Evaluation of the role played by multiple scattering on the radiative properties of soot fractal aggregates

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Soot particles are well known for their toxicological impact due to their ultrafine size and content in carcinogenic molecules (PAH) and more recently for their strong influence on the global warming (Bond, et al., 2013). These particles appear like aggregates composed of primary spheres with optical properties similar to the graphitic carbon. Their specific spatial layout permits to establish a fractal like behaviour, *i.e.* a power law relationship between the aggregate mass and its size.

There is a rise in the use of optical diagnostics for *in-situ* characterization of soot particles. Indeed, measurement of light extinction by soot particles is often considered as a reliable way to determine the soot volume fraction. Laser induced incandescence is widely used to measure soot volume fraction in many applications or to evaluate the wavelength dependence of soot radiative properties. Angular light scattering is also used to determine the particle size or fractal dimension d_f (morphological parameter). Because of the complex morphology of these particles, the evaluation of their exact radiative properties is difficult and time consuming. For this reason, it is highly desirable to employ a simple yet adequately accurate theory, namely the Rayleigh Debye Gans theory for fractal aggregates (RDG-FA) in the interpretation of the detected angle resolved scattering signals or extinction measurements to infer morphological parameters.

The RDG-FA theory predicts that light absorption is related to the number of primary spheres N_p in the aggregates (proportional to the particle volume) and the light scattering to N_p^2. The angular dependence of the scattered light is described by a structure factor f (red curve in figure 1) which depends on the particle size and fractal dimension (Dobbins and Megaridis, 1991). This theory appears to be reliable relatively to the experimental uncertainties (Chakrabarty, et al., 2007); however, several important hypotheses are made including that radius of the primary sphere has to be smaller than the wavelength (Rayleigh regime), each primary spheres is supposed to be exposed to the incident light source and multiple scattering inside the aggregate is neglected. Although the role played by multiple scattering has been observed (Liu and Smallwood, 2010; Yon, et al., 2008), to our knowledge, how these assumptions affect the evaluation of the soot radiative properties has not been investigated. The present study was motivated by this observation.

In this study, the absorption and scattering properties of 150 aggregates generated by diffusion limited cluster-cluster aggregation (DLCA) are calculated by the discrete dipoles approximation (DDSCAT) (Draine and Flatau, 2010). The results are compared to those determined by RDG-FA in order to demonstrate the influence of multiple scattering and the shielding effect on the soot radiative properties for 7 wavelengths (from 266 to 1064 nm). For example, figure 1 presents the comparison between the structure factor numerically determined for each aggregates for the wavelength 266 nm (with multiple scattering) and the corresponding RDG-FA one (without).

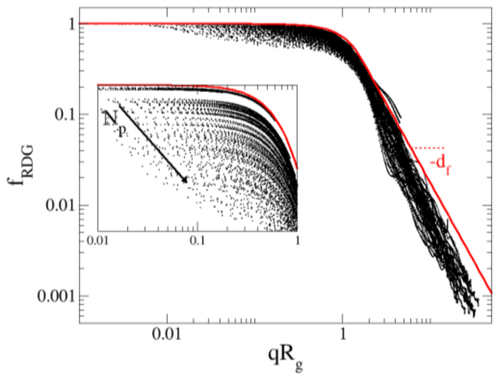

Figure 1. Comparison between structure factors evaluated by DDA approach and RDG-FA (red curve).

This figure shows that multiple scattering can affect the slop of the structure factor in the power-law regime and consequently the determination of the fractal dimension. This deviation from the RDG-FA theory is explained by a phenomenological approach involving the scaling approach introduced by Sorensen (q-region, 2013) by adding the role played by multiple scattering. We finally show that multiple scattering also has a strong impact on the evaluation of absorption and total scattering cross sections.

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