Soot morphology, light scattering and direct radiative forcing

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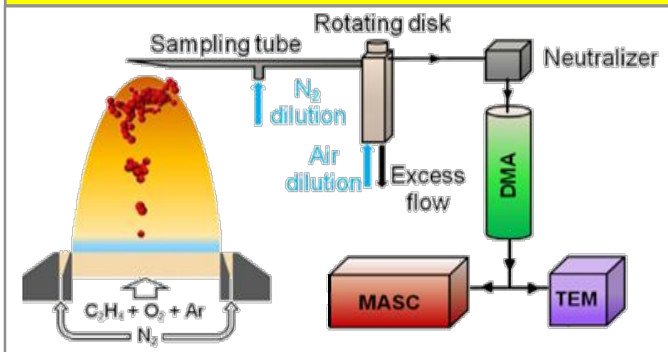
Motivation

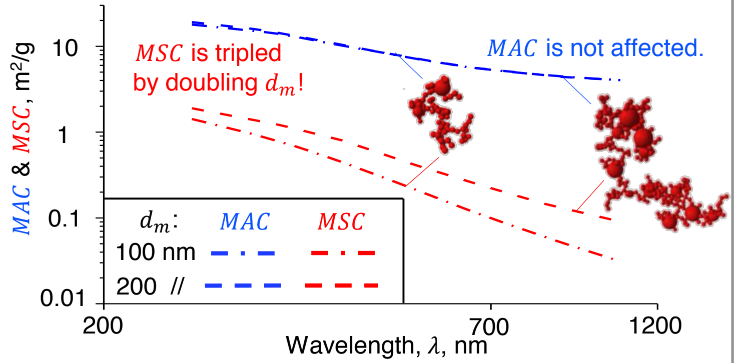
Soot optical properties are calculated typically by Mie theory for spheres[1] or Rayleigh Debye Gans (RDG) theory,[2] neglecting the ramified agglomerate structure consisting of aggregated and polydisperse primary particles (PPs) and impeding accurate estimation of soot environmental impact. Here, soot morphology and radiative properties are investigated experimentally and numerically accounting for surface growth, aggregation[3] and agglomeration[4] by Discrete Element Modeling (DEM) coupled with Discrete Dipole Approximation (DDA).[5]

Soot light scattering measurements



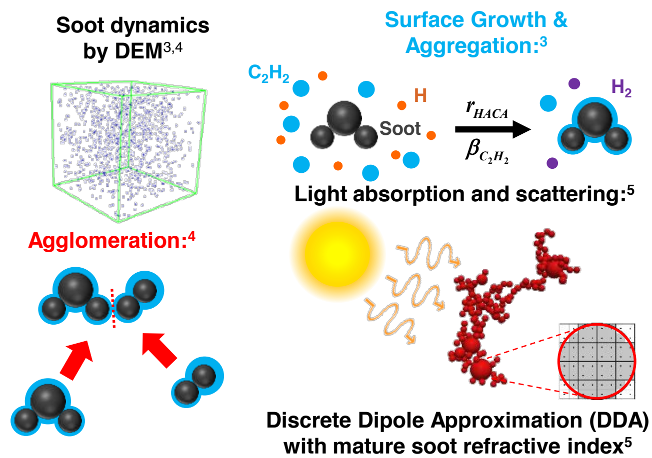
Soot agglomerates are sampled and diluted above a premixed ethylene flame with equivalence ratio of 2.1. The diluted sample is directed to a differential mobility analyzer (DMA) and then to a multiple angle scattering chamber (MASC) or collected for transmission electron microscopy (TEM).

Mature soot *MAC* & *MSC*

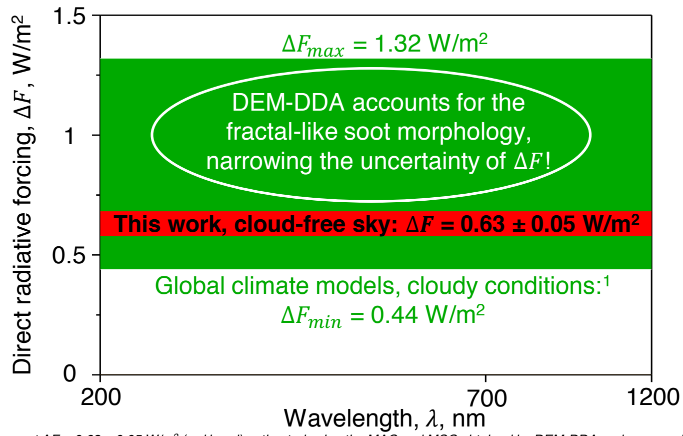


Mass Absorption, MAC, and Scattering Cross-sections, MSC of DEM-derived mature soot agglomerates with d_m = 100 (dot-broken lines) and 200 nm (broken lines) estimated by DDA as function of λ.

Modeling soot structure and optical properties

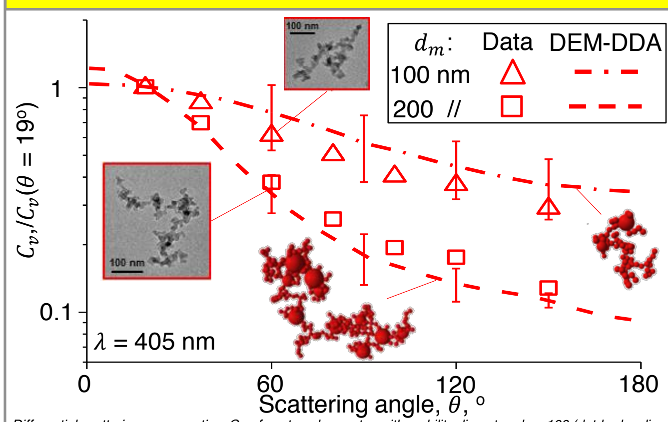


Soot contribution to global warming [6]



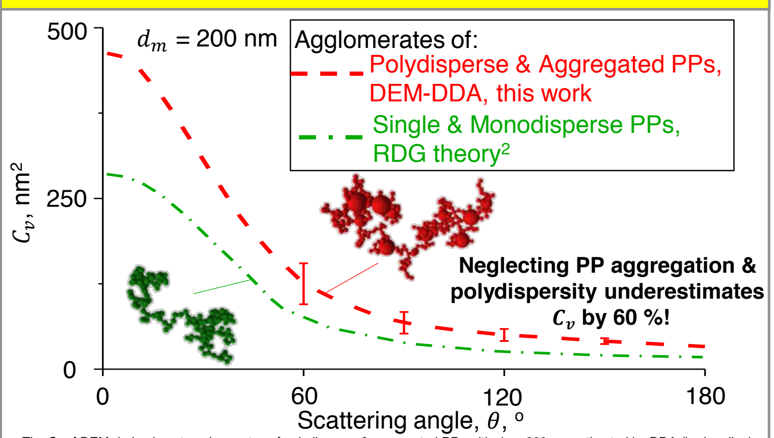
The soot ΔF = 0.63 ± 0.05 W/m² (red band) estimated using the MAC and MSC obtained by DEM-DDA and averaged over d_m = 50-250 nm is compared to the minimum, ΔF_{min}, and maximum, ΔF_{max}, from global climate models (green bands).[1]

Validation of soot light scattering model



Differential scattering cross-section, C_v, of soot agglomerates with mobility diameter, d_m = 100 (dot-broken line, triangles) and 200 nm (broken line, squares) estimated by DEM-DDA (lines) or measured in premixed flames (symbols) and normalized with respect to scattering angle, θ = 19 ° at wavelength, λ = 405 nm.

Comparison to RDG theory



The C_v of DEM-derived soot agglomerates of polydisperse & aggregated PPs with d_m = 200 nm estimated by DDA (broken line) compared to the RDG theory for agglomerates of single & monodisperse PPs with the same d_m (dot-broken line).

References

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Conclusions

1. Soot light scattering simulated for the first time by DEM-DDA is in good agreement with measurements in premixed ethylene flames.
2. The soot *MSC* is enhanced up to a factor of 3 by doubling d_m, while the *MAC* is not sensitive to soot agglomerate size.
3. The RDG theory neglecting PP aggregation and polydispersity underestimates up to 60 % the C_v calculated by DEM-DDA.
4. The average *ΔF* = 0.63 ± 0.05 W/m² estimated here for a cloud-free sky is in between the global climate model predictions using the Mie/ RDG theories for cloudy conditions. This indicates that *ΔF* might be overpredicted by these models that need to account for soot structure and polydispersity.