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COATING MATERIAL-DEPENDENT DIFFERENCES IN MODELLED LIDAR-MEASURABLE QUANTITIES FOR HEAVILY COATED SOOT PARTICLES

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PLEASE NOTE:

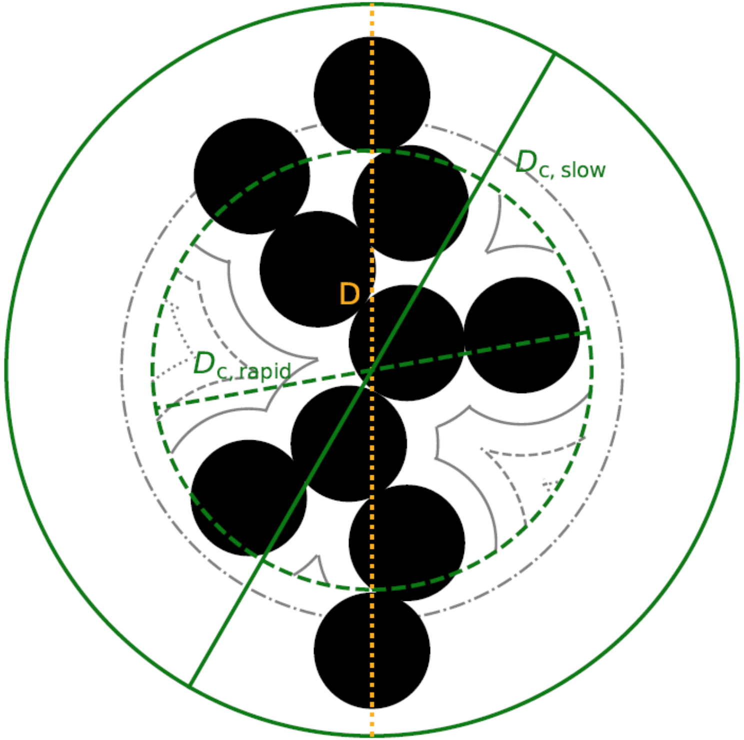
The presentation is based on the following Open Access publication, which covers technical details more in-depth than these slides.

Franz Kanngiesser and Michael Kahnert, "Coating material-dependent differences in modelled lidar-measurable quantities for heavily coated soot particles," Opt. Express **27**, 36368-36387 (2019)

https://doi.org/10.1364/OE.27.036368

INTRODUCTION

- Proposed coating model with tuneable transition between film-coating and spherical-shell coating
- Transition after sphere defined by D_c is filled
- Uncertainty estimate and further details in 2018 article: https://doi.org/10.1016/j.jqsrt.2018.05.014

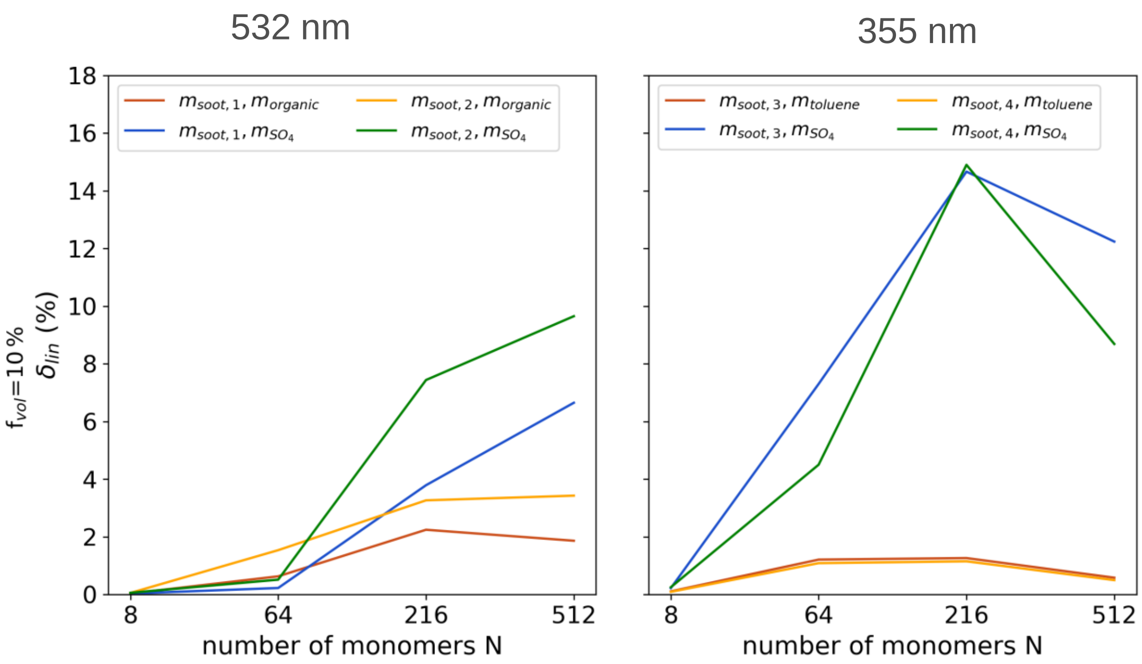


adapted from
Kanngießer & Kahnert, 2018 JQSRT

INTRODUCTION

Impact of changes in refractive index of both soot and coating at 532 nm (left) and 355 nm (right) on linear depolarisation ratio of heavily coated soot particles

Coating refractive index was one of the largest sources of uncertainty for model on previous slide



adapted from Kanngießer & Kahnert, 2018 JQSRT

INTRODUCTION – SCIENTIFIC QUESTION

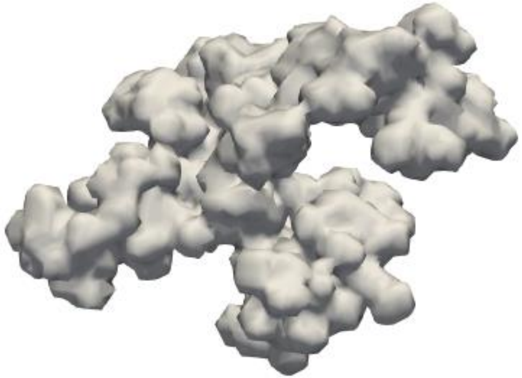
Based on the rather large uncertainty associated with coating material:
Can depolarisation ratio and extinction-to-backscatter ratio be potentially used to distinguish between different coating materials?
I.e. can the coating material be considered a source of information rather than uncertainty?

CONSTRUCTING BARE AGGREGATES

Aggregates follow fractal scaling relation (used for constructing):

$$N = k_0 \left(\frac{R_g}{a}\right)^{D_f}$$

aggregate size controlled by number of monomers,
size increases with ΔN=26



number of monomers **N=26-1508**

fractal dimension **D_f=2.2**

(describes compactness, for sphere D_f=3)

fractal prefactor **k_0=1.625**

(describes packing density

along branch, the higher k_0 the denser the packing)

monomer radius **a=28 nm**

overlap factor **C_{ov}=0.33**

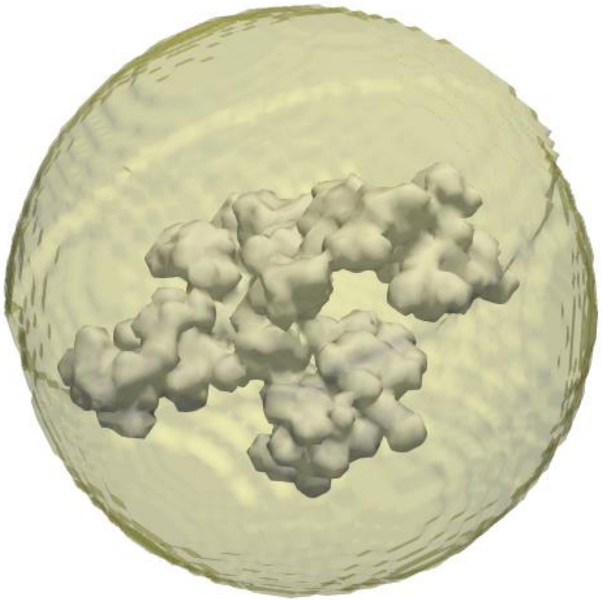
(quantifies overlap between monomers,

C_{ov}=0 point-contact, C_{ov}=1, full overlap)

Input values from:
China et al., 2013 Nature; Adachi et al., 2010 JGR

CONSTRUCTING COATED AGGREGATES

adding coating material layer-by-layer onto the aggregate, until a predefined soot volume fraction f_{vol}=0.07 is reached

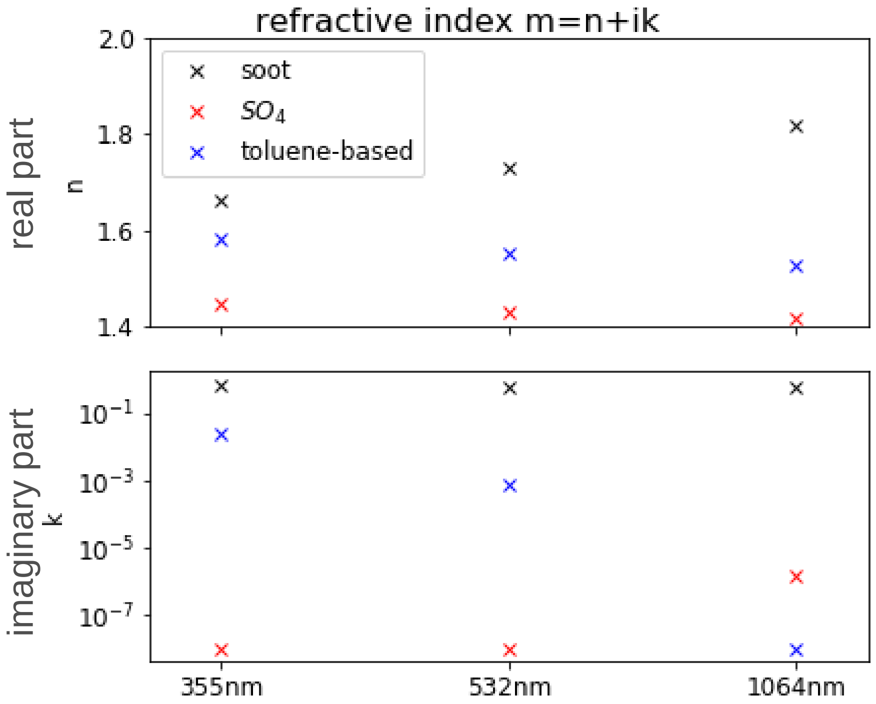


$$f_{vol} = \frac{V_{soot}}{V_{soot} + V_{coat}}$$

REFRACTIVE INDICES

Spectral behaviour of refractive indices used

Coated aggregates only differ with respect to the coating material (sulphate or a toluene-based material)



Hess et al.,1998 BAMS; Chang and Charampopoulos, 1990 Proc R Soc Lond; Liu et al., 2015 ACP

CALCULATIONS

ADDA version 1.2 (Yurkin and Hoekstra, 2007 JQSRT)

Provides Scattering matrix F, optical cross sections C_{ext}, C_{abs}

Linear backscattering depolarisation ratio:

$$\delta_l = \frac{F_{11}-F_{22}}{F_{11}+F_{22}}$$

$\delta_l = 0$ for rotationally symmetric shapes, like homogeneous spheres

Extinction-to-backscatter ratio (lidar ratio)

$$S = 4\pi \frac{C_{ext}}{C_{sca}F_{11}}$$

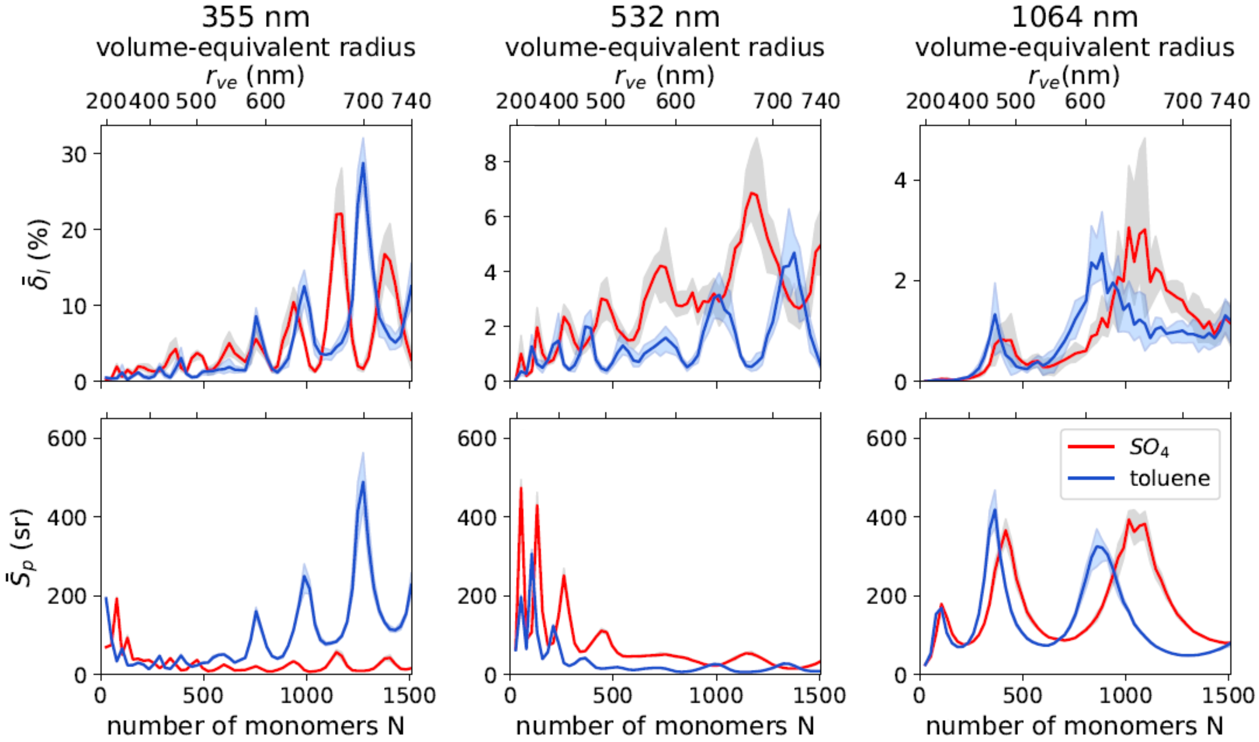
with F_{11}, F_{22} elements of normalized Stokes scattering matrix in backscattering direction

SIZE-DEPENDENT RESULTS

Depolarisation ratio
(Note different ranges for y-axes!)

Extinction-to-backscatter ratio

Solid lines represent arithmetic mean over 5 different aggregate realisations, shaded areas the entire range

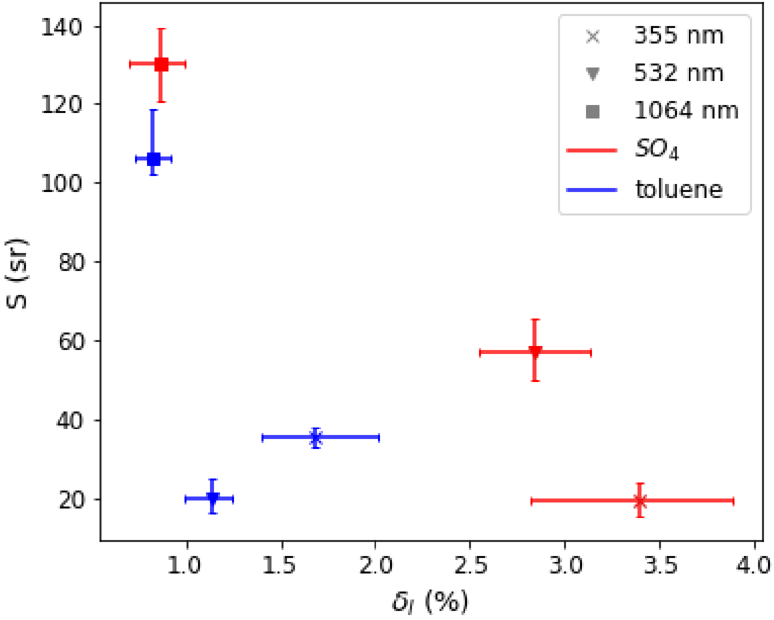


adapted from Kanngiesser and Kahnert, 2019 OE

SIZE-AVERAGED RESULTS

- Applying different log-normal distributions based on number of monomers (N)
- $300 \leq m \leq 700$, $300 \leq s \leq 1350$ (values from field measurements: m=498, s=995 (China et al., 2013 Nature))

extinction-to-backscatter ratio



depolarisation ratio

Error bars include uncertainties from aggregate realisation and varying the log-normal distribution's shape

LIMITATIONS

So far: well controlled numerical experiment

Atmospheric soot: coating thickness, aggregate geometry, chemical composition vary → further uncertainties

SUMMARY

- Calculations of depolarisation ratio and extinction-to-backscatter ratio for thickly coated soot aggregates with 26 – 1508 monomers and two different coating materials
- Distinct coating-material dependent differences in depolarisation and extinction-to-backscatter ratio

CONCLUSION

Based on the rather large uncertainty associated with coating material:
Can depolarisation ratio and extinction-to-backscatter ratio be potentially used to distinguish between different coating materials?
I.e. can the coating material be considered a source of information rather than uncertainty?

Depolarisation ratio and extinction-to-backscatter ratio can potentially be used to distinguish between coating materials of heavily coated soot particles

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