

Transformation of Black Carbon Aerosol in the Atmosphere – Observations from Smog Chamber Studies and Ambient Air

J.H. Pagels¹, A.C. Eriksson², J. Rissler¹, E.Z. Nordin¹, C. Wittbom², P.T. Nilsson¹, A. Kristensson², P. Roldin², B. Svenningsson² and E. Swietlicki²

¹Ergonomics and Aerosol Technology, Lund University, Lund, SE-22100, Sweden

²Division of Nuclear Physics, Lund University, Lund, SE-22100, Sweden

Keywords: soot transformation, BC, SP-AMS, DMA-APM, smog chamber

Presenting author email: Joakim.pagels@design.lth.se

Atmospheric processing of soot ultimately leads to transformation of the agglomerated soot structure to close to spherical particles with an embedded elemental carbon core. These processes affect health and climate relevant (e.g. optics and cloud formation) and properties but the dynamics and time scales of the processes are poorly known (Zhang et al. 2008). The vacuum aerodynamic size distribution measured with the Soot Particle Aerosol Mass Spectrometer (SP-AMS) is a powerful tool to separate fresh and aged soot. In this work we investigated the transformation of diesel soot in detail in a smog chamber and contrast these results with recent street side and rural background measurements.

The composition of the soot core, its coatings and vacuum aerodynamic size distribution (Dva) was analyzed with an SP-AMS (Aerodyne Research). The soot morphology and size dependent condensed mass fraction was investigated using a Differential Mobility Analyzer-Thermal Denuder-Aerosol Particle Mass Analyzer (DMA-TD-APM) combined with TEM.

Controlled experiments were first carried out in a 6 m³ Teflon (FEP) smog chamber using black lights to initiate OH-chemistry. Diesel exhaust from an idling Euro II light duty vehicle was injected using a heated inlet. Varying concentrations of the anthropogenic SOA precursors toluene and m-xylene were added to the smog to investigate different degrees of aging. Soot transformation time scales are calculated based on the secondary aerosol mass flux to the particle phase in the chamber. Atmospheric background measurements were carried out at the Vavihill supersite in south Sweden. Urban street-side measurements were carried out in downtown Copenhagen adjacent to a busy street (60 km south-west of the background site).

During condensation of SOA, the effective density increased from low values typical for fresh diesel agglomerates towards 1.4 g/cm³ and the dynamic shape factor decreased towards 1.0, which showed that the soot agglomerates had been transformed to essentially spherical particles. The results have been summarized in a parameterization of the dependence of particle morphology (dynamic shape factor and effective density) on the condensed mass fraction and particle size during aging in the smog chamber. Humidifying the particles from the smog chamber to 90% RH using a hygroscopic tandem DMA, showed that uptake of water vapor leads to progressed compaction of the soot core.

Vacuum aerodynamic sizes of soot particles detected with the SP-AMS increased from about 100 nm up to about 400 nm when the organic fraction increased

from about 10% to 95% with aging in the chamber. This was due to the dual effects of increased volume and more compact shape leading to increased Dva (figure 1).

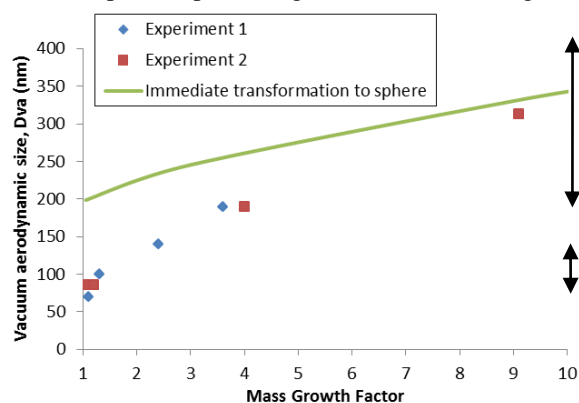


Figure 1. Mass median Dva vs mass growth factor measured in the smog chamber. To illustrate the large impact of particle morphology on the Dva, calculated results assuming immediate transformation to spheres are given for comparison. Arrows indicate the range of the two modes of soot containing particles at street side measurements.

In the street side measurements two main particle types were found, more massive particles with effective densities of about 1.4 g/cm³ nearly independent of particle size (interpreted as long range transport) and less massive particles with effective density decreasing with size. The SP-AMS showed one mode consistent with fresh agglomerated soot (Dva~100 nm) and another mode of larger soot containing particles.

At the background station at eastern wind directions, the more massive particles strongly dominated, while during south-western winds there was in addition a mode of less massive particles with effective densities equal to or higher than for fresh diesel soot, indicative of urban influence from the Copenhagen-Malmö metropolitan area (population: 2.6 Million).

We have shown that the combination of the SP-AMS and DMA-TD-APM is a powerful tool to investigate soot transformation in various environments. The vacuum aerodynamic size distribution measured with SP-AMS is a powerful tool in separating fresh and aged soot on a quantitative mass basis. However, more information is needed to investigate the transformation of soot from other sources such as biomass combustion.

This work was financed by the Swedish research council FORMAS, grant nbr. 2010-1678

Zhang, Khalizov, Pagels et al. (2008) PNAS 30, 10291