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Gas-particle interactions above a Dutch heathland: I. Surface exchange fluxes of NH_3, SO_2, HNO_3 and HCl

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Abstract. A field measurement campaign was carried out over a Dutch heathland to investigate the effect of gas-to-particle conversion and ammonium aerosol evaporation on surface/atmosphere fluxes of ammonia and related species. Continuous micrometeorological measurements of the surface exchange of NH_3, SO_2, HNO_3 and HCl were made and are analyzed here with regard to average fluxes, deposition velocities (V_d), canopy resistances (R_c) and canopy compensation point for NH_3. Gradients of SO_2, HNO_3 and HCl were measured with a novel wet-denuder system with online anion chromatography. Measurements of HNO_3 and HCl indicate an R_c of 100 to 200 s m^{-1} during warm daytime periods, probably at least partly due to non-zero acid partial pressures above NH_4NO_3 and NH_4Cl on the leaf surfaces. Although it is likely that this observation is exacerbated by the effect of the evaporation of airborne NH_4^+ on the gradient measurements, the findings nevertheless add to the growing evidence that HNO_3 and HCl are not always deposited at the maximum rate. Ammonia (NH_3) fluxes show mainly deposition, with some periods of significant daytime emission. The net exchange could be reproduced both with an R_c model (deposition fluxes only) using resistance parameterizations from former measurements, as well as with the canopy compensation point model, using parameterizations derived from the measurements. The apoplastic ratio of ammonium and hydrogen concentration ($\Gamma_s = [NH_4^+]/[H^+]$) of 1200 estimated from the measurements is large for semi-natural vegetation, but smaller than indicated by previous measurements at this site.

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