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Structure and Composition of Air-Plane Soots and Surrogates Analyzed by Raman Spectroscopy and Laser/Ions Desorption Mass Spectrometry

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Aviation alters the composition of the atmosphere globally and can thus drive climate change and ozone depletion [1]. An aircraft exhaust plume contains species emitted by the engines, species formed in the plume from the emitted species and atmospheric species that become entrained into the plume. The majority of emitted species (gases and soot particles) are produced by the combustion of kerosene with ambient air in the combustion chamber of the engine. Emissions of soot particles by air-planes produce persistent contrails in the upper troposphere in ice-supersaturated air masses that contribute to cloudiness and impact the radiative properties of the atmosphere. These aerosol-cloud interactions represent one of the largest sources of uncertainty in global climate models [2]. Though the formation of atmospheric ice particles has been studied since many years [3], there are still numerous opened questions on nucleation properties of soot particles [4], as the ice nucleation experiments showed a large spread in results depending on the nucleation mode chosen and origin of the soot produced. Most likely one of the reasons behind these discrepancies resides in the different physico-chemical properties (composition, structure) of soot particles produced in different conditions, e.g. with respect to fuel or combustion techniques.
In this work, we use Raman microscopy (266, 514 and 785 nm excitation) and ablation techniques (SIMS, Secondary Ions Mass Spectrometry, and Laser Desorption Mass Spectrometry) to characterize soot particles produced from air-plane at different engine regimes simulating a landing and taking-off (LTO) cycle.
First, the spectral parameters of the first-order Raman band of various soot samples, collected from three different sources in the frame of the MERMOSE project (http://mermose.onera.fr/): PowerJet SaM-146 turbofan (four engine regimes), CAST generator (propane fuel, four different global equivalence ratios), and Kerosene laboratory flame are provided. The spectra are analyzed by performing a de-convolution using the approach described by Sadezky et al. (2005). The soot obtained at different engine regimes presents very similar spectra, with the only exception of the soot obtained at 30% engine regime. In this case, the contribution of D2 band is similar to the contribution of D3 band, while for the samples obtained at 70%, 85% and 100% engine regimes D3 contribution is larger. The results point to a very little impact of engine regime on the generated soot structure. In contrast, surrogate soots show a dependence on the initial combustion parameters and collection conditions.
Second, the surface chemical composition of the soot particles with special focus on PAHs are analyzed by two-Step (Desorption/Ionization) Laser Mass Spectrometry (L2MS) and Time of Fight Secondary Ion Mass Spectrometry (ToF-SIMS) techniques. In L2MS, the adsorbed phase is probed by nanosecond laser desorption (λd=532nm), then the ejected molecules are ionized with a second ns laser (λi=266nm) and further mass-separated by ToF-MS. In both techniques the spectra are obtained using positive polarity, which is better suited for detection of PAHs. A good agreement was obtained between the two techniques for the total PAH content of the analyzed samples. Moreover, the total PAH content followed the same trend as the OC/EC ratio measured with a thermo-optic analyzer (Improve protocol): the 30% engine regime soot presents a high concentration of PAHs and a high OC content, while the three other regimes give a relatively low content of PAHs and OC.
References

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