Mixing state of airborne particles in central Copenhagen and rural background – Particles effective densities for lung dose estimations

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Aerosol instruments based on the Differential Mobility Analyzer (DMA) are widely used characterising ambient submicron particle number size distributions. The DMA classifies particles according to their equivalent mobility diameter \(d_m\), which has been shown to be the determining property for the deposition in the respiratory tract, for spherical as well as non-spherical particles (<400 nm, Rissler et al., 2012).

In traffic-dense urban environments, soot (black carbon) is often dominating the emissions by mass. The soot cores carry different degrees of coatings (commonly organic). As soon as the particles are emitted to the atmosphere, the atmospheric aging process begins. When estimating exposure, or dose to the lung, with respect to surface area or mass from number size distributions, information about the particle shape and effective density are needed.

In this study, mass mobility relationship and particle shape is studied in a traffic-dense urban environment using a DMA-Aerosol Particles Mass analyzer (DMA-APM) technique (Park et al., 2003). From the data the effective density can be determined, defined as:

\[
\rho_{eff}(d_m) = \frac{6m}{\pi d_m^3}
\]

During the study the chemical and physical properties of the particles were further characterized with an SMPS, an AMS, TEM-image analysis, OC/EC, and H-TDMA. For comparison, measurements were also performed in the rural background (Vavihill, Sweden).

The measurement showed that the particles were externally mixed, with one group of dense particles with effective density ~1.4 g/cm\(^3\), and one group of aggregated particles, mainly soot. The aggregates had the typical behavior of decreasing effective densities with size, ranging from 0.94 g/cm\(^3\) for 50 nm particles down to 0.26 g/cm\(^3\) for 400 nm (Figure 1). The porous structure of aggregates may lead to overestimating the dose to the lung with respect to particle mass, while underestimating surface area – if assuming spherical particles.

The particle effective densities found at the street level were almost identical to those found for freshly emitted diesel particles generated under well controlled lab-conditions (Park et al., 2003; Rissler et al., 2013), with no signs of atmospheric aging under winter conditions (January-February).

Whereas the particle effective densities were stable over time, the relative number fraction of the two types of particles varied considerably. Two types of time variations were observed; that over time of day, and that depending on the air mass origin. The daily variation in agglomerated soot particle concentration was associated with the traffic pattern – showing the lowest concentrations at night (00:00-04:00). The fraction of ‘dense’ particles increased during occasions with trajectories from polluted areas. Thus, these particles are likely dominated by long range transport, while the soot particles are from local sources.

By introducing a thermo denuder between the DMA and the APM it was shown that the dense particles were volatile at 300 °C, losing >85% of their mass. The aggregates only lost ~10% of their mass, corresponding to a coating layer of ~2nm for 100 nm particles.

The results for the dense particles associated with long range transport were confirmed by the AMS and DMA-APM measurements at the rural background site.

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