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Diurnal variability in semi-volatile inorganic aerosol concentration and implications for regional radiative effects

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Ammonium nitrate forms a substantial component of the sub-micron particulate composition in polluted regions of Europe and is projected to become more influential as a pollutant and climate forcing agent in future decades. We evaluate simulated diurnal cycles in semi-volatile inorganic aerosol concentrations at Cabauw, the Netherlands, during an enhanced pollution episode in May 2008, where ammonium nitrate is known to have had a large impact on the aerosol optical thickness. We use the UM-UKCA composition-climate model, including the GLOMAP interactive aerosol microphysics module and a recently developed 'hybrid' dissolution solver (HyDis) to accurately represent size-resolved partitioning of ammonia and nitric acid to the particle phase.

Our simulated ammonium nitrate concentrations are consistent with both ground-based and airborne Aerosol Mass Spectrometer measurements from the European Integrated Project on Aerosol and Cloud Climate Air Quality Interactions (EUCAARI) field campaign. We show that simulated size-resolved surface ammonium nitrate concentrations exhibit strong diurnal cycles during this particular episode at Cabauw that are in good agreement with surface observations. Changes in the simulated gas-particle partitioning indicate that temperature is a key driver of this diurnal cycle, with nitric acid favouring the gas phase during the day and particulate phase at night. Moreover, the UM-UKCA captures key features in the observed mid-day vertical distribution of ammonium nitrate, which are known to be important for calculations of the direct aerosol radiative effect, and suggests that the altitude of maximum nitrate concentration varies diurnally due to aerosol semi-volatility. This relatively rapid change in the vertical aerosol distribution may not be represented in models that simplify or omit treatments for semi-volatile gas-to-particle partitioning.

We calculate the influence of this strong diurnal variability in nitrate aerosol vertical distribution on the regional radiative effect at Cabauw and highlight the importance of considering these effects in all regions influenced by semi-volatile aerosol behaviour.