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The heterogeneous reaction of NO_3 on laboratory flame soot.

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Uptake experiments of NO_3 on decane flame soot were carried out under continuous molecular flow conditions at 298 \pm 2K using the thermal decomposition of N₂O₅ as a NO₃ source. Two different types of soot were produced: soot originating from a rich flame at a high fuel/oxygen ratio ('grey' soot) and soot generated from a lean flame at a low fuel/oxygen ratio ('black' soot). In situ laser detection using Resonance Enhanced Multiphoton Ionization (REMPI) was used in addition to mass spectrometry in order to specifically detect NO₂ in the presence of N₂O₅ and NO₃. At [NO₃] = (2.7 ± 1.0) x 10^{11} cm⁻³ we found a steady state uptake coefficient γ_{ss} of (0.2 ± 0.02) for both types of soot with γ_{ss} decreasing as [NO₃] increased. Adsorbed NO₃ led to an enhanced uptake of NO2 compared to pure NO2 uptake. For soot originating from a rich flame HONO is released at yields of up to 80 % on large quantities of soot and $[NO_3] =$ $(2.3 \pm 0.5) \times 10^{12} \text{cm}^{-3}$ whereas no HONO was formed on soot originating from a lean flame. The HONO yield obtained from the source emitting a mixture of NO2 and NO_3 at small $[NO_3]$ was smaller by a factor of two than pure NO_2 flow of comparable magnitude. For both grey and black soot we observed production of NO independent of the amount of soot present and of $[NO_3]$. The disappearance of NO₃ was in part accompanied by the formation of N_2O_5 according to $NO_3(ads) + NO_2(g)$? $N_2O_5(ads)$? $N_2O_5(g)$ probably due to the presence of adsorbed NO₃ on the substrate.