The climate effect of soot particles caught in act

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The interaction of water with atmospheric aerosol particles is a key aspect in determining their effect on the Earth's climate. Condensed phase water associated with aerosol particles affects their optical properties, i.e., scattering and absorption properties, as well as their ability to act as cloud condensation nuclei. Here, we present X-ray microspectroscopic investigations of water uptake to individual wood and diesel combustion particles that have undergone aging in the PSI smog chamber (Heringa et al., 2011; Chirico et al., 2011; Zelenay et al., 2011a).

Exhaust from two vehicles, a Euro-2 van and a Euro-3 passenger car, and a wood stove was injected into the PSI smog chamber. Samples were taken before and during processing of the exhaust by simulated solar radiation. Different online particle analysis instruments were operated during the experiments, such as a scanning mobility particle sizer (SMPS) and a hygroscopicity tandem differential mobility analyzer (H-TDMA) (Tritscher et al., 2011). Individual particles were analyzed in a microreactor mounted at the POLLUX microscope at the Swiss Light Source (SLS) at PSI. This microscope allows performing X-ray absorption spectroscopy in the soft X-ray range with high spatial resolution down to below 40nm (Raabe et al., 2008). The microreactor allows spectroscopic and microscopic analysis of deposited submicron particles in situ while exposed to controlled temperature and humidity levels (Huthwelker et al., 2010; Zelenay et al., 2011b). Particles were directly sampled onto the inside of the silicon nitride membrane window of the microreactor. Near edge X-ray absorption fine structure spectroscopy (NEXAFS) probes unoccupied molecular orbitals and therefore carries significant chemical information.

Carbon functional group composition varied with type of exhaust aftertreatment technology in diesel soot particles (Euro 2 vs Euro 3) and responded clearly to aging of the soot particles in the smog chamber, for both diesel soot and wood combustion particles. These changes observed via NEXAFS spectroscopy at the carbon K-edge (Figure 1, right) reflect the competing effects of gas phase oxidation and gas to particle conversion versus particle phase oxidation processes. The water uptake behavior observed in situ at the individual particle level through NEXAFS spectroscopy at the oxygen K-edge (Figure 1, left) could be directly related to these chemical fingerprints obtained from the same particles.

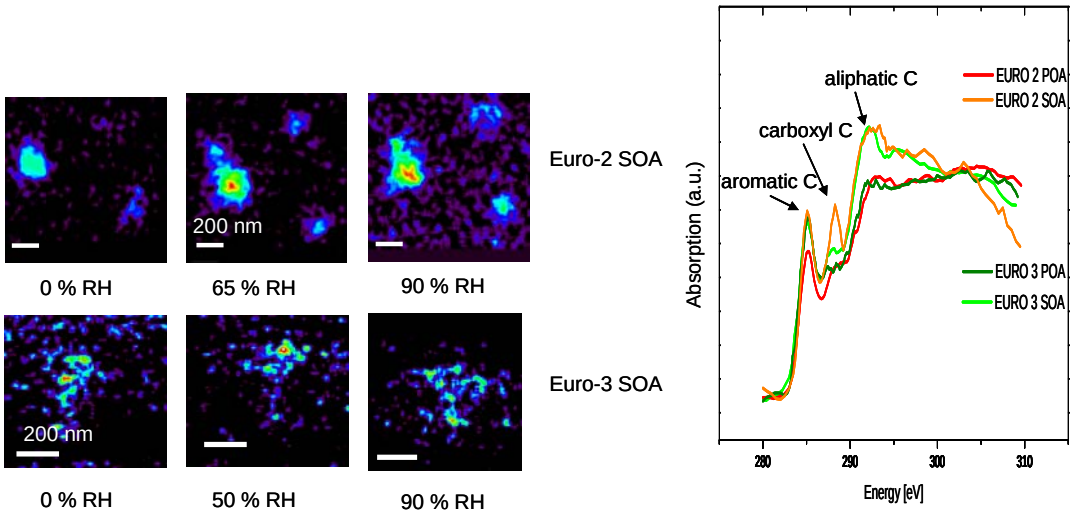

Figure 1: Left: Series of water uptake images based on absorption measurements at 550 eV at different relative humidity (RH) for the Euro-2 van and the Euro-3 passenger car. The amount of water associated with the Euro-2 derived particles at 90% RH is equivalent to roughly 10 nm liquid water. Right: Carbon K-edge spectra from primary (POA) and processed (SOA) particles from the same sources. Spectra were normalized at 305 eV for the purpose of comparison.

Acknowledgement

This work was supported by the Center of Competence in Energy and Mobility (CCEM.CH) through the project NEADS (Next generation diesel exhaust aftertreatment devices)

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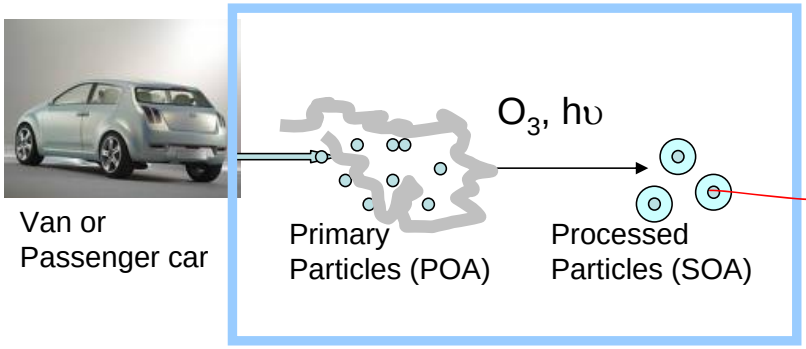
Climate effect of soot particles

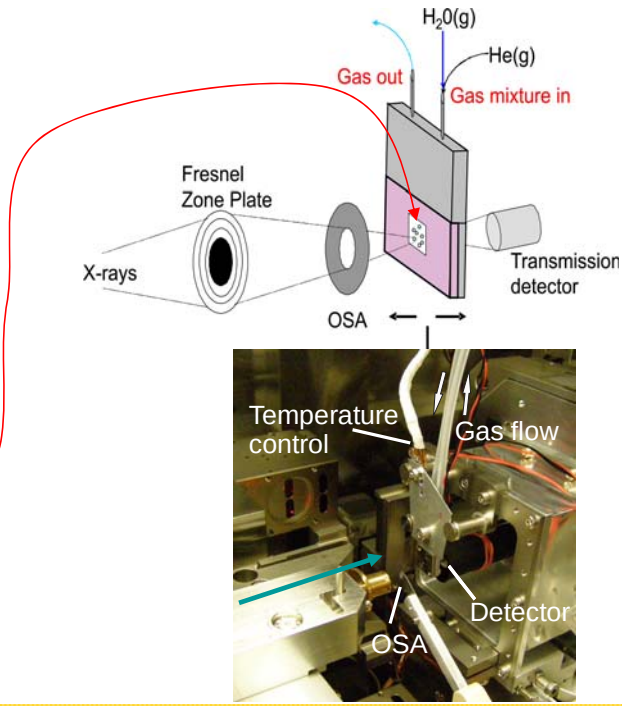
- Direct effect: scattering and absorption of short and long wave radiation
- Indirect effect: aerosols modify the microphysical and hence the radiative properties of clouds

Critical aspect:
Relation between chemistry and ability to interact with water vapor

PSI smog chamber



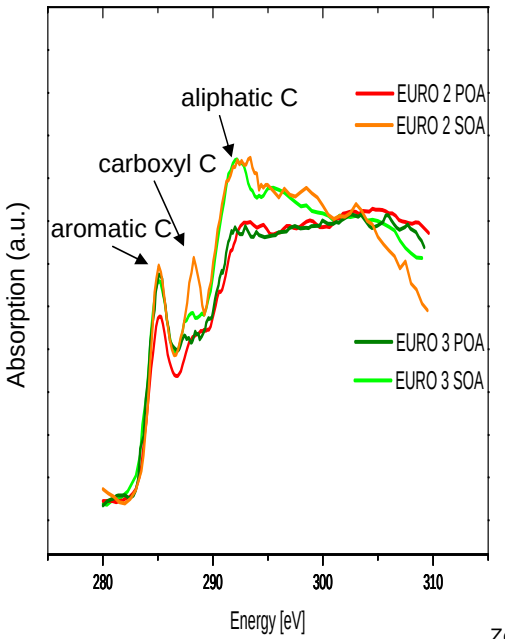
X-ray Transmission Microspectroscopy



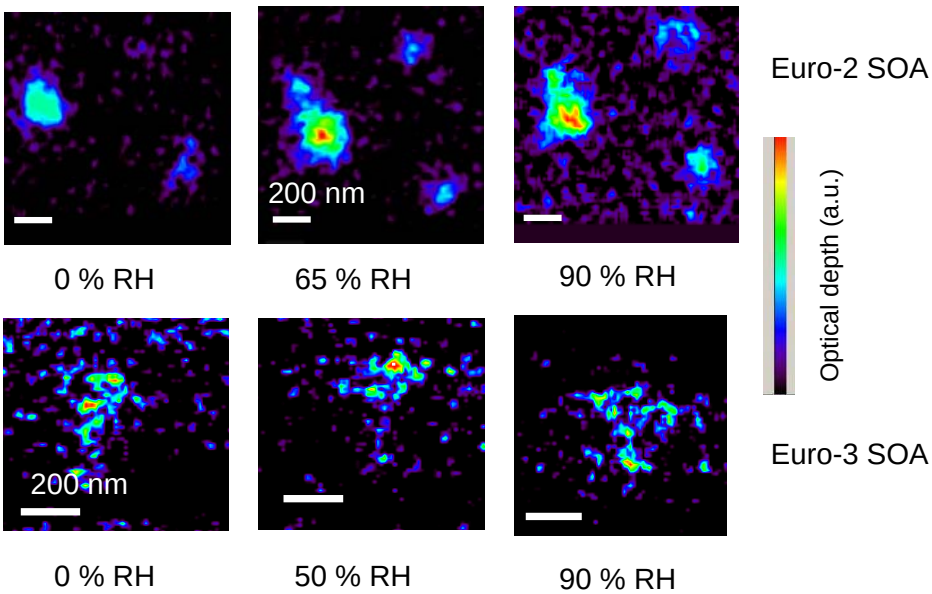
Conclusion

Chemical composition and climate effect of diesel soot particles depend on exhaust aftertreatment technology and aging history in the atmosphere

Carbon functional group composition from C-edge X-ray absorption spectra of individual soot particles



Maps of water uptake from spatially resolved images of X-ray absorption at 550 nm as a function of relative humidity (RH)



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