Advancements in the incorporation of complex soot morphology within atmospheric sciences

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Theoretical foundation

- Soot fractal particles form through combustion and age in stages, altering their size, shape, and composition over time [11,12].
- Numerical simulations of ~6,000 fractal soot particles with varying shapes, sizes, and compositions were conducted to quantify changes in their radiative properties.

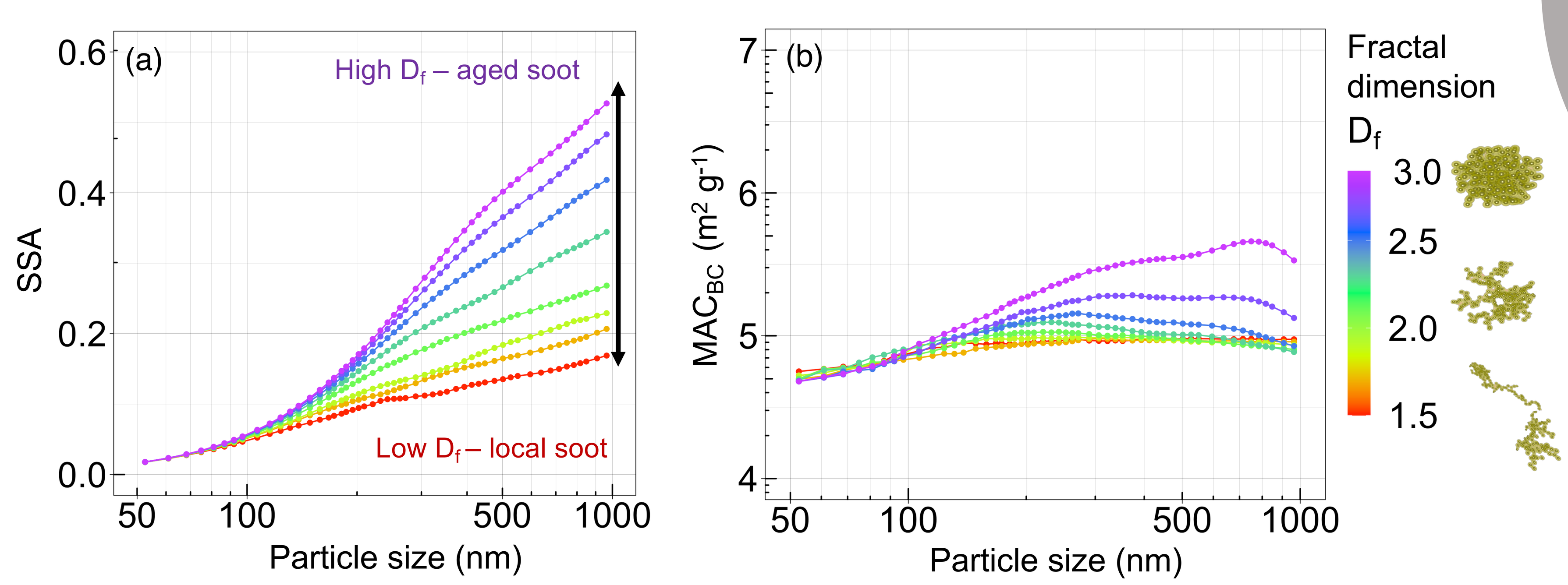

Fig. 1. Significant variation in single scattering albedo (a) and mass absorption cross-section (b) of soot as a function of particle size (X-axis), with different morphologies characterized by fractal dimension D_f (legend on right). An aged compact soot particle has a D_f of 3, whereas, a freshly emitted soot particle has a lower D_f starting from ~ 1.5.

Table 1. Extent of change in soot optical properties with ageing.

Optical property	Factor of change due to morphology
SSA	1.4 – 3.5
g	1.0 – 2.6
MAC	1.1 – 1.5
E_{abs}	1.5
AAE	1.3

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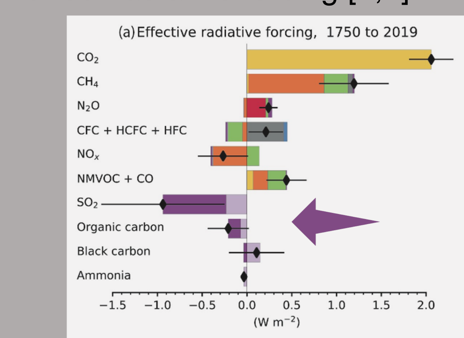
Radiative properties of soot can significantly vary with changes in their morphology.

Background

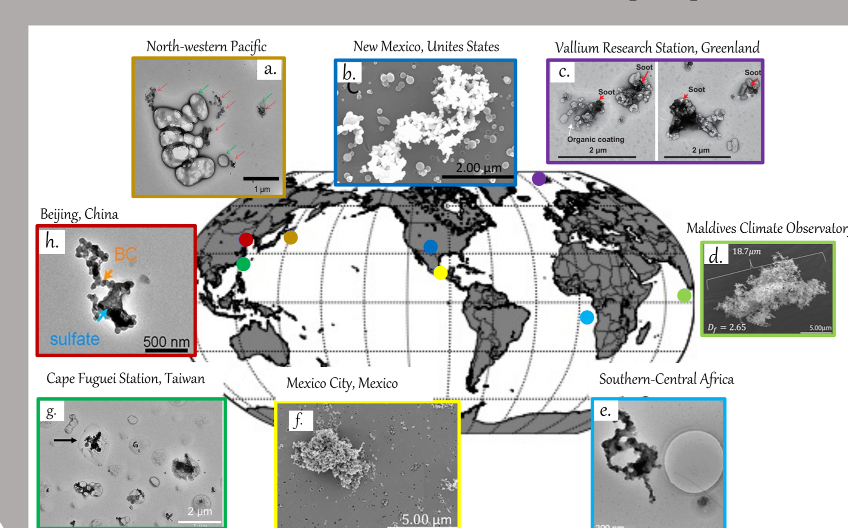
Soot particles are known for their strong light absorption and role in direct radiative forcing [1,2].



Still large uncertainties in their forcing.

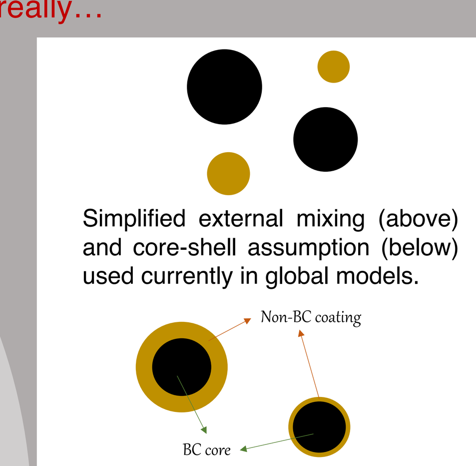
Did you know that soot is actually emitted in the form of fractal-like particles?

Did you know they can retain their fractal shape even 1000kms away from their source of emission? [3-7]



Do you think global models take their complex fractal morphology into account? [8-10]

Not really…



Simplified external mixing (above) and core-shell assumption (below) used currently in global models.

Insights from laboratory measurements

- We investigated the importance of accurately representing size and morphology when modeling the optical properties of black carbon, through a comparison of laboratory measurements and model simulations.
- A series of eleven laboratory experiments were carried out to generate soot with diverse properties, which were then measured, modeled, and systematically compared.

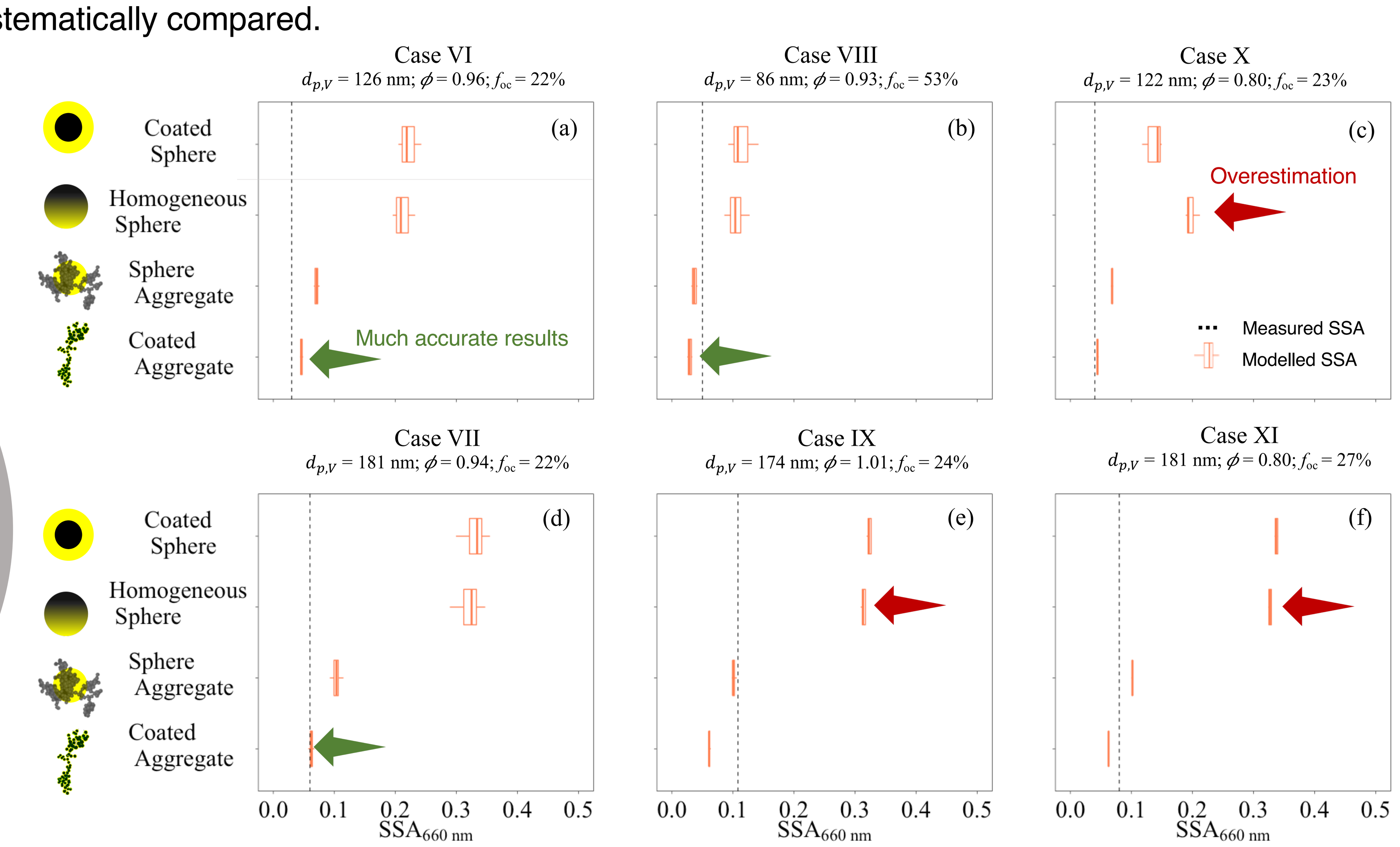


Fig. 2. Comparison between modelled and measured single scattering albedo (SSA) of soot particles. The box plots represent modelled SSA using four morphological assumptions: coated sphere and homogeneous sphere (commonly used simplified representations in global models), and sphere aggregate and coated aggregate (more realistic representations of soot morphology). The dashed line indicates the measured SSA. Panels (a) to (f) display results from six distinct experiments, each with different particle sizes and compositions, as noted at the top of each panel.

Laboratory experiments also showed that accounting for the soot's fractal morphology reduces modeling uncertainties.

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Evidence from polluted environment

- This study confirms that incorporating fractal morphology is crucial for reducing overestimation in aerosol light absorption calculations in urban polluted environments.
- In Delhi, during periods of high anthropogenic BC emissions, light absorption is overestimated by 50–200% when using external or core-shell internal mixing assumptions.

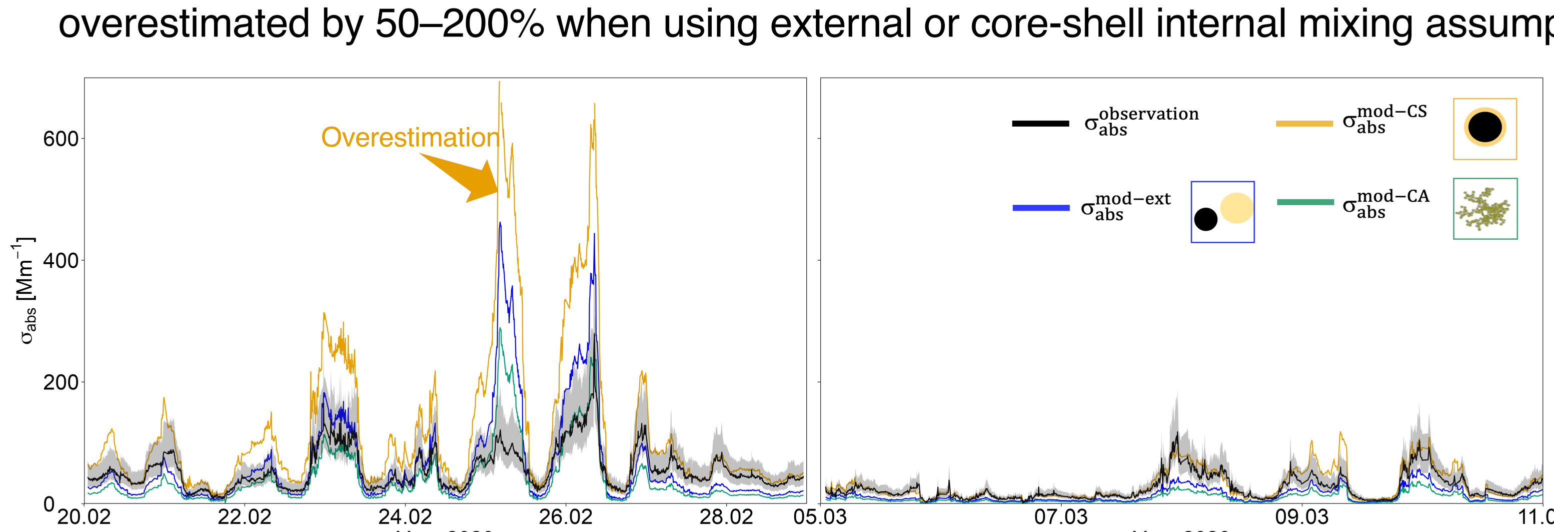
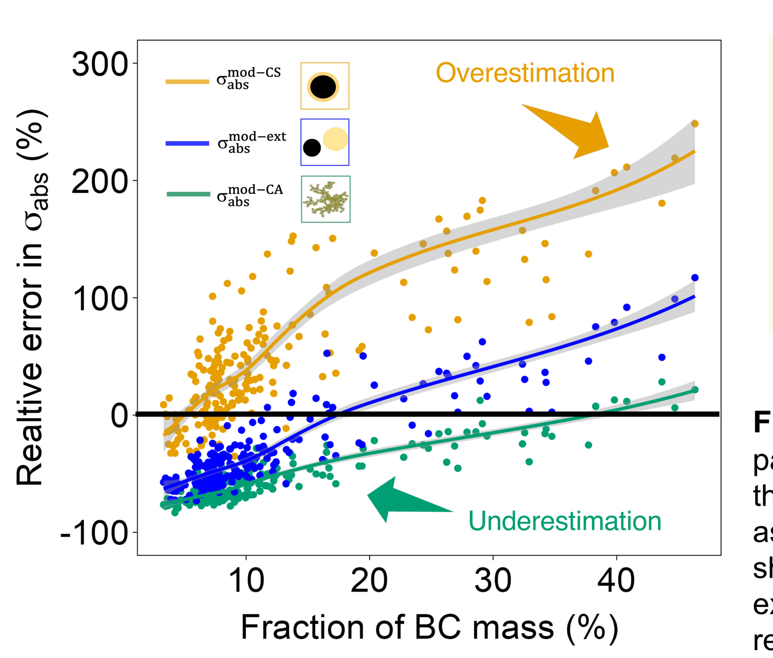


Fig. 3. Time series of modelled and measured particle light absorption coefficient (σ_{abs}) in a polluted urban environment in New Delhi. The measured σ_{abs} is shown as a black line, while the modelled values are represented using three different morphological assumptions: simplified core-shell (orange), simplified external mixing (blue), and realistic coated fractal (green).



Modelled light absorption shows large discrepancies across all morphology models, with performance strongly influenced by the BC mass fraction, which may indicate the dominant particle morphology.

Fig. 4. Relative error in the particle light absorption due to three different morphological assumptions: simplified core-shell (orange), simplified external mixing (blue), and realistic coated fractal (green).

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Take home message

- Soot radiative properties are highly sensitive to morphological changes during ageing, proving simplified core-shell assumptions inadequate.
- Laboratory studies show that fractal morphology is essential for representing fresh black carbon, while in ambient environments, its use depends on BC mass fraction—higher fractions favor fractal over spherical representations
- A machine learning algorithm was developed to predict radiative properties of BC fractal aggregates across all ageing stages.
- Morphology Index (MI) provides a practical metric for capturing soot complexity in climate models, streamlining implementation and reducing uncertainties.

Improved adaptability for global models

- The two extreme morphology models were used to derive an index for morphology (MI):

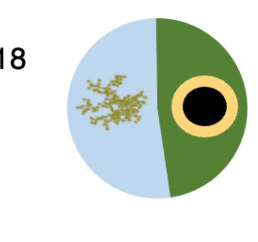
$$\sigma_{abs}^{internally-mixed} = \sigma_{abs} \cdot \mathrm{MI} + \sigma_{abs} \cdot (1 - \mathrm{MI}) + \epsilon$$

$$0 < \mathrm{MI} < 1$$

Urban polluted environment

MI = 0.48 ± 0.18

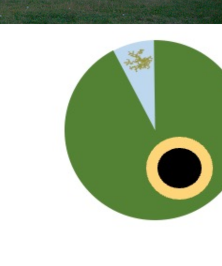
f_{BC} = 21%



Rural European environment

MI = 0.80 – 1.0

f_{BC} < 5%



Morphology Index (MI) offers a promising global metric for representing complex soot morphology in climate models, simplifying implementation while reducing associated uncertainties.

- To directly estimate the radiative properties of fractal soot without relying on computationally intensive simulations, a fast and accurate machine learning–based tool was developed. Artificial neural networks (ANN) and kernel ridge regression (KRR) were used.

Table 3. Absolute errors in the ML predictions and experiments for all optical properties.

Optical property	Random split		Interpolation split		Extrapolation split		Feature range
	KRR	ANN	KRR	ANN	KRR	ANN	
Q_{abs}	0.0022	0.0039	0.0122	0.0287	0.0329	0.0354	0 – 2
Q_{sca}	0.0019	0.0031	0.0224	0.0466	0.0393	0.0939	0 – 2
g	0.0044	0.0038	0.0429	0.0289	0.0879	0.0485	0 – 1

Table 2. Significant reduction in time.

ML model	Training time	Prediction time
KRR	33.3s	0.0006s
ANN	1770s	0.0005s

ML algorithm was developed which generated the radiative properties of BC fractal aggregates for all stages of ageing.

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