Effect of organic coatings on optical properties of black carbon aerosol: Insights from Mie theory based model simulations

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Abstract

Black carbon (BC), play a crucial role in climate change due to their significant impact on radiative forcing. This study investigates the influence of organic carbon (OC) coatings on the optical properties of BC aerosols using Mie theory-based calculations. We have examined a range of core diameters and coating thicknesses to assess changes in both absorption and scattering cross-sections. The results reveal that coatings consistently enhance both absorption and scattering properties, with the enhancement in scattering being higher in magnitude than enhancement in absorption. The study compares the optical properties of coated BC with that of OC particle showing that as the coating thickness increases, the optical behavior of the coated BC particle converges towards that of OC. The calculations for simple radiative forcing efficiency showed that as the coating on BC increases, its forcing efficiency decreases, implying that heavily coated BC may have a reduced radiative impact in the atmosphere.

Keywords: black carbon, organic carbon, enhancement ratio, radiative forcing, mie theory

Synopsis: Inclusion of enhanced scattering due to organic carbon coatings on black carbon can impact its climate forcing assessments

1. Introduction

Aerosol plays a significant role in atmospheric radiative forcing, but at the same time, it is marked by large uncertainty in its estimates.[1] Though major aerosol types have a negative contribution, BC, due to its strong absorptive properties, contributes positively to radiative forcing. It is among the top anthropogenic contributor, along with CO_2 and methane, to climate forcing.[1,2] BC radiative forcing from fossil fuels and biomass burning has been estimated to be approximately +0.4 W m^{-2}.[2] However, sensitivity tests suggest that the mixing state and morphology of BC aerosols can significantly affect the absorption of BC and thus its radiative forcing potential.[3,4,5] OC, the other type of carbonaceous aerosol, is predominantly scattering in nature with negligible absorption. Thus, it is estimated to contribute negatively to net aerosol radiative forcing.[6]

In the atmosphere, BC is typically co-emitted with OC during combustion, which leads to external and internal mixing states. In internal mixing, the OC vapors can get condensed on the BC substrate, leading to the complete or partial encapsulation of BC. In external mixing, BC and OC remain separated but get mixed externally. The evaluation of BC optical properties remains restricted due to the significant uncertainties associated with BC morphologies and mixing states.[5,7,8] The internal mixing state of BC has been extensively investigated for its light absorption properties.[9,10] Theoretical calculations using the Mie core-shell model have shown that when BC is coated with OC, then the shell acts as a lens to increase the overall absorption properties of BC. This is termed the lensing effect.[11] Experimentally, thermal heating methods have been used to study this absorption enhancement.[12] Heating the aerosol at preset temperature leads to the removal of coatings from BC substrate. The ratio of absorption coefficients measured before and after removal of organic coatings provides the absorption enhancement ratio (E_{abs}). Both laboratory experiments and field measurements show increased absorption due to coatings on BC, leading to E_{abs} greater than 1.[12,13,14,15] However, the coatings on BC, along with its absorption, are expected to alter its scattering properties as well. The changes in scattering properties due to coating are comparatively less explored. Few laboratory measurements have reported enhancement in BC's light scattering upon coating.[16,17,18] Similar to case of absorption enhancement, the enhancement in light scattering can be expressed in the form of scattering enhancement ratio (E_{sca}). In terms of absolute magnitude, E_{sca} observed in these experimental studies was higher than E_{abs}. In order to estimate radiative forcing potential, along with absorption, scattering as well plays an equally important role. Climate models, which include enhancement in BC absorption due to its mixing state, often assume an E_{abs} value of ~1.5 - 2 for its radiative forcing estimation.[10,19] However, the changes in the scattering properties of coated BC particles and their potential effect on radiative forcing capabilities remain contentious and unaddressed.

In this study, we have performed Mie theory calculations on the core-shell model to estimate changes in both absorption and scattering properties of coated BC spheres. The study focuses on the implications of OC coating on the overall optical properties of BC, providing insights into how these changes may affect radiative forcing potential. By examining both absorption and scattering enhancements, could help in improving the accuracy of radiative forcing estimates and contribute to a better understanding of BC's role in climate change.

2. Methods

2.1. Model configurations

The spherical core shell model was investigated using Mie theory based PyMieScatt computational package.[20] The package implements Mie theory solutions for scattering and absorption of light by spherical particles. For our simulation study, we have considered internally mixed aerosol comprising of BC as core and OC as shell. The core shell configuration is plausible, as it could be produced by organic species condensing on the insoluble BC core once emitted during combustion or at a later stage during aging. Many of the previous studies have utilized the core shell model to investigate the interaction of light with BC aerosol. In this study, the optical cross-sections for different combinations of core and shell thickness were calculated. The core diameter was varied from 10 nm to 500 nm, and the shell thickness varied from 10 nm to 1000 nm. The wide range of core and shell thicknesses considered here is expected to include most of the observable size range of BC in the atmospheric aerosol.[10] The parameters considered for the simulation are compiled in Table S1. The study primarily discusses the optical properties of the coated BC at 550 nm, which is near the center of the visible light solar spectrum. Most of the optical measurements of aerosols are reported at 550 nm, so it makes simulation findings relevant and easily comparable with a large body of existing literature. However, the calculations were also performed for other wavelengths at 350 nm, 450 nm, and 650 nm, and compiled as supplementary information. The refractive index for BC and OC at 550 nm was taken as 1.95 + 0.79i and 1.65 + 0.08425i respectively.[21,22] Here the optical properties of OC is expected to represent mixture of brown carbon and purely scattering organic part.[22]

2.2. Enhancement calculations

Based on the calculated absorption cross-section, the E_{abs} for a given BC core was estimated using equation 1.

$$E_{abs} = \frac{C_{abs\_coated}}{C_{abs\_uncoated}} \qquad \text{eq. 1}$$

Here, C_{abs_coated} is the absorption cross-section of coated BC sphere and $C_{abs\_uncoated}$ is the absorption cross-section of uncoated BC sphere with core diameter remaining unchanged.

On similar lines the enhancement in scattering due to organic coatings was estimated in terms of E_{sca} using equation 2.

$$E_{sca} = \frac{C_{sca\_coated}}{C_{sca\_uncoated}} \qquad \text{eq. 2}$$

Here, C_{abs_coated} is the scattering cross-section of coated BC sphere and $C_{abs\_uncoated}$ is the scattering cross-section of uncoated BC sphere with core diameter remaining unchanged.

It’s important to note here that we are comparing coated BC with the uncoated BC for the same core size. However, in order to compare the effect of coating on the standalone optical properties of BC, it is pertinent to remove the contribution of the shell to absorption and scattering. Some studies have calculated shell contribution to absorption using linear algebraic methods.[15,23,24] However, they are approximations and do not assign an accurate contribution for the shell contribution to absorption. In the case of shell contribution to scattering properties, it's even trickier due to the stronger dependence of scattering properties on size as compared to absorption.[25] In order to accurately assess the standalone optical properties of BC, it is crucial to conduct further research and develop more precise methods for determining the shell contribution to both absorption and scattering. Nevertheless, for our case, we are going to compare the consolidated properties of coated and uncoated BC particles. This research will help us better understand the optical behavior of coated BC particles. Further in this study, the simulation is extended to compare coated BC with organic aerosol for different size configurations.

2.3 **Radiative forcing implications**

The effect of change in scattering potential due to coatings on BC aerosol can significantly impact its radiative forcing potential. The simple forcing efficiency (SFE) equation as stated in equation 3, proposed by Bond and Bergstrom (2006) provides a way to estimate the sensitivity of different model inputs.[21]

$$SFE = -\frac{1}{4} F_T \, \tau^2 (1 - A_c)[2(1 - R_s)^2 \beta . MSC - 4R_s . MAC] \qquad \text{eq. 3}$$

Here, F_T is the solar irradiance, τ is the atmospheric transmission, A_c is the cloud fraction, R_s is the surface albedo which will depend on the location of the aerosols, β is the backscatter fraction estimated using Mie calculation, and MSC and MAC are the mass scattering and mass absorption cross sections per gram, respectively. MSC and MAC can be estimated from the scattering and absorption cross-sections by normalizing them by their mass. The mass of coated and uncoated BC particle was estimated using the density provided in Table S1. The SFE equation can be framed to include wavelength dependence as stated in equation 4.

$$\frac{d(SFE)}{d\lambda} = -\frac{1}{4}\frac{dF_T(\lambda)}{d\lambda}\tau^2(\lambda)(1 - A_c)[2(1 - R_s)^2\beta(\lambda).MSC(\lambda) - 4R_s.MAC(\lambda)] \quad \text{eq. 4}$$

The above equation is used to investigate the effects of changes in absorption and scattering properties due to coatings on BC sphere at 550 nm. Here, $\frac{dF_T(\lambda)}{d\lambda}$ was obtained from the ASTM G173-03 Reference Spectra. The cloud fraction A_c was taken as 0.6 and surface albedo R_s was taken as 0.19.[22] The results of this study will provide valuable insights into the impact of BC coatings on radiative forcing potential, helping to improve climate models and predictions.

3. Results and discussion

3.1. Enhancement in absorption and scattering cross-section

The Mie theory calculated optical cross-sections were used to estimate E_{abs} and E_{sca} values using equation 1 and equation 2, respecively. The E_{abs} was typically greater than 1 for all non zero coating thicknesses considered in this study. The variation in E_{abs} for various combinations of core and coated thickness is shown in Figure 2. The increase in absorption cross-section for coated BC spheres is due to the lensing effect.[23] For a given core size, E_{abs} increased with the increase in coat thickness. The smallest core size and largest coating thickness configuration exhibited the highest E_{abs}. This suggests that the thickness of the coating on the BC sphere plays a significant role in enhancing absorption. Similar patterns in abosprtion enhancement dependence on coat thickness were predicted by Bond et al. (2006) using the Mie theory for the coated BC particles with the lognormal size distribution.[10] The absolute magnitude of the E_{abs} obtained in this study is very high compared to those reported from experimental observations. The values obtained in this study are theoretical estimations for change in absorption cross-section due to coating for a single sphere. However, for an experimental setup in real scenarios, the values of E_{abs} represent an ensemble of coated particles with complex morphologies. For more accurate resemblance to a real-world scenario, advanced simulation methods need to be explored, which is beyond the scope of the present investigation. However, the core shell model with Mie theory solutions is widely employed and it does provide valuable insights that can suggest accurate further studies.

Similar to absorption cross-section, the scattering cross-section of coated BC was also found to be higher than the uncoated BC sphere. The increase in scattering cross-section is due to an increase in physical dimension caused by organic coating which is predominantly scattering in nature. The increase in scattering cross-section with coating thickness indicates that the presence of a coating can enhance the scattering of radiation by BC particles in the atmosphere. The net effect was the E_{sca} being greater than 1 for non zero coating thickness. The data presented in Figure 3 illustrates the relationship between core and coating thickness on the E_{sca}. The variation pattern of E_{sca} is similar to that observed for E_{abs}. The E_{sca} typically increased with an increase in coat thickness for any given core size. Small core size and large coating thickness exhibited the highest E_{sca}. The E_{sca} was equal to 1 for no coating configuration across all the core sizes as expected. Compared to E_{abs}, the magnitude of E_{sca} was higher. Figure 3 illustrates how the ratio of E_{sca} to E_{abs} changes with core size and coating thickness. For most configurations of core size and coating thickness, this ratio was typically greater than 1. However, in cases of high core size combined with very low coating thickness, the enhancements in absorption and scattering were comparable. This indicates that the organic coating on BC can significantly impact scattering properties as compared to absorption properties. The overall change in absorption and scattering cross-section due to coatings provides valuable insights for understanding the optical behavior of coated BC particles in the atmosphere.

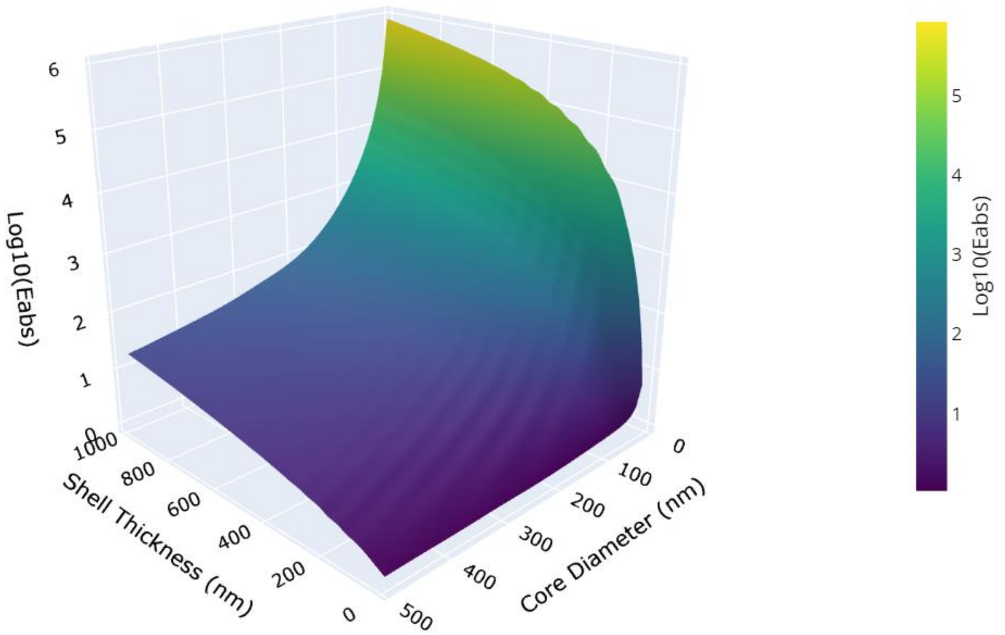

Figure 1 Enhancement in absorption cross-section due to organic coating on black carbon core

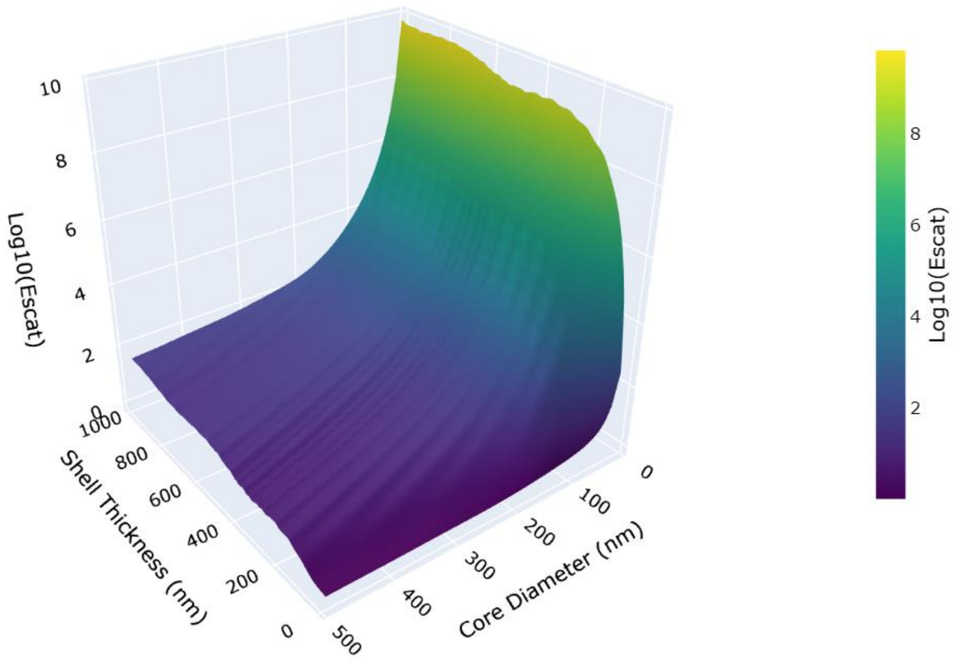

Figure 2 Enhancement in scattering cross-section due to organic coating on black carbon core

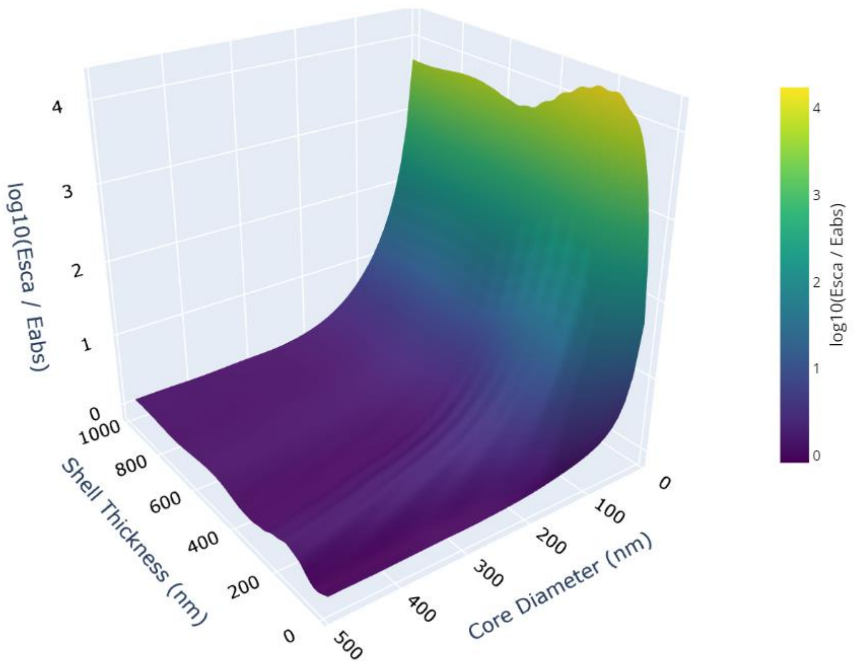

Figure 3 Comparision of enhancement in scattering and absorption cross-section due to organic coating on black carbon core

3.2. Comparison of coated BC sphere with OC sphere

In order to gain further insights into the optical properties of coated BC, its optical properties were compared with OC spheres having the same physical dimension. For this comparison, the ratio (R_{abs}) of absorption cross-section of the coated BC and that of OC with the same diameter was considered. Similarly, the ratio (R_{sca}) of scattering cross-section of the coated BC and that of pure OC with the same diameter was estimated. Coating thickness can be expressed in terms of coating factor (CF), which can be defined as the ratio of the diameter of the coated sphere to the uncoated sphere. Figure 4, shows the changes in R_{abs} with CF for different core BC diameters. It can be seen that as the coating thickness is increased, the R_{abs} approaches 1, irrespective of the core diameter. Figure 5, shows the changes in R_{sca} with CF for different core BC diameters. Similar to R_{abs}, R_{sca} also approaches 1 with the increase in CF. The results show that for a given BC core diameter, as the OC coating increases, the optical properties of the overall particle converge to those of the OC particle having the same dimension. It implies that the core BC gradually loses its optical significance as the OC coating increases. As seen from Figure 4, when the CF increases above 15, the absorption properties of coated BC and pure OC are almost alike. Similarly, the difference in scattering properties of coated BC and pure OC becomes insignificant when CF increases above 4. In real scenarios, such high coating factors could be plausible for very small BC core sizes. These findings suggest that as the OC coating on BC thickens, the optical properties of the two materials become more similar. Therefore, in practical applications where very thick organic coatings are observed, the optical significance of the core BC is expected to diminish significantly.

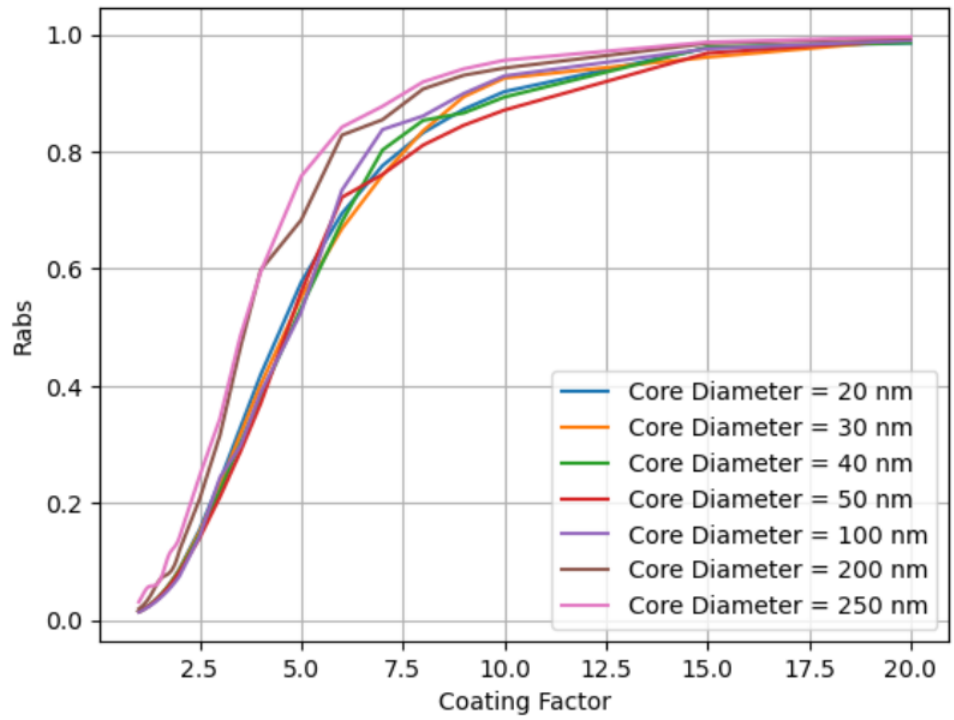

Figure 4 Effect of coating factor on ratio (R_{abs}) of absortion crosssection of organic carbon and coated black carbon

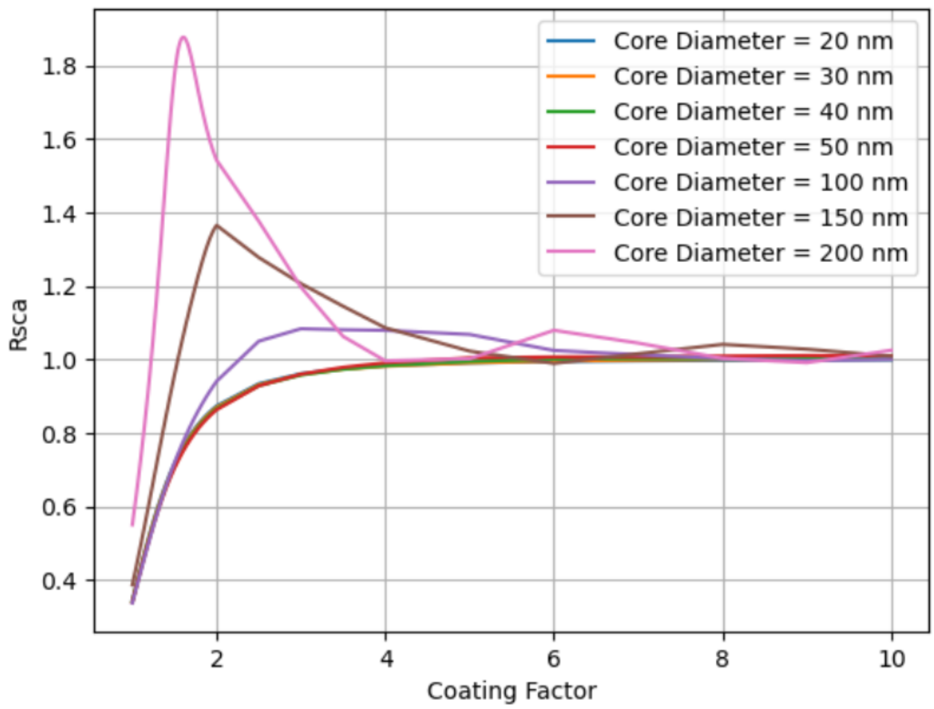

Figure 5 Effect of coating factor on ratio (R_{sca}) of scattering crosssection of organic carbon and coated black carbon

3.3. Implications on radiative forcing potential

The SFE at 550 nm was estimated for coated BC spheres for various configuration of core and coat thicknesses using equation 4. MAC and MSC required for the calculation of SFE were estimated using Mie calculations by normalizing the optical cross-section by their respective mass. Figures S3 and S4 illustrate the variations in MAC and MSC, respectively, as a function of the coating factor. MAC value for uncoated BC sphere (CF = 1) was found to be vary from 4.8 to 7.1 m^2 g^{-1} depending on the BC diameter. This range aligns with the MAC values reported for BC calculated using Mie theory.[21,27,28] MAC for uncoated BC obtained in this study was found to be increasing with the diameter. The trend is consistent with existing literature, as MAC is expected to increase with diameter up to certain diameter before decreasing in magnitude.[28,29] As it can be seen in Figure S3, the MAC for the coated BC sphere decreased with increase in organic coating thickness. Conversely, it was found that MSC increased with thicker coatings as seen in Figure S4. This behaviour can be attributed to the altered optical properties resulting from the introduction of organic coatings, which modifies the overall composition of the particle.

Figure 6 shows the variation of SFE with coating factor for different BC core diameters. It can be seen that as the thickness of the OC coating on BC increased, the SFE of the coated BC sphere decreased irrespective of core diameter. The resultant decrease in SFE is due to an increase in overall scattering cross-section caused by organic coating, as discussed in section 3.1. For the present model calculation, the SFE turns from positive to negative for CF of around 3 and above. This suggests that in real-world scenarios where BC particles are coated with thick organic material, their impact on radiative forcing may be less significant than previously thought. The net effect of the organic coating is to reduce the radiative forcing potential of the coated BC particle. Neglecting enhancement in scattering properties due to organic coatings can potentially lead to overestimation of radiative forcing capacity of coated BC. These findings have important implications for climate models and the understanding of aerosol effects on Earth's energy balance.

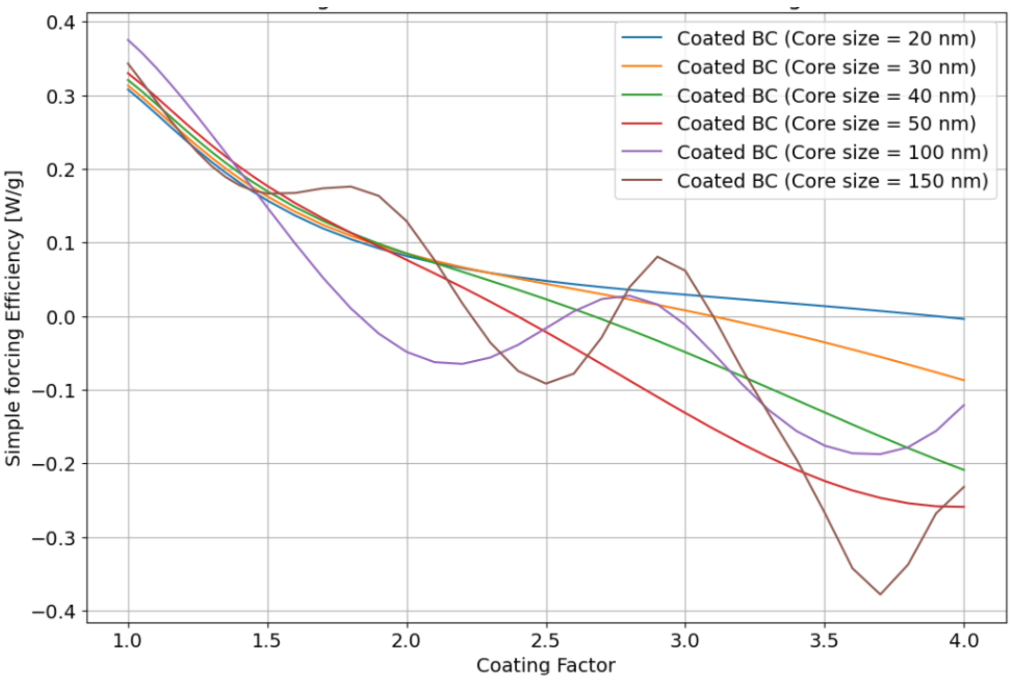

Figure 6 Variation in simple forcing efficiency with changes in coating factor for given black carbon core diameters

4. Conclusion

The study demonstrates the significant influence of coating thickness and core size on the optical properties of BC aerosols. The results reveal that both the absorption and scattering cross-sections of coated BC particles are enhanced compared to uncoated BC, primarily due to the lensing effect and the increased physical cross-section from the coating. The enhancement in scattering was more pronounced compared to absorption, highlighting the critical role of coating thickness in modulating the scattering properties of BC aerosols. The study finds that as the organic coating on BC increases, the optical properties of the coated BC particles begin to resemble those of pure OC particles with the same dimensions. This convergence suggests that thick coatings can significantly diminish the optical significance of the BC core, reducing its contribution to overall absorption and scattering. The SFE of coated BC was found to decrease with increasing coating thickness, which suggest less pronounced impact on radiative forcing in real-world scenarios where BC particles are coated with organic material. Overall, these insights emphasize the need for careful consideration of coating effects on the optical properties of BC for assessment of its impact on radiative forcing.

Author statement

T. D. Rathod: Conceptualization, Analysis and preparation of manuscript.

S. K. Sahu: Conceptualization, Supervision and preparation of manuscript.

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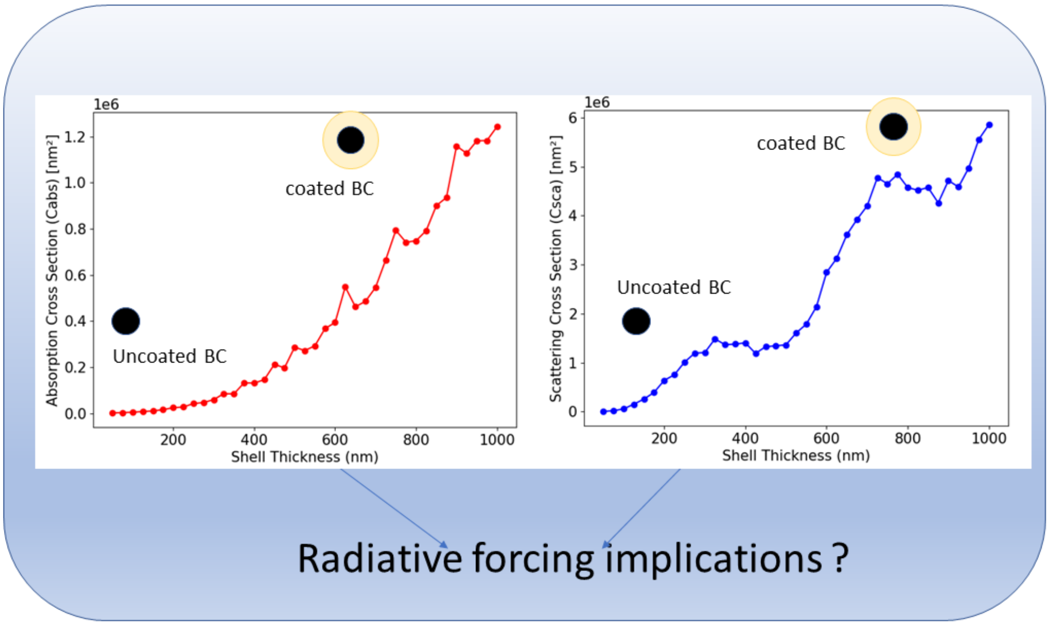
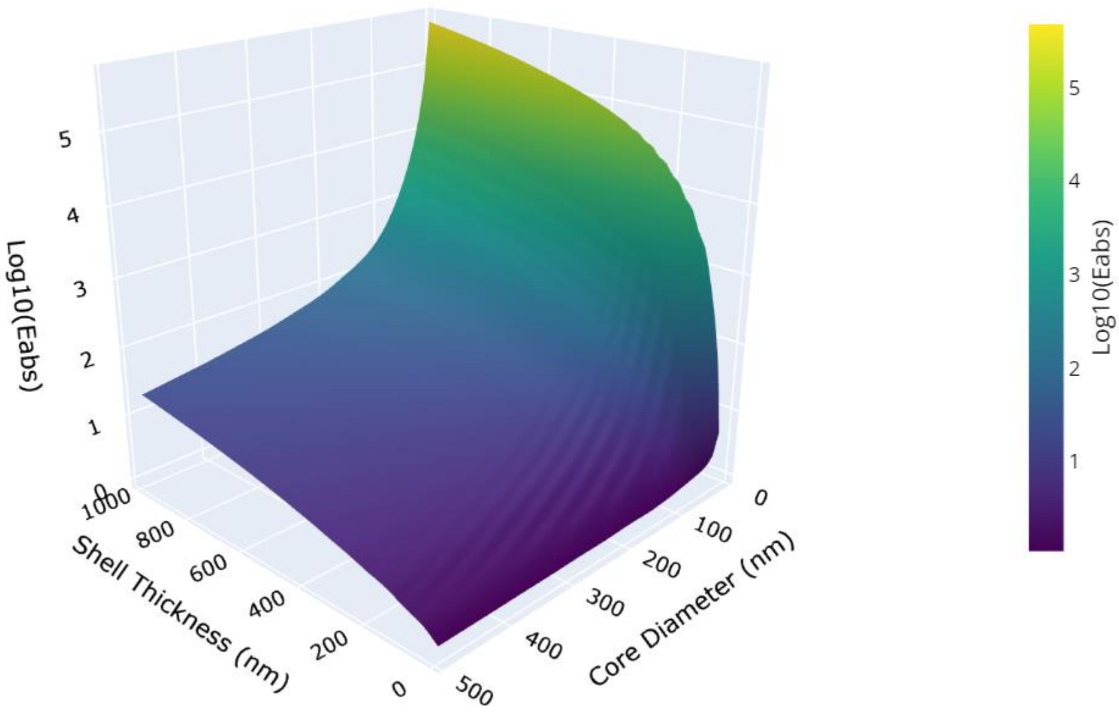


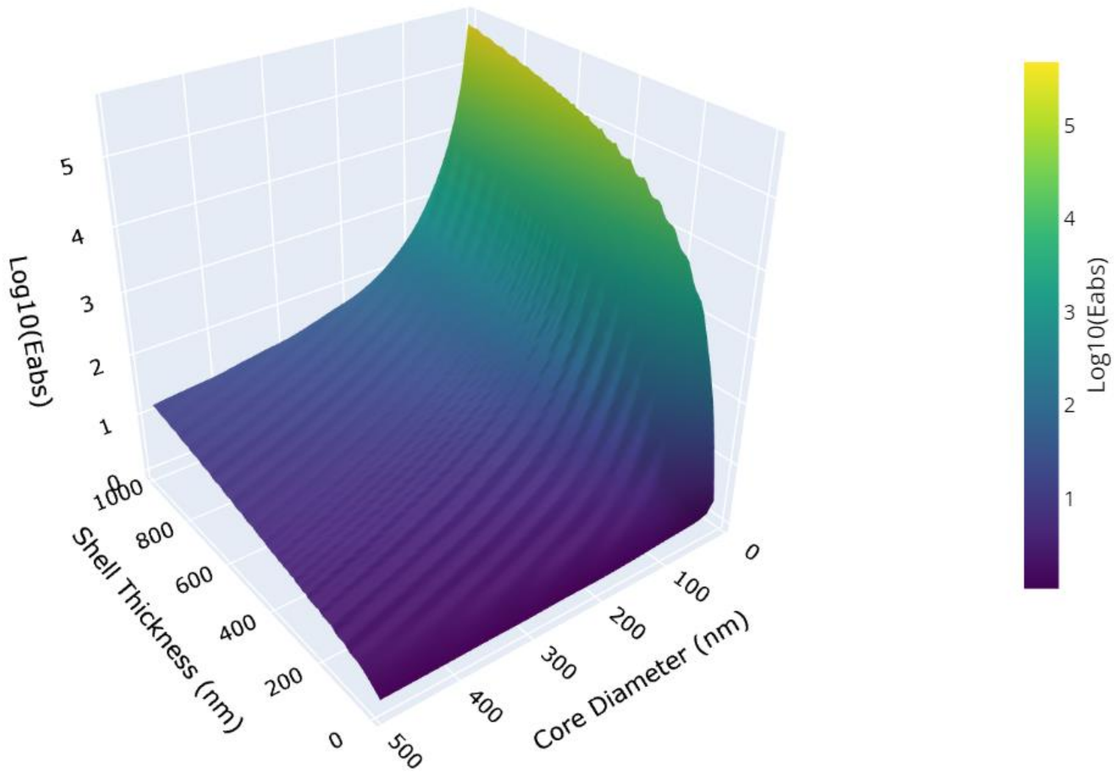
SUPPORTING INFORMATION

Table S1 List of parameters used for Mie theory calculations in this study

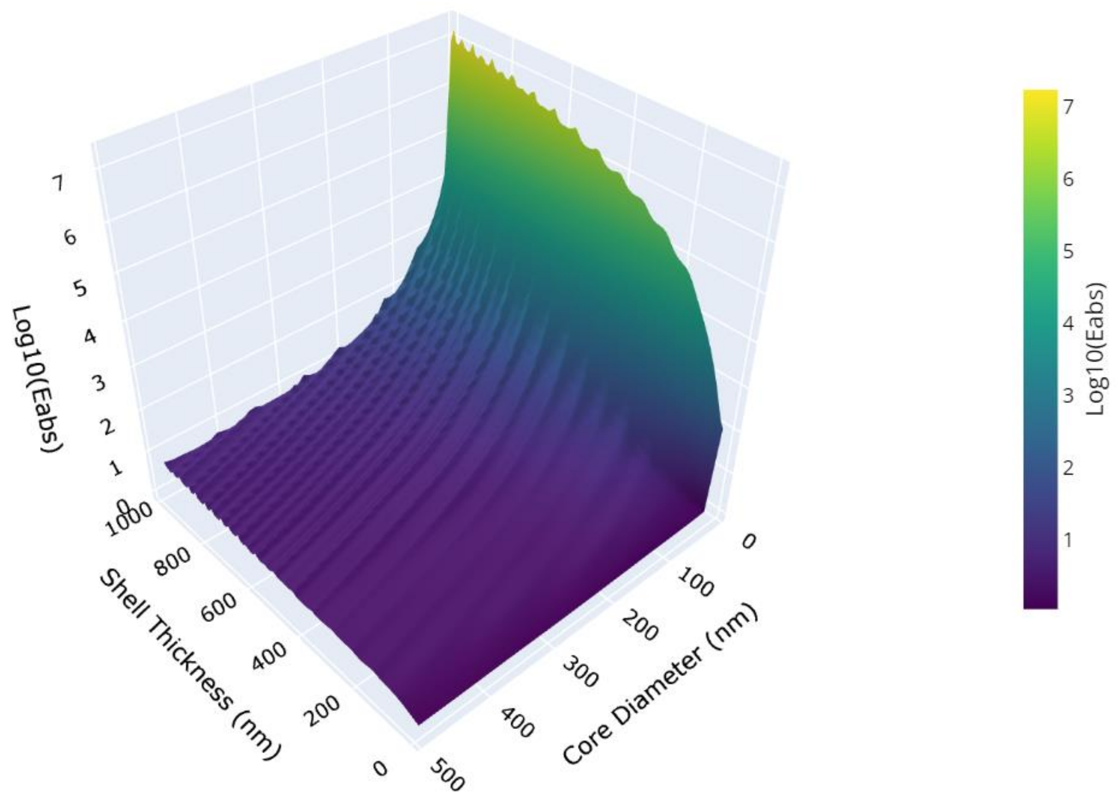
Parameters	Value	Reference
Core diameter range	10 nm to 250 nm	-
Shell thickness (ST) range	10 nm to 1000 nm	-
Wavelength	550 nm	-
Refractive Index (550 nm)		-
BC	1.95 + 0.79 i	Bond and Bergstrom, 2006.[21]
OC	1.65 + 0.08425 i	Rathod et al., 2017.[22]
Density		
BC	1.8 g cm^{-3}	Bond et al., 2006.[10]
OC	1.1 g cm^{-3}	Schkolnik et al., 2007.[26]



(a) 350 nm

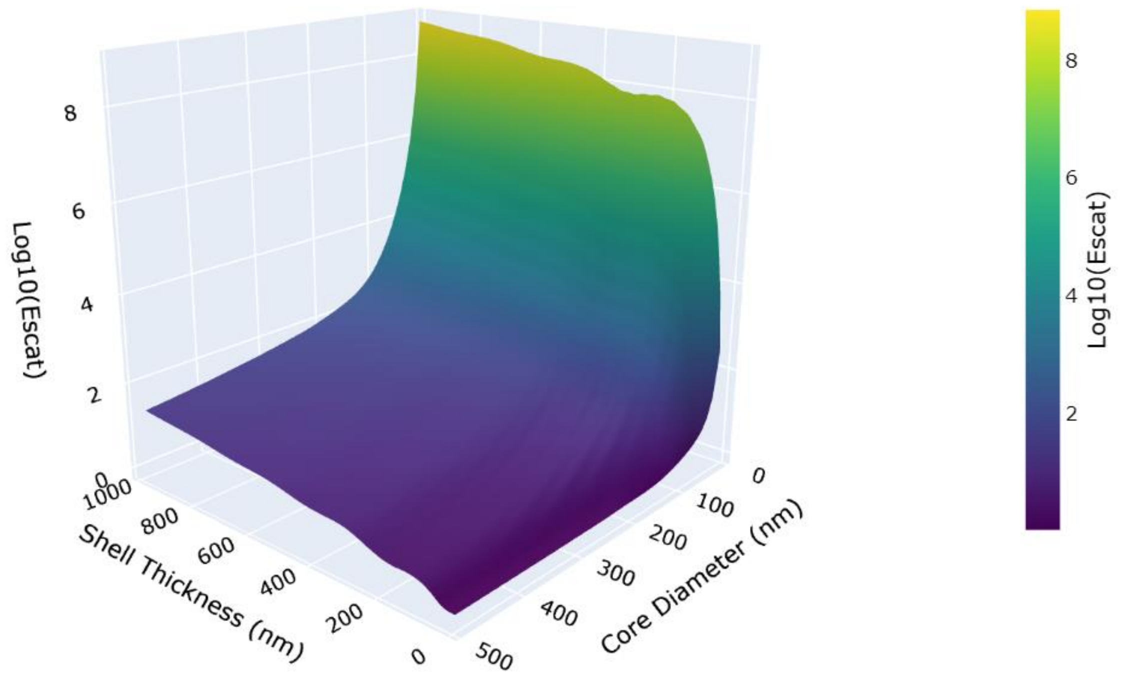


(b) 450 nm

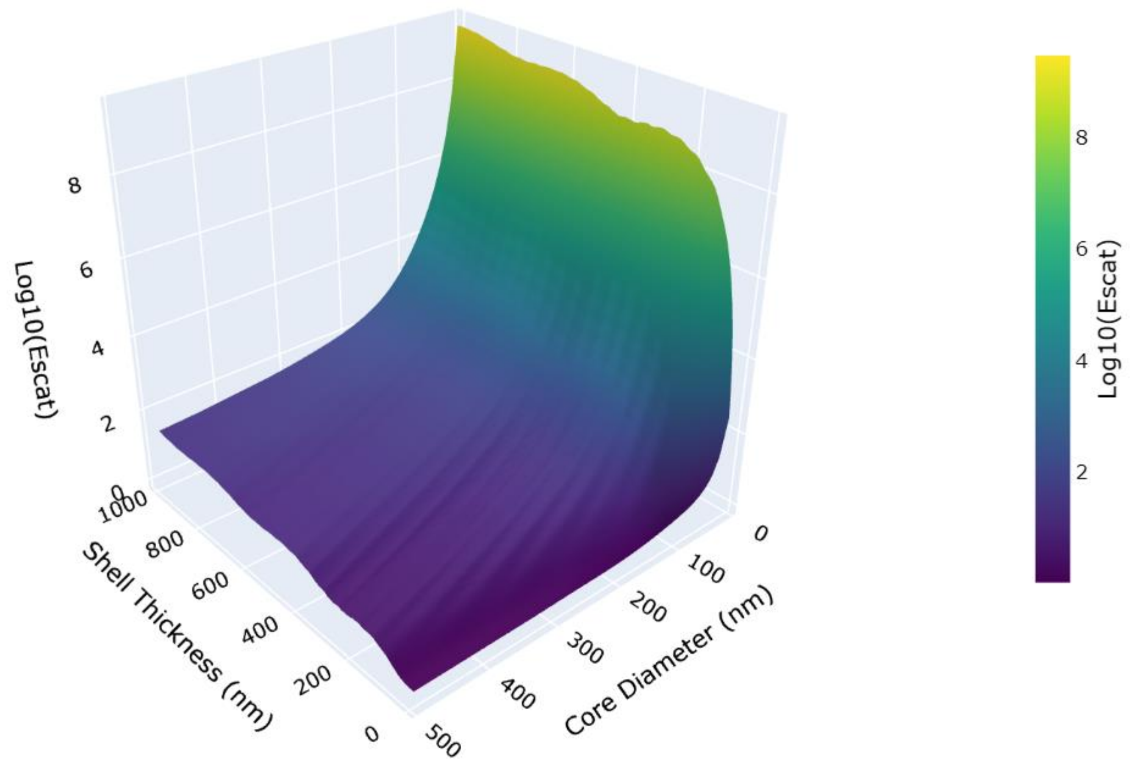


(c) 650 nm

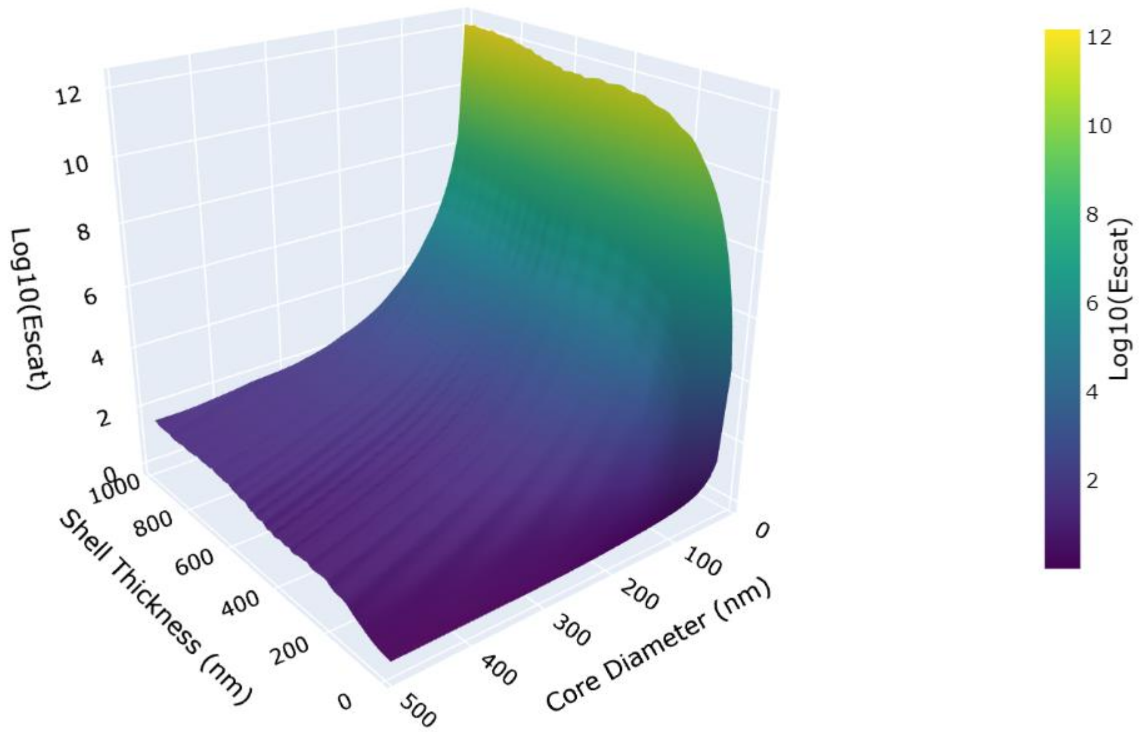
Fig. S1 Enhancement in absorption cross-section due to organic coating on black carbon core at different wavelengths



(a) 350 nm



(b) 450 nm



(c) 650 nm

Fig. S2 Enhancement in scattering cross-section due to organic coating on black carbon core at different wavelengths

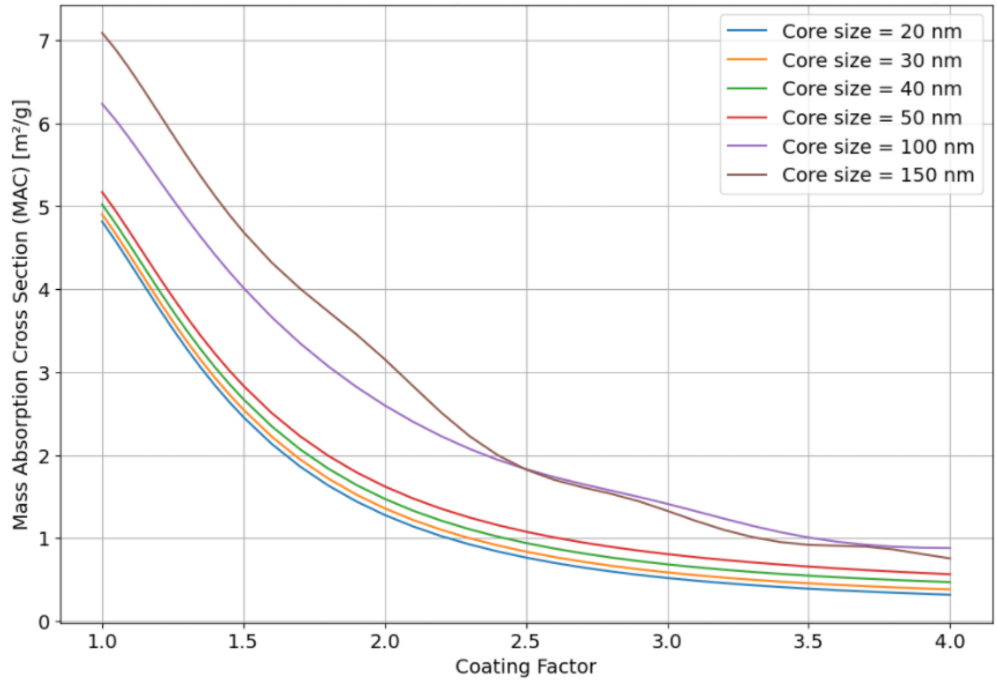


Figure S3 Variation in mass absorption cross-section with changes in coating factor for given black carbon core diameters

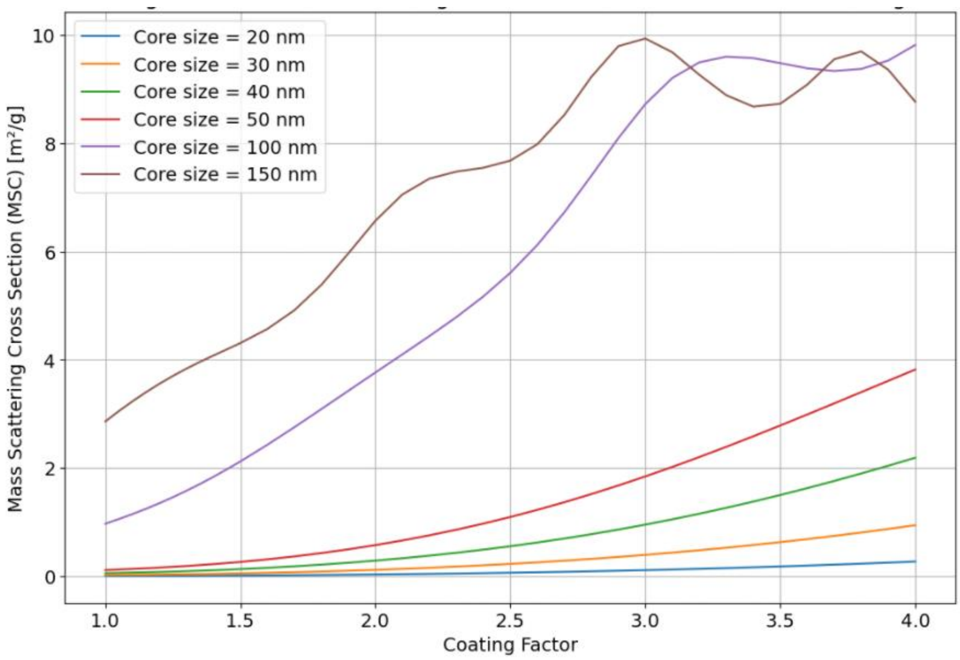


Figure S4 Variation in mass scattering cross-section with changes in coating factor for given black carbon core diameters