Response to the comments of the Reviewer #2

(The responses are highlighted in blue)

First of all, we would like to thank the three anonymous reviewers for their thoughtful review and valuable comments to the manuscript. In the revision, we have accommodated all the suggested changes into consideration and revised the manuscript accordingly. All changes are highlighted in the revised manuscript in **BLUE** in the revision.

In this response, the questions and comments of reviewers are in black font, and responses are highlighted in **BLUE**. The changes made in the revised manuscript are marked in RED font.

Black carbon (BC) particles mixing with brown carbon (BrC) coatings are simulated by two morphologies, including: thinly and thickly coated states. Light absorption properties of BC-containing particles are calculated using the superposition T-matrix method. The sensitivity of the imaginary part of BrC refractive index on the light absorption is investigated for the realizations with the aerosol ensembles. The authors showed some interesting results, but the effects they presented are not clear and the simulations are not validated by the measurements. While you revise the paper, please take the following into consideration.

Response: Thanks for your valuable comments. Please see the point-to-point response below to the concerns raised for this manuscript.

1. In this study, the absorption enhancement, lensing effect, blocking effect, sunglasses effect, and strengthening effect are discussed, but they are confused. In the previous studies, e.g. Liu (2017), "lensing effect is that the addition of non-black-carbon materials to black-carbon particles may enhance the particles' light absorption by 50 to 60% by refracting and reflecting light". The clear definitions of these effects are important, because brown coating also absorbs solar radiation itself. In Equation (3), the effect of brown coating on absorption enhancement is not considered, and may generate an unreasonable E_{abs} value, such as 5.4 (Line 5 in abstract). It would more appropriate to compare the absorption of BC coated by BrC with an external mixture of BrC and BC, rather than bare BC alone. In Equation (4), how to calculate far-field results of C_{abs_BrC} (total_size) and C_{abs_BrC} (bare_size) in the BC-BrC mixtures, and do you considered the complex morphologies of BrC in 'total size' cases? In bare BC, the BrC coating may be not exist. The 'lensing effect' is widely used in the climate studies, thus, Equation (5) may be potentially misleading. It is necessary to clearly explain these

effects.

Referece: Liu, Dantong, et al. "Black-carbon absorption enhancement in the atmosphere determined by particle mixing state." Nature Geoscience 10.3 (2017): 184.

Response: Thanks for your comments and valuable suggestions. We agree that the definition of $E_{abs\_lensing}$ in the previous version of the manuscript is not clear. The sunglass effect, BC absorption enhancement, BrC enhancement, and the lensing effects should be clearly explained.

Usually, "lensing effect is widely used in the climate studies", and they contribute all absorption enhancement of internally mixed BC to the "lensing effect" of coatings. From a physics point of view, we think lensing effects is suitable for non-absorbing coatings, the definition is as follows:

$$E_{lensing} = \frac{C_{abs\_coated\_non\text{-}absorbing}}{C_{abs\_bare}}$$

The lensing effect defined in Equation (5) of previous version is the comparison of the absorption of BC coated by BrC with an external mixture of BrC and BC. It is resulted from the interaction of lensing effect and sunglass effect we defined.

Liu et al. (2017a) defined the lensing effect as the absorption enhanced by addition of non-black carbon. However, from the physical point of view, for BC with BrC coatings, the definition may be not clear, and it can be confused with E_{abs}. Therefore, we redefined the lensing effect as the absorption enhanced by addition of non-absorbing coatings in the revised manuscript. In addition, we assume that the lensing effect of BC with absorbing coatings is the same as those with non-absorbing coatings. We believe this is a reasonable assumption since the BrC and nonabsorbing coating have a similar value of real part of refractive index.

We agree that it is a valuable suggestion to compare the absorption of BC coated by BrC with an external mixture of BC and BrC. Actually, the $E_{abs\_lensing}$ defined in the previous version of the manuscript has compared the absorption of BC coated by BrC with an external mixture of BC and BrC. However, the comparison is not in a direct manner. Therefore, we define a new parameter, $\boldsymbol{E_{abs\_internal}}$, to represent the ratio between the absorption of BC coated by BrC and an external mixture of BC and BrC.

However, in the revised manuscript, the E_{abs} was compared with measurement results in the literatures, while the $E_{abs\_internal}$ was not. It is because the E_{abs} were commonly measured while usually $E_{abs\_internal}$ results were not available by measurements.

The absorption of BrC shell is calculated by the absorption of BrC that is with the same shape as the coated BC subtracting the absorption of BrC that is with the same shape as the bare BC, as shown in Equation (4) of previous version. The calculation of BrC shell is illustrated in Figure 1 in this response (Figure S1 in the revised manuscript). In this process, we assume that the absorption of BrC with the same shape as the coated BC is identical as the external mixture of BrC with the same shape as bare BC and BrC shell. We must clarify that this process neglects the blocking effect and lensing effect of outer BrC shell on the internal BrC. However, as the BrC absorption is significantly less than the BC absorption with identical shape, the absorption caused by the blocking effect and lensing effect of outer BrC on the internal BrC is relative small compared with the BC absorption. Therefore, it is reasonable to make some simplifications.

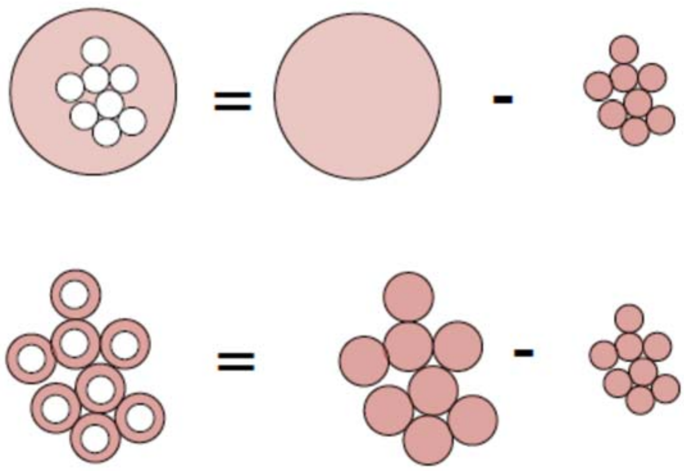


Figure 1 Calculation of the absorption of BrC shell.

Therefore, we indeed considered the complex morphologies of BrC in 'total size' cases. We think "$C_{abs\_BrC\_(total\_size)}$" and "$C_{abs\_BrC\_(bare\_size)}$" may be a little confusing. For this reason, we have changed "$C_{abs\_BrC\_(total\_size)}$" and "$C_{abs\_BrC\_(bare\_size)}$" into "$C_{abs\_BrC\_(coated\ shape)}$" and "$C_{abs\_BrC\_(bare\ shape)}$". It is true that the BrC coating don't exist in the case of bare BC. However, Equation (4) is aimed to calculate the BrC shell of absorption.

We clearly defined the sunglass effect in the revised manuscript. We contribute E_{abs} of BC with BrC coatings to lensing effect, BrC absorption enhancement and sunglass effect. Therefore:

$$E_{Sunglass} = -\frac{C_{abs\_\mathrm{coated}} - C_{abs\_BrC\_shell} - C_{abs\_\mathrm{non-absorbing}}}{C_{abs\_bare}}$$

The negative sign represents that the sunglass effect can cause the decrease of total absorption. Combining all the definitions, we can easily obtain the relation that the absorption of BC coated with BrC is less than that of an external mixture of BrC and BC when $E_{Sunglass} > E_{abs\_lensing} - 1$.

One may confused about the calculation of coated BC with the BrC shell. Coated BC was calculated directly using MSTM. The calculation of the absorption caused by the Sunglass effect is shown in Figure 2 of this response.

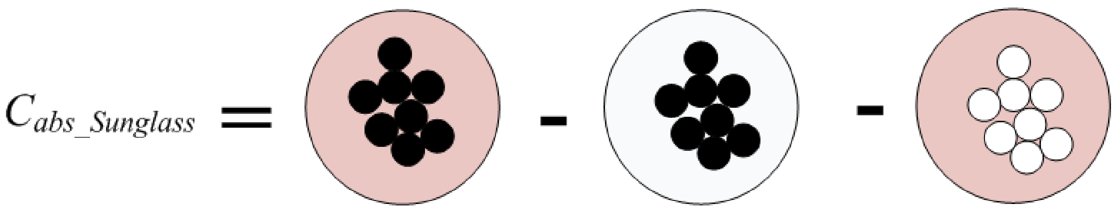


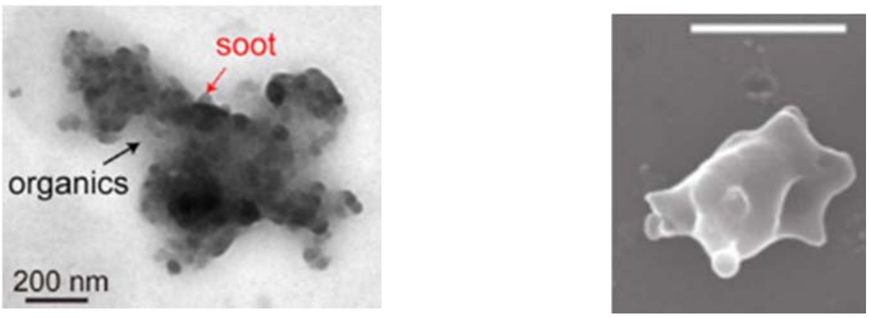
Figure 2 Calculation of the absorption caused by the sunglass effect. $C_{abs\_Sunglass}$ represents the absorption cross-section caused by the sunglass effect.

2. Line25-27, Page 2. "Nevertheless, in the atmosphere, there is a type of organic carbon that absorbs the radiation in the range of the ultraviolet and visible spectra, which is known well as brown carbon (BrC); BC can also be mixed with BrC." Please give the references about the morphologies and mixing states of BC-BrC mixtures to support this simulation.

Response: Thanks for your comments. As the BC ages in the atmosphere, BC becomes more compact and other materials (such as sulfate and organic substances) can condense onto the particles. The organic coating can be POA or SOA. BC can be embedded in a non-BC shell (Wang et al., 2017) (China et al., 2013). When non-BC fraction is low, BC can still present fractal structure, as demonstrated in the Figure 1 (a-3) of Wang et al. (2017) (Figure 3a in this response). As BC is further coated, BC becomes more compact and the coating shell becomes more spherical (Lewis et al., 2009), as shown in the Figure 3 of China et al. (2013) (Figure 3b in this response).

Eventually, BC aggregates are collapsed into more compact and spherical clusters when fully engulfed in coating material (referred as thickly-coated BC in this study) (Zhang et al., 2008b). We added the references about the morphologies and mixing states of BC in the introduction of revised manuscript:

"Freshly emitted BC commonly presents fractal structures. As the BC ages in the atmosphere, BC becomes more compact and OC materials can condense onto the particles. Therefore, BC can be embedded in an OC shell (China et al., 2013a; Wang et al., 2017). When non-BC fraction is low, BC can still present near fractal structure (referred as thinly coated in this study) (Wang et al., 2017). As BC is further coated, BC aggregates are collapsed into more compact and spherical clusters when fully engulfed in coating material (referred as thickly-coated BC in this study) (Zhang et al., 2008b)."



(a) (b)

Figure 3 Typical morphologies of coated BC. (a) Thinly-coated BC with fractal structure (Wang et al., 2017); (b) Coated BC with more spherical coating (China et al., 2013).

3. In Section 3.2 Bulk radiative properties, it is suggested to estimate the absorption enhancements of BC aerosols by the mass absorption cross section (MAC) rather than the cross section (Cabs), because of the normalization of BC mass. Moreover, the simulations of MAC can be validated by the previous measurements.

Response: Thanks for your valuable suggestions. The calculation of E_{abs} was not estimated by mass absorption cross section (MAC), the reason is that the absorption enhancement is defined as the amplification of total absorption but not the amplification of mass absorption. Although many studies used the MAC to estimate E_{abs}, MAC is calculated as the mass absorption per unit mass of BC but not per unit mass of total BC-containing aerosols. Therefore, the MAC can be calculated using:

$$MAC_{coated/bare} = C_{abs\_coated/bare} / m_{BC}$$

Where m_{BC} represents the mass of BC.

The E_{abs} can be expressed as:

$$E_{abs} = \frac{MAC\_abs\_coated}{MAC\_abs\_bare} = \frac{C_{abs\_coated}}{C_{abs\_bare}}$$

This is consistent with the definition of E_{abs} in our work.

We do agree that our calculations should be validated by measured results. We compared the modeled E_{abs} and MAC with the results of Liu et al. (Liu et al., 2015), and the comparison is supplemented in support information, as shown in the Figure S1. The measured E_{abs} values were estimated from Figure 1 of Liu et al. (Liu et al., 2015). In this work, we assume the E_{abs} and MAC at λ=700 nm and 781 nm do not deviate largely. The comparison of measured and modeled MAC is shown in Figure S7 and Figure S8 in the revised manuscript. When BC mass density is assumed to be 1.5g/cm^3, the calculated MAC and E_{abs} are relatively well in agreement with the measurements (see Figure 5 in this response or Figures S8-S9 in the revised manuscript). As for why assuming BC mass density to be 1.5g/cm^3, we have explained the reason in the revised manuscript (page 11 in the revised manuscript):

"In this work, we assume ambient BC mainly composed of primary organic matter with a low degree of oxidation. Based on the study of Nakao et al. (2013), an OC mass density range of 1-1.2 g/cm^3 has been used by Liu et al. (2017). ρ_{BrC} = 1.1 g/cm^3 is assumed in this work. For the BC mass density, the study of Horvath (1993) gives values of ρ_{BC} = 0.625 g/cm^3 and ρ = 1.125 g/cm^3. However, Fuller et al. (1999a) pointed out that the values may be not representative for BC in the atmosphere. Medalia and Richards (1972) and Janzen (1980) suggested ρ_{BC} in the range of 1.8-1.9 g/cm^3, while Bergstrom (1972) found that the ρ_{BC} value of 1.9-2.1 g/cm^3. Bond and Bergstrom (2006) suggested to use a value of 1.8 g/cm^3. Figure S8 compares the computations with measurements by assuming ρ_{BC} = 1.8 $g/cm^{3.}$ We assume that E_{abs} and MAC at λ = 0.7 *um* do not deviate largely with those at λ = 0.781 *um*. Modeled E_{abs} at λ = 0.7 *um* agrees well with the measurements. Although E_{abs} at λ = 0.404 *um* seems to be relatively higher than the measurements, it dose not deviate largely with the measurements. However, modeled MAC is a little smaller than the measured MAC. Similar results were obtained for bare BC ((Kahnert, 2010b), (Liu and Mishchenko, 2005)) . Therefore, ρ_{BC} = 1.8g/cm^3 may be a little high for estimation of MAC.

Bond and Bergstrom (2006) concluded that MAC value of 7.5 ± 1.2 m^2/g for bare BC can be assumed at λ = 0.55*um* by reviewing 21 publications of MAC measurements. However, our calculated MAC of 6.02-6.2 m^2/g (see Table 2) at λ =0.532 *um* lies below the range of MAC values suggested by Bond and Bergstrom (2006). Similar conclusions were drawn by Kahnert (2010b) and Liu and Mishchenko (2005). However, our calculated MAC agrees well with the calculated MAC of 6.0 ± 0.1 m^2/g by Kahnert (2010b) at λ = 0.55 *um*. As MAC depends significantly on BC mass density, to agree with measurements, Liu and Mishchenko (2005) used ρ_{BC} = *1.0 g/cm³*. However, as pointed by Kahnert (2010b), the measured MAC and modeled MAC were not at the same wavelength, therefore leading to too low retrived ρ_{BC}. To raise the computed MAC values to the average observed value of MAC = (7.5 ± 1.2) m^2/g, ρ_{BC} = 1.3-1.4 g/cm^3 was suggested by Kahnert (2010b). However, this ρ_{BC} value is rather drastic smaller than the value suggested by Bond and Bergstrom (2006). Therefore, Kahnert (2010b) suggested to assume ρ_{BC} = 1.5 ~ 1.7 g/cm^3 to raise the computational MAC results to the lower bound of the observations. By assuming ρ_{BC} = 1.5 g/cm^3, the comparison of modeled MAC and E_{abs} with measurements is shown in Figure S9. Overall, the modeled MAC and E_{abs} agree relatively well with the measurement by assuming ρ_{BC} = 1.5 g/cm^3. Therefore, ρ_{BC} = 1.5 g/cm^3 is assumed in this study."

Some E_{abs} values (eg. E_{abs}=5.4, see Figure 11 in the revised manuscript) are scarcely observed in the atmosphere. The most likely reason is the results for BC with thicker BrC coating is unavailable at ultraviolet wavelength. For sensitivity analysis, the BC volume fraction is independent on the BC size. Therefore, to gain E_{abs}=5.4, the BC volume fraction should be 5% for all BC. However, in the atmosphere, not all BC is thickly coated. Limited measurements were conducted at ultraviolet wavelength, therefore the measurements are not available for all circumstance. In fact, our calculated results are in general agreement with the measurements in the visible wavelengths. The E_{abs} can reach approximately 3.96 at λ =532*nm* when f_{BC}=5% in this work, which is consistent with the reported E_{abs} value of 2.6-4.0 at Beijing for λ =470*nm*, China (Xu et al., 2016).

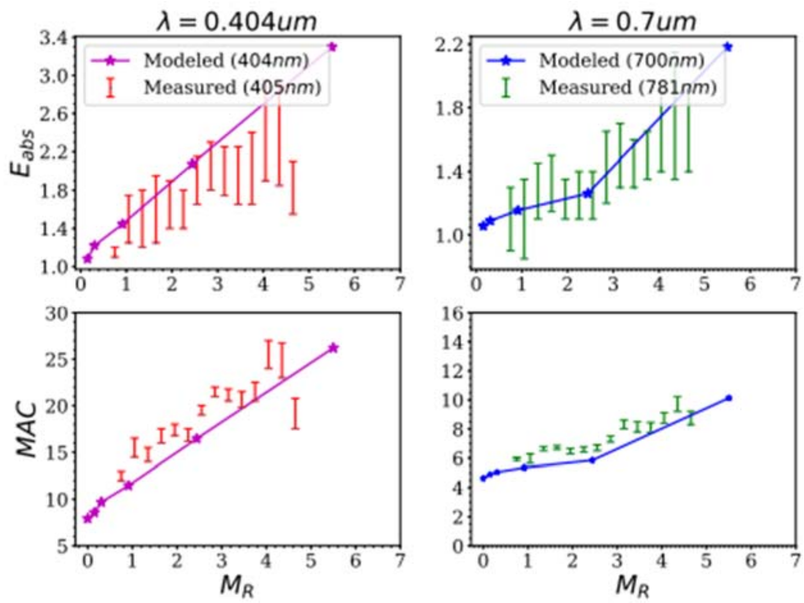


Figure 4 (Figure S8 in the revised manuscript) Comparison of modeled E_{abs} and MAC with measurements, BC mass density is assumed to be 1.8g/cm3, and the measured results are derived from the study of Liu et al. (2015).

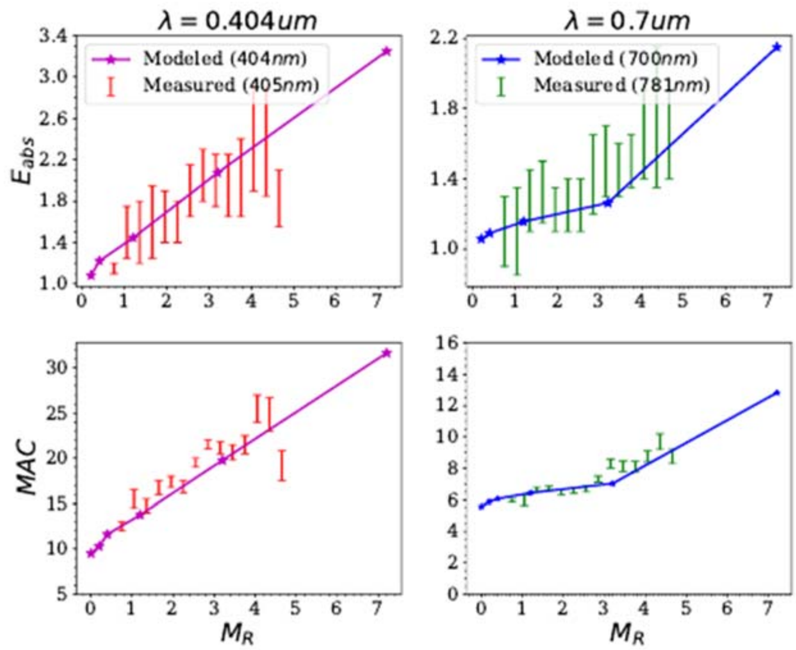


Figure 5 (Figure S9 in the revised manuscript) Comparison of modeled Eabs and MAC with measurements, BC mass density is assumed to be 1.5g/cm3, and the measured results are derived from the study of Liu et al. (2015).

Table 1 (Table 2 in the revised manuscript) MAC (m^2/g) for bare BC at different D_f (r_g = 0.06 um, σ_g = 1.5)

λ(nm)	D_f=1.8	D_f=2.2	D_f=2.6
350	9.30	9.03	8.48
404	8.14	7/95	7.60
532	6.20	6.11	6.02
700	4.68	4.64	4.65

4. In the abstract, please define the 'C_{abs}', 'K_{BrC}' before use them.

Response: Thanks for pointing it out. We have corrected it in the revised manuscript.

5. In Figure 6, 7 and 11, the range of color bar is suggested to be unified.

Response: Thanks for your suggestions. The range of color bar has been unified.

References

Adachi, K., Chung, S. H., and Buseck, P. R.: Shapes of soot aerosol particles and implications for their effects on climate, J Geophys Res-Atmos, 115, 2010.
Bergstrom, R. W.: Predictions of the spectral absorption and extinction coefficients of an urban air pollution aerosol model, Atmospheric Environment (1967), 6, 247 – 258, 1972.
Bond, T. C. and Bergstrom, R. W.: Light absorption by carbonaceous particles: An investigative review, Aerosol Science and Technology, 40, 27–67, 2006.
China, S., Mazzoleni, C., Gorkowski, K., Aiken, A. C., and Dubey, M. K.: Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles, Nat Commun, 4, 2013.
Fuller, K. A., Malm, W. C., and Kreidenweis, S. M.: Effects of mixing on extinction by carbonaceous particles, Journal of Geophysical Research-Atmospheres, 104, 15 941–15 954, 1999a.
Horvath, H.: Atmospheric Light-Absorption - a Review, Atmospheric Environment Part a-General Topics, 27, 293–317, 1993.
Janzen, J.: Extinction of Light by Highly Nonspherical Strongly Absorbing Colloidal Particles - Spectrophotometric Determination of Volume Distributions for Carbon-Blacks, Applied Optics, 19, 2977–2985, 1980.
Kahnert, M.: On the Discrepancy between Modeled and Measured Mass Absorption Cross Sections of Light Absorbing Carbon Aerosols, Aerosol Science and Technology,

44, 453–460, <GotoISI>://WOS:000277436300006, 2010b.
Lewis, K. A., Arnott, W. P., Moosmuller, H., Chakrabarty, R. K., Carrico, C. M., Kreidenweis, S. M., Day, D. E., Malm, W. C., Laskin, A., Jimenez, J. L., Ulbrich, I. M., Huffman, J. A., Onasch, T. B., Trimborn, A., Liu, L., and Mishchenko, M. I.: Reduction in biomass burning aerosol light absorption upon humidification: roles of inorganically-induced hygroscopicity, particle collapse, and photoacoustic heat and mass transfer, Atmos Chem Phys, 9, 8949-8966, 2009.
Liu, D. T., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S. F., Williams, P. I., Ting, Y. C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G., Coe, H., and Allan, J. D.: Black-carbon absorption enhancement in the atmosphere determined by particle mixing state, Nature Geoscience, 10, 184–U132, 2017.
Liu, L. and Mishchenko, M. I.: Effects of aggregation on scattering and radiative properties of soot aerosols, Journal of Geophysical ResearchAtmospheres, 110, 2005.
Medalia, A. I. and Richards, L. W.: Tinting Strength of Carbon-Black, Journal of Colloid and Interface Science, 40, 233–&, 1972.
Nakao, S., Tang, P., Tang, X. C., Clark, C. H., Qi, L., Seo, E., Asa-Awuku, A., and Cocker, D.: Density and elemental ratios of secondary organic aerosol: Application of a density prediction method, Atmospheric Environment, 68, 273–277, 2013.
Wang, Y. Y., Liu, F. S., He, C. L., Bi, L., Cheng, T. H., Wang, Z. L., Zhang, H., Zhang, X. Y., Shi, Z. B., and Li, W. J.: Fractal Dimensions and Mixing Structures of Soot Particles during Atmospheric Processing, Environ Sci Tech Let, 4, 487-493, 2017.
Xu, X. Z., Zhao, W. X., Zhang, Q. L., Wang, S., Fang, B., Chen, W. D., Venables, D. S., Wang, X. F., Pu, W., Wang, X., Gao, X. M., and Zhang, W. J.: Optical properties of atmospheric fine particles near Beijing during the HOPE-J(3)A campaign, Atmos Chem Phys, 16, 6421-6439, 2016.