Geophysical Research Abstracts
Vol. 18, EGU2016-15905-1, 2016
EGU General Assembly 2016

Characterization of Air Plane Soot Surrogates using Raman spectroscopy and laser ablation techniques

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Aviation alters the composition of the atmosphere globally and can thus drive climate change and ozone depletion [1]. Aircraft exhaust plumes contain species (gases and soot particles) produced by the combustion of kerosene with ambient air in the combustion chamber of the engine. Soot particles emitted 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 for 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. The reasons behind these discrepancies reside 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 (514 and 785 nm excitation wavelengths) and ablation techniques (Secondary Ions Mass Spectrometry, and Laser Desorption Mass Spectrometry) to characterize soot particle surrogates produced from a CAST generator (propane fuel, four different global equivalence ratios). They are produced as analogues of air-plane soot collected at different engine regimes (PowerJet SaM-146 turbofan) simulating a landing and take-off (LTO) cycle (MERMOSE project (http://mermose.onera.fr/)) [6].

The spectral parameters of the first-order Raman bands of these soot samples are analyzed using a de-convolution approach described by Sadezky et al. (2005) [5]. A systematic Raman analysis is carried out to select a number of parameters (laser wavelength, irradiance at sample, exposure time) that will alter the sample and the resulting spectra. If much literature discussed the most appropriate values of these parameters for analyzing graphite, coals, or amorphous carbon, only few works have dealt with soot, and no published studies have proposed the optimal parameter values for air-plane soot surrogates.

In this work, we present the effect of three Raman parameters (laser wavelength, irradiance at sample, exposure time) on air-plane soot surrogates. The obtained results qualitatively indicate higher reactivity of soot samples collected in specific air-oxidation conditions. 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 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. CAST samples are further processed by Raman spectroscopy to get complementary information on their structure.

References

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