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Heterogeneous conversion of NO₂ and NO on HNO₃ treated soot surfaces: atmospheric implications

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Abstract. In the present study, the heterogeneous conversion of nitrogen oxide (NO) and nitrogen dioxide (NO₂) was studied at atmospheric humidity levels on flame soot surfaces treated with gaseous nitric acid (HNO₃). In addition, the heterogeneous reaction of HNO₃ on soot was investigated at atmospheric humidity.

For the treatment of soot by pure HNO₃ only reversible uptake with a surface coverage of $\sim 1\text{--}2 \times 10^{14}$ HNO₃ cm⁻² was observed for HNO₃ mixing ratios in the range 250-800ppbv. Only for higher HNO₃ mixing ratios of >800ppbv the formation of NO and NO₂ was observed. The results were not affected by the addition of NO. In none of the experiments with HNO₃ the formation of nitrous acid (HONO) was observed. For HNO₃ mixing ratios <600ppbv the upper limit yields for HONO, NO₂ and NO were found to be <0.2%, <0.5% and <1%, respectively. Compared to untreated soot, the product formation of the reaction of NO₂ with soot was not significantly affected when the soot surface was treated with gaseous HNO₃ prior to the experiment. Only for high surface coverage of HNO₃ the formation of HONO was suppressed in the initial phase of the reaction, probably caused by the blocking of active sites by adsorbed HNO₃.

Under the assumption that the experimental findings for the used model flame soot can be extrapolated to atmospheric soot particles, the results show that the reactions of HNO₃ and HNO₃+NO on soot surfaces are unimportant for a "renoxification" of the atmosphere and do not represent an atmospheric HONO source. In addition, the integrated HONO yield of ca. 10¹⁴cm⁻² in the reaction of NO₂ with soot is not significantly influenced by simulated atmospheric processing of the soot surface by HNO₃, and is still too small to explain HONO formation in the atmosphere.

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