Point-by-point response to review comments on manuscript acp-2019-654 "Amplification of black carbon light absorption induced by atmospheric aging: temporal variation at seasonal and diel scales in urban Guangzhou"

By Cheng WU on behalf of all authors

We thank the two anonymous reviewers for their valuable time and constructive comments to improve the manuscript. Our point-by-point responses to the review comments are listed below as shown in blue. Changes to the manuscript are marked in teal blue in the revised manuscript. The marked manuscript is submitted together with this response document.

Anonymous Referee #1

General comments:

This paper reports a field measurement about the amplification of light absorbing property of black carbon aerosol by coating using a statistical method. The results of the light property measurement from this study is interesting and quite comprehensive, which provides some valuable insights on light absorbing amplification of black carbon aerosols in highly polluted urban region in China.

Author's Response: We appreciate the valuable time spent and efforts from the referee to improve the manuscript. Please see below for the point-by-point response to reviewers' comments.

Major comments:

R1-Q1. The authors need to clearly state what is new in this paper and highlight which are the major new findings. I feel that the current writing style makes this paper look more like a data report of some measurement in a different location.

Author's Response: Thanks for the suggestion. We have rewritten and reorganized many parts of the manuscript to make it more scientific-question-orientated.

For example, part of the abstract had been rewritten as follows.

Black carbon (BC) aerosols had been widely recognized as a vital climate forcer in the atmosphere. Amplification of light absorption can occur due to coatings on BC during atmospheric aging, an effect that remains uncertain in accessing the radiative forcing of

BC. Existing studies on absorption enhancement factor (E_{abs}) have poor coverage on both seasonal and diurnal scales. In this study, we applied a recently developed minimum R squared (MRS) method, which can cover both seasonal and diurnal scales, for E_{abs} quantification. Using field measurement data in Guangzhou, the aims of this study is to explore: 1) the temporal dynamics of BC optical properties at seasonal (wet season, July 31–September 10; dry season, November 15, 2017–January 15, 2018) and diel scales (1-hour time resolution) in the typical urban environment; 2) the influencing factors on E_{abs} temporal variability.

This result suggests that during the wet season, lensing effect was more likely dominating the AAE diurnal variability rather than the contribution from Brown Carbon (BrC). Secondary processing can affect E_{abs} diurnal dynamics. The E_{abs520} exhibited a clear dependency on secondary organic carbon to organic carbon ratio (SOC/OC), confirming the contribution of secondary organic aerosols on E_{abs}. E_{abs520} correlated well with nitrate and showed a clear dependence on temperature. This new finding implies that gas-particle partitioning of semi-volatile compounds may potentially play an important role in steering the diurnal fluctuation of E_{abs520}.

R1-Q2. It seems that one novelty of this paper is applying a newly developed MRS method to estimate the light absorption enhancement. If this is the key novelty, then I would recommend to add a separate section to discuss the difference between the current results from this method and those from other methods.

Author's Response: Thanks for the suggestion. We have rewritten and reorganized abstract and introduction to emphasize the novelty of study clearly.

One rewritten example in introduction is shown below.

As summarized in Table 1, the TD approach has high time resolution but limited sampling duration, while the AFD approach potentially has long sampling duration coverage but low time resolution. As a result, E_{abs} studies with both high time resolution and long sampling duration remained limited, leading to a lack of knowledge on E_{abs} variability on both seasonal and diel scales. To fill this knowledge gap, the aims of this study include: 1) to explore the temporal dynamics of E_{abs} on both seasonal and diel scales using the recently developed MRS approach; 2) to investigate the influencing factors on E_{abs} temporal variability, including photochemical aging, biomass burning (BB) and BC mixing state. In this study, filed measurements with one-hour time resolution were conducted in urban Guangzhou, a typical megacity in southern China in both wet (July 31–September 10, 2017) and dry (November 15, 2017–January 15, 2018) seasons.

We added a new section to discuss the comparison between E_{abs} by this study and previous studies.

3.2 Comparison of E_{abs} with previous studies

Filed measurements of E_{abs} values around the world are summarized in Table 2. Studies using the TD approach can achieve sub-hour time resolution, but studies using the TD approach had limited temporal coverage (normally less than a month). The AFD approach can potentially provide long-term E_{abs} results as long as filter samples are

available. However, the measurement duration of existing AFD studies was less than one month as shown in Table 2. The limited temporal coverage of existing AFD studies was likely due to the intense labor involved in filter treatment. In addition, the time resolution of existing AFD studies (8–24 hour) was not sufficient to fully resolve E_{abs} diurnal pattern. As a result, diurnal variations of E_{abs} values at different seasons were not covered in previous studies. In comparison, the MRS approach is a good alternative to explore the E_{abs} variations at both seasonal and diurnal scales. As shown in Table 2, low E_{abs} values were found in California (1.06@532 nm) (Cappa et al., 2012) and Japan (1.06@532 nm) (Ueda et al., 2016). Liu et al. (2017) observed a moderate E_{abs} in UK (1.0-1.3@532 nm) and suggested that the small E_{abs} observed by Cappa et al. (2012) was a result of mixing state diversity. A recent study in California (Cappa et al., 2019) found moderate E_{abs} at Fresno (1.22@532 nm) but low E_{abs} at Fontana (1.07@532 nm), which was partially associated with unequal distribution of coating between different BC-containing particle types (Lee et al., 2019). In general, higher E_{abs} values had been observed in more polluted urban areas, such as France (Paris, 1.53@880 nm) (Zhang et al., 2018a) and India (Kanpur, 1.8@781 nm) (Thamban et al., 2017). High E_{abs} values had been reported in various locations in China. The E_{abs} value in the wet season in our study (1.51) is higher than that in Nanjing (1.42) (Ma et al., 2019) but lower than those in central China (Shouxian, 2.3) (Xu et al., 2018), eastern China (Jinan, 1.9) (Bai et al., 2018) and northern China (Yuncheng, 2.25) (Cui et al., 2016b). The E_{abs} value in the dry season in our study (1.29) is lower than those in other locations in China, such as Beijing (1.66-4.0) (Xu et al., 2016; Zhang et al., 2018b), Nanjing (1.6) (Cui et al., 2016a), Xi'an (1.8) (Wang et al., 2014) and Jinan (2.07) (Chen et al., 2017). Since the collocated comparison of the three E_{abs} methods do not exist, a direct comparison between the three methods remain difficult. Nevertheless, a few studies, which conducted at the same city but during different periods, yielded comparable E_{abs} values. For example, E_{abs} in Nanjing by MAE method (1.6) (Cui et al., 2016a) was higher than that by the TD method (1.42) (Ma et al., 2019). This difference in E_{abs} might not only due to the different E_{abs} determination methods, but could also be a result of seasonal variations of E_{abs}.

R1-Q3. The introduction part is like a mini review rather than an introduction. It provides too many details, some of which are not quite relevant to this study. In addition, this introduction appears not to reflect the significance of raised issue/science questions.

Author's Response: Thanks for the suggestion. We agree that the introduction is longer than typical studies. Since the E_{abs} determination by MRS approach is relatively new to most readers, we try to provide sufficient information to help the readers to understand the motivation of using MRS instead of conventional approaches. This background information also helps the readers to understand the working principle and advantage of MRS. Nevertheless, we agree that there are room for improving the logic flow. We have made substantial revisions in the introduction to make the manuscript more scientific-question-orientated.

R1-Q4. The authors need to clearly state what is new in this paper and highlight which are the major new findings. I feel that the current writing style makes this paper look more like a data report of some measurement in a different location.

Author's Response: Thanks for the suggestion. We rewritten some of the contents to show the new findings clearly.

An example in abstract is shown below.

E_{abs520} correlated well with nitrate and show a clear dependence on temperature. This new finding implies that gas-particle partitioning of semi-volatile compounds may potentially play an important role in steering the diurnal fluctuation of E_{abs520}.

R1-Q5. When analyzing diurnal patterns of BC particles, I would always recommend to separate weekdays and weekends.

Author's Response: Thanks for the suggestion. Now we included Figure S10 to show the weekday/weekend effect on diurnal patterns. Corresponding discussions were added and also shown below.

To explore the effect of traffic volume, the weekday/weekend effect was investigated in Figure S10. The evening EC peak reduced substantially during weekend, implying that traffic volume has a strong influence on shaping the diurnal pattern of EC.

The difference of diurnal OC/EC pattern between weekday and weekend is negligible (Figure S10), suggesting that the portion of different vehicle type (e.g. diesel vs. gasoline) is relatively constant between weekday and weekend.

R1-Q6. This paper is quite long and not well written. I think the authors should make it more concise and improve its writing.

Author's Response: Thanks for the suggestion. To shorten the length of the manuscript, we had moved back trajectory and wind rose discussions to the supplement. We had also made necessary revisions to improve the conciseness of the manuscript.

Specific comments:

R1-Q7. Eq3: the denominator "EC" needs to be defined first.

Author's Response: Thanks for pointing out. The following content is added.

EC in Eq. 3 and 4 represents elemental carbon mass concentration determined by the thermal optical analysis method (Wu et al., 2012), which can be considered as a surrogate of BC mass concentration.

R1-Q8. L104: "to low" should be "too low"

Author's Response: Thanks for pointing out the typo. Revision made.

R1-Q9. L108: Consider revising the sentence "Third, the TD is not the ideal time machine for reversing the morphology transformation of BC."

Author's Response: Thanks for the suggestion. The sentence had been rewritten as shown below.

Third, the TD approach cannot perfectly reverse the morphology transformation of BC from aged state back to freshly emitted state.

R1-Q10. L169: what are these "MAE values"?

Author's Response: Thanks for the suggestion. We add Table S1 to show theses literature values and the sentence had been rewritten as shown below.

MAE values from study by Drinovec et al. (2015) was adopted for σ_{abs_total} back-calculations from eBC at different wavelengths as shown in Table S1.

Table S1. MAE values from study by Drinovec et al. (2015) was adopted for σ_{abs_total} back-calculations at different wavelengths.

Wavelength (nm)	MAE (m^2g^{-1})
370	18.47
470	14.54
520	13.14
590	11.58
660	10.35
880	7.77
950	7.19

R1-Q11. L269: The sentence "The MAE at the minimum R2 of the EC vs. …" is confusing and needs to rewrite.

Author's Response: Thanks for the suggestion. The sentence had been rewritten as shown below.

In MRS calculation, the correlation (R^2) between measured EC and estimated hypothetical σ_{abs_aging} is examined as a function of a series of hypothetical MAE_p (MAE_{p_h}). Since σ_{abs_aging} was resulted from secondary processing while EC was coming from primary emissions, a MAE_{p_h} that leads to a minimum R^2(EC, $\sigma_{abs\_aging\_h}$) can best represents the independent nature between EC and σ_{abs_aging}. As a result, MAE_{p_h} at minimum R^2(EC, $\sigma_{abs\_aging\_h}$) corresponds to the authentic MAE_p.

R1-Q12. L620: The sentence "As a result, the increasing nitrate might potentially affect BC's radiative forcing in China." should be rewritten.

Author's Response: Thanks for the suggestion. The sentence had been rewritten as shown below.

If the nitrate fraction in the coating materials on BC increases, the diurnal pattern of E_{abs} for BC may be affected by the fluctuation of nitrate content in aerosol particles. As a result, the increasing concentration of nitrate might potentially affect radiative forcing by BC in China.

Anonymous Referee #2

General comments:

The manuscript presents a comprehensive study on the black carbon light absorption enhancement (Eabs) in urban China. They used a newly developed method for Eabs determination, which utilizes measurements from a filter-based absorption instrument and a thermal-optical analysis OC/EC analyzer. The seasonal and diurnal patterns of Eabs were analyzed, and the potential influencing factors were discussed. This manuscript includes sufficient originality, and the topic seems to fit the scope of ACP. In general, the overall quality of the manuscript is good yet the logic of some contents, especially the introduction, can be improved. I believe that the points below should be addressed. I therefore recommend a Minor Revision before publication in ACP.

Author's Response: We appreciate the valuable time spent and efforts from the referee to improve the manuscript. Please see below for point-by-point response to reviewers' comments.

Major comments:

R2-Q1. The introduction is long but the motivation of this study seems missing. The authors should state clearly what's the scientific question that this study is trying to answer.

Author's Response: Thanks for the suggestion. We have made substantial revisions in the introduction section by emphasizing the motivation and scientific question.

The motivation of this study is emphasized as shown below.

As summarized in Table 1, the TD approach has high time resolution but limited sampling duration, while the AFD approach potentially has long sampling duration coverage but low time resolution. As a result, E_{abs} studies with both high time resolution and long sampling duration remained limited, leading to a lack of knowledge on E_{abs} variability on both seasonal and diel scales. To fill this knowledge gap, the aims of this study include: 1) to explore the temporal dynamics of E_{abs} on both seasonal and diel scales using the recently developed MRS approach; 2) to investigate the influencing factors on E_{abs} temporal variability, including photochemical aging, biomass burning (BB) and BC mixing state. In this study, filed measurements with one-hour time resolution were conducted in urban Guangzhou, a typical megacity in southern China in both wet (July 31–September 10) and dry (November 15, 2017–January 15, 2018) seasons.

R2-Q2. Since this study uses a new method for Eabs quantification, a comparison with previous studies should be given in more details. Table 2 provides a useful summary but corresponding discussions seems too simple in the current manuscript.

Author's Response: Thanks for the suggestion. We added a separated section to discuss the comparisons of the current study and previous studies.

3.2 Comparison of E_{abs} with previous studies

Filed measurements of E_{abs} values around the world are summarized in Table 2. Studies using the TD approach can achieve sub-hour time resolution, but studies using the TD approach had limited temporal coverage (normally less than a month). The AFD approach can potentially provide long-term E_{abs} results as long as filter samples are available. However, the measurement duration of existing AFD studies was less than one month as shown in Table 2. The limited temporal coverage of existing AFD studies was likely due to the intense labor involved in filter treatment. In addition, the time resolution of existing AFD studies (8–24 hour) was not sufficient to fully resolve E_{abs} diurnal pattern. As a result, diurnal variations of E_{abs} values at different seasons were not covered in previous studies. In comparison, the MRS approach is a good alternative to explore the E_{abs} variations at both seasonal and diurnal scales. As shown in Table 2, low E_{abs} values were found in California (1.06@532 nm) (Cappa et al., 2012) and Japan (1.06@532 nm) (Ueda et al., 2016). Liu et al. (2017) observed a moderate E_{abs} in UK (1.0-1.3@532 nm) and suggested that the small E_{abs} observed by Cappa et al. (2012) was a result of mixing state diversity. A recent study in California (Cappa et al., 2019) found moderate E_{abs} at Fresno (1.22@532 nm) but low E_{abs} at Fontana (1.07@532 nm), which was partially associated with unequal distribution of coating between different BC-containing particle types (Lee et al., 2019). In general, higher E_{abs} values had been observed in more polluted urban areas, such as France (Paris, 1.53@880 nm) (Zhang et al., 2018a) and India (Kanpur, 1.8@781 nm) (Thamban et al., 2017). High E_{abs} values had been reported in various locations in China. The E_{abs} value in the wet season in our study (1.51) is higher than that in Nanjing (1.42) (Ma et al., 2019) but lower than those in central China (Shouxian, 2.3) (Xu et al., 2018), eastern China (Jinan, 1.9) (Bai et al., 2018) and northern China (Yuncheng, 2.25) (Cui et al., 2016b). The E_{abs} value in the dry season in our study (1.29) is lower than those in other locations in China, such as Beijing (1.66-4.0) (Xu et al., 2016; Zhang et al., 2018b), Nanjing (1.6) (Cui et al., 2016a), Xi'an (1.8) (Wang et al., 2014) and Jinan (2.07) (Chen et al., 2017). Since the collocated comparison of the three E_{abs} methods do not exist, a direct comparison between the three methods remain difficult. Nevertheless, a few studies, which conducted at the same city but during different periods, yielded comparable E_{abs} values. For example, E_{abs} in Nanjing by MAE method (1.6) (Cui et al., 2016a) was higher than that by the TD method (1.42) (Ma et al., 2019). This difference in E_{abs} might not only due to the different E_{abs} determination methods, but could also be a result of seasonal variations of E_{abs}.

R2-Q3. Figure 6 shows a clear dependency of E_{abs} on SOC/OC. However, the correlation between organics and E_{abs} is not that good as expected. The authors should explain why a good dependency was observed in Figure 6 but meanwhile a low r^2 was found in Figure 7.

Author's Response: Thanks for the comments. There are several possibilities for the lower R^2 of organics in Figure 7. First, Organics shown in Figure 7 contain both primary and secondary organics, two types or organics that might have different impacts on E_{abs} values. Second, the dependency of E_{abs} on SOC/OC might not necessarily be reflected in the form of correlation on a diurnal scale.

The following contents in section 3.6 were rephased.

It should be noted that a good E_{abs} dependence on SOC/OC observed in Figure 6 does not necessarily lead to a good diurnal correlation between E_{abs} and SOC/OC (e.g. Figure 4a). In other words, the dependency of E_{abs} on SOC/OC might not necessarily be reflected in the form of correlation on a diurnal scale. Thus, the poor diurnal correlation between SOC/OC and E_{abs520} observed in the wet season (Figure 4a) cannot rule out the contribution of SOC on E_{abs520}.

The following contents in section 3.7 were rephased.

There are two possibilities for the lower R^2 (Organics, E_{abs}) in Figure 7d comparing to R^2(SOC/OC, E_{abs}) in Figure 4b. First, organics shown in Figure 7d contain both primary and secondary organics, while SOC/OC shown in Figure 4b represents the secondary portion only. Second, poor diurnal correlations do not necessarily rule out the contribution of organics and sulfate to E_{abs} by analogy with the SOC correlation with E_{abs} as discussed in Section 3.6.

R2-Q4. As related to comment # 3) above, one of the most interesting findings of this study is that Eabs exhibits dependency on SOC/OC ratio and has good correlation with nitrate. For Eabs dependency on SOC/OC ratio, one might believe that it is the "concentration" of SOC in total OC that affects the absorption enhancement. For good correlation between nitrate and Eabs, is there any possible reason other than partitioning behavior that would potentially contribute to the good correlation?

Author's Response: Thanks for the suggestion. Besides lensing effect of non-absorbing coatings (e.g. nitrate) induced light absorption enhancement, formation of light-absorbing components could also lead to light absorption enhancement. For example, nitro-aromatic compounds (NACs) are secondary formed light-absorbing components that contribute light absorption enhancement. Since NAC formation is strongly associated with NO_x, a good correlation between NACs and nitrate had been observed (Chow et al., 2016). However, NACs are part of Brown Carbon (BrC), thus have strong wavelength dependency. If a considerable fraction of NACs is present in $PM_{2.5}$, an elevated AAE would be observed. The AAE observed in this study was <2, implying a weak signature of BrC. In our study, the abundance of NACs was not measured, therefore E_{abs} contribution from NACs remains unclear. But considering the fact that typical NAC concentration in $PM_{2.5}$ was low (<10 ng m^{-3}) (Chow et al., 2016) and their contribution to light absorption was small (<1%@370 nm) (Teich et al., 2017), the contribution of NACs to R^2 (Nitrate, E_{abs}) is expected to be very small.

R2-Q5. How measurement uncertainties would affect E_{abs} determination by MRS method?

Author's Response: The measurement uncertainties of MRS method for E_{abs} determination had been systematically examined in our previous study (Wu et al., 2018). Thus, only a brief introduction is given here. Three scenarios are considered using the data from our previous study (Wu et al., 2018). Scenario A represents systematic biases in Aehtalometer and OCEC measurements in the form of multipliers. The biased data are marked as σ'_{abs550} and EC', respectively, as shown below:

$$\sigma'_{abs550} = \sigma_{abs550} \times 2 \quad \text{(R-1)}$$

$$EC' = EC \times 0.7 \quad \text{(R-2)}$$

The E_{abs} remain the same in scenario A, suggesting that MRS is not sensitive to the systematic biases that are in the form of multipliers.

In scenario B, EC by different TOA protocols are compared to investigate the effect of different EC determination approaches while σ_{abs550} remains unchanged. EC by IMPROVE TOR protocol is calculated from NIOSH TOT EC following an empirical formula for suburban sites derived from a 3-year OCEC dataset in PRD (Wu et al., 2016):

$$EC_{IMP\_TOR} = 2.63 \times EC_{NSH\_TOT} + 0.05 \quad \text{(R-3)}$$

In scenario B, EC uncertainty induced E_{abs} change is small (from 1.44 to 1.40) as shown in Figure R2, suggesting that the MRS is not sensitive to the systematic biases that contain both multipliers and offsets.

Scenario C examines the impact of sample-dependent bias as a function of E_{abs}. Unlike the proportional bias in Scenario A and B that is the same for all data points, the bias in Scenario C depends on the E_{abs550} of individual samples, which are parametrized by Eqs. (R-4) and (R-5).

$$\sigma'_{abs550} = \sigma_{abs550} + \sigma_{abs550} \times (k \times E_{abs550} - k) \quad \text{(R-4)}$$

$$EC' = EC - EC \times (k \times E_{abs550} - k) \quad \text{(R-5)}$$

As shown in Eqs. (R-4) and (R-5), the positive bias of σ_{abs550} and negative bias of EC are proportional to E_{abs550}. The magnitude of E_{abs550}-dependent bias is regulated by the factor k. Since σ'_{abs550} and EC' are biased in different directions, resulting a further amplification in MAE biases, which could be considered as the extreme case. As shown in Figure R3, for k=10% (corresponding to a bias of 10% when E_{abs}=2), the bias of MRS-derived E_{abs} is very small (1%). For k=20%, the MRS-derived E_{abs} changes from 1.44 to 1.66, leading to a bias of 15%. These results imply that if the measurement bias follows the same form as demonstrated in scenario C, the bias is not negligible but still acceptable. If the impact only affects σ_{abs} or EC rather than impacting both, the bias is expected to be smaller than the estimation shown in Scenario C.

In summary, E_{abs} determination by MRS method is not sensitive to measurement uncertainties in light absorption or EC mass.

Scenario A

Original data → Systematically biased data

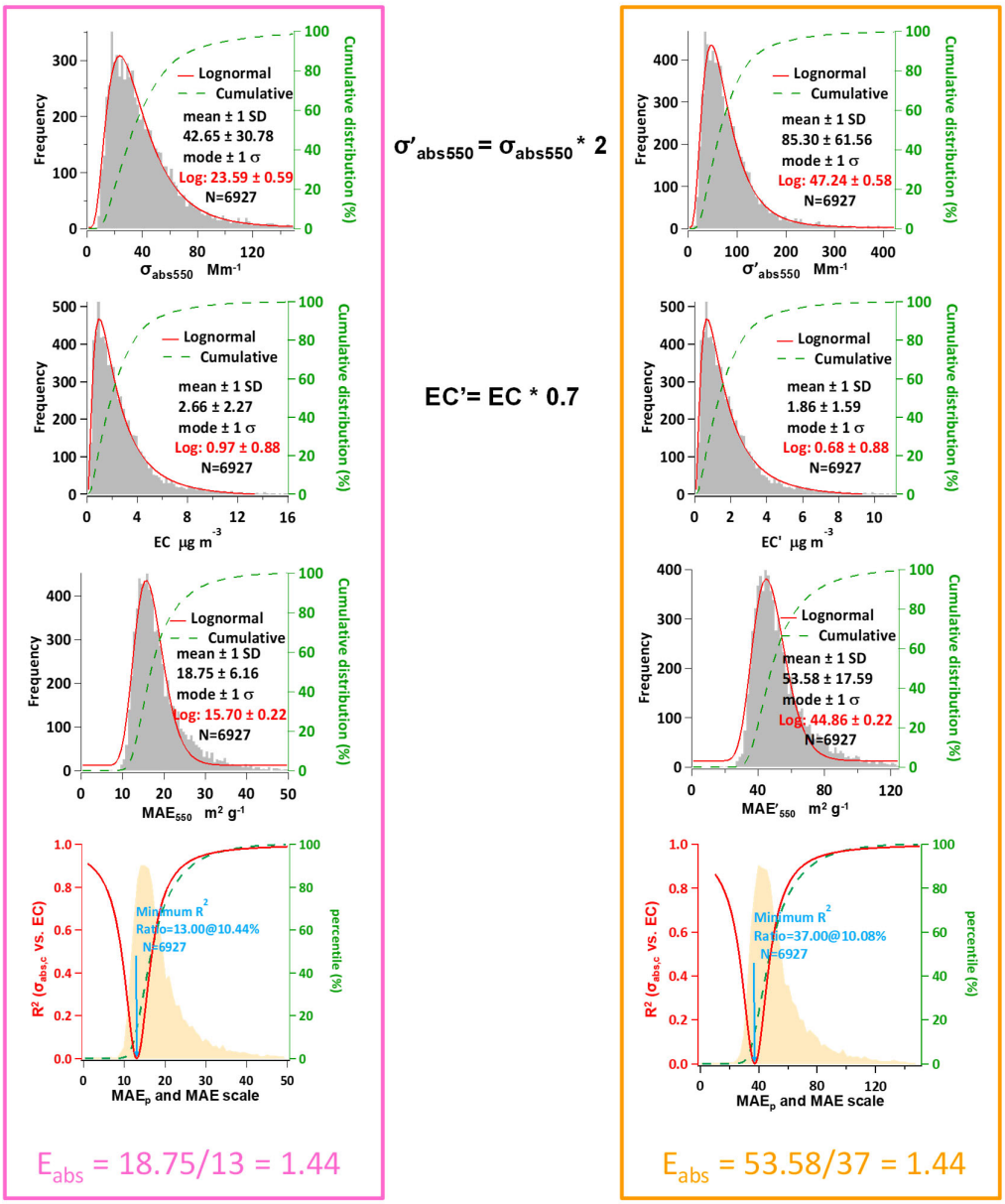

Figure R1. Comparison of E_{abs} from original data and systematically biased data (Scenario A). It should be noted that the E_{abs} shown here is ratio of averages, which is different form the annual average E_{abs} calculated from average of ratios.

Scenario B

Original data → Systematically biased data

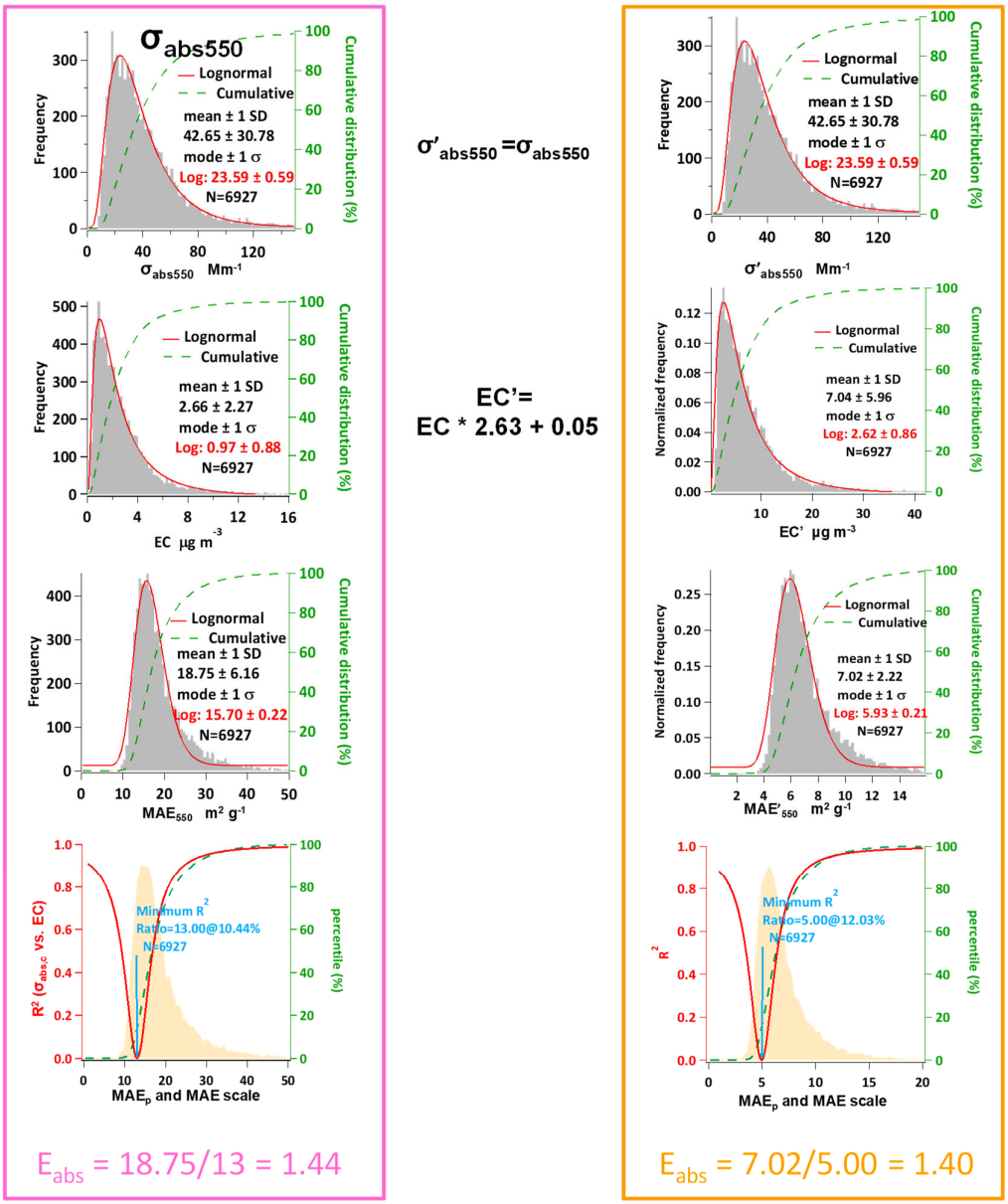

Figure R2. Comparison of E_{abs} from data using NIOSH EC and data using IMPROVE EC (Scenario B). It should be noted that the E_{abs} shown here is ratio of averages, which is different form the annual average E_{abs} calculated from average of ratios.

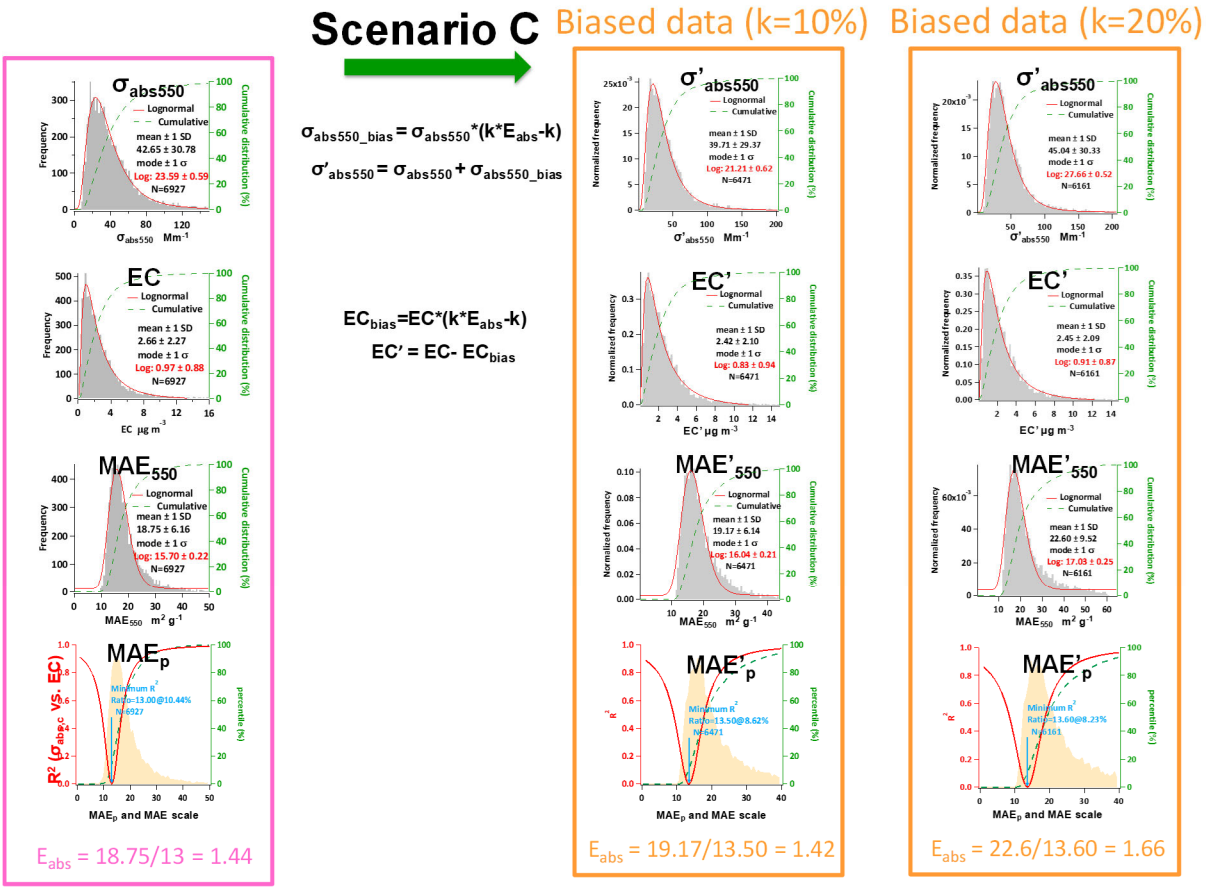

Figure R3. Comparison of E_{abs} from data using original data and E_{abs} depended biased data (Scenario C). It should be noted that the E_{abs} shown here is ratio of averages, which is different form the annual average E_{abs} calculated from average of ratios.

R2-Q6. The MAE values reported in this study seems higher than those reported in the literature. Any reasons?

Author's Response: Two possible factors can lead to high MAE values. The first possibility is associated with measurement uncertainty of MAE, which can be further attributed to two uncertainties: σ_{abs} determination uncertainty (AE33) and EC determination uncertainty (OCEC analyzer). Both overestimation of σ_{abs} or underestimation of EC can lead to overestimation of MAE. Over estimation of σ_{abs} could happen when the AE33 correction factor C_{ref} is underestimated, since C_{ref} could vary by locations and changed over time. EC determination could be different by a factor of 2-4 due to different analysis protocols (Chow et al., 2004; Wu et al., 2012). The second possibility is that the particles measured in this study were highly aged, leading to higher MAE values than those in previous studies. Nevertheless, the focus of this study is E_{abs}, whose determination procedure by MRS is not sensitive to measurement biases in MAE, as discussed in response to R2-Q5.

R2-Q7. The authors suggest that the correlation between Eabs and nitrate was associated with the volatility of nitrate. If that is the case, would that be applied to the organics that have a volatility similar to nitrate?

Author's Response: We agreed with the reviewer that organics with a volatility similar to nitrate could potentially contribute to the diurnal variability of E_{abs}. This possibility is mentioned in section 3.7.

By analogy with nitrate, organic compounds with a volatility similar to nitrate might potentially involve in shaping the diurnal pattern of E_{abs} in the wet season.

Technical comments:

R2-Q8. Line 27. "exhibit" should be "exhibited"

R2-Q9. Line 30. "were" should be "was"

R2-Q10. Line 37. "exhibit" should be "exhibited"

R2-Q11. Line 104. "to low" should be "too low"

R2-Q12. Line 592. "a Aethalometer" should be "an Aethalometer"

R2-Q13. Line 606. "two component" should be "two-component"

Author's Response: Thanks for the technical suggestions. Revisions made accordingly.

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Amplification of black carbon light absorption induced by atmospheric aging: temporal variation at seasonal and diel scales in urban Guangzhou

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Abstract. Black carbon (BC) aerosols had been widely recognized as a vital climate forcer in the atmosphere. Amplification of light absorption can occur due to coatings on BC during atmospheric aging, an effect that remains uncertain in accessing the radiative forcing of BC. Existing studies on absorption enhancement factor (E_{abs}) have poor coverage on both seasonal and diurnal scales. In this study, we applied a recently developed minimum R squared (MRS) method, which can cover both seasonal and diurnal scales, for E_{abs} quantification. Using field measurement data in Guangzhou, the aims of this study is to explore: 1) the temporal dynamics of BC optical properties at seasonal (wet season, July 31–September 10; dry season, November 15, 2017–January 15, 2018) and diel scales (1-hour time resolution) in the typical urban environment; 2) the influencing factors on E_{abs} temporal variability. Mass absorption efficiency at 520 nm by primary aerosols (MAE_{p520}) determined by MRS exhibited a strong seasonality (8.6 m^2g^{-1} in the wet season and 16.8 m^2g^{-1} in the dry season). E_{abs520} was higher in the wet season (1.51±0.50) and lower in the dry season (1.29±0.28). Absorption Ångström exponent ($AAE_{470\text{-}660}$) in the dry season (1.46±0.12) was higher than that in the wet season (1.37±0.10). Collective evidence showed that the active biomass burning (BB) in dry season effectively altered the optical properties of BC, leading to elevated MAE, MAE_p and AAE in dry season comparing to those in the wet season. Diurnal E_{abs520} was positively correlated with $AAE_{470\text{-}660}$ (R^2=0.71) and negatively correlated with the AE33 aerosol loading compensation parameter (k) (R^2=0.74) in the wet season, but these correlations were significantly weaker in the dry season, which may be related to the impact of BB. This result suggests that during the wet season, the lensing effect was more likely dominating the AAE diurnal variability rather than the contribution from Brown Carbon (BrC). Secondary processing can affect E_{abs} diurnal dynamics. The E_{abs520} exhibited a clear dependency on secondary organic carbon to organic carbon ratio (SOC/OC), confirming the contribution of secondary organic aerosols on E_{abs}. E_{abs520} correlated well with nitrate and showed a clear dependence on temperature. This new finding implies that gas-particle partitioning of semi-volatile compounds may potentially play an important role in steering the diurnal fluctuation of E_{abs520}. In the dry season, the diurnal variability of E_{abs520} was associated with photochemical aging as evidenced by the good correlation (R^2=0.69) between oxidant concentrations (O_x=O_3+NO_2) and E_{abs520}.

1. Introduction

Atmospheric aerosols have received great attention in recent years due to their global climatic effects and environmental effects (Anderson et al., 2003). Carbonaceous aerosols account for a large fraction of the global aerosol mass as the main light-absorbing materials in aerosols (Kanakidou et al., 2005; Bond and Bergstrom, 2006). Black carbon (BC), which originated from incomplete combustion of hydrocarbon fuels (Johansson et al., 2018), is the dominating fraction of light absorbing carbonaceous aerosols. BC had been widely recognized not only as an air pollutant that poses threat to the public health (Grahame et al., 2014; Apte et al., 2015), but also an essential climate forcer (Chung and Seinfeld, 2002). The BC burden in the atmosphere increased substantially in the recent years as evidenced by the ice core samples (Ruppel et al., 2014) and sediment cores from eastern China marginal seas (Fang et al., 2018). The growing abundance of BC in the atmosphere leads to elevated environmental impacts. BC had been regarded as the third most important climate forcer after carbon dioxide and methane (IPCC, 2013). On a global scale, BC can heat the atmospheric directly owing to its strong light absorption across the solar spectrum (Bond and Bergstrom, 2006), thus contributes to the warming effect (Bond et al., 2013). On a regional scale, BC deposition on ice and snow can reduce surface albedo, leading to glacier melting (Gertler et al., 2016; Kopacz et al., 2011; Flanner et al., 2007; He et al., 2018; Hansen and Nazarenko, 2004), especially at high-altitude regions such as the Tibetan Plateau (Ming et al., 2008). On a local scale, BC can modify planetary boundary layer meteorology that leads to the "dome effect", and thus enhance local pollution indirectly (Ding et al., 2016; Wilcox et al., 2016). In microscale, BC was found playing a key role in the photochemical aging of soot by initiating the oxidation of OC (Li et al., 2018c). In addition, BC can indirectly affect the climate by altering cloud formation and cloud cover (Nenes et al., 2002; Koch and Del Genio, 2010; Kaufman and Koren, 2006; Albrecht, 1989).

However, large uncertainties still exist in estimating the radioactive forcing of BC (Bond et al., 2013). The gap largely arises from the limited characterization of BC mixing state and optical properties evolution in the atmosphere (Fuller et al., 1999; Jacobson, 2001; Nordmann et al., 2014). BC is chemically inert, but morphology transformation is unavoidable once emitted into the atmosphere. A recent study suggested that BC restructuring during aging can be divided into two steps (Pei et al., 2018). First, the void of the BC particles will be filled by the aging induced materials. Once filled, further accumulation of organic and inorganic coating materials leads to the growth of particle size. Ma et al. (2013) reported soot restructuring during water evaporation in a laboratory study. Such morphology transformation leads to alternation of BC optical properties, as evidenced by a number of numerical studies (Fuller et al., 1999; Bond et al., 2006; Liu et al., 2016a; Zhang et al., 2017; Lefevre et al., 2019), laboratory experiments (Schnaiter et al., 2005; Zhang

et al., 2008; Xue et al., 2009; Shiraiwa et al., 2010; Metcalf et al., 2013; Wei et al., 2013; Chen et al., 2015) and field studies (Knox et al., 2009; Cappa et al., 2012; Lack et al., 2012b; Liu et al., 2015; Liu et al., 2017a; Liu et al., 2019c). The presence of coating materials on BC leads to the increase of mass absorption efficiency (MAE) through the lensing effect (Schwarz et al., 2008b). Besides coating thickness, the magnitude of light absorption enhancement by the lensing effect also depends on the optical properties of the coating materials. A coating of brown carbon (BrC) can further amplify the light absorption comparing to a transparent coating (Lack and Cappa, 2010). Recent studies suggested that BC mixing state diversity also affects the bulk E_{abs} (Fierce et al., 2016; Matsui et al., 2018; Cappa et al., 2019).

The total BC light absorption (σ_{abs_total}) after aging can be segregated into primary absorption (σ_{abs_pri}) by the BC core and the additional absorption (σ_{abs_aging}) due to the presence of coating:

$$\sigma_{abs\_total} = \sigma_{abs\_pri} + \sigma_{abs\_aging} \quad (1)$$

The key parameter for light absorption enhancement, E_{abs}, can be calculated from:

$$E_{abs} = \frac{\sigma_{abs\_total}}{\sigma_{abs\_pri}} = \frac{MAE_t}{MAE_p} \quad (2)$$

where MAE_t is the MAE of coated BC:

$$MAE_t = \frac{\sigma_{abs\_total}}{EC} \quad (3)$$

and MAE_p represent the MAE of BC when fleshly emitted:

$$MAE_p = \frac{\sigma_{abs\_pri}}{EC} \quad (4)$$

EC in Eq. 3 and 4 represents elemental carbon mass concentration determined by the thermal optical analysis method (Wu et al., 2012), which can be considered as a surrogate of BC mass concentration. Atmospheric aging process can lead to BC E_{abs} larger than 1 due to the increase of MAE_t.

Three technical approaches had been applied for E_{abs} quantification as summarized in Table 1. The first approach is to use a thermal denuder (TD) upstream of the instrument that measures σ_{abs} (e.g. PAS, photo-acoustic spectrometer). By measuring the denuded and ambient sample in rotation with a desired interval (e.g. 5 min), σ_{abs_total} and σ_{abs_pri} can be obtained to determine E_{abs} following Eq.2. Particle loss in TD is unavoidable and need to be accounted for (Burtscher et al., 2001). The advantage of TD is its ability to obtain high-time-resolution data (Cappa et al., 2012; Lack et al., 2012b; Liu et al., 2015; Liu et al., 2017a). But TD has its own limitations. First, TD is not suitable for long-term measurements (e.g. most studies last for a few weeks as shown in Table 2). Second, the selection of working temperature is sample depended and varied by

sampling sites, which can largely affect the E_{abs} measurement results. As a result, a universal optimal TD working temperature does not exist. If the temperature is to low, the coating materials cannot be fully vaporized. Study by Ma et al. (2019) shown that coating materials still account for 60% of particle mass after thermodenuding at a temperature of 280°C, implying the incomplete removal of coating materials. On the other hand, if the temperature is too high, pyrolysis would occur (Irwin et al., 2013), leading to a biased E_{abs} measurement. For example, Li et al. (2018a) explore the variability of TD temperature on E_{abs} determination in Hong Kong. For a TD temperature of 50°C to 200°C, E_{abs} ranges from 1.02 to 1.20. E_{abs} reaches 1.6 for a TD temperature 280°C. Third, the TD approach cannot perfectly reverse the morphology transformation of BC from aged state back to freshly emitted state. Previous studies have shown that the chain-like-aggregate morphology of nascent BC cannot be restored after thermodenuding of the coatings on the reconstructed BC core, which tends to be more compact and spherical (Bambha et al., 2013; Ghazi and Olfert, 2013). In addition, the high cost of TD-PAS system thwarts its wider applications in field studies.

The second approach for E_{abs} determination is aerosol filter filtration-dissolution (AFD). AFD remove coatings on BC using water and organic solvents (Cui et al., 2016b). The advantage of AFD is that this method can be applied on historical filters archived by long-term/large-scale speciation sampling networks. It opens up a new path to retrieve the historical E_{abs} from datasets with large temporal and spatial coverage. The limitation mainly arises from the AFD treatment process, which only removes the soluble part of the coating. The AFD treatment process is also labor intensive. The time resolution of E_{abs} by AFD depends on the interval of filter sampling, which has a typical sampling time of 24 hr, making it difficult to study the diurnal pattern of E_{abs}.

The third approach is MAE method. E_{abs} is quantified from the ratio of MAE_t to MAE_p as shown in Eq. 2. Since MAE_t can be obtained from ambient measurements, the determination of MAE_p is the key to this approach. One way is to adopt empirical MAE_p in the literature (Cui et al., 2016a) (abbreviated as MAE+empirical approach hereafter). Since the real-world MAE_p could be highly diverse by different sources and varies temporally and spatially (Roden et al., 2006; Adler et al., 2010; Shen et al., 2013; McMeeking et al., 2014; Healy et al., 2015; Cheng et al., 2016; Weyant et al., 2016; Dastanpour et al., 2017; Radney et al., 2017; Conrad and Johnson, 2019), empirical MAE_p at one site might not be applicable at other sites.

Another method to determine MAE_p is combing σ_{abs_total} measurement with single particle measurement to provide the mixing state of BC, e.g. Single-Particle Soot Photometer (SP2) or Soot Particle Aerosol Mass Spectrometer (SP-AMS). This method is abbreviated as MAE+SP hereafter for easy reference. The lag time between the incandescence signal and scattering in SP2 can be used to differentiate thickly

coated BC and bare BC. The intercept of linear regression between MAE (y axis) against the number fraction of aged BC (r_{aged}, x axis) represents MAE_p (Lan et al., 2013; Wang et al., 2014). This method only considers E_{abs} dependency on the number fraction of aged particles and ignores the coating thickness of the aged particles, thus is only valid for a limited period of time when coating thickness and size distribution is relatively stable. An improved method for MAE_p determination by SP2 is utilizing the rBC size distribution to calculate the MAE_p by Mie model (Liu et al., 2017a; Wang et al., 2018a; Wang et al., 2018b).

A recently developed approach, Minimum R Squared method (MRS) can be applied to MAE_p determination using elemental carbon (EC) as an tracer (Wu et al., 2018). MRS is a statistic approach and MAE_p can be determined in a quantitative manner that minimizes the arbitrariness in MAE_p estimation by the traditional approach. Application of MRS for E_{abs} determination is abbreviated as MAE+MRS approach for easy reference. As summarized in Table 1, the TD approach has high time resolution but limited sampling duration, while the AFD approach has long sampling duration coverage but low time resolution. As a result, E_{abs} studies with both high time resolution and long sampling duration remained limited, leading to a lack of knowledge on E_{abs} variability on both seasonal and diel scales. To fill this knowledge gap, the aims of this study include: 1) to explore the temporal dynamics of E_{abs} on both seasonal and diel scales using the recently developed MRS approach; 2) to investigate the influencing factors on E_{abs} temporal variability, including photochemical aging, biomass burning (BB) and BC mixing state. In this study, filed measurements with one-hour time resolution were conducted in urban Guangzhou, a typical megacity in southern China in both wet (July 31–September 10, 2017) and dry (November 15, 2017–January 15, 2018) seasons. Abbreviations used in this paper are listed in Table A1 in the appendix for easy reference.

2. Field measurements and data analysis methods

2.1 Characteristics of the observation site

As shown in Figure 1, sampling of this study was conducted at Jinan University (JNU) atmospheric super site (113.35°E, 23.13°N, 40 meters above sea level), which located in Tianhe District, downtown Guangzhou. The site is on top of the library building and surrounded by teaching and residential areas. The campus was surrounded by three busiest road of the city (Figure S1) and traffic emission is a major source of primary emissions. Guangzhou is located in the southern China and is also the geographical center of Guangdong Province. There are limited industrial pollution sources around the sampling site, thus this site can represent the typical urban environment in the Pearl River Delta (PRD) region.

The subtropical climate of PRD is strongly affected by two monsoon systems: South China Sea (SCS) monsoon and Northeast monsoon. April to May is the transition period of the Northeast monsoon to the SCS

monsoon. June to September is the SCS monsoon-dominated period (wet season). The southern prevailing wind brings the clean and humid air masses from the vast ocean. October is the transition period of the SCS monsoon to the Northeast monsoon. November to March is the Northeast monsoon-dominated period (dry season). The northeastern prevailing wind brings polluted air masses from the more economically-developed regions in the eastern Asia. This study included two sampling periods: July 31–September 10 2017 and November 15 2017–January 7 2018, corresponding to wet and dry seasons, respectively.

2.2 Light absorption measurements

A dual–spot Aethalometer (Model AE33, Magee Scientific Company, Berkeley, CA, USA) was used for σ_{abs_total} determination. Aethalometer sampling was performed at a flow rate of 5 L min^{-1} with a 2.5 μm cyclone inlet. A Nafion dryer was used to maintain the RH<40%. The data logging time resolution is 1 minute. AE33 reports results in the form of equivalent BC mass (eBC), which can be used to back-calculate the σ_{abs_total}. MAE values from study by Drinovec et al. (2015) was adopted for σ_{abs_total} back-calculations from eBC at different wavelengths as shown in Table S1. A multiple scattering correction factor C_{ref}=3.29 was used according to a recent study in this region (Qin et al., 2018). As a filter-based method, deposition of light absorbing particles on filter leads to the attenuation of filter transmittance signal, which is proportional to the BC mass concentration. However, as the particle deposition layer gradually increase, light was block at the upper particle layer before reaching the underneath particle layer, resulting the well-known artifact: loading effect. Since the lower layer particles did not contribute to the light attenuation, the linear relationship between BC mass concentration and light attenuation signal was distorted.

The AE33 adopted the "dual spot" design to minimize the loading effect (Drinovec et al., 2015), which is an improvement of the traditional "single spot" correction (Virkkula et al., 2007). Two spots perform the sampling simultaneously. The correction can be implemented for each wavelength by the following two equations,

$$eBC1_{raw} = eBC_{\text{compensated}} \cdot (1 - k \cdot ATN1) \quad (5)$$

$$eBC2_{raw} = eBC_{\text{compensated}} \cdot (1 - k \cdot ATN2) \quad (6)$$

Where $eBC1_{raw}$ and $eBC2_{raw}$ are the uncorrected eBC mass determined by the two spots. $eBC_{\text{compensated}}$ is the corrected eBC concentration to be determined. k is the empirical compensation parameter. $ATN1$ and $ATN2$ are the light attenuation measured at the two spots. The flows of the two spots were maintained at a ratio of 2:1 to achieve differential increase of ATN in a set window of time (e.g. 1 min). Since $BC1_{raw}$, $BC2_{raw}$, $ATN1$ and $ATN2$ are all known variables, $eBC_{\text{compensated}}$ and k can be

calculated for each measurement following Eqs. (5) & (6). As shown in Figure S2, $eBC1_{raw}$ and $eBC2_{raw}$ exhibit discontinuity once filter was moved to the next position, which implies biases induced by the loading effect. After the dual spot correction, the discontinuity was minimized substantially as shown in Figure S2.

It is worth noting that in the single spot correction, k was a constant in each spot cycle, which means all $eBC_{\text{compensated}}$ within the same cycle (e.g. a cycle last for several hours) have to share the same k. In contrast, time-resolved k can be determined for individual $eBC_{\text{compensated}}$ in the dual spot correction, which is a useful indicator for the mixing state (Drinovec et al., 2017). Zero test was conducted monthly for data quality control purpose.

The absorption Ångström exponent (AAE) can be determined by the multiwavelength measurement of AE33. AAE is a useful parameter to quantify the wavelength dependency of BC light absorption, as defined by the following equation (Moosmüller et al., 2011):

$$\frac{\sigma_{abs,\lambda 1}}{\sigma_{abs,\lambda 2}} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-AAE} \quad (7)$$

where $\sigma_{abs,\lambda 1}$ and $\sigma_{abs,\lambda 2}$ are the light absorption coefficients at the wavelengths of λ_1 and λ_2. The AAE of freshly emitted soot from vehicular emissions is close to 1 (Bond and Bergstrom, 2006; You et al., 2016). An increase of AAE could occur due to the coating of either BrC or non-absorbing materials. Samples that strongly influenced by BB, which are generally rich in primary BrC, can inflate AAE larger than 2 (Reid et al., 2005; Lewis et al., 2008; McMeeking et al., 2009; Pokhrel et al., 2016). Beside BB influence, an increase of AAE up to 1.5 due to coating of non-absorbing materials on the BC particles had also been observed in both model simulations (Lack and Langridge, 2013) and laboratory experiments (You et al., 2016).

2.3 OC and EC measurements

A filed carbon analyzer (Model RT-4, Sunset Laboratory Inc, Tigard, Oregon, USA) was used for OC and EC determination. The detailed sampling procedures can be found in our previous study (Wu et al., 2019), and only a brief description is given here. The sample was collected in the first 45 minutes of each hour at a flow rate of 8 L min^{-1}. The sample was analyzed in the next 15 minutes using thermo-optical analysis (Huntzicker et al., 1982). In the first stage, OC was vaporized by step-wise heating under helium (He) that provides an oxygen-free environment. In the second stage, carrier gas was shifted to oxygen (2% O_2 in He) to oxidize EC on the filter. The decomposition products of these two stages were converted to carbon dioxide (CO_2) by a manganese dioxide (MnO_2) catalyst, then detected by a non-dispersive infrared absorption (NDIR) detector. The instrument blank was analyzed on a daily basis. Filter was changed every 6 days to minimized the bias due to the accumulation of refractory materials on the filter (Jung et al., 2011).

2.4 Single particle mass spectrometry measurements

In the wet season, a Single Particle Aerosol Mass Spectrometer (SPAMS, Hexin Analytical Instrument Co., Ltd., China) was deployed at Jinan university atmospheric super site during 11 to 18 August 2017. In dry season, SPAMS data (15 November 2017 to 27 December 2017) from Guangdong environmental monitoring center (GDEMC) was used to characterize the EC-containing particles. The GDEMC site was located south to the JNU site (4 km). The operation principle of SPAMS had been introduced previously (Li et al., 2011), and only a brief introduction is given. The particles are introduced into the vacuum system through an 80 μm critical orifice, and then focused into a particle beam by the aerodynamic lens. As a result, the particles are accelerated to a size-dependent terminal velocity. The flight time of a known distance (6 cm) for individual particles is then detected by two orthogonally-orientated continuous laser beams (Nd:YAG, 532 nm) for particle size determination. Sized particles are individually vaporized and ionized by a 266 nm pulsed laser (Nd:YAG, 0.6 mJ). The generated positive and negative ions are then detected by a Z-shaped bipolar time-of-flight mass spectrometer. SPAMS data analysis was performed by the Computational Continuation Core (COCO, V3.2) toolkit based on the MATLAB software. In total, 327,453 and 2,212,688 particles with both positive and negative mass spectra were determined by the SPAMS in wet and dry season, respectively. Based on the ion marker criteria shown in Table S2, 120,351 and 595,180 EC-containing particles were identified in wet and dry season, respectively. EC-containing particles accounting for 37% and 27% of total detected particles in wet and dry season, respectively, which is comparable with a previous SPAMS study in Guangzhou (Zhang et al., 2015). EC-containing particles were further grouped into two categories, EC-fresh and EC-aged. EC-aged particles were extracted from EC-containing particles using the ion markers with the relative peak area (RPA) threshold listed in Table S2, including -97 $[HSO_4]^-$, -62 $[HNO_3]^-$, -46$[NO_2]^-$, 43$[C_2H_3O]^+$, etc. Once EC-aged particles were defined, the remaining EC-containing particles are considered as EC-fresh particles.

Despite the limitations in chemical composition quantification that associated with the matrix effects induced by laser desorption/ionization, SPAMS is a unique technique that can provide chemical composition on a single particle level. The major advantage of single particle analysis by SPAMS enables the characterization of coating martials exclusively on soot particles (Li et al., 2018b), while bulk analytical techniques are incapable of distinguishing whether the non-EC materials are internally or externally mixed with EC. Relative peak area (RPA), which was defined as the peak area of each marker ion divided by the peak area of total ions, has been recognized as an indicator of the relative amount of a species on a particle (Gross et al., 2000; Jeong et al., 2011; Hatch et al., 2014; Zhou et al., 2016). Therefore, RPA is used in this

255 study for SPAMS data analysis.

2.5 Auxiliary measurements

NO_2 was determined by a chemiluminescence analyzer (Model 42iTL, Thermo Scientific), while O_3 was measured by UV photometric analyzer (Model 49i, Thermo Fisher Scientific, Waltham, MA, USA). Span and zero calibrations for the gas analyzers were performed automatedly on a weekly basis. Meteorological factors
260 were measured by a multi-parameter sensor (Model WXT 520, Vaisala, Vantaa, Finland). The planetary boundary layer height (PBLH) measurements was conducted by a micro-pulse lidar (Sigma Space Co., USA) at the Guangzhou Meteorological Bureau (GMB, 23.00° N, 113.32° E, elevation: 43 m). Hourly backward trajectories for the past 72 hours were calculated using NOAA's HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory, version 4) model (Draxier and Hess, 1998) for both dry wand wet seasons.
265 Backward trajectory cluster analysis was conducted using MeteoInfo (Wang, 2014, 2019). Fire count data from Visible Infrared Imaging Radiometer Suite (VIIRS) on board the Suomi NPP weather satellite (Csiszar et al., 2014) was downloaded from the NASA FIRMS website (https://firms.modaps.eosdis.nasa.gov/) to generate the fire count map.

2.6 MAE_p estimation by MRS method

270 MAE_p is the key parameter in the E_{abs} calculation. In this study, MAE_p was determined by the newly developed MRS method (Wu et al., 2018), using EC as a tracer. The atmospheric aging induced additional light absorption (σ_{abs_aging}), which can be calculated by subtracting the absorption coefficient of primary aerosols, as shown in Eq.8 (a combination of Eqs. 1&4):

$$\sigma_{abs\_aging} = \sigma_{abs\_total} - MAE_p \times EC \quad (8)$$

275 In MRS calculation, the correlation (R^2) between measured EC and estimated hypothetical σ_{abs_aging} is examined as a function of a series of hypothetical MAE_p (MAE_{p_h}). Since σ_{abs_aging} was resulted from secondary processing while EC was coming from primary emissions, a MAE_{p_h} that leads to a minimum $R^2(EC, \sigma_{abs\_aging\_h})$ can best represents the independent nature between EC and σ_{abs_aging}. As a result, MAE_{p_h} at minimum $R^2(EC, \sigma_{abs\_aging\_h})$ corresponds to the authentic MAE_p. The detailed method evaluation
280 of MRS can be found in our previous paper (Wu et al., 2018) . Only a brief description on the calculation steps is provided here. EC from the Sunset carbon analyzer and σ_{abs_total} from AE33 are used as input variables. During the calculation of MAE_p by MRS, MAE_{p_h} is varied continuously in a reasonable range. At each MAE_{p_h}, corresponding hypothetical σ_{abs_aging} ($\sigma_{abs\_aging\_h}$) values are calculated for the dataset and

a correlation coefficient value (R^2) of EC vs. $\sigma_{abs\_aging\_h}$ (i.e., R^2(EC, $\sigma_{abs\_aging\_h}$)) is obtained. By searching the MAE_{p_h} in a desired range (e.g. from 0.1 to 50 with an interval of 0.1), a series of R^2(EC, $\sigma_{abs\_aging\_h}$) values are then plotted against the MAE_{p_h} values (Figure 2), which curve only has a single minimum point.

The σ_{abs_pri} is the part of light absorption from primary emitted soot particles. As a result, σ_{abs_pri} is well correlated with EC mass. In contrast, σ_{abs_aging} is the part of light absorption gained during the aging processes after emission. The variability of σ_{abs_aging} mainly depends on the coating thickness of the soot particles. Consequently, σ_{abs_aging} is independent of EC mass and the MAE_{p_h} corresponding to the minimum R^2(EC, $\sigma_{abs\_aging\_h}$) would then represent the authentic MAE_p.

It is worth noting that MAE_p by MRS represents the MAE_p at the emission source, which is conceptionally different from the MAE_p by the TD method. First, the morphology and optical properties of freshly emitted BC particles (chain-like aggregates) is different from that of thermally denuded BC particles (compact aggregates). Second, most of the coatings are removed for TD denuded BC particles, but freshly emitted BC particles usually come with a thin coating of OC formed from condensation of organic vapors due to the temperature gradient from the flame to the ambient air. As a result, the MRS-derived MAE_p is expected to be higher than the MAE_p by the TD method.

2.7 Secondary organic carbon (SOC) estimation by MRS method

OC can be separated into two categories based on the formation nature. Primary organic carbon (POC) can be emitted from traffic emission (Huang et al., 2014), biomass burning (Simoneit, 2002), trash burning and cooking (Mohr et al., 2009). Secondary organic carbon (SOC) can be formed through oxidation of volatile organic compounds (VOCs) or semi-volatile POC (Hallquist et al., 2009). The EC tracer method had been used extensively for SOC estimation (Turpin and Huntzicker, 1995):

$$POC = (OC/EC)_{pri} \times EC + OC_{non-comb} \quad (9)$$

$$SOC = OC_{total} - POC \quad (10)$$

Combing Eqs. (9)&(10):

$$SOC = OC_{total} - (OC/EC)_{pri} \times EC - OC_{non-comb} \quad (11)$$

$(OC/EC)_{pri}$ represents the overall OC/EC ratio of aerosols from primary emission sources, while $OC_{non\text{-}comb}$ represents primary OC from non-combustion process. $OC_{non\text{-}comb}$ can be determined from the intercept of OC

vs. EC linear regression. In this study, weighted orthogonal distance regression (WODR) was used to account for errors in both x and y variables (Wu and Yu, 2018). By grouping the data into percentile subsets using OC/EC ratio from the lowest to the highest (1–100%, with an interval of 1%), a series of intercepts were obtained as a function of OC/EC percentile (Figure S3). The intercept term in the OC vs. EC WODR is very small (-0.88 – -0.05) throughout the percentile range (1–100%). Since this term is small, $OC_{non\text{-}comb}$ was set to zero for SOC estimation in this study.

$(OC/EC)_{pri}$ is the key parameter for SOC calculation in the EC tracer method. In MRS method, the correlation (R^2) between measured EC and estimated SOC (from Eq. 10) was examined as a function of a series of hypothetical $(OC/EC)_{pri}$ ($(OC/EC)_{pri\_h}$). The OC/EC ratio at the minimum R^2 (EC vs. SOC) corresponds to the authentic primary OC/EC ratio (Millet et al., 2005). The detailed calculation steps can be found in our previous paper (Wu and Yu, 2016). Only a brief description is given here. In MRS calculation, $(OC/EC)_{pri\_h}$ was varied continuously in a reasonable range (e.g. from 0.1 to 10 with an interval of 0.1). Hypothetical SOC (SOC_h) values were calculated at individual $(OC/EC)_{pri\_h}$ for the whole dataset. A series of R^2 values of EC vs. SOC_h (i.e., $R^2(EC,SOC_h)$) were generated and then plotted against the $(OC/EC)_{pri\_h}$ values. Based on the assumption that variations of EC and SOC are independent, the $(OC/EC)_{pri\_h}$ corresponding to the minimum $R^2(EC,SOC_h)$ would then indicate the authentic $(OC/EC)_{pri}$ ratio.

In our previous work, numerical studies were performed and the results showed that the minimum R squared method (MRS) is more robust in SOC estimation than the minimum OC/EC and percentile OC/EC method (Wu and Yu, 2016). As a result, the MRS method had been gradually adopted for SOC estimation in recent studies (Xu et al., 2018b; Bian et al., 2018; Ji et al., 2018; Ying et al., 2018; Ji et al., 2019; Wu et al., 2019).

An Igor Pro (WaveMet rics, Inc. Lake Oswego, OR, USA) based computer program (Wu and Yu, 2016) was used to implement MRS calculation. Another two Igor Pro-based computer programs, Histbox (Wu et al., 2018) and Scatter Plot (Wu and Yu, 2018), were used for generating the box plots and scatter plots presented in this study. These computer programs (with operation manuals) can be downloaded freely from https://sites.google.com/site/wuchengust.

3. Results and discussions

3.1 Seasonality of carbonaceous aerosols concentrations and optical properties

The time series of EC, OC, optical properties and supporting measurements during the wet and dry seasons are shown in Figure S4. The hourly EC concentrations ranged from 0.43 to 7.40 and 0.54 to 12.04 μgC m^{-3} in wet and dry seasons, respectively. As for OC, the hourly average ranged from 0.32 to 13.84 and 0.51 to

25.31 μgC m^{-3} in wet and dry seasons, respectively. The hourly OC/EC ratios ranged from 0.25 to 6.92 and 0.33 to 8.69 in wet and dry seasons, respectively. In wet season, the wind direction is southeasterly dominated, bringing the relatively clean background air masses from the vast ocean. In dry season, the northeasterly wind prevails, which promotes the long-range transport of air pollutants from the east and central China.

As shown in Figure 1, EC, OC and OC/EC $AAE_{470\text{-}660}$ all exhibit clear seasonality. Average EC concentrations (with 1 standard deviation, hereafter) were 1.94±0.93 and 2.81±2.01 μgC m^{-3} in wet and dry seasons, respectively. The EC level was comparable to the measurements made in 2012 at a Guangzhou suburban site (1.67±1.35 μgC m^{-3} in wet season, 3.47±2.75 μgC m^{-3} in dry season) (Wu et al., 2019). The average concentrations of OC doubled in dry season (7.02±5.19 μgC m^{-3}) comparing to those in wet season (3.38±1.93 μgC m^{-3}), leading to elevated OC/EC ratio in dry season (2.56±0.94) in contrast to wet season (1.78±0.83). The hourly $AAE_{470\text{-}660}$ ranged from 1.14 to 1.67 and 1.07 to 1.76 in wet and dry seasons, respectively. As shown Figure S5a, $AAE_{470\text{-}660}$ observed in dry season (1.46±0.12) was significantly ($P<0.001$) higher than that in wet season (1.37±0.10).

Measured MAE_{520} in dry season (18.47±5.49 m^2 g^{-1}) is significantly ($P<0.001$) higher than that in wet season (10.73±4.96 m^2 g^{-1}), as shown in Figure S5b. The elevated MAE during dry season was likely a result of BB influences, which will be discussed in detail in Section 3.3.

The MAE_{p520} values determined by MRS were 8.6 and 16.8 m^2g^{-1}, for wet and dry seasons, respectively (Figure 2a & b). Similar to MAE_{520}, the increase of MAE_{p520} in the dry season was also likely a result of BB influence, which could lead to larger BC cores (Ditas et al., 2018) and with thicker primary coatings (Schwarz et al., 2008a; Kondo et al., 2011; Lack et al., 2012a; Liu et al., 2014). More details of the BB influences will be discussed in Section 3.3. Consequently, light absorption enhancement was found to be more pronounced in wet season (E_{abs520}=1.51±0.50, Table 2) than in dry season (1.29±0.28), because E_{abs} depends on the ratio of MAE to MAE_p, not their absolute values.

In summary, as evidenced by $AAE_{470\text{-}660}$ and MAE results, carbonaceous aerosols exhibit a strong seasonality in urban Guanghzou. This seasonality was associated with two seasonal factors, including contrasted direction of the prevailing wind, and diverse primary BC optical properties induced by seasonal BB influence.

3.2 Comparison of E_{abs} with previous studies

Filed measurements of E_{abs} values around the world are summarized in Table 2. Studies using the TD approach can achieve sub-hour time resolution, but studies using the TD approach had limited temporal coverage (normally less than a month). The AFD approach can potentially provide long-term E_{abs} results as

long as filter samples are available. However, the measurement duration of existing AFD studies was less than one month as shown in Table 2. The limited temporal coverage of existing AFD studies was likely due to the intense labor involved in filter treatment. In addition, the time resolution of existing AFD studies (8–24 hour) was not sufficient to fully resolve E_{abs} diurnal pattern. As a result, diurnal variations of E_{abs} values at different seasons were not covered in previous studies. In comparison, the MRS approach is a good alternative to explore the E_{abs} variations at both seasonal and diurnal scales. As shown in Table 2, low E_{abs} values were found in California (1.06@532 nm) (Cappa et al., 2012) and Japan (1.06@532 nm) (Ueda et al., 2016). Liu et al. (2017a) observed a moderate E_{abs} in UK (1.0-1.3@532 nm) and suggested that the small E_{abs} observed by Cappa et al. (2012) was a result of mixing state diversity. A recent study in California (Cappa et al., 2019) found moderate E_{abs} at Fresno (1.22@532 nm) but low E_{abs} at Fontana (1.07@532 nm), which was partially associated with unequal distribution of coating between different BC-containing particle types (Lee et al., 2019). In general, higher E_{abs} values had been observed in more polluted urban areas, such as France (Paris, 1.53@880 nm) (Zhang et al., 2018a) and India (Kanpur, 1.8@781 nm) (Thamban et al., 2017). High E_{abs} values had been reported in various locations in China. The E_{abs} value in the wet season in our study (1.51) is higher than that in Nanjing (1.42) (Ma et al., 2019) but lower than those in central China (Shouxian, 2.3) (Xu et al., 2018c), eastern China (Jinan, 1.9) (Bai et al., 2018) and northern China (Yuncheng, 2.25) (Cui et al., 2016b). The E_{abs} value in the dry season in our study (1.29) is lower than those in other locations in China, such as Beijing (1.66-4.0) (Xu et al., 2016; Zhang et al., 2018b), Nanjing (1.6) (Cui et al., 2016a), Xi'an (1.8) (Wang et al., 2014) and Jinan (2.07) (Chen et al., 2017). Since the collocated comparison of the three E_{abs} methods do not exist, a direct comparison between the three methods remain difficult. Nevertheless, a few studies, which conducted at the same city but during different periods, yielded comparable E_{abs} values. For example, E_{abs} in Nanjing by MAE method (1.6) (Cui et al., 2016a) was higher than that by the TD method (1.42) (Ma et al., 2019). This difference in E_{abs} might not only due to the different E_{abs} determination methods, but could also be a result of seasonal variations of E_{abs}.

3.3 Influence of biomass burning on the BC optical properties during dry season

Evidence from particle chemical compositions showed that BB influence was more intense in the dry season. Levoglucosan had been widely accepted as the tracer for BB in the $PM_{2.5}$ (Engling et al., 2006; Bhattarai et al., 2019). As shown in Figure S6, levoglucosan concentrations in Guangzhou were elevated by one order of magnitude during the dry season (159.33 ng m^{-3}) comparing to those in the wet season (35.93 ng m^{-3}). Besides levoglucosan, primary OC/EC ratio can also be used as an indicator of BB influence since the BB influence samples has a higher OC/EC ratio than that from traffic emissions (Schmidl et al., 2008; Pokhrel et al., 2016).

In this study, $(OC/EC)_{pri}$ determined by MRS in dry season (2.31) was higher than that in wet season (1.49), as shown in Figure S7. In addition, the northeasterly wind prevailed during the dry season, which favors long-range transport of aerosols from BB from central and eastern China to the PRD region. Remote sensing results also confirmed the more intense BB in dry season, as shown by gridded fire count map (Figure S8) determined by VIIRS.

As a result, the optical properties of BC were largely affected by the intense BB influences during the dry season. First, as shown in Figure S5b, significantly ($P<0.001$) higher MAE_{520} was observed in dry season (18.47±5.49) comparing to that in wet season (11.28±9.88). A previous field study at a suburban site in Guanghzou also reported the influence of BB on MAE, which observed a positive correlation between K^+ and MAE (Wu et al., 2018). High MAE from BB had been reported in BB emission studies as well (Roden et al., 2006; Schmidl et al., 2008; Levin et al., 2010; Wang et al., 2018a). Single particle soot photometer (SP2) studies have shown that BB influenced BC particles are more likely to have larger BC cores (Ditas et al., 2018), and with thicker initial coatings than those from vehicular emissions (Schwarz et al., 2008a; Kondo et al., 2011; Lack et al., 2012a; Liu et al., 2014). This is in good agreement with the MAE_{p520} obtained in the present study, which was almost doubled in dry season (16.8 m^2g^{-1}) comparing to that in wet season (8.6 m^2g^{-1}).

In dry season the E_{abs} showed little wavelength dependence (Figure 2d) despite the influence from BB. In this sense, the BB influence did substantially alter the optical properties of primary BC in the dry season, but the contribution of secondary BrC on E_{abs} was likely limited. The weak wavelength dependence of E_{abs} was also observed a previous study at a suburban site in Guanghzou (Wu et al., 2018). A previous study in Guangzhou also found that the seasonal difference of BrC light absorption contribution at 405 nm between dry season (15-19%) and wet seasons (12-15%) was small (Li et al., 2018d). In addition, the small seasonal difference of AAE between wet (1.37±0.10) and dry (1.46±0.12) seasons observed in this study also implies that secondary BrC contribution was not the dominating driver for AAE deviation from 1, which was the typical AAE for fresh soot without atmospheric aging. The results found in PRD were in contrast to a study in Paris, which found systematic higher E_{abs370} than E_{abs880} at wintertime due to the influence of BB (Zhang et al., 2018a).This discrepancy implies the complex linkage between BB and BrC optical properties.

The complex relationship between AAE and BrC can be affected by a variety of factors. First, the optical properties of primary BrC from BB exhibit large diversity in previous studies (Martinsson et al., 2015; Tian et al., 2019a) , which can be affected by fuel type and combustion conditions (Reid et al., 2005; Roden et al., 2006). Second, atmospheric aging can lead to AAE elevation through the formation of secondary BrC from

a variety of pathways (Moise et al., 2015; Laskin et al., 2015), including nitration of aromatic compounds (Jacobson, 1999), reaction of ammonia (Bones et al., 2010), bond-forming reactions between SOA constituents (Shapiro et al., 2009), reactions of BB products (Gilardoni et al., 2016; Kumar et al., 2018), photo-enhancement (Hems and Abbatt, 2018; Liu et al., 2016b; Ye et al., 2019), and aqueous-phase reactions (Lin et al., 2015; Tang et al., 2016; Xu et al., 2018a).On the other hand, AAE decrease could also occur during atmospheric aging (Romonosky et al., 2019), either induced by photo-bleaching of BrC (Adler et al., 2011; Zhong and Jang, 2011, 2014; Lee et al., 2014; Canonaco et al., 2015; Lin et al., 2016; Sumlin et al., 2017; Bhattarai et al., 2018; Fortenberry et al., 2018; Hems and Abbatt, 2018; Browne et al., 2019; Dasari et al., 2019; Li et al., 2019a; Wong et al., 2019; Cai et al., 2019; Liakakou et al., 2019; Ray et al., 2019), or aqueous-phase BrC degradation in the absence of light (Santos and Duarte, 2015; Santos et al., 2016b; Santos et al., 2016a; Fan et al., 2019). The relative contribution of secondary BrC formation and BrC degradation on the total BrC light absorption budget is still poorly understood. BrC degradation could be one of the reasons of the small seasonal AAE difference observed in the PRD region. More studies are needed by incorporating both time-resolved optical measurements and time-resolved detailed chemical speciation measurements to better understand the balance of BrC formation and degradation.

3.4 Diurnal dynamics of carbonaceous aerosols concentrations and optical properties

The diurnal variations of EC, OC, OC/EC, SOC, $AAE_{470-660}$ and E_{abs520} in wet and dry seasons are shown in Figure 3. Two peaks can be observed for EC (Figure 3a), one in the early morning (7:00) and the other in the evening (19:00), which reflects local traffic emissions in two rush hours. The lowest EC was found in the afternoon (14:00), likely associated with two factors considering the nature of EC source exclusive from primary emissions. The first factor is planetary boundary layer (PBL) height. As shown in Figure S9, the diurnal maximum PBL height was at 14:00 and 15:00 for wet and dry seasons, respectively. The fully developed PBL would help dilute the concentrations of primary pollutants (Deng et al., 2016; Liu et al., 2019a; Williams et al., 2019). The second factor is the diurnal variations of traffic volume. Previous studies (Yao et al., 2013; Xie et al., 2003) showed that traffic volume during 12:00 – 15:00 is lower than those in the morning and evening rush hours. To explore the effect of traffic volume, the weekday/weekend effect was investigated in Figure S10. The evening EC peak reduced substantially during weekend, implying that traffic volume has a strong influence on shaping the diurnal pattern of EC. The combination of these two factors leads to the reduced EC concentrations in the afternoon. The diurnal pattern of EC is similar between wet and dry seasons, but the magnitude was greatly elevated in the dry season.

OC exhibits a bimodal distribution (Figure 3b), peaking at 13:00 and 19:00, respectively. OC can be both primary and secondary, making its diurnal pattern different from that of EC. OC/EC also has two peaks as shown in Figure 3c. The first peak appeared at 13:00 and the second peak showed up at 17:00. It is worth noting that in wet season the afternoon OC/EC peak was higher than that in the evening OC/EC peak, while in dry season the reverse is true. The difference of diurnal OC/EC pattern between weekday and weekend is negligible (Figure S10), suggesting that the proportion of different vehicle types (e.g. diesel vs. gasoline) is relatively constant between weekday and weekend.

As shown in Figure 3d, two SOC peaks are observed in wet season, with the first SOC peak at 13:00 and the second SOC peak at 19:00. While in dry season the afternoon SOC peak was merged into the broadened evening peak. Despite the higher SOC concentrations observed in dry season, SOC formation was more active during the wet season as evidenced by the diurnal SOC/OC ratios (Figure 4a). The diurnal SOC/OC in wet season was always higher than that in dry season. It is worth noting that in wet season, despite that the SOC evening peak was comparable to the afternoon peak as shown in Figure 3d, the SOC/OC evening peak was smaller than the afternoon peak (Figure 4a). This observation implies that the SOC evening peak in the wet season was a result of the combination of pollutant accumulation (e.g. PBL decrease after sunset) and SOC formation, rather than the formation process alone. The small evening peak of SOC/OC in summer (19:00-21:00) would also be likely a result of condensation of semi-volatile organic compound (Warren et al., 2009; Pathak et al., 2008; Liang et al., 1997) due to the temperature decrease after sunset.

SOC/OC dependence on RH was investigated (Figure S11) to explore the effect of aqueous-phase secondary organic aerosol formation. During wet season, SOC/OC decreases as RH increases and the results were the same for both daytime and nighttime (Figure S11a&b). During nighttime when no solar radiation was supplied, higher RH leads to a lower SOC/OC (Figure S11b). This piece of evidence suggests that aqueous-phase reactions were unlikely the dominating pathway for SOC formation during the wet season. In dry season, SOC/OC did not show clear dependence on RH, suggesting that SOC formation is not sensitive to RH in dry season.

The diurnal trend of $AAE_{470\text{-}660}$ was similar between wet and dry seasons, which is higher in the evening and lower during the midday, but the magnitude of $AAE_{470\text{-}660}$ slightly increased during the dry season. The E_{abs520} exhibit different diurnal patterns between the wet and dry seasons. As shown in Figure 5, elevated E_{abs520} was found during nighttime in wet season but in dry season inflated E_{abs520} was observed in the afternoon. In addition, the degree of light absorption enhancement was more pronounced in the wet season. The influencing factors of dynamics are discussed in the following sections.

3.5 The diurnal correlations between AAE, *k* and E_{abs}

The loading effect correction factor used in AE33, *k*, has been found to be a useful indicator for the light absorption enhancement of BC (Drinovec et al., 2017). As shown in Figure 5a, in wet season a good anti-correlation was found between k_3 (*k* value for 520 nm) and E_{abs520} with a R^2 of 0.74. In dry season, such anti-correlation was substantially weakened (R^2 = 0.20) as shown in Figure 5b, likely due to the influence of BB. These results agree well with the findings reported by Drinovec et al. (2017) that *k* can be used as a BC particle coating indicator without the influence of BB. As shown in Figure 5a, a good correlation was found between $AAE_{470\text{-}660}$ with E_{abs520} (R^2=0.71). Considering the weak BB influence in wet season as discussed in Section 3.3, atmospheric aging induced coating on BC particles was more likely the dominating driver of $AAE_{470\text{-}660}$ dynamics during the wet season in the PRD region. The presence of coating of BC could also explained that despite the BB influence is small in wet season, the observed average AAE (1.37±0.10) was significantly higher than the AAE of fresh BC (~1). This result is also consistent with previous studies that found non-light-absorbing coating can lead to elevated AAE up to 1.5 (Lack and Cappa, 2010; Lack and Langridge, 2013). In dry season, the variability of AAE was governed by both coating thickness and BB influence, thus leading to a degraded R^2 (0.22) between $AAE_{470\text{-}660}$ and E_{abs520} as shown in Figure 5b. A recent study showed that the diurnal pattern of BrC was moderately correlated with a BB tracer K^+ in the PRD region during the dry season (Li et al., 2019c), implying that BB did have considerable influence on AAE viability during dry season.

The spectral fingerprints of *k* were shown in Figure S12. Observations in Europe showed that the presence of BrC could lead to increased *k* at longer wavelengths (Drinovec et al., 2017). Our observations showed that the seasonal difference in spectral fingerprints of *k* between wet and dry seasons is small. Considering the limited increase of AAE in dry season and similarity of seasonal spectral fingerprints of *k*, these results suggest that, in the PRD region, despite the BB influence in dry season effectively altered the optical properties of BC aerosols, there was likely limited secondary BrC contribution on E_{abs} during the dry season, which is in agreement with discussions in Section 3.3.

3.6 The influence of secondary processing on E_{abs} diurnal dynamic

Photochemical reactions play an important role in the aging process of black carbon, leading to the modification of BC morphology and optical properties as evidenced by laboratory studies (Saathoff et al., 2003; Schnaiter et al., 2005; Martinsson et al., 2015; Pei et al., 2018) and quasi-atmospheric chamber studies (Peng et al., 2017; Peng et al., 2016). Filed studies at various locations have also showed that photochemical processing can promote the light absorption enhancement of BC, including in Beijing (Liu et al., 2019b),

Yangtze River Delta (Xu et al., 2018c), Xi'an (Wang et al., 2017c), Los Angels (Krasowsky et al., 2016) and Toronto (Knox et al., 2009). The concentration of odd oxygen ($O_x = O_3+NO_2$) proposed by Liu (1977) and (Levy II et al., 1985) have been widely used as the indicator of photochemical aging. In this study, the diurnal correlations between O_x and E_{abs520} were investigated to explore the effect of photochemical processing. As shown in Figure 4a, in wet season O_x and E_{abs520} peaked at 15:00 and 0:00, respectively. The O_x experienced a continuous decline since 15:00 until the sunrise of the next day, but the growth of E_{abs} extended to midnight. The nighttime E_{abs520} peak suggests that the increase of coating can be achieved without the presence of solar radiation. These differences in the diurnal patterns led to a low correlation between O_x and E_{abs} (R^2=0.01). This result implies that in wet season the diurnal variability of E_{abs520} was unlikely dominated by photochemical reactions, despite that O_x was more pronounced in the wet season. As for dry season (Figure 4b), both O_x and E_{abs520} peaked at 17:00 leading to a good correlation with a R^2 of 0.69, suggesting that photochemical reactions could be one of the main drivers for E_{abs} diurnal variations. This result strongly indicates that BC light absorption can be markedly amplified through photochemical reactions. Our dry-season results are consistent with a previous study in Northern China (Wang et al., 2017c), which also showed the dependence of light absorption enhancement on O_x during the wintertime.

In the meantime, the formation of SOC also contributes to light absorption enhancement of BC, which had been observed in both field studies (Moffet and Prather, 2009; Wang et al., 2017a; Zhang et al., 2018a) and laboratory studies (Schnaiter et al., 2005; Lambe et al., 2013; Saliba et al., 2016). In this study, the effect of SOC formation on E_{abs} was investigated using SOC/OC as the indicator rather than using SOC alone. The advantage of using SOC/OC is that the SOC variations induced by non-secondary-formation process (e.g. PBLH shallowing) can be minimized, thereby focusing the analysis on the effect of secondary formation processes. A good diurnal correlation between SOC/OC and E_{abs520} were observed in dry season (R^2=0.53), but no correlation was found in wet season (R^2=0.01). The E_{abs} dependence on SOC/OC was examined in Figure 6. E_{abs} dependence on SOC/OC was found in both wet and dry seasons, but a clearer dependence was observed in the dry season. It should be noted that a good E_{abs} dependence on SOC/OC observed in Figure 6 does not necessarily lead to a good diurnal correlation between E_{abs} and SOC/OC (e.g. Figure 4a). In other words, the dependency of E_{abs} on SOC/OC might not necessarily be reflected in the form of correlation on a diurnal scale. Thus, the poor diurnal correlation between SOC/OC and E_{abs520} observed in the wet season (Figure 4a) cannot rule out the contribution of SOC on E_{abs520}. A study in Paris (Zhang et al., 2018a) found that more oxidized oxygenated organic aerosols (MOOOA) and less oxidized OOA (LO-OOA), which are surrogates of SOA, were the dominating contributors for E_{abs}, especially in summertime. In the present study,

due to the lack of quantitative chemical speciation data, quantification of contributions from different chemical species on E_{abs} is not possible. A recent study in Guangzhou (Wu et al., 2019) found that traffic-derived SOC could be a significant source of SOC in the urban area, which can account for half of the total SOC. In that sense, traffic emissions are expected to have a considerable contribution to BC light absorption enhancement in both wet and dry seasons.

The temperature effect on E_{abs} was examined in Figure 6c&d for wet and dry seasons, respectively. In the wet season, a positive response of E_{abs} on temperature was observed for the temperature range of 24 ±□30 ºC, implying a favorable condition for coating formation on BC particles. However, further temperature increment beyond 30 ºC led to decline of E_{abs}, which might be associated with the evaporation of the coating materials on BC. In the dry season, E_{abs} was not sensitive to temperature for the range of 12 ±□24 ºC. The volatility effect of coating materials on E_{abs} will be discussed in more details in the next section.

3.7 The influence of semi-volatile compounds on E_{abs} diurnal variations

The SPAMS data from both wet (August 11-18, 2017) and dry season (15 November 2017 to 27 December 2017) were analyzed to explore the mixing state of EC-containing particles from a single-particle perspective. The average EC-fresh and EC-aged mass spectra are shown in Figure S13 for both wet and dry seasons. The domination of EC-aged particles in EC-containing particles number fraction suggest that most of the EC particles are internally mixed with other species (Table S3). This result agrees with previous studies in this region (Zhang et al., 2013; Zhang et al., 2014).

To study the relative abundance of coating materials on EC particles, we investigate the ratios of RPA by different species (organics, sulfate and nitrate) to RPA by EC in both wet and dry seasons (Figure 7). In wet season, organics and sulfate on EC-containing particles demonstrated similar diurnal trends that both peaked at 13:00, implying the association with photochemical reactions. The timing of the organic peak by SPAMS shown in Figure 7a also agrees well with the bulk measurements of SOC/OC (Figure 4a). However, the diurnal variations of organics and sulfate were poorly correlated with E_{abs520} as shown by the low R^2 in the scatter plot in Figure 7a&b. There are two possibilities for the lower R^2 (Organics, E_{abs}) in Figure 7d comparing to R^2(SOC/OC, E_{abs}) in Figure 4b. First, organics shown in Figure 7d contain both primary and secondary organics, while SOC/OC shown in Figure 4b represents the secondary portion only. Second, poor diurnal correlations do not necessarily rule out the contribution of organics and sulfate to E_{abs} by analogy with the SOC correlation with E_{abs} as discussed in Section 3.6. Although the quantitative contribution estimation of sulfate and SOA to E_{abs} is not possible in this study, a rough estimation can be projected. Considering the typical annual average SOC concentration (3 μgC m^{-3})(Wu et al., 2019), typical SOA/SOC mass ratio (1.8)

(Li et al., 2017), and sulfate concentration (8 μg m^{-3})(Liu et al., 2017b) in the PRD region, SOA and sulfate would likely have comparable contributions to the $E_{abs,}$ according to the E_{abs} dependency on sulfate-to-SOA mass ratio results by Zhang et al. (2018a). Summertime nitrate was low in daytime and high in nighttime (Figure 7c), which agrees with measurements at the roadside site in Hong Kong (Lee et al., 2015) and Shanghai (Li et al., 2018b). Temperature-dependent gas-particle partitioning would be one of the possible reasons for the observed nitrate diurnal pattern (Appel et al., 1981; Xue et al., 2014; Griffith et al., 2015). Higher temperature during the daytime (Figure S14) favors HNO_3 partitioning into the gas-phase in wet season. The diurnal pattern of nitrate correlates well with that of E_{abs520} (R^2=0.59) as shown in Figure 7c, suggesting that E_{abs520} was likely affected by temperature-induced gas-particle partitioning during wet season. Studies in Nanjing (Ma et al., 2019) and Beijing (Xie et al., 2019) also observed nitrate evaporation induced E_{abs} decrease, which is in agreement with our study. A previous chamber study had shown the decrease of E_{abs} due to SOA evaporation (Metcalf et al., 2013). By analogy with nitrate, organic compounds with a volatility similar to nitrate might potentially involve in shaping the diurnal pattern of E_{abs} in the wet season.

In dry season, organics were moderately correlated with E_{abs520} (R^2=0.38) as shown in Figure 7d. The improved correlation of organics in dry season was in agreement with O_x and SOC/OC results as shown in Figure 4b. Sulfate was still poorly correlated with E_{abs520}. Since the contribution of sulfate on E_{abs520} cannot be ruled out, one possible explanation is that the contribution of sulfate on E_{abs520} was not reflected on the diurnal time scale.

5. Conclusions and implications

This study explored the temporal dynamics of optical properties of carbonaceous aerosols in urban Guangzhou, a typical megacity in southern China, focusing on the atmospheric aging induced light absorption enhancement of BC. Field measurements were conducted at an urban site during wet season (July 31–September 10, 2017) and dry season (November 15, 2017–January 15, 2018). A newly developed approach, the minimum R squared (MRS) method (Wu et al., 2018), was successfully applied to determine the light absorption enhancement factor, E_{abs}, using data from an Aethalometer and a field-deployable semi-continuous carbon analyzer. The MRS approach avoids specialized instrument setup (e.g. thermal denuder and photo-acoustic spectrometer) for E_{abs} determination, hence has a great potential for expending data pool of E_{abs}, considering the fact that collocated Aethalometer and field carbon analyzer measurements have been widely deployed around the world.

A strong seasonality of BC was observed. The average concentration of EC was 1.94±0.93 and 2.81±2.01 μgC m^{-3} in the wet and dry seasons, respectively. Collective evidence from remote sensing fire counts and

ground measurements of levoglucosan showed that biomass burning (BB) was more active in the dry season. Consequently, optical properties of BC were effectively altered, leading to elevated MAE (dry season: 18.47±5.49 m^2 g^{-1}, wet season: 10.73±4.96 m^2 g^{-1}), MAE_p (dry season: 15.8 m^2 g^{-1}, wet season: 8.1 m^2 g^{-1}) and AAE (dry season: 1.46±0.12, wet season: 1.37±0.10) in dry season comparing to those in wet season. However, little dependence of E_{abs} on wavelength was observed in dry season despite the influence from BB. The diurnal correlation analysis between AAE, *k* and E_{abs} revealed different results between wet and dry seasons. During the wet season when BB influence was small, AAE was well correlated with E_{abs}, implying that coating was likely the main driver for AAE>1. In other words, the two-component AAE model might not be suitable for BrC absorption estimation under such circumstance. The aethalometer loading effect correction factor, *k*, was confirmed to be a useful E_{abs} indicator owing to its good correlation with E_{abs} during the wet season. In dry season, the weak correlation between AAE and E_{abs} implies the contribution from BB on AAE. In dry season, the BB influence leads to poor correlation between *k* and E_{abs}, confirming that *k* can only be used as the coating indicator when BB influence is small.

The effect of atmospheric aging on E_{abs} diurnal pattern was examined. O_x and SOC/OC were found well correlated with E_{abs} during the dry season but no correlation was observed in the wet season. However, further analysis showed E_{abs} dependence on SOC/OC in both wet and dry season. This observation implies that poor diurnal correlation in wet season does not necessarily rule out the E_{abs} contribution from SOC. In other words, the SOC contribution on E_{abs} in wet season was not necessarily be reflected in mere diurnal correlation.

In the wet season, high temperature (> 32ºC) induced E_{abs} decline was observed. In addition, a good diurnal correlation between nitrate and E_{abs} was found. These evidences imply the potential role of semi-volatile coatings on BC in regulating the diurnal dynamics of E_{abs}. In China, the sulfate problem had been effectively mitigated by the reduction measures implemented in recent years (Xia et al., 2016; Wang et al., 2017b). In contrast, nitrate increased substantially in the recent years (Xu et al., 2019; Tian et al., 2019b; Li et al., 2019b; Wang et al., 2019). If the nitrate fraction in the coating materials on BC increases, the diurnal pattern of E_{abs} for BC may be affected by the fluctuation of nitrate content in aerosol particles. As a result, the increasing concentration of nitrate might potentially affect radiative forcing by BC in China.

Appendix

Table A1. Abbreviations.

Abbreviation	Definition
$AAE_{470-660}$	Ångström absorption exponent between 470 and 660 nm
AFD	aerosol filter filtration-dissolution
Aeth	Aethalometer
BB	biomass burning
BC	black carbon
BrC	brown carbon
E_{abs520}	light absorption enhancement factor at 520 nm
σ_{abs520}	light absorption coefficient at 520 nm
σ_{abs_total}	total light absorption coefficient of a coated particle
σ_{abs_pri}	primary light absorption coefficient attributed to the soot core alone of a coated particle
σ_{abs_aging}	extra light absorption other than σ_{abs_pri}, including those from the lensing effect arise from non-absorbing coating on the soot core and secondary brown carbon during atmospheric aging
eBC	equivalent BC mass concentration determined by optical methods (e.g. aethalometer)
GDEMC	Guangdong environmental monitoring center
$k_1, k_2 \ldots k_7$	compensation factors (Eqs. 5 & 6) at 7 wavelengths (370,470,520,590,660, 880 and 950 nm).
MAE_{520}	mass absorption efficiency at 520 nm, also known as mass absorption cross section (MAC)
MAE_p	primary MAE of freshly emitted soot particles
MAE_{p_h}	a series of hypothetical MAE_p tested in MRS calculation
MAE+emprical	MAE approach for E_{abs} quantification using emprical MAE_p
MAE+SP	MAE approach for E_{abs} quantification using single particle measurements for MAE_p determination
MAE+MRS	MAE approach for E_{abs} quantification using MRS for MAE_p determination
MRS	minimum R squared method
OC	organic carbon
PRD	Pearl River Delta region, China
rBC	refectory black carbon (commonly used for reporting BC detected by SP2)
r_{aged}	the ratio of aged particles to fresh particles determined by SP2
RPA	relative peak area
SP2	Single-Particle Soot Photometer
SPAMS	Single Particle Aerosol Mass Spectrometer
SP-AMS	Soot Particle Aerosol Mass Spectrometer
SSA	single-scattering albedo
TD	thermodenuder

Author contributions

C.W. designed the study. J.Y.S. and C.W. performed the experiments. J.Y.S., C.W., C.C., Q.Z. and Y.L. conducted the data analysis. J.Y.S. and C.W. wrote the paper with the inputs from all authors.

Data availability

OC, EC, and σ_{abs} data used in this study are available from corresponding authors upon request.

Competing interests

The authors declare that they have no conflict of interest.

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Table 1. Comparisons of three E_{abs} determination approaches.

Approach		Time resolution	Temporal coverage	E_{abs} determination	Instrument	Advantages	Limitations
TD		minutes	weeks	$\boldsymbol{E_{abs}} = \frac{\sigma_{abs\_total}}{\sigma_{abs\_pri}}$	TD+PAS	very high time resolution	TD temperature selection is tricky; denuded particle morphology different from emission
AFD		daily	years		Filter sampler + off-line OCEC	can be applied on archived filter samples	labor intensive; only remove soluble coating; low time resolution
MAE	**MAE + MRS**	hourly	years	$\boldsymbol{E_{abs}} = \frac{MAE_t}{MAE_p}$	Aeth + online OCEC	high time resolution	MRS has minimum data points requirement, not suitable for a small dataset
	MAE + empirical	hourly	months		Aeth + online OCEC		empirical MAE_p could be unrealistic for the sampling site
	MAE + SP	hourly	weeks		PAS/Aeth + SP2/SP-AMS		expensive instruments; limited sampling duration

Table 2. Comparisons E_{abs} measurements in various field studies.

Approach	Location	Sampling period	Sampling duration	Time resolution	λ nm	E_{abs}	Reference
MAE + MRS	Guangzhou,China (Urban)	Jul–Sep 2017	6 months	1 hr	520	1.51±0.50	This study
		Nov 2017–Jan 2018				1.29±0.28	
	Guangzhou,China (Suburban)	Feb 2012–Jan 2013	1 year	1 hr	550	1.50±0.48	(Wu et al., 2018)
MAE + emprical	Nanjing, China (Suburban)	Nov 2012	15 days	12 hr	532	1.6	(Cui et al., 2016a)
	Beijing, China (Suburban)	Nov 2014–Jan 2015	2 months	1 hr	470	2.6–4.0	(Xu et al., 2016)
MAE + SP	Beijing, China (Urban)	Nov 2014	14 days	1 hr	/	1.66-1.91	(Zhang et al., 2018b)
	Manchester, UK (Urban)	Oct–Nov 2014	13 days	5 min	532	1.0–1.3	(Liu et al., 2017a)
	Paris, France (Urban)	Mar 2014–Mar 2017	3 years	24 hr	880	1.53±0.39	(Zhang et al., 2018a)
	Kanpur, India (Urban)	Jan–Feb 2015	2 months	1 hr	781	1.8	(Thamban et al., 2017)
	Xi'an, China (Urban)	Dec 2012–Jan 2013	1 month	20 min	870	1.8	(Wang et al., 2014)
TD	Shouxian, China (Rural)	Jun-Jul 2016	8 days	10 min	532	2.3 ± 0.9	(Xu et al., 2018c)
	Beijing, China (Urban)	Jun 2017	10 days	20 min	630	1.59±0.26	(Xie et al., 2019)
	Sacramento, USA (Urban)	Jun-Jul 2010	13 days	20 min	532	1.06± 0.01	(Cappa et al., 2012)
	Fresno, USA (Urban)	Dec 2014–Jan 2015	19 days	10 min	532	1.22± 0.15	(Cappa et al., 2019)
	Fontana, USA (Urban)	July 2015	23 days		532	1.07± 0.22	
	Suzu, Japan	April-May 2013	26 days	20 min	532	1.06	(Ueda et al., 2016)
	Nanjing, China (suburban)	Aug–Sep 2014	16 days	2 hr	405	1.41± 0.39	(Ma et al., 2019)
					532	1.42± 0.40	
					781	1.35± 0.38	
AFD	Jinan, China (Urban)	Feb 2014	13 days	8 hr	678	2.07± 0.72	(Chen et al., 2017)
	Yuncheng, China (Rural)	Jun–Jul 2014	18 days	12 hr	678	2.25± 0.55	(Cui et al., 2016b)
	Jinan, China (urban)	Jul–Jul 2016	30 days	24 hr	678	1.9± 0.7	(Bai et al., 2018)
	Mt. Tai, China (Rural)	July–Aug 2014	31 days	24 hr	678	2.0± 0.3	

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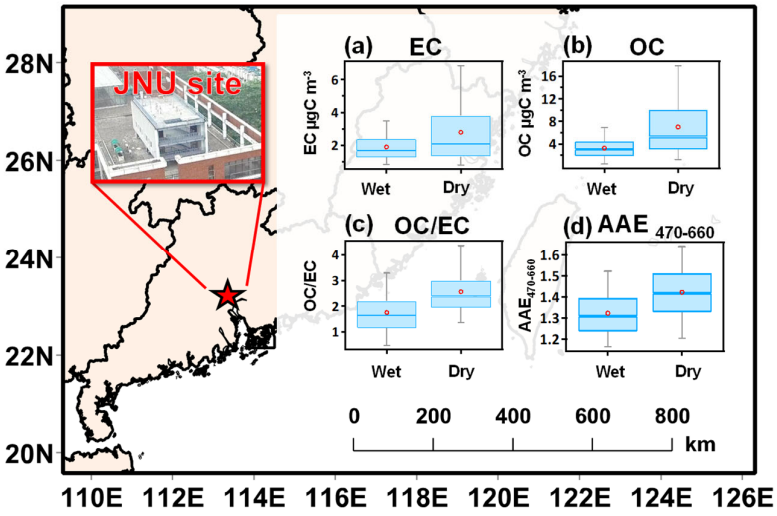

Figure 1. The location of the observation site (JNU). (a), (b), (c) and (d) show the box plots of EC, OC, OC/EC and $AAE_{470\text{-}660}$ during wet (July– September) and dry (November–January) seasons, respectively. Red circles represent the seasonal average. The line inside the box indicates the median. Upper and lower boundaries of the box represent the 75th and the 25th percentiles; the whiskers above and below each box represent the 95th and 5th percentiles.

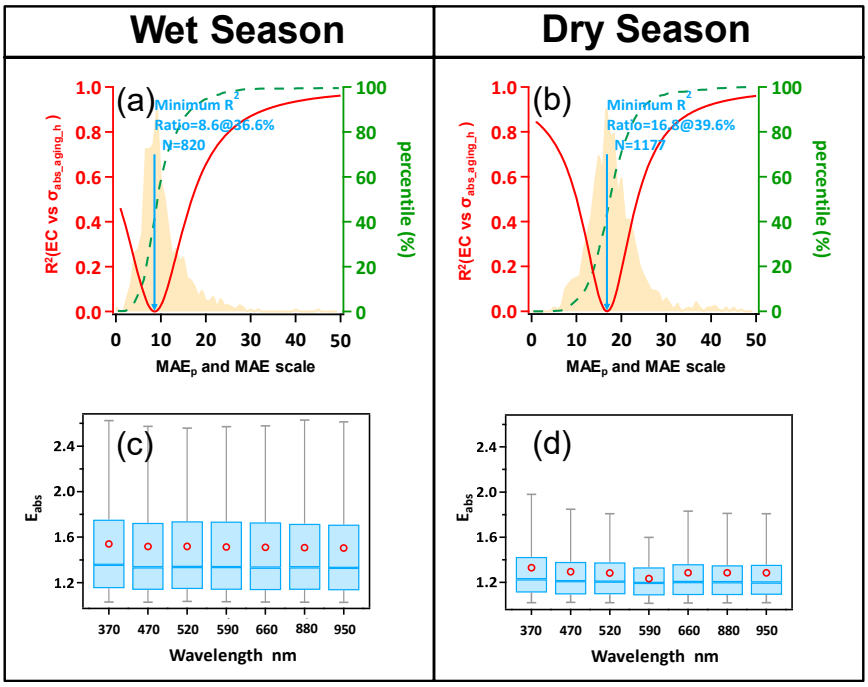

Figure 2. E_{abs} determination by MRS. (a) Wet season MAE_p determined by MRS at 520 nm. The red curve represents the correlation coefficient (R^2) between hypothetical σ_{abs_aging} (σ_{abs_total} – EC * MAE_p) and EC mass as a function of MAE_{p_h}. The shaded area in light tan represents the frequency distribution of observed MAE. The dashed green line is the cumulative distribution of observed MAE. (b) same as (a) but for dry season. (c) Spectral E_{abs} determined by MRS in wet season. Red circles represent the average values. The line inside the box indicates the median. Upper and lower boundaries of the box represent the 75th and the 25th percentiles; the whiskers above and below each box represent the 95th and 5th percentiles. (d) Spectral E_{abs} in dry season.

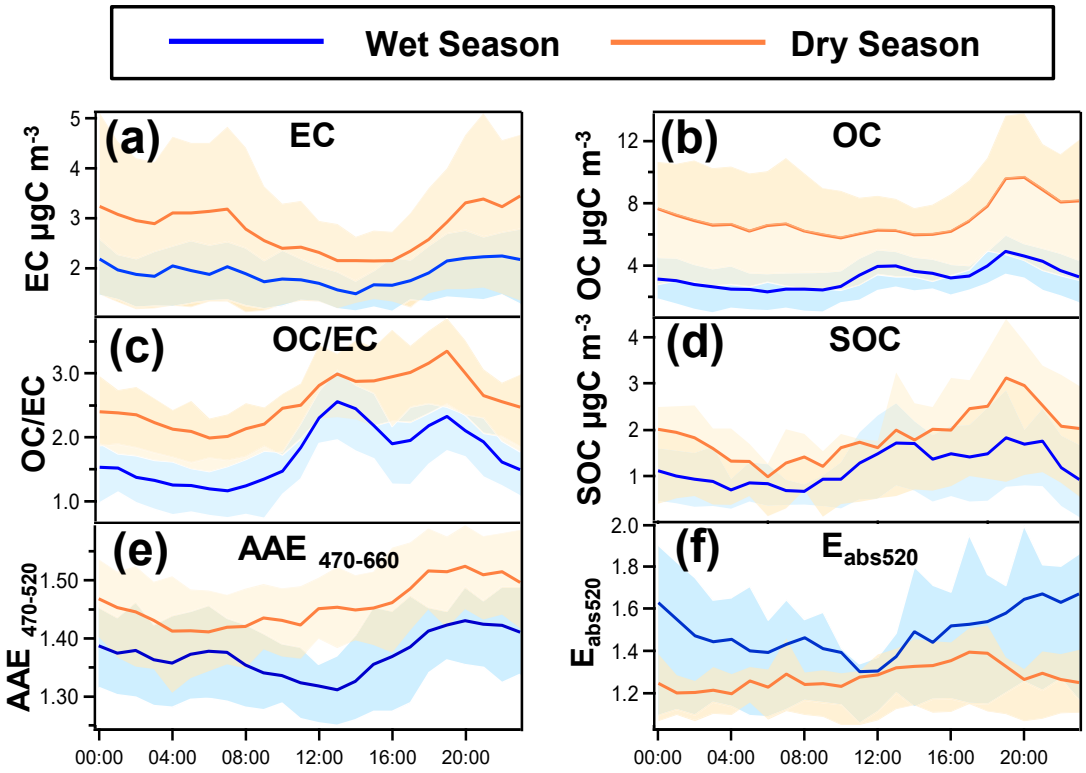

Figure 3. Diurnal pattern carbonaceous aerosols in wet and dry season. The solid lines represent hourly averages and the shaded areas represent 25th% and 75th% percentile. (a) EC. (b) OC. (c) OC/EC ratio. (d) SOC. (e) $AAE_{470-660}$. (f) E_{abs520}.

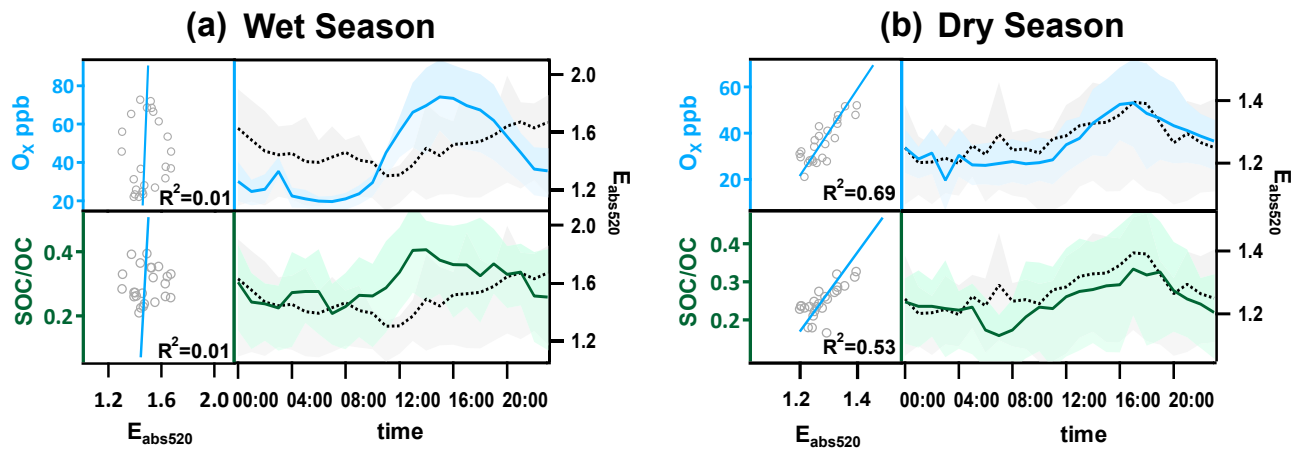

Figure 4. The effect of secondary process on E_{abs520}. (a) Diurnal pattern of E_{abs520} and O_x in wet season. The lines (O_x in blue, SOC/OC in green and E_{abs520} in black dot lines) represent hourly averages and the shaded areas represent 25th% and 75th% percentile. (b) Same as (a) but in dry season.

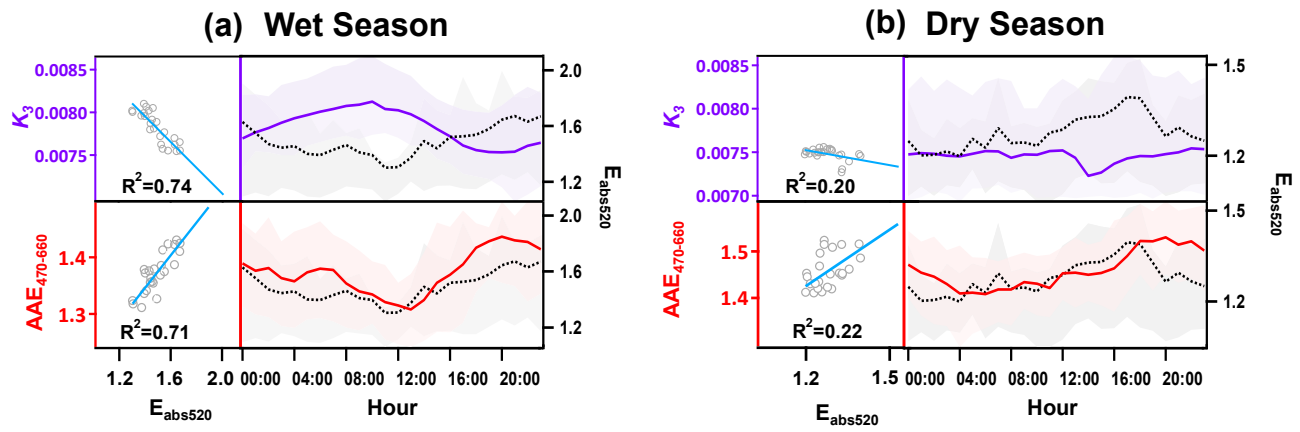

Figure 5. diurnal patterns of E_{abs520} $AAE_{470-660}$ and k_3. The lines (k_3 in purple, $AAE_{470-660}$ in red and E_{abs520} in black dot lines) represent hourly averages and the shaded areas represent 25^{tho}% and 75^{tho}% percentile. (a) Wet season. (b) Dry season.

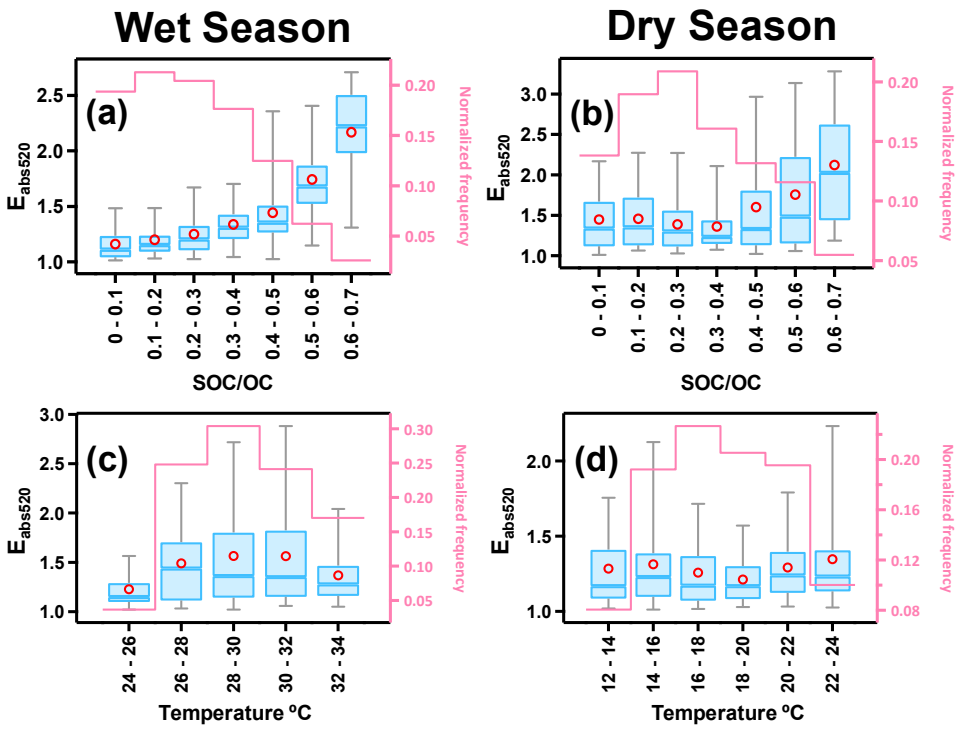

Figure 6. E_{abs520} dependency on SOC/OC ratio and temperature during wet and dry seasons. Red circles represent the average values. The line inside the box indicates the median. Upper and lower boundaries of the box represent the 75th and the 25th percentiles; the whiskers above and below each box represent the 95th and 5th percentiles. The pink lines represent normalized frequency of data points in each bin.

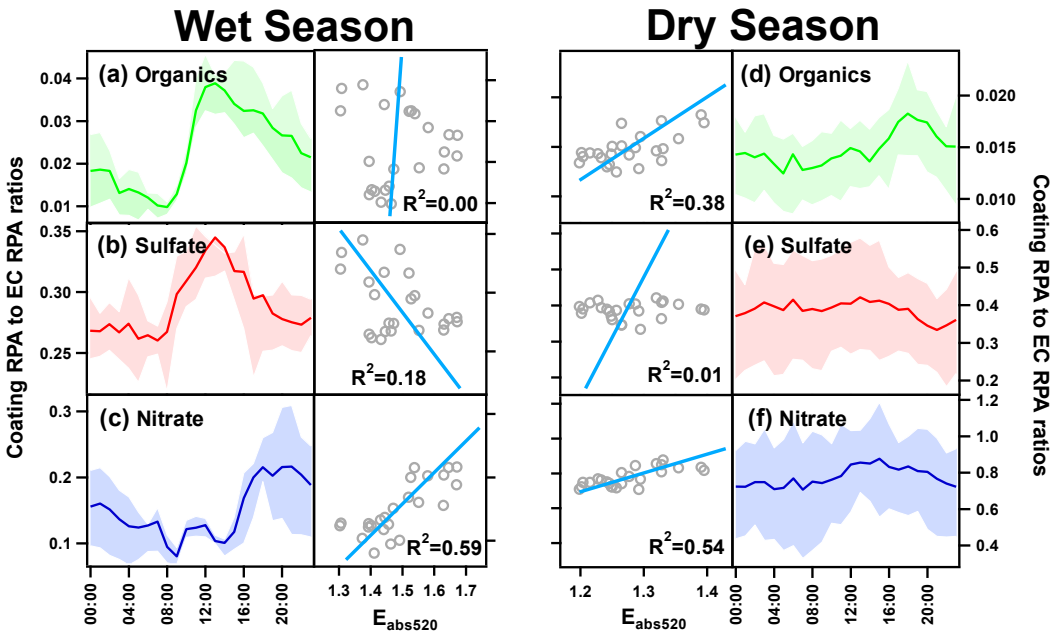

Figure 7. Diurnal variations of coating (including organics, sulfate and nitrate) RPA to EC RPA ratios of EC-aged particles measured by SPAMS in wet and dry seasons. The solid lines represent hourly averages and the shaded areas represent 25th% and 75th% percentile. (a) ~(c) In wet season, organics, nitrate and sulfate RPA to EC PRA ratios. The scatter plots show the corresponding correlations with E_{abs520}. The scatter plots share the same y axis scale with the diurnal plots. (d) ~(f) are the same as (a) ~(c) but for the dry season. Following ions are used: EC (m/z +12$[C]^+$, +24$[C_2]^+$, +36$[C_3]^+$, +48$[C_4]^+$), organics (m/z +43$[C_2H_3O]^+$), sulfate (m/z -97 $[HSO_4]^-$, -80$[SO_3]^-$) and nitrate (m/z -62 $[NO_3]^-$, -46$[NO_2]^-$).