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Nitrate Dynamics in UK Urban Environments

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Nitrate makes a significant contribution to the concentration of $PM_{2.5}$ and PM_{10} with important implications for human health and regional climate forcing. This is particularly true for NW Europe, where large emissions of NH_3 and NO_x combine with comparably low temperatures and high relative humidities to create conditions that favour the production of NH_4NO_3. Despite its importance, most atmospheric chemistry models are still not modelling NH_4NO_3 very well, indicating that dynamics are still not represented correctly. In addition, due to its changing equilibrium, there are processes which operated at a sub-grid scale and are therefore difficult to simulate.

In this presentation we review new evidence on nitrate dynamics in urban environments, drawing on data from recent urban field studies, mainly, but not exclusively, in the UK. This new look is enabled through the use of new measurement technology such as Aerosol Mass Spectrometry (AMS), urban eddy-covariance flux measurements of aerosol chemical compounds and long-term nitrate measurements.

We quantify the relative contribution NH_4NO_3 to the UK aerosol, reviewing the existing UK AMS database and data from UK and European denuder/filter-pack networks.

Paired long-term measurements of aerosol concentrations in and outside of two UK urban areas (London & Edinburgh; Tang *et al*., 2008), have provided information on the urban NO_3^- increment. The measurements indicate that, on average:

$$NO_3^-(\text{urban}) = 1.13 \times NO_3^-(\text{rural}) + 0.58\ \mu g\ m^{-3},$$

with somewhat larger increments during the winter months than during summer.

Measurements of the size-distributions of sub-micron non-refractory NO_3^- by aerosol mass spectrometry frequently show periods of a NO_3^- size mode in the range 100 to 300 nm, in addition to the accumulation mode at 300 to 800 nm. This suggests that NO_3^- is formed by condensation on the combustion mode which is prevalent near traffic sources. The fine NO_3^- mode is sporadically observed, and appears to correlate with cold, humid conditions and atmospheric inversions.

The role of urban areas in producing NH_4NO_3 is further supported by the growing database of aerosol chemical compounds above urban areas, by aerosol mass spectrometry (e.g. Nemitz et al., 2008), which suggests emission of NO_3^- from most cities, which is nevertheless highly variable between days (unlike the emission of organic aerosol).

Vertical gradient measurements above the city centre of London during the REPARTEE campaign (comparing measurements on the Telecom tower at 165 m with ground-based urban background measurements) show higher NO_3^- concentrations on the tower, possibly due to colder temperatures at higher heights shifting the gas/aerosol equilibrium towards the aerosol phase.

We also present evidence that the fate of NH_4NO_3 is affected by its dissociation potential. In warm conditions, NH_4NO_3 volatilises during the deposition process to semi-natural vegetation near the ground, where temperatures are raised and concentrations of NH_3 and HNO_3 lowered due to deposition. This greatly increases the effective deposition rate of NH_4NO_3 aerosol and greatly decreases its atmospheric lifetime. Since this volatilisation near the ground cannot be resolved by current CTMs, it is suggested that effective deposition rates need to be incorporated into models to account for this effect.

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