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Gas-aerosol cycling of ammonia and nitric acid in The Netherlands

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Atmospheric ammonia and nitric acid are present over NW Europe in large abundance. Observations made during the IMPACT measurement campaign (May 2008, Cabauw, The Netherlands) show a pronounced diurnal cycle of aerosol ammonium and nitrate on relatively dry days. Simultaneously, AERONET data show a distinct diurnal cycle in aerosol optical thickness (AOT). We used a global aerosol-climate model (ECHAM5-HAM) and a detailed aerosol-cloud column model to help analyse the observations from this period. The study shows that the diurnal cycle in AOT is partly associated with particle number concentration, with distinct peaks in the morning and evening. More important is relative humidity (RH). RH maximizes in the night and early morning, decreases during the morning and increases again in the evening. The particle wet radius, and therefore AOT, changes accordingly. In addition, the RH variability also influences chemistry associated with ammonia and nitric acid (formation of ammonium nitrate, dissolution in aerosol water), resulting in the observed diurnal cycle of aerosol ammonium and nitrate. The additional aerosol matter increases the hygroscopicity of the particles, and this leads to further swelling by water vapor condensation and a further increase of AOT. During the day, as RH decreases and the particles shrink, aerosol ammonium and nitrate are again partly expelled to the gas phase. This behaviour contributes significantly to the observed diurnal cycle in AOT, and it illustrates the complexity of using AOT as a proxy for aerosol concentrations in aerosol climate studies in the case of heavily polluted areas.