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Light changes the atmospheric reactivity of soot

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Soot particles formed by incomplete combustion processes comprise a significant portion of the fine aerosol loading (below 1micron). These particles affect the radiative forcing contributing to global warming and have increased toxicity relative to larger particles because they may settle and persist in the deeper respiratory tract, and can even cross biological barriers. In addition, they are often coated with polycyclic aromatic hydrocarbons (PAHs). Soot particles exhibit a large specific surface area, approx. 100 m2 g-1, which suggests a potential for heterogeneous interactions with atmospheric trace gases. Consequently, soot was suggested to be an important sink for some atmospheric oxidants such as O3 or NOx and its heterogeneous chemistry has been largely investigated in the past years. However, its atmospheric impact was suggested to be negligible due to a rapid soot surface deactivation under atmospheric conditions. As previous studies were done under dark conditions, we decided to investigate the effect of light on the heterogeneous reaction of NO2 on various soot samples. As well, we studied the effect of O3 and light exposure on the aging of soot samples.
The heterogeneous reaction between soot particles and NO2 was studied by means of a coated flow tube equipped with near-UV emitting lamps (300-420 nm). The effect of O3 and light on the wettability of soot was studied by contact angle measurements. The characterization of the soot particles was performed by TEM, ESEM and AMS. Different combustion conditions were used to produce the soot samples, which were generated with a mini-CAST soot generator using propane as fuel.
We determined the uptake coefficients for different gas phase NO2 concentrations (15-120 ppbv) under near UV irradiation. The results showed that the heterogeneous reaction of NO2 and soot under irradiation leads to NO and HONO production with different yields according to the combustion conditions of the generated soot particles. The conversion of NO2 to HONO leads to persistent reactivity over long times (7 hours). Uptake coefficients increased linearly with the irradiation intensity indicating that the number of reactive sites at the soot surface is proportional to the number of photoactivated species.
We suggest that nitrogen-containing organic compounds are also produced on the soot surface as a consequence of the heterogeneous reaction with NO2 under irradiation. These compounds can then be photolyzed and release NO and HONO in a NOx-free atmosphere. An estimation of the HONO production rate indicates that heterogeneous soot photochemistry may contribute to the daytime HONO concentration (1). When soot particles are exposed to high concentrations of O3 under irradiation there is an increase in hydrophobicity as it was previously observed for organic surface films(2).

(1)ME Monge, B D'Anna, L Mazri, A Giroir-Fendler, M Ammann, D. J. Donaldson, and C George. Light changes the atmospheric reactivity of soot. PNAS, 2010, doi:10.1073/ pnas.0908341107 In press

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