Impact of volatile organic carbon on soot light absorption

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The light absorption of soot contributes significantly to global warming (Bond & Bergstrom, 2006) and is essential for soot characterization by laser induced incandescence and light extinction (Desgroux *et al*., 2013). The optical properties of soot depend on its size (e.g., mobility diameter, d_m) and maturity (e.g., C/H, optical band gap, E_g, non-volatile, refractory organic, OC, and elemental carbon, EC) (Bond & Bergstrom, 2006). Non-refractory, volatile OC (VOC) may also form, coagulate with or adsorb onto soot during combustion (Schnaiter *et al*., 2006), as well as condense during atmospheric aging (Bond *et al*., 2006). Advanced experimental and numerical techniques have focused on the enhancement of soot light absorption in the atmosphere through the lensing effect of condensed VOC (ibid), neglecting though any VOC adsorption taking place during combustion. Thus, understanding the impact of VOC adsorption on soot light absorption is essential for characterization of freshly-emitted soot by optical diagnostics and can improve the accuracy of climate models.

Here, the light absorption of soot containing VOC is investigated by discrete element modeling (DEM) coupled with discrete dipole approximation (DDA; Kelesidis & Pratsinis, 2019). So, DEM-derived nascent and mature soot agglomerates are formed in the absence of VOC by surface growth and agglomeration (Kelesidis *et al*., 2017). During subsequent VOC adsorption, the soot d_m is increased based on the VOC mass fraction, w_{VOC}, by a mass balance. Figure 1 shows that the DEM-derived d_m distributions of soot mixed with VOC (line) are in good agreement with those measured from diffusion flames with w_{VOC} = 0.2 (symbols; Maricq, 2014).

The refractive index of VOC is commonly averaged with that of mature soot that consists mostly of EC to estimate the overall mass absorption cross section, *MAC*. This overestimates up to 160 % the *MAC* measured from premixed and diffusion flames where young soot containing non-volatile OC and VOC (without EC) is formed. The refractive index of soot derived here varies with E_g and VOC content. Using this refractive index, the *MAC* of DEM-derived young soot estimated by DDA decrease up to 50 % in the presence of VOC that predominantly scatters light. The *MAC* of DEM-derived soot containing up to 50 % of VOC is in excellent agreement with data from premixed (Russo *et al*., 2017) and diffusion flames (e.g., CAST soot generator: Schnaiter *et al*., 2006) with E_g = 0.25 - 0.6 eV. This confirms that the maturity of freshly emitted soot is inversely proportional to its VOC composition and determines the light absorption of their composite.

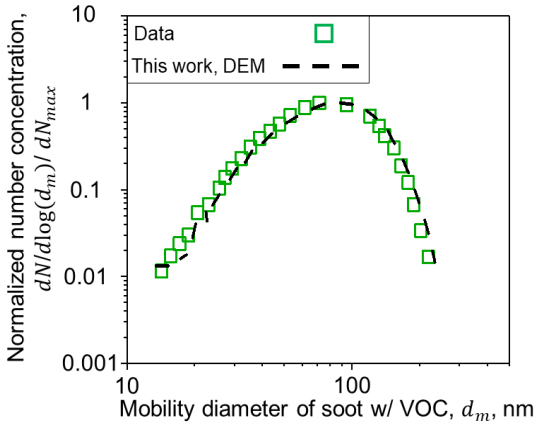

Figure 1. Normalized d_m distribution of soot having average w_{VOC} = 0.2 estimated (line) and measured (symbols) from diffusion flames.

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