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Soot Particle AMS measurement in smog chamber experiments

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There is considerable uncertainty regarding the effects of soot containing aerosols on climate and human health. Investigating these effects is complicated by the fact that airborne soot is notoriously difficult to quantify. This is partly due to the ambiguity of 'soot'. Various techniques exist which utilize the effective light absorption or low volatility as means of quantification of airborne soot. The operationally defined quantities measured by these techniques are not necessarily comparable. Furthermore, it is desirable to measure soot as one chemical category among others rather than as a stand alone parameter.

Aerosol Mass Spectrometry has recently emerged as a powerful technique for on-line measurements of particulate composition. The most widely used device, the Aerodyne Aerosol Mass Spectrometer (AMS), operates by means of flash vaporization due to impaction on a heated (600 C) surface. The low volatility of soot makes it a refractory species, i.e. it is not measured by the standard AMS. In order to address this problem a Soot Particle (SP) module was developed, which vaporizes soot by means of a 1064 nm Nd-YAG laser.

In this study an SP-AMS was used to measure soot in diesel exhaust from a light duty vehicle. The exhaust was artificially aged by adding SOA precursors such as toluene and m-xylene and exposing the mixture to UV irradiation in a 6 m^3 smog chamber (Nordin et al 2011) for five hours. The resulting SOA condensation enabled investigations into the difference in instrumental response between fresh and aged soot. Soot particles produced by means of a diffusion flame propane soot generator were also included in the study, to determine the suitability of these as substitute for exhaust particles.

Two complementary techniques were used to validate the measured soot content of the aerosol, Particulate Soot Absorption Photometer (PSAP 3λ, Radiance Research and PSAP 1 λ, custom built) and Aerosol Particle Mass Analyser (APM, model 3600, Kanomax). The PSAP measures light absorption of a filter onto which particles are deposited. From this quantity a mass loading of soot is inferred, taking into account light scattering simultaneously measured with a Nephelometer (Ecotech, Aurora 3000, 3 λ). With an APM one determines the mass of charged aerosol particles through balancing centrifugal and electrostatic forces. In these experiments a Differential Mobility Analyzer (DMA, TSI, model 3071) and thermo-denuder (TD, custom built) were used in combination with the APM. By denuding the sampled aerosol at 300 C, the mass fraction of soot in particles can be deduced.

Throughout the campaign, the SP-AMS was used with both the standard vaporization device and the soot vaporisation module present. The laser was typically engaged in five minute intervals separated by an hour, while the standard device was engaged continuously. This is a compromise which enables soot mass measurements while retaining the capabilities of a normal AMS, producing data which is comparable with that of other studies. The soot and SOA signal generated were qualitatively similar to PSAP and APM signals.

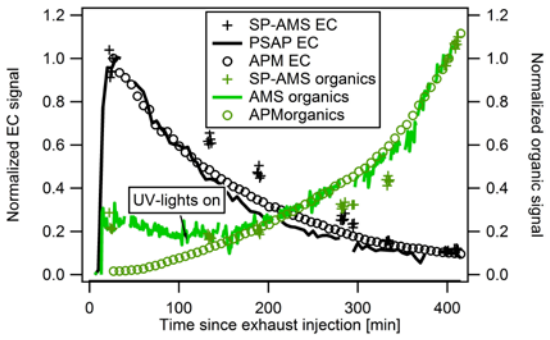

Figure 1. Soot and organic signal during an experiment were diesel exhaust spiked with toluene and m-Xylene was irradiated with UV.

When quantifying chemical species by AMS, one needs to determine their ionization efficiency (IE). This aspect of data analysis is to our knowledge not investigated for soot. There are two key differences from conventional AMS, the molecular structure of the soot, and the mode of vaporization. Preliminary results indicate that the IE of soot is low compared to that of non-refractory species. This has important implications for soot quantification. In the data plotted above, it results in an over prediction of soot mass loading by ~120% at t=400 (where TD-DMA-APM indicates 90% of particulate mass is SOA) if one fails to account for pure C_n fragments formed by the SOA, even though these contribute <3% of SOA signal.

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Nordin, E. Z. *et al*. (2011). *Smog Chamber Studies on SOA Formation from Gasoline Exhaust and Pure Precursors.* EAC proceedings.