RESPONSE LETTER (EGUSPHERE-2024-1000)

Title: Retrieval of refractive index and water content for the coating materials of aged black carbon aerosol based on optical properties: a theoretical analysis

Dear Referee:

We have revised our manuscript based on the comments. The corrections and modifications have been included in the revised manuscript and the details are listed as follows. The responses are highlighted in blue font. The changes made in the revised manuscript are marked in red font.

Comments:

This study is a theoretical analysis about the influence of water content of coatings on complex refractive index of aged black carbon. Three models are used, which are closed-cell model, partially coated model and coated-aggregate model. Varied simulations were made since different values of the most noticeable parameters involved were selected. For instance, the influence of relative humidity at different wavelengths, or particle radius was investigated. Consequently, the paper may be accepted for publication in Atmospheric Chemistry and Physics after the introduction of some minor changes.

Response:

Thanks a lot for reviewing our manuscript and all these constructive comments. We have responded to the comments point by point and modified related descriptions in the revised manuscript.

The main inconvenience that authors should correct is the lack of citations in the results section. Some references should be introduced to indicate that these results are not isolated and can be compared with those from previous analyses. This comparison is useful to discuss the results of this research.

Response:

Thank you very much for the valuable comments and suggestions! Essential investigation results in previous research have been analyzed and compared with our results, we observed that our results agreed well with previous observation and simulation investigations, the related references have been cited in the revised manuscript along with necessary descriptions as follows:

"The retrieved CRIs also decrease with the RHs for different morphological models at both 532 and 1064 nm, Levoni et al. (1997) also revealed the downtrend of the retrieved refractive indices during the hygroscopic growth."

"With the increase of atmosphere humidity, the retrieval results are more and more reasonable, a larger amount of real part values range in 1.33-1.50, which are the refractive indices of water and sulfate respectively. Field observations conducted by Zhao et al. (2020) at areas with high concentrations of sulfate also showed that the retrieved CRIs ranged from about 1.37 to 1.51, and research by Zhang et al. (2013) showed that the real part of aerosol refractive index fluctuated around 1.50. When RHs are smaller than 30%, most of the retrieved real parts of CRIs for partially-coated and coated-aggregate models are larger than 1.50."

"With RHs increasing from 30% to 95%, the retrieved water contents gradually increase, due to the enhanced water absorbing capacity (Li et al., 2024; Bian et al., 2014)."

Moreover, a link with real situations, for instance, real cases with high concentration of black carbon and high relative humidity where this theory could be applied, should be encouraged. Similar examples should reveal that presented calculations respond to real conditions.

Response:

Thank you for the constructive comments!

Our study was designed and conducted in the framework of the single scattering albedo spectrometer, for the sake of improving the retrieval accuracy of aerosol refractive index based on the optical observation from the spectrometer. With the assistance of a single scattering albedo spectrometer, Xu et al. (2016) monitored the optical properties of $PM_{1.0}$ particles during the winter heating season in Beijing, which has high concentration of black carbon aerosols, the retrieved real part of aerosol refractive index based on Mie theory is 1.40±0.06. Zhou et al. (2020) measured the scattering, absorption, extinction coefficients and SSA at 532 nm using a humidifier cavity enhanced albedometer (H-CEA) in the laboratory, the relative humidity of H-CEA could increase from 10% to 88%. Tan et al. (2013) conducted field observation with a hygroscopic tandem differential mobility analyzer (H-TDMA) under high humidity (about 90%) at Pearl River Delta, which also has high BC concentration. In order to characterize the aerosol liquid water contents (ALWC) at North China Plain, Kuang et al. (2018) employed a three-wavelength humidified nephelometer system to measure optical properties at different RHs and showed that the measured ALWC was in good agreement with the calculated results of thermodynamic model. Therefore, according to the previous field and laboratory investigations, we conducted the numerical simulations of BC aerosols with the RH ranges from 30% to 95%.

We have added related descriptions in the revised manuscript as follows:

"Refractive index of aerosols, m=n+ki, can be determined from the scattering and absorption properties, the real part is related to the former and the imaginary part is related to the latter. Tan et al. (2013) conducted field observation with a hygroscopic tandem differential mobility analyzer (H-TDMA) under high humidity (about 90%) at Pearl River Delta, which also has a high BC concentration. In order to characterize the aerosol liquid water contents (ALWC) at North China Plain, Kuang et al. (2018) employed a three-wavelength humidified nephelometer system to measure optical properties at different RHs and they stressed that the measured ALWC was in good agreement with the calculated results of thermodynamic model. Zhou et al. (2020) measured the scattering, absorption, extinction coefficients and SSA at 532 nm using a humidifier cavity-enhanced albedometer (H-CEA) in the laboratory, the relative humidity of H-CEA could increase from 10% to 88%.

Zhao et al. (2020) measured absorption coefficient, scattering coefficient, size distribution and size-resolved mixing state of aerosols in eastern China, CRIs of BC containing and BC free aerosols were investigated separately based on Mie theory for sphere and core-shell structure, the corresponding CRIs were 1.67±0.67i and 1.37-1.51. Xu et al. (2016) monitored the optical properties of PM1.0 particles during the winter heating season in Beijing, which has high concentration of black carbon aerosols, the retrieved real part of aerosol refractive index based on Mie theory is 1.40±0.06."

Minor remarks.

Figure-1 caption should be revised and corresponding labels should be introduced.

Response:

Thank you very much for the comments, we have modified the caption of Figure 1 in the revised manuscript as follows:

"Figure 1: Morphological models of coated black carbon with 150 monomers."

Dots after "Figure" in figure captions should be suppressed (from Figure 2 to Figure 9).

Response:

Thank you very much for pointing this out, we have suppressed the dots in the revised manuscript as follows:

"Figure 2: Comparison of preset and retrieved real parts of CRIs based on different optical properties of coated-aggregate BC models with D_f=2.60 and V_f=0.10 at different RHs."

"Figure 3: Comparison of preset and retrieved real part of CRIs of coated BC aerosols at different RHs and wavelengths. (a) Closed-cell model; (b) Partially-coated model; (c) Coated-aggregate model."

"Figure 4: Retrieved real parts of CRIs of coated-aggregate models with different fractal dimensions and BC volume fractions during the hygroscopic process. (Retrieved results are shown in points and fitted in lines)"

"Figure 5: Retrieved real parts of CRIs of partially-coated models with different fractal dimensions and BC volume fractions during the hygroscopic process. (Retrieved results are shown in points and fitted in lines)"

“Figure 6: Retrieved real parts of CRIs of closed-cell model with different fractal dimensions and BC volume fractions during the hygroscopic process. (Retrieved results are shown in points and fitted in lines)”

“Figure 7: Retrieved refractive indices of closed-cell model, partially-coated mode, and coated-aggregate model under different RHs.”

“Figure 8: Retrieved water content in coatings for coated-aggregate models with different D_f and V_f during the hygroscopic growth. The dotted lines are the 1:1 dividing lines.”

“Figure 9: Retrieved water content in coatings for partially-coated models with different D_f and V_f during the hygroscopic growth. The dotted lines are the 1:1 dividing lines.”

Some dots after “Figure” and figure number in text should be suppressed. For instance, L. 188, L. 208, L. 220, L. 239.

Response:

Thanks for the suggestion, we have checked the full text and suppressed the dots in the revised manuscript as follows:

“Figure 3 illustrates the retrieved real part of CRIs of coated BC with closed-cell, partially-coated, and coated-aggregate models with V_f=0.10 at different RHs and wavelengths.”

“Figure 4 describes the variation of retrieved real parts of CRIs of coated-aggregate models with different BC core sizes, BC volume fractions and fractal dimensions.”

“Figure 5 shows the variation of retrieved real parts of CRIs of partially-coated models at different RHs. Similar to coated-aggregate modes, the retrieved real parts of CRIs also increase at first and then decrease, but the maximum values are significantly affected by relative humidity.”

“Figure 7 illustrates the comparisons of real parts of optical retrieved CRIs and the corresponding preset values during the hygroscopic process, all coated particles with different fractal dimensions, BC volume fractions, and BC core sizes are considered.”

L. 214. Introduce one space between “(b)” and “and”.

Response:

Thanks a lot for the comments, we have modified this in the revised manuscript as follows:

“As shown in Fig. 4(b) and 4(d), the retrieved real parts of CRIs are smaller for larger BC cores with larger fractal dimensions.”

References should follow the journal style.

Response:

Thank you very much for the comments. We have modified the references according to the style of Atmospheric Chemistry and Physics.

Furthermore, other detailed revisions are listed below.

LOCATION	**REVISED MANUSCRIPT**	**ORIGINAL MANUSCRIPT**
Abstract	performs best	has the best performance
Introduction paragraph 2	increased slowly	increase slowly
Introduction paragraph 3	cross-sections	cross sections
Section 3.1, paragraph 1	Figure 2	Fig. 2.
Section 3.1, paragraph 2	decrease	decreases
Section 3.3, paragraph 1	Figure 8	Fig. 8.
	Figure 9	Fig. 9.

References

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