

Light Scattering and Absorption by Soot Aerosols with Different Morphologies and Coating Distributions

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Soot and Its Climate Impacts

- Soot particles, which are fractal carbon aggregates, are released upon combustion of fossil fuels and stay suspended in the atmosphere for days
- Soot aggregates can absorb and scatter sunlight
- Absorption causes warming of the atmosphere and scattering leads to cooling of the atmosphere
- The ratio of scattering to absorption depends on size, composition, and morphology of soot particles
- Composition of soot aggregates changes in time



Contribution of soot to radiative forcing. IPCC Fifth Assessment Report: Climate Change 2013, Ch: 8, p 698

Interaction of Soot With Atmospheric Chemicals



Compaction of soot aggregate due to condensation

- Presence of a coating on an aggregate affects scattering and absorption
- Condensation causes compaction of the fractal core, which also has an impact on optical properties
- Optical properties of spherical particles can be calculated exactly
- Models of fractal soot exist, but do not always capture all the intricacies
- Experiments are required

- Our group's previous work has shown that two types of coating distributions can form on soot aggregates
- The type of coating distribution depends on vapor pressure and supersaturation of the condensing material



Uniform and pendular-ring coating distributions

* Enekwizu, O., Singh, D., & Khalizov, A. (2020). Absorption and scattering of light by soot aggregates with uniform and pendular ring coatings. Journal of Aerosol Science, 147, 105583–. https://doi.org/10.1016/j.jaerosci.2020.105583

Goal and Objectives

- The goal of the study was to investigate the dependence of absorption and scattering of coated soot aggregates on coating mass and coating distribution experimentally
- The objectives were:
 - Separate the contributions from the addition of coating and aggregate restructuring caused by coating
 - Compare the two possible coating distributions: uniform and pendular-ring
- We have some interesting findings, and I will focus on them

Experimental Setup



- Soot was generated, sized, and aged in a continuous flow system
- Dioctyl sebacate (DOS) was used to produce uniform coatings and triethylene glycol (TEG) was used to produce pendular-ring coatings
- Coated soot was "thermally denuded" to separate the contributions from coating addition and coating-induced restructuring

Fractal Nature of Soot

- Condensation of vapor on soot particles first causes a decrease in mobility diameter
- Diameter starts increasing only after maximum compaction is reached
- Before maximum compaction, both backbone restructuring and coating layer growth contribute to change in optics
- After maximum compaction has been reached, optical properties change only because of coating layer getting thicker



Growth of a spherical particle



Growth of a soot aggregate

Scattering and Absorption After DOS Condensation

- Scattering increases with coating mass both due to restructuring and presence of a liquid layer
- Lensing causes absorption of coated particles to increase
- Compaction of the core causes a small decrease in absorption
- We speculate that the decrease in absorption is due to breaking of necks between monomers

Scattering enhancement, 240 nm soot



Absorption enhancement, 240 nm soot



Decrease in Absorption and Breaking of Necks



Illustration of breaking necks

- DDA calculations showed that breaking necks in a trimer leads to 0.96 absorption enhancement
- Breaking of necks was simulated by rotating monomers with the adjacent neck material

- DDA simulations have been performed by our group to quantify the effects of necks on absorption cross section
- They have shown that a decrease in necking fraction (fraction of neck material) causes a decrease in Mass Absorption Cross section (MAC) in case the necks are made of graphitic carbon



Mass absorption cross section vs. necking fraction, 40 monomers. Ogochukwu Enekwizu: dissertation

Conclusions and Future Work

- Coated soot:
 - Experimental results supported theoretical predictions
 - 7x increase in scattering was observed (x3.6 mass increase)
 - An increase in absorption was observed
- Coated-denuded soot:
 - 1.5x increase in scattering was observed (x2.2 mass increase and larger)
 - The decrease in absorption is novel and will be investigated further

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