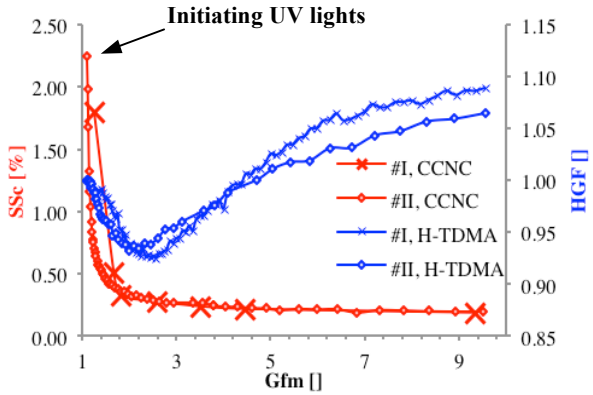

Figure 1.Ageing results in an increase in the mass growth factor (Gfm, x-axis), which is the ratio of total particle mass divided by the soot core mass. During ageing, the critical supersaturation (SS_c) decreases (red, left y-axis) and the corresponding hygroscopic growth factor (HGF, at 90% RH) increases (blue, right y-axis). Results are for diesel soot particles ($d_{m,dry}$=150 nm) from two experiments, Campaign #1 and #2.

Fresh diesel soot particles show no hygroscopic growth (HGF, Fig.1). Due to the morphology, the mobility diameter is not a relevant size measure for predicting cloud activation of fresh soot particles and is thereby not an accurate input parameter when calculating neither HGF (Fig.1) nor when modelling the SS_c. The cloud droplet activation starts long before the particles show any hygroscopic growth at all at 90% relative humidity (RH) (Fig.1). The soot particles gradually collapse to spheres up to a coating of 50-60% condensed organic material (HGF≈0.93; Gfm≈2-2.5, Fig.1). These results indicate that diesel soot particles, when aged, do affect the cloud forming process in the atmosphere.

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