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A comparison of measured HONO uptake and release with calculated source strengths in a heterogeneous forest environment

Matthias Sörgel (1), Ivonne Trebs (2), Dianming Wu (1), Andreas Held (3,4)

(1) Max Planck Institute for Chemistry, Biogeochemistry, Mainz, Germany (m.soergel@mpic.de), (2) Luxembourg Institute of Science and Technology, Environmental Research and Innovation (ERIN) Department , Esch/Alzette, Luxembourg, (3) University of Bayreuth, Atmospheric Chemistry, Bayreuth, Germany, (4) Bayreuth Center of Ecology and Environmental Research, Bayreuth, Germany

Vertical mixing ratio profiles of nitrous acid (HONO) were measured in a clearing and on the forest floor in a rural forest environment (in the south-east of Germany) by applying a lift system to move the sampling unit of the LOng Path Absorption Photometer (LOPAP) up and down. For the forest floor, HONO was found to be predominantly deposited, whereas net deposition was dominating in the clearing only during nighttime and net emissions were observed during daytime. For selected days, net fluxes of HONO were calculated from the measured profiles using the aerodynamic gradient method. The emission fluxes were in the range of 0.02 to 0.07 nmol m^{-2} s^{-1}, and, thus were in the lower range of previous observations. These fluxes were compared to the strengths of postulated HONO sources and to the amount of HONO needed to sustain photolysis in the boundary layer. Laboratory measurements of different soil samples from both sites revealed an upper limit for soil biogenic HONO emission fluxes of 0.025 nmol m^{-2} s^{-1}. HONO formation by light induced NO_2 conversion was calculated to be below 0.03 nmol m^{-2} s^{-1} for the investigated days, which is comparable to the potential soil fluxes. Due to light saturation at low irradiance, this reaction pathway was largely found to be independent of light intensity, i.e. it was only dependent on ambient NO_2.

We used three different approaches based on measured leaf nitrate loadings for calculating HONO formation from HNO_3 photolysis. While the first two approaches based on empirical HONO formation rates yielded values in the same order of magnitude as the estimated fluxes, the third approach based on available kinetic data of the postulated pathway failed to produce noticeable amounts of HONO. Estimates based on reported cross sections of adsorbed HNO_3 indicate that the lifetime of adsorbed HNO_3 was only about 15 min, which would imply a substantial renoxification. Although the photolysis of HNO_3 was significantly enhanced at the surface, the subsequent light induced conversion of the photolysis product NO_2 did not produce considerable amounts of HONO. Consequently, this reaction might occur via an alternative mechanism. Therefore, the semi-volatile weak acid HONO can serve as an example for the complexity of exchange processes as multiple sources and sinks are coexisting, such as (photo-) chemical formation, microbial formation, adsorption/desorption and (stomatal-) uptake.