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Processing of Soot by Controlled Sulfuric Acid and Water Condensation – Mass and Mobility Relationship and Morphology

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Soot particles are ubiquitous in the atmosphere and are of interest in studies related to adverse effects on human health and climate forcing. An important property of soot is its structure - or morphology. The morphology and mass-mobility relationship of fresh soot, such as diesel exhaust has been studied previously (e.g. Park et al. 2003). However, processing of soot undergoing condensation, simulating ageing in the atmosphere, has received much less attention.

We investigated the influence on morphology and the mass-mobility relationship of airborne soot particles upon coating with sulphuric acid and water. The main components of the experimental set-up were: 1. A Santoro burner, using propane as fuel to generate soot; 2. A Differential Mobility Analyzer (DMA) for selecting particles with given mobility size; 3. An "aerosol conditioner," which simulates processing similar to that which may occur in the atmosphere; 4. Either a second DMA or an APM (Aerosol Particle Mass Analyzer) to measure the altered mobility size or mass, respectively. This information also enabled us to determine the effective density and fractal dimension of fresh and processed soot. In the aerosol conditioner sulfuric acid was condensed onto the soot particles as described by Zhang and Zhang (2005). A heater was used to evaporate condensed material. A few experiments involved high RH cycling (a humidifier to increase the RH to 90% and a drier to reduce the RH to 5% before measurement).

$$\rho_{eff} = \frac{6m}{d_B^3\pi} \quad (1), \quad m \propto d_B^{Df} \quad (2)$$

Typically the mass (m) increased and the mobility diameter (d_B) decreased as sulphuric acid was condensed onto the particles. The combined effects lead to an increased effective density (ρ_{eff} ; Eq. 1) and fractal dimension (Df ; Eq. 2). The influence on morphology depended strongly on the mass fraction of condensed material in the processed soot particles. Coating the particles with a low sulphuric acid mass fraction (~20%) resulted in a moderate influence on ρ_{eff} and Df . Results for a higher H_2SO_4 mass fraction (~55%) are given in Figure 1. The fractal dimension increased from 2.11 to 2.54 upon coating with sulphuric acid. An additional high RH

cycling further increased Df to 2.80. For sizes below 100 nm, effective densities approached the estimated bulk density and dynamic shape factors approached 1, indicating a transformation from highly agglomerated to compact, nearly spherical particles.

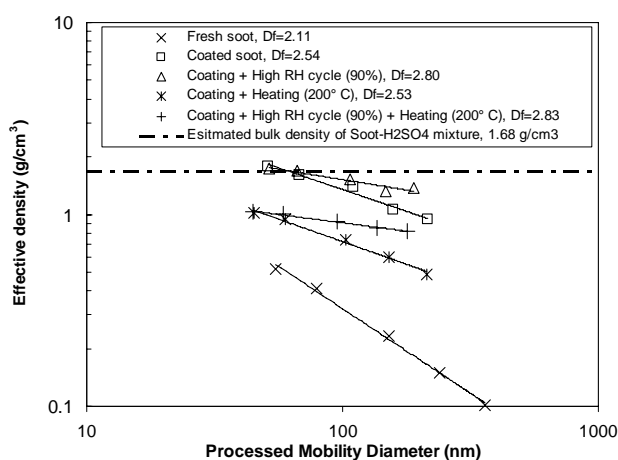


Figure 1. Effective density of fresh and processed soot.

Increased ρ_{eff} and Df were also observed when sulphuric acid was condensed on agglomerates and then removed by heating, indicating restructuring of the soot core. Soot with a hydrophilic coating, but not fresh hydrophobic soot, experienced restructuring upon RH cycling.

The atmospheric implication is that fresh (hydrophobic) soot will remain agglomerated when exposed to high RH cycles at subsaturation or upon low amounts of condensation. However as more mass condenses a gradual compaction takes place, which leads to full compaction at a mass ratio ($m_{processed}/m_{fresh}$) of 2-3. If the condensate is hygroscopic then a high RH cycle even at RH below 100% can significantly progress the compaction.

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