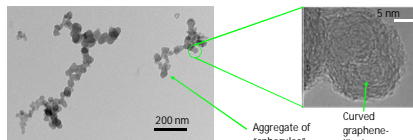


The size-dependent mass absorption cross-section (MAC) of soot particles from various sources

Joel C. Corbin^{1†}, Tyler J. Johnson², Fengshan Liu¹, Timothy A. Sipkens¹, Mark Johnson³, Prem Lobo¹, and Greg Smallwood¹
¹ Metrology Research Centre, National Research Council Canada, Ottawa, ON, Canada
² Atmos Ltd., Edmonton, Canada; ³ Rolls Royce Corp., Derby, UK
 † joel.corbin@nrc-cnrc.gc.ca

1. Background

- Soot black carbon (soot BC) particles are flame-synthesized nano-aggregates of nearly graphitic carbon spherules [1,2]:



- The MAC is used in pollution and emissions measurements to quantify soot mass concentrations, and in atmospheric modelling to predict the radiative effects of soot on climate.
- The mass absorption cross-section (MAC) is perhaps soot's most important property. The MAC is the light absorption per unit mass of a soot sample. Essentially, soots "blackness":

$$MAC = (\text{light absorption coefficient at wavelength } \lambda) / (\text{mass conc.})$$

2. Objective

- We performed measurements, modelling, and a literature review to demonstrate that the MAC of soot particles generally decreases for smaller particles, and elucidate the reasons why [3].

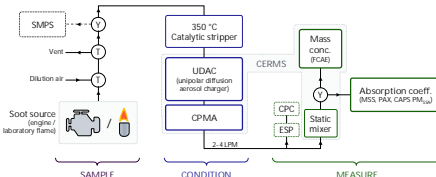
3. Models

- We modelled all existing literature hypotheses related to size-dependent MACs. Each model represents a different physical hypothesis. These models included generalized Mie models (GMMs), which represent the soot morphology shown above with negligible assumptions, as well as simplified Rayleigh-Debye-Gans models, which treat individual soot spherules as non-interacting.

Model	Hypothesis	Reference
RDG-0	Null (no size dependence)	-
GMM-1	Aggregate internal scattering	Sorensen, JQSRT 2018
GMM-2	Monomer-aggregate size correlation	Dastanpour & Rogak, AS&T 2014; Dastanpour et al. Carbon 2017
RDG-3	Quantum confinement	Kelesidis & Pratsinis, PCI 2019; C. Liu, ..., H. Wang, PNAS 2019
(none)	Size-dependent maturity	Various, see Corbin et al., Carbon 2021

4. Experimental methods

We used the CERMIS approach [4] to determine the MAC:

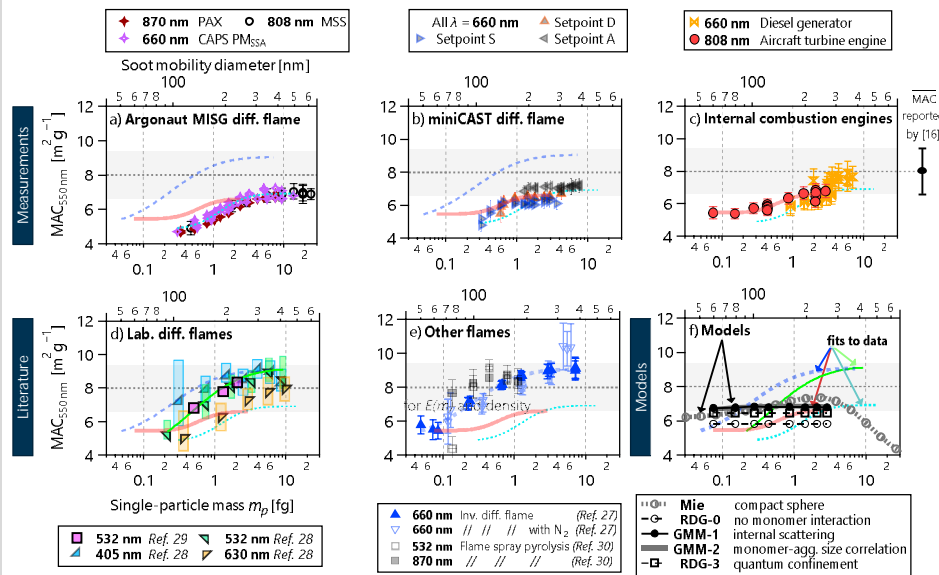


In the CERMIS, a centrifugal particle mass analyzer (CPMA) transmits particles of a known mass-to-charge ratio before a Faraday cup aerosol electrometer (FCAE) measures the total current. The total mass concentration is therefore accurately known. The average mass of individual particles m_p is then calculated by modelling particle charging in the UDAC. The uncertainty in this calculation is considered negligible. It does not affect the MAC trends we report.

We corroborated our conclusions using studies from literature, which also varied the conditioning and measurement steps, and measurement system.

5. Experimental results

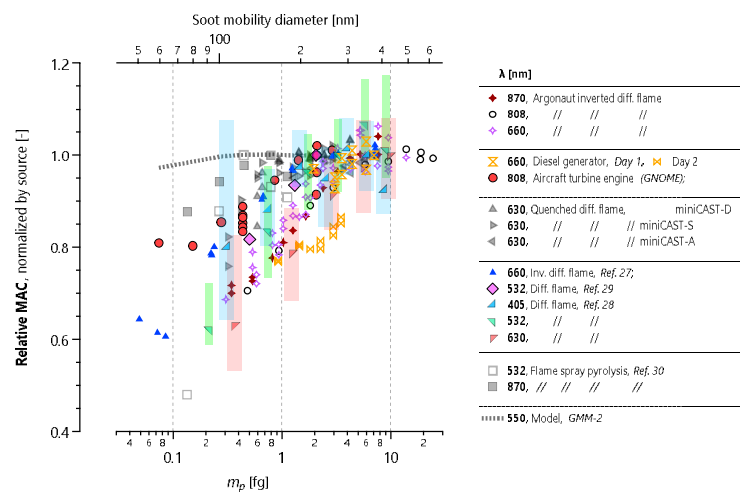
We observed a sigmoidal size dependence for four different samples. Four other studies using a variety of techniques also observed similar in our study (panels a-c below). This trend was observed using various trends (panels d-e), although our novel experimental approach light wavelengths, various instruments, and various combustion systems generated a uniquely large data set.



6. Results cont'd

Combining and normalizing the results of our study and literature (right) makes it clear that the size-dependent MAC follows a different trend for different sources.

Note that we use the term "size" to encompass measurements that are based both on single-particle mass m_p and on its conversion to soot mobility-equivalent diameter following the parameterization of Ref. [5]. The accuracy of this parameterization does not affect the trends discussed here.



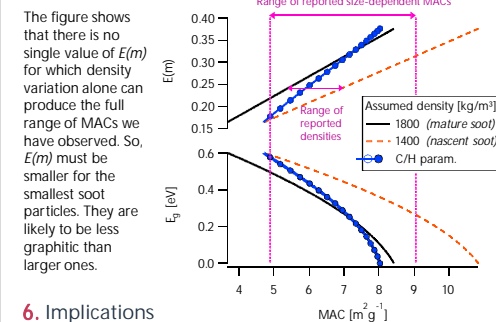
3. Model results

The null model from the table in Section 3 is:

$$MAC_{RDG} = \frac{6\pi E(m)}{\text{wavelength} \times \text{density}}$$

Where $E(m)$ is the absorption function, which depends only on the refractive index. This model includes several important simplifications. However, Panel f (left) shows that it is sufficient for our discussion, since none of the literature models showed a greater size dependence than the null model (RDG-0). Therefore, the size-dependent MAC is due to some other property than those varied in the table (left).

The only hypotheses not explored by the literature models are that smaller soot particles absorb less light due to a lower density or due to a difference in $E(m)$. The figure below tests the density hypothesis. It illustrates the null model evaluated across the full range of reasonable [4] literature values for $E(m)$ and density.



6. Implications

The size-dependence of the MAC influences the measurement accuracy of common instruments and has implications for all of the applications given in Section 1. We illustrate this influence using calibration data from an aircraft-turbine engine, where a 10% variability in the calibration result (slope) is observed as a function of particle size (indicated by the CPMA setpoint).

