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Aging soot particles at atmospherically relevant ozone concentrations and after coating with α-pinene for 16h in a well-mixed continuous flow aerosol chamber.

Franz Friebel (1) and Amewu Mensah A (2)

(1) ETH Zurich, Institute for Atmospheric and Climate Science, Department of Environmental Systems Science, Zürich, Switzerland (franz.friebel@env.ethz.ch), (2) ETH Zurich, Institute for Atmospheric and Climate Science, Department of Environmental Systems Science, Zürich, Switzerland (amewu.mensah@env.ethz.ch)

Freshly emitted soot particles are known to be poor cloud condensation nuclei (CCN) but from atmospheric measurements, it can be deduced that some soot particles act as CCNs. Soot particles have an average atmospheric lifetime of one week. During this time their properties can be changed due to different aging processes such as heterogeneous oxidation (e.g. with O_3 or OH-radicals) or coating (e.g. with oxidized and hygroscopic organic compounds). The investigation of such processes in the laboratory is an experimentally challenging task due to the long time span, which has to be covered to achieve comparability to atmospheric aging.
At ETH we performed aging experiments in which we exposed 100 nm size-selected soot particles to different atmospherically relevant conditions (O_3-oxidation and/or α-pinene coating) for up to 16 h. The long aging time was achieved by applying the continuous flow stirred tank reactor (CSTR) concept. This concepts allowed us to conduct experiments in a rather small aerosol chamber (3 m3) while keeping the particle concentration below 1000/cm3 permitting for size selection prior to the aging steps. For the retrieval of kinetic data from these CSTR-experiments we applied the activation time concept. This is a newly developed mathematical framework that allows for the analysis of non-gradual transitions such as CCN-activity.

Two soot types were produced with a miniCAST soot generator. An organic rich soot - CAST brown (CBw) and a soot low organics content - CAST black (CBk). Both soot types were either directly exposed to 200 ppb O_3, or exposed to α-pinene vapor beforehand. For the coating step, the freshly produced soot aerosol was mixed with a 0.5 ml/min saturated α-pinene air flow in a premix volume. The α-pinene remaining in the gas phase was removed with a charcoal denuder prior to size selection. The remaining α-pinene-concentration in the gas phase was below the 1 ppb detection limit of the sensor.

In the case of heterogeneous oxidation with O_3 CBw particle became CCN-active after 1h:50min - at 1.4% super saturation (SS) and after 8h:30min at 0.6% SS. In contrast, CBk-particles had to be exposed 5 to 6 times longer to reach the similar CCN-activity at the same conditions. In the case of coating with α-pinene the CCN-activity for both soot types was equally high (35min aging for 1.4% SS and 3h:45min for 0.6% SS) which is an increase by factor 3 and 15 for CBw and CBk, respectively.

Parallel to the CCN activity we measured the mobility particle diameter and the single particle mass. We found that exposure to 200 ppb O_3 lead to a fast (< 15 min) increase in mass (CBw: +20%; CBk: +10%) and diameter (CBw: +4 nm; CBk: +3nm) due to O_3 adsorption. A secondary increase in mass (+80%) and diameter (+10nm) over several hours of aging was detected in all α-pinene experiments. The overall increase in CCN-activity correlates well with the secondary mass increase.