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Heterogeneous conversion of NO_2 and NO on HNO_3 treated soot surfaces: atmospheric implications

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Abstract. In the present study, the heterogeneous conversion of nitrogen oxide (NO) and nitrogen dioxide (NO_2) was studied at atmospheric humidity levels on flame soot surfaces treated with gaseous nitric acid (HNO_3). In addition, the heterogeneous reaction of HNO_3 on soot was investigated at atmospheric humidity.
For the treatment of soot by pure HNO_3 only reversible uptake with a surface coverage of ~1-2x10^{14} HNO_3 cm^{-2} was observed for HNO_3 mixing ratios in the range 250-800ppbv. Only for higher HNO_3 mixing ratios of >800ppbv the formation of NO and NO_2 was observed. The results were not affected by the addition of NO. In none of the experiments with HNO_3 the formation of nitrous acid (HONO) was observed. For HNO_3 mixing ratios <600ppbv the upper limit yields for HONO, NO_2 and NO were found to be <0.2%, <0.5% and <1%, respectively. Compared to untreated soot, the product formation of the reaction of NO_2 with soot was not significantly affected when the soot surface was treated with gaseous HNO_3 prior to the experiment. Only for high surface coverage of HNO_3 the formation of HONO was suppressed in the initial phase of the reaction, probably caused by the blocking of active sites by adsorbed HNO_3.

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 HNO_3 and HNO_3+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. $10^{14}cm^{-2}$ in the reaction of NO_2 with soot is not significantly influenced by simulated atmospheric processing of the soot surface by HNO_3, and is still too small to explain HONO formation in the atmosphere.

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