Linking structural and chemical characteristics of soot particles to their optical absorption in the UV-visible

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Aerosols affect the climate system through various physical processes as they can scatter and absorb solar radiation, emit thermal radiation, or act as cloud condensation nuclei that modify the cloudiness coverage, changing then its albedo. Carbonaceous solid aerosols resulting either from anthropogenic processes or biomass burning are one of the most significant contributors to global climate change (Lohmann, 2005) with respect to their impact on radiative forcing. It is nevertheless not clear how their nanoscopic structural and chemical characteristics can affect their optical properties in the UV-visible spectrum. In this work, we generate soot particles having various structural and chemical properties and we investigate the link between these characteristics at the molecular level and the soot optical properties.

Soot samples are produced using a miniCAST-5201C generator running at various controlled propane-nitrogen flow rates. It allows producing soot particles with organic carbon/total carbon (OC/TC) ratio ranging from 4 to 87%. Structural characteristics are established from HRTEM measurements, which reveal differences in the size distribution of soot primary particles as well as for carbonaceous crystallites' length that compose of these particles. The chemical speciation of carbon and oxygen is undertaken by X-ray photoelectron spectroscopy (XPS) and near-edge X-ray absorption fine structure (NEXAFS) measurements. These techniques also provide crucial informations on the nature of structural and chemical defects existing within the various soot samples (Parent, 2016). Ultraviolet photoemission spectroscopy (UPS) and specific extinction measurements in the near UV-visible spectrum are also performed in order to determine optical properties of soot particles (Bescond, 2016).

NEXAFS and UPS measurements enable us to determine the interband electronic transitions and their relation to structural defects and chemistry of soot particles. We propose an assignment of the resonances observed in the UV-visible spectrum on the basis of these interband transitions, which can be therefore understood in term of structural and chemical properties of the soot (Fig.1).

We show that NEXAFS in combination with UPS, HRTEM and specific extinction measurements enable us to get a better understanding of soot aerosols' UV-visible optical properties that strongly depend on structural and chemical properties at the molecular level.

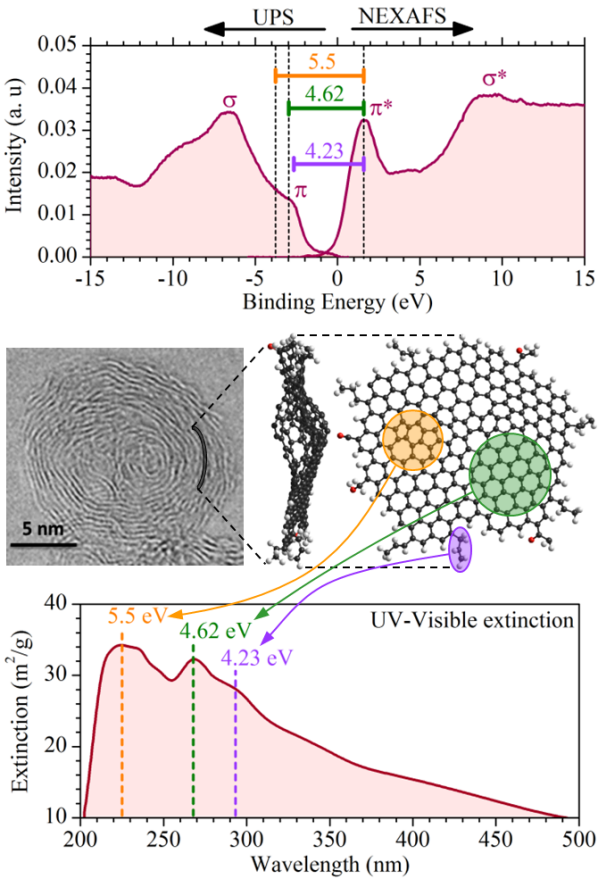

Figure 1. Interband electronic transitions and UV-Visible spectrum of a soot particle made of nanometric carbonaceous crystallites.

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