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Gas-particle interactions above a Dutch heathland: II. Concentrations and surface exchange fluxes of atmospheric particles

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Abstract. Size-dependent particle number fluxes measured by eddy-covariance (EC) and continuous fluxes of ammonium (NH_4^+) measured with the aerodynamic gradient method (AGM) are reported for a Dutch heathland. Daytime deposition velocities (V_d) by EC with peak values of 5 to 10 mm s^{-1} increased with particle diameter (d_p) over the range 0.1–0.5 µm, and are faster than predicted by current models. With a mean V_d of 2.0 mm s^{-1} (daytime: 2.7; night-time 0.8 mm s^{-1}) NH_4^+ fluxes by AGM are overall in agreement with former measurements and NH_4^+-N dry deposition amounts to 20% of the dry input of NH_3-N over the measurement period. These surface exchange fluxes are analyzed together with simultaneous gas-phase flux measurements for indications of gas-particle interactions. On warm afternoons the apparent fluxes of acids and aerosol above the heathland showed several coinciding anomalies, all of which are consistent with NH_4^+ evaporation during deposition: (i) canopy resistances for HNO_3 and HCl of up to 100 s m^{-1}, (ii) simultaneous particle emission of small particles (D_p<0.18 µm) and deposition of larger particles (D_p>0.18 µm), (iii) NH_4^+ deposition faster than derived from size-distributions and size-segregated EC particle fluxes. These observations coincide with the observations of (i) surface concentration products of NH_3 and HNO_3 well below the thermodynamic equilibrium value and (ii) Damköhler numbers that indicate chemical conversion to be sufficiently fast to modify exchange fluxes. The measurements imply a removal rate of volatile NH_4^+ of $3–30\times10^{-6}$ s^{-1} averaged over the 1 km boundary-layer, while NH_3 deposition is underestimated by typically 20 ng m^{-2} s^{-1} (28%) and flux reversal may occur.

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