

## Reviewer #01

### General comments:

Che et al. reported sunphotometer measurements of aerosol microphysical and optical properties and modeling of aerosol surface and top-of-the-atmosphere (TOA) radiative forcing (RF) at several ground stations of eastern China. This manuscript is poorly written and needs a major overhaul. In many places, discussions on the results are groundless without providing any backing evidence or appropriate references to the literature. Most of the discussions are too superficial to provide any value or interest to the aerosol-climate-change or air-pollution communities. The logical flow is confusing and unnatural, with potential for improvement in the organization of sections. Some sections need to be rewritten and reorganized to remove redundancy, while additional information must be provided in a few places.

Response: Thanks for the reviewer's important and constructive comments and suggestions, the major overhaul has been done in some sections which need to be rewritten and reorganized according to the helpful suggestions. The discussions and the logical flow improvement in the organization in the revised manuscript have been rewritten and reorganized and some redundancy section has been removed and concise.

## Reviewer 1

1. Title of the manuscript needs to be changed. Title implies that the study reports aerosol instantaneous radiative forcing, while monthly and annual RF are shown instead. Remove "instantaneous". "Temporospatial" is awkward. Consider using "spatiotemporal" instead. Even worse, the author mentioned nothing whatsoever in the manuscript about the purpose or the advantage of such high temporospatial data (more comments on this later on). "Seven sites" should not be treated as "high spatial resolution". Consider writing the full name of CARSNET in the title.

Response: Thanks for the suggestions. The title of the manuscript has been changed as "Aerosol optical properties and radiative forcing based on synchronous measurements of China Aerosol Remote Sensing Network (CARSNET) over eastern China" according to the reviewer's important comments.

2. Abstract needs to be rewritten. The common flow of logic is to discuss the aerosol microphysical properties (size, refractive index) first, then optical properties (Kext, FMF, SSA, etc), and finally RF. This comment also applies to the main body text. I understand that the author is trying to follow the way how these variables are derived from the AERONET inversion algorithm. I do not think this is necessary. The result subsections need to be reorganized, combined, and shortened to make the paper more concise and less confusing.

Response: According to the helpful suggestions of reviewer, the section of Abstract has been rewritten in the revised manuscript. In the Abstract, the aerosol microphysical properties of volume size distribution has been described firstly, then the aerosol optical properties including AOD, SSA, refractive index, AAOD and finally RF has been written. This order has also applies to the main body text to make the paper more concise and less confusing.

3. L38: what wavelength is the AOD value? If it is 440 nm, why not use 500 nm or 550nm to make the result more useful for the community and more easily comparable to past studies? Optical properties are wavelength dependent. Provide this information when introducing any variables at the first time.

Response: The wavelength of AOD is at 440nm in the sentences of "*The aerosol optical depth (AOD<sub>440nm</sub>) varied from 0.68 to 0.76,*" in line 45-46 in Abstract. Consider of the suggestions of reviewers, we added the section of monthly and diurnal AOD at 500 nm in section 3.2 from line 364 to 384 as well as figure 4 and 5 as follows: "*The monthly and diurnal cycle of AOD at 500nm has also been discussed in Fig.4 and Fig.5. The annual values of AOD<sub>500nm</sub> over the seven urban, suburban and rural sites in this study varied from 0.53 (ChunAn) to 0.68 (Hangzhou). The results show that two peaks of AOD at 500nm occurs in June and September with values of  $1.25 \pm 0.59$  and  $1.00 \pm 0.42$  in the urban site of Hangzhou, respectively which has the similar pattern as the other sites. The increase of AOD at 500nm in June is not corresponding to the same increase pattern of EAE (about 1.5) which indicates the aerosols types may be relatively constant in this region. The Fig.5 depicts the diurnal patterns of AOD at 500nm in this megacity area of eastern China. We can see that there are two types of diurnal patterns in this region. The daily AOD has been found increased in early morning (08:00 hr to 09:00 hr) and afternoon (12:00 hr to 14:00 hr) about the value of 0.60 to 0.70 in Hangzhou,*

Xiaoshan, Fuyang and Linan, while the decreasing of daily AOD has been observed from 0.70 to 0.50 during the daytime (from 07:00 hr to 16:00 hr) in Tonglu, Jiande and ChunAn. The high AOD during 07:00~09:00 in the urban area may be due to the anthropogenic activities and aerosol emissions from the morning rush hour. The decreased AOD with the value of  $0.37 \pm 0.36$  occurred in the suburban cities of Tonglu, Jiande and ChunAn may be due to the meteorological conditions more than anthropogenic effects. During the day, the aerosols in the near-surface may spread into vertical as a result of turbulence due to the more and more unstable atmosphere by the continuous strengthening of solar radiation.”.

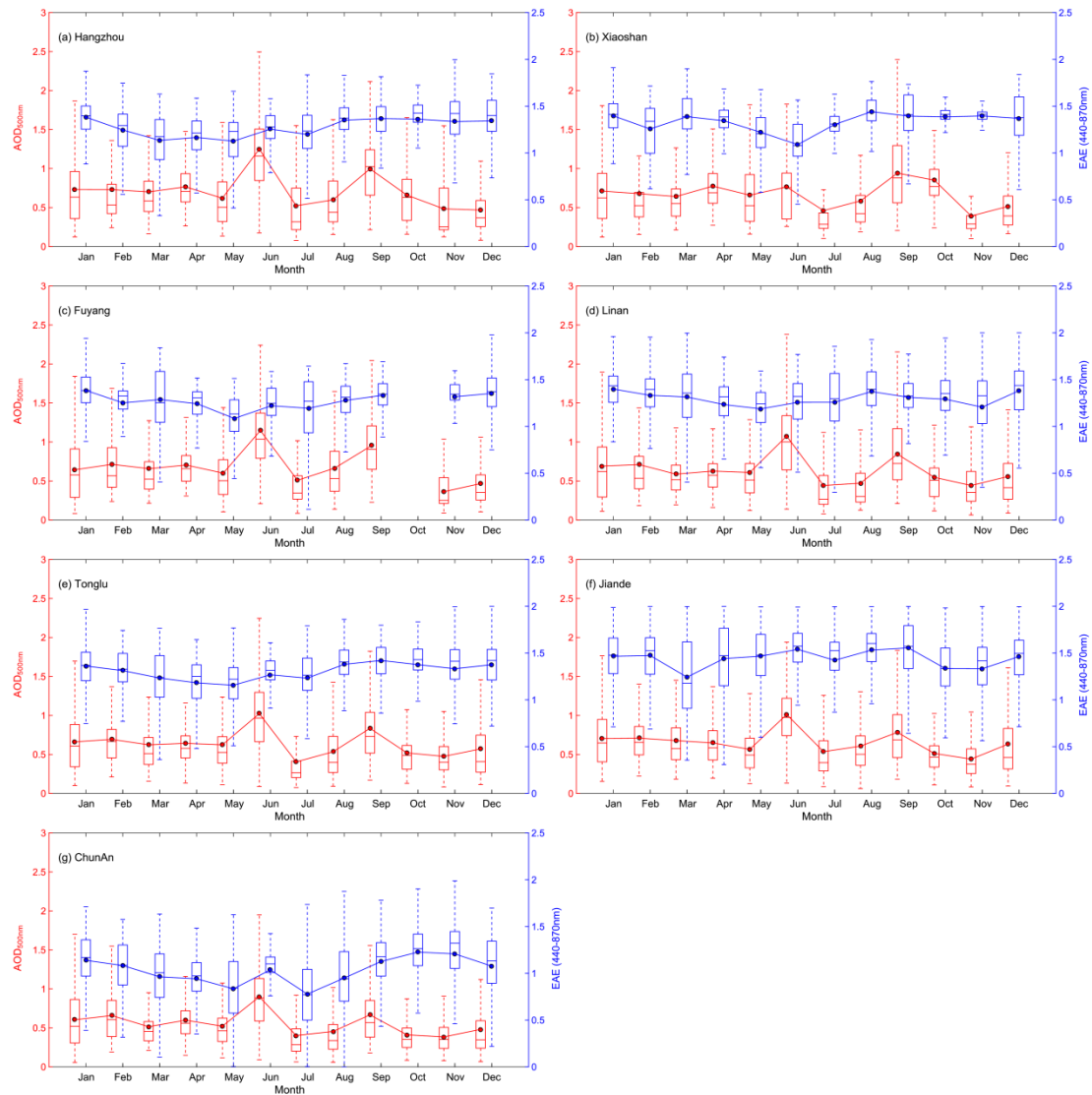


Fig.4. Variation in the AOD at 500nm & EAE at 440–870 nm over (a) Hangzhou, (b) Xiaoshan,

(c) Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn. The boxes represent the 25th to 75th percentile distribution, while the dots and solid lines within each box represent the mean and median, respectively.

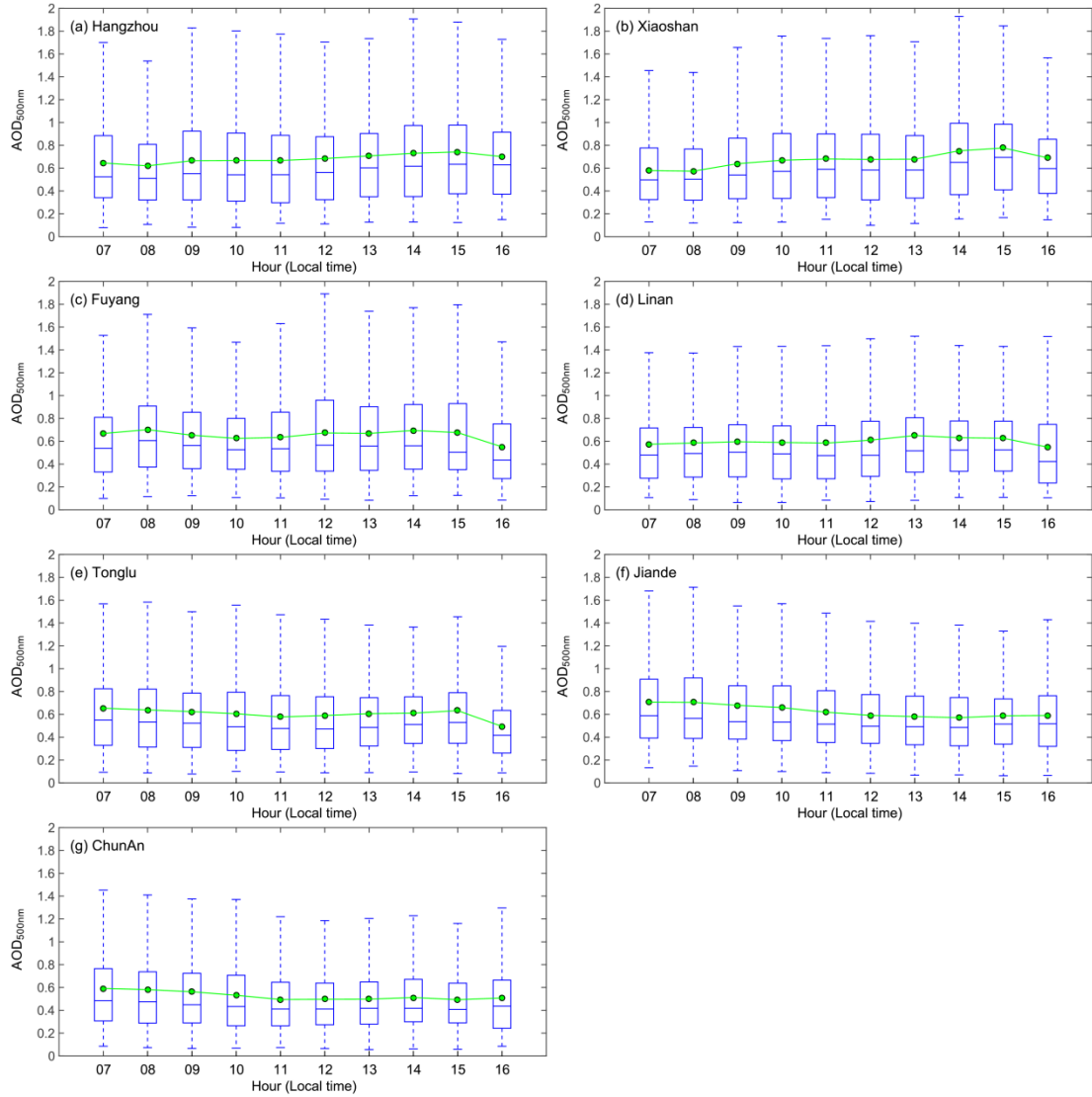


Fig.5. Variation of diurnal cycle in the AOD at 500 nm over (a) Hangzhou, (b) Xiaoshan, (c) Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn. The boxes represent the 25th to 75th percentile distribution, while the dots and solid lines within each box represent the mean and median, respectively.

4. L40: use the term “fine mode fraction” if that is what you mean here.

Response: The words “The ratio of the AOD of fine-mode particles” has been changed as “The fine mode fraction” in Abstract in line 47 and this change applied to the rest of the manuscript.

5. L43: The Pearson correlation coefficient is noted as R, not R<sup>2</sup> (which is called coefficient of determination).

Response: Thanks for the suggestions of the reviewer. The sentences in line 43 of “The correlation coefficients (R<sup>2</sup>) between the MODIS C6 AOD data and the values measured on the ground were ~0.73–0.89.” has been modified as “The MODIS/Terra C6 retrieval AOD values was generally more stable in the YRD region compared with the MODIS/Aqua product with the two Deep Blue (10km) and Dark Target (3km and 10km) methods against ground-based observations.” in the revised manuscript in Abstract on line 50-55. In the section 3.2, the corresponding correlation coefficients (R) values has been revised as “*The correlation coefficients (R) between the MODIS/Aqua and MODIS/Terra between by the Dark Target methods at 3km and sun photometer AOD (550 nm) values were about 0.84 to 0.92 and 0.85 to 0.94 in the YRD region, respectively.*” in line 411-415, “*The correlation coefficients (R) of the MODIS/Aqua and MODIS/Terra between sun photometer AOD (550 nm) values by the Deep Blue and Dark Target methods at 10km were about 0.81 to 0.90, 0.85 to 0.90, 0.69 to 0.91 and 0.85 to 0.93 in the YRD region, respectively.*” in line 432-line 435, “*In particular, the biases of the correlation coefficients (R) occurred in LinAn and Jiande has decreased from 0.94 and 0.90 to 0.87 and 0.88.*” in line 437-439.

6. L49-50, use volume fraction instead.

Response: According the reviewer’s suggestions, the sentences in line 49-50 “The fine-mode radii in the Yangtze River Delta region were ~0.2–0.3  $\mu\text{m}$  with a volume of 0.10–0.12  $\mu\text{m}^3$  and the coarse-mode radii were ~2.0  $\mu\text{m}$  with a volume close to 0.07  $\mu\text{m}^3$ .” has been modified as “The fine-mode radii in the Yangtze River Delta region were ~0.2–0.3  $\mu\text{m}$  with a volume fraction of 0.10–0.12  $\mu\text{m}^3$  and the coarse-mode radii were ~2.0  $\mu\text{m}$  with a volume fraction close to 0.07  $\mu\text{m}^3$ .” in line 39-41. This changes has also been revised in section 3.1 line 229-231 as “*The fine-mode radii were ~0.2–0.3  $\mu\text{m}$  in the YRD with a volume fraction of 0.10–0.12  $\mu\text{m}^3$  and the coarse-mode radii were ~2.0  $\mu\text{m}$  with a volume fraction close to 0.07  $\mu\text{m}^3$ .*”.

7. L52-54: reads like a method description (remove it), and what are the key findings?

Response: Thanks for the important suggestions. The sentences *“The absorption Angström exponent and the extinction Angström exponent were used to classify the different types of aerosol and the components of mixtures.”* in line 52-54 has been modified in the revised manuscript as *“The absorption Angström exponent and the extinction Angström exponent shows that the “mostly dust” category was very low in the suburban and rural sites (<0.01%) and also less in the urban site (~0.24%)”* to highlight the key findings in line 63-66.

8. L54-56: this statement is too general to be included in an abstract. Abstract is to present the most original and important findings of the study. Do not simply report the results as general statements. Writing an interesting abstract is a critical step to pass a peer review.

Response: Thanks for the helpful suggestions. This statement in the abstract has been revised to present the most original and important findings of the study. The sentences in L54-56 of *“The aerosols caused negative radiative forcing both at the Earth’s surface and at the top of the atmosphere all year round in the Yangtze River Delta region of eastern China.”* has been changed as *“The aerosols caused negative radiative forcing both at the Earth’s surface and at the top of the atmosphere all year round in the Yangtze River Delta region with the lower surface albedo in an unique geographical climate conditions of better vegetation in the YRD region than in north/northeast China.”* to show the special surface features in this distinct in line 66-70.

## Section 1

9. Introduction needs to polish. Always write out the acronyms at their first appearance. This comment applies to the rest of the manuscript.

Response: According to the reviewers suggestion, the introduction part has been revised. The acronyms such as “AERONET”, “SKYNET”, “EARLINET” and “GAW-PFR Network” has been written out the full name at their first appearance as “AERONET (Aerosol Robotic Network)”, “SKYNET (SKYrad Network)”, “EARLINET (European aerosol Lidar network)” and “GAW-PFR Network (Global Atmosphere Watch Programmer-Precision Filter Radiometers)” in Introduction and throughout the manuscript.

10. L92: it looks like the author wants to emphasize the advantage of using multi-site, multiyear, as well as three-minute-interval (L35) or high-frequency (L97) data; however, not in a single place of the manuscript the author discusses the advantage of such data. After all, all sites seemed to have similar aerosol characteristics. And all results are presented as monthly or annual means. Consider adding analysis of diurnal cycle and interannual variations, or exploring other unique aspects of the data.

Response: Thanks for the constructive comments and suggestions. We added the analysis of monthly and diurnal cycle of AOD at 500nm in section 3.2 to display a unique aspect of the data in this region. The result is as follows *“The monthly and diurnal cycle of AOD at 500nm has also been discussed in Fig.4 and Fig.5. The annual values of AOD<sub>500nm</sub> over the seven urban, suburban and rural sites in this study varied from 0.53 (ChunAn) to 0.68 (Hangzhou). The results show that two peaks of AOD at 500nm occurs in June and September with values of 1.25±0.59 and 1.00±0.42 in the urban site of Hangzhou, respectively which has the similar pattern as the other sites. The increase of AOD at 500nm in June is not corresponding to the same increase pattern of EAE (about 1.5) which indicates the aerosols types may be relatively constant in this region. The Fig.5 depicts the diurnal patterns of AOD at 500nm in this megacity area of eastern China. We can see that there are two types of diurnal patterns in this region. The daily AOD has been found increased in early morning (08:00 hr to 09:00 hr) and afternoon (12:00 hr to 14:00 hr) about the value of 0.60 to 0.70 in Hangzhou, Xiaoshan, Fuyang and Linan, while the decreasing of daily AOD has been observed from 0.70 to 0.50 during the daytime (from 07:00 hr to 16:00 hr) in Tonglu, Jiande and ChunAn. The high AOD during 07:00~09:00 in the urban area may be due to the anthropogenic activities and aerosol emissions from the morning rush hour. The decreased AOD with the value of 0.37±0.36 occurred in the suburban cities of Tonglu, Jiande and ChunAn may be due to the meteorological conditions more than anthropogenic effects. During the day, the aerosols in the near-surface may spread into vertical as a result of turbulence due to the more and more unstable atmosphere by the continuous strengthening of solar radiation.”.*

Section 2.

11. L130: Given the high AOD (comparable to urban site), ChunAn is nothing like a

background site. (L177)

Response: Thanks for the suggestions. Considering to the AOD level of ChunAn comparable to urban site, we changed the description of ChunAn as “rural site” in Table 1 and “clean site” as “*The rural site of ChunAn can be regarded as a representative clean site less affected by local and regional pollution.*” In section 2 line 145 and “*The site at LinAn is regarded as the clean suburban site in eastern China with an average AOD about  $0.73 \pm 0.44$ ,*” in section 4 on line 813-814 and “*e.g. particles from urban/industrial activities) over the clean rural sites than over urban or suburban sites.*” In Section 3.3 on lines 516-518 instead of the background site in this manuscript.

12. L146-157: describe the assumptions of the inversion algorithms, some of which may not apply for eastern China. Also describe the accuracies of the inversion variables (which are essentially a remote sensing product).

Response: Thanks for the important suggestions. The assumption of the inversion algorithm is “*The inversion algorithm is under an assumption of homogeneous nonsphericity aerosol particles distribution according to Dubovik (2006).*” and “*Real and imaginary parts of refractive index at 4 wavelengths (440, 675, 870, and 1020 nm) were retrieved from sky radiance and were confined in the range of 1.33–1.60 and 0.0005–0.50, respectively (Dubovik and King, 2000; Che et al., 2015b). Also retrieved were aerosol volumes of 22 size bins within the 0.05 - 15  $\mu\text{m}$  radius range.*” as mentioned in section 2 on line 171-173 and line 177-180. The accuracies of the inversion variables has been described in section 2 as “*The accuracies of SSA is  $\sim 0.03$ , and the errors are about 30%–50%/0.04 for the imaginary/real part of the complex refractive index under the conditions of  $\text{AOD}_{440\text{nm}}$  larger than 0.4 with the solar zenith angle more than  $50^\circ$ .*” in line 173-175. The assumption of the inversion algorithms has also been used in Taihu, Nanjing, etc in eastern China in AERONET sites (Holben et al., 1998).

13. L159-161: Need more descriptions on the technical details of radiative transfer calculations. For example, how is the atmospheric profile treated and where are the profile data coming



from? How is cloud scattering treated? How are clouds and aerosols vertically distributed?  
How is the surface and TOA radiative forcing defined?

Response: Thanks for the reviewer's constructive comments. More descriptions of atmospheric profile, cloud scattering treated, aerosols vertically distributed and the surface and TOA radiative forcing has been added in section 2 as follows "*The ARF (aerosol radiative forcing) data were calculated by the radiative transfer module used by the AERONET inversion (García et al., 2012) under the assumption of cloud-free consideration. In this code, the aerosol vertical properties have been considered into a homogeneous atmosphere layers because of the weak dependent of ground radiances on the whole atmospheric column with minor uncertainties (Dubovik et al., 2000). The broadband fluxes from 0.20 to 4.0 $\mu$ m were calculated according to the radiative transfer model GAME (Global Atmospheric ModEI) (Dubuisson et al., 1996, 2006; Roger et al., 2006). The size distribution, complex refractive index, and spherical particles fraction has been retrieved from the almucantar plane in the measurements. The SA (surface albedo) is obtained from the MODIS albedo product (MCD43C3) with the interpolation value of 440, 670, 870, and 1020 nm. The water vapor at 940 nm has been retrieved by the sun photometer. The ozone content was obtained from NASA Total Ozone Mapping Spectrometer measurements from 1978 to 2004. And other atmospheric gaseous data came from the US standard 1976 atmosphere model. In this study, the two parameters of ARF at the surface (ARF-BOA) and at the top of the atmosphere (ARF-TOA) have been calculated to describe the aerosol direct radiation effect to account for the changes of the solar radiation by calculating the difference energy between the aerosols presentation and absentation.*" In Line 188-211.

14. L162-172: Why not use Aqua? I understand that MODIS/Aqua is more stable and has better calibration than Terra (corrects me if I am wrong). It seems that the author calculated AOD at 550 nm in order to validate MODIS. If this is the case, why not show AOD at this wavelength for the entire manuscript (e.g. Table 1)?

Response: The reviewer's suggestions is very important. The Aqua product has been added to validate the MODIS AOD at 550nm according to the suggestions in section 3.2. The MODIS C6 aerosol optical thickness products refined by Levy et al. (2013) were evaluated against our

ground-based observations by the Deep Blue and Dark Target methods at 3km and 10km separately in section 3.2 as follows in line 400-476:

*The product of MODIS/Terra and MODIS/Aqua with Deep Blue (at 10km) and Dark Target (at 3km and 10km) methods has been evaluated against by ground-based observations separately in Fig. 6-8. We use the better estimated data of Quality flag = 3 and Quality flag=2, 3 for DT and TB methods, respectively. The systematic performance of the MODIS/Terra C6 retrieval AOD values was generally more stable in the YRD region compared with the MODIS/Aqua product with the two Deep Blue and Dark Target methods, which most of the plots scattered around the 1:1 regression line.*

*The correlation coefficients (R) between the MODIS/Aqua and MODIS/Terra between by the Dark Target methods at 3km and sun photometer AOD (550 nm) values were about 0.84 to 0.92 and 0.85 to 0.94 in the YRD region, respectively. The linear regression fitting performed better at the suburban sites of LinAn and Jiande according to the product of MODIS/Terra by the Dark Target methods at 3km. The fitting curve was almost consistent with the 1:1 reference line, which suggests that the aerosol properties were well defined for the MODIS C6 products. A large part of the MODIS retrieval AOD value was outside the expected error envelope of  $\pm (0.05 + 20\%T_{CARSNET})$ , especially for AOD values < 0.80 in Hangzhou and Xiaoshan. This indicates that the MODIS retrieval algorithm could still be improved, especially in urban areas. The MODIS retrieval AOD performed better at the other five sites (Fuyang, LinAn, Tonglu, Jiande and ChunAn) in the YRD; most of the retrieved AOD values for these sites fell within the expected error envelope. The MODIS/Aqua retrievals with Dark Target methods at 3km were underestimated while the MODIS/Terra retrievals with Dark Target methods at 3km were overestimated except Hangzhou, Tonglu and Jiande. The small deviation at the suburban sites suggested that the MODIS C6 retrieval using the DT method was suitable for capturing the optical properties of aerosols in suburban areas with dense vegetation coverage of the YRD. However, this method may have larger difference in the urban areas with less vegetation such as Hangzhou. The correlation coefficients (R) of the MODIS/Aqua and MODIS/Terra between sun photometer AOD (550 nm) values by the Deep Blue and Dark Target methods at 10km were about 0.81 to 0.90, 0.85 to 0.90, 0.69 to 0.91 and 0.85 to 0.93 in the YRD region,*

respectively. The MODIS/Aqua and MODIS/Terra retrievals with Deep Blue and Dark Target methods at 10km were underestimated except Hangzhou and Xiaoshan. In particular, the biases of the correlation coefficients ( $R$ ) occurred in LinAn and Jiande has decreased from 0.94 and 0.90 to 0.87 and 0.88. The validation results indicate a good MODIS/Terra matching with better fitting correlation at 3km rather than 10km products.

The AOD overestimation retrieved using Dark Target (DT) and Deep Blue (DB) methods are more influenced by the SSA and the phase function of aerosol in eastern China with AOD > 0.4 (Tao et al. 2015). Therefore, the detailed ground-based observation in this work is more helpful to the calibration of MODIS retrievals in eastern China.

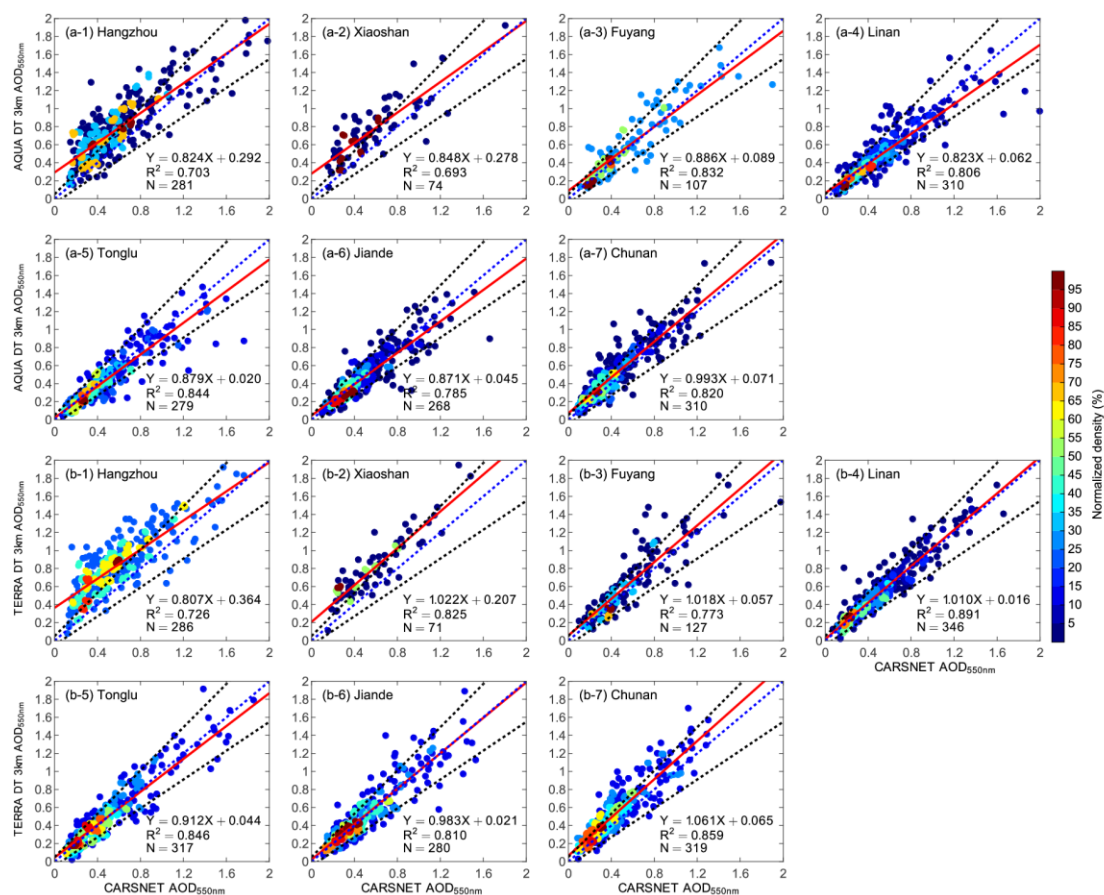


Fig.6. Comparison of MODIS/Aqua Dark Target (DT) AOD at 550 nm with the CARSNET AOD at 3km in (a-1) Hangzhou, (a-2) Xiaoshan, (a-3) Fuyang, (a-4) LinAn, (a-5) Tonglu, (a-6) Jiande, (a-7) ChunAn and MODIS/Terra DT AOD at 550 nm with the CARSNET AOD at 3km in (b-1) Hangzhou, (b-2) Xiaoshan, (b-3) Fuyang, (b-4) LinAn, (b-5) Tonglu, (b-6) Jiande, (b-7)

ChunAn. The red solid line represents the linear regression. The two black dotted lines represent the expected errors in the MODIS retrievals.

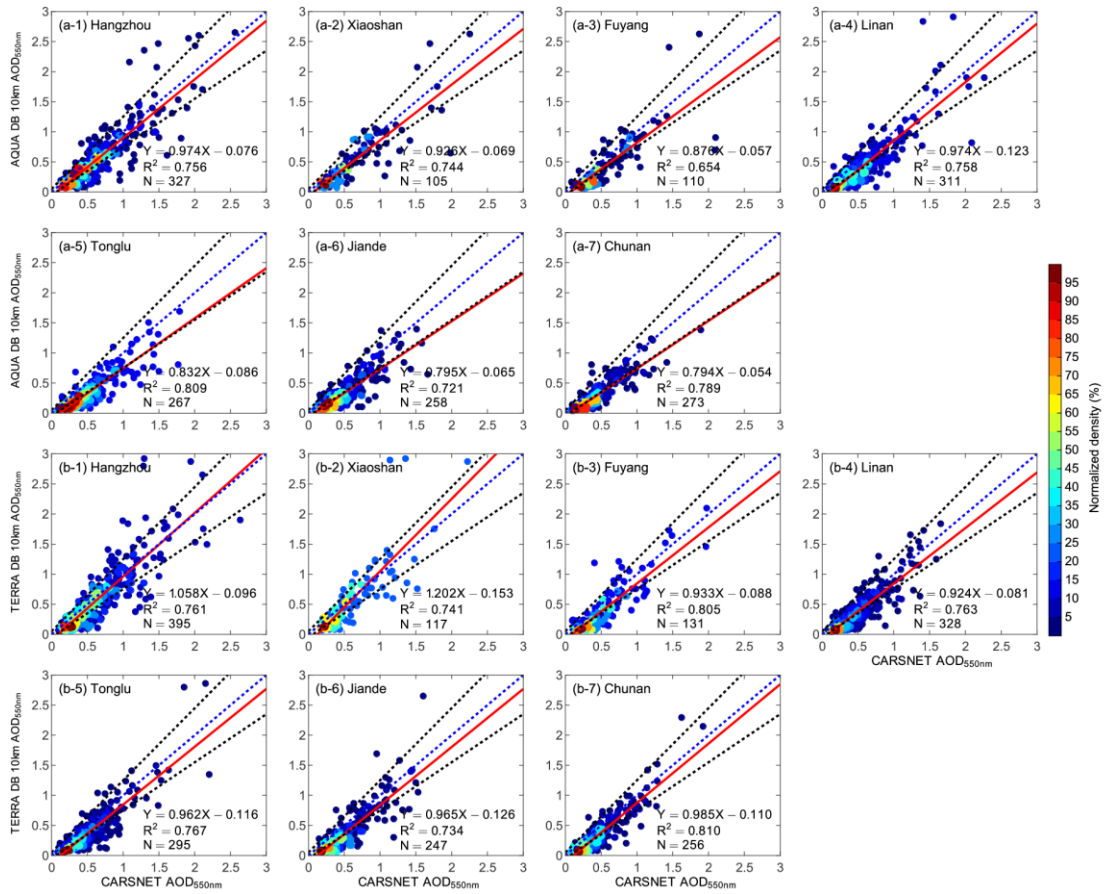
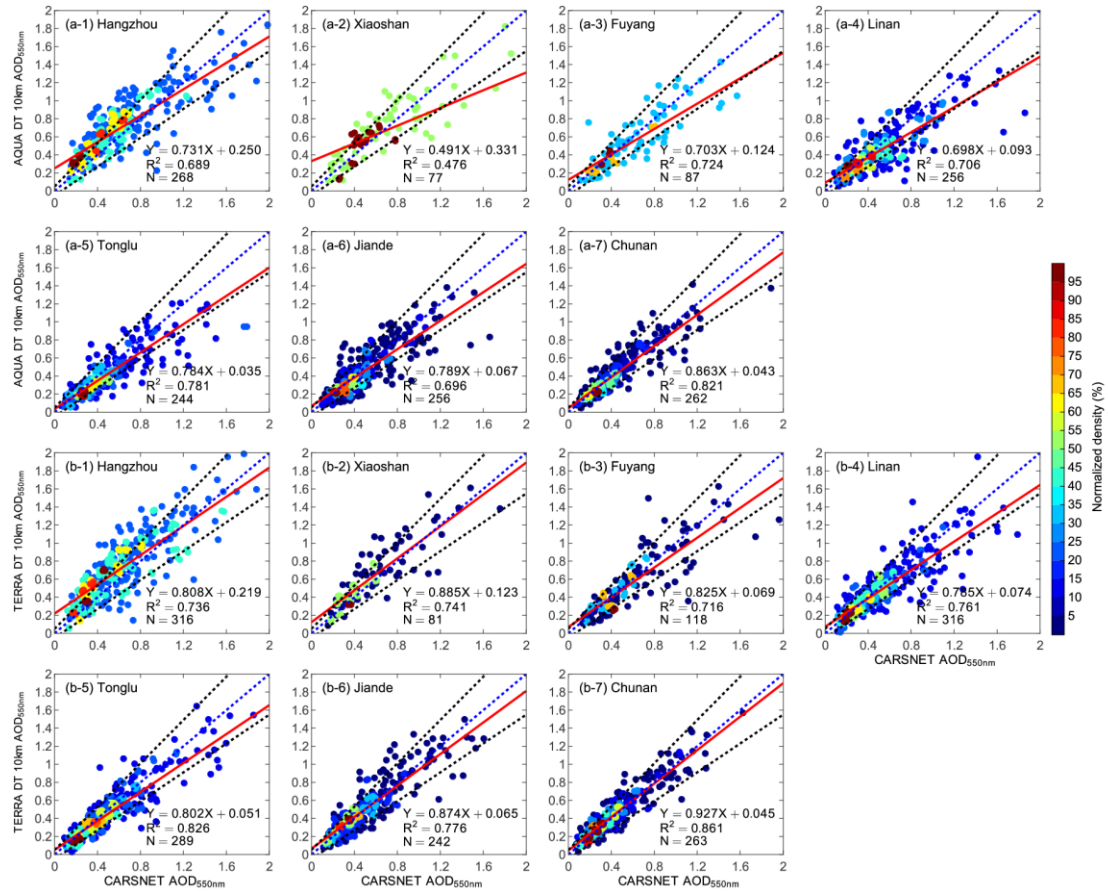


Fig.7. Comparison of MODIS/Aqua Deep Blue (DB) AOD at 550 nm with the CARSNET AOD at 10km in (a-1) Hangzhou, (a-2) Xiaoshan, (a-3) Fuyang, (a-4) LinAn, (a-5) Tonglu, (a-6) Jiande, (a-7) ChunAn and MODIS/Terra AOD DB at 550 nm with the CARSNET AOD at 10km in (b-1) Hangzhou, (b-2) Xiaoshan, (b-3) Fuyang, (b-4) LinAn, (b-5) Tonglu, (b-6) Jiande, (b-7) ChunAn. The red solid line represents the linear regression. The two black dotted lines represent the expected errors in the MODIS retrievals.



*Fig.8. Comparison of MODIS/Aqua AOD DT at 550 nm with the CARSNET AOD at 10km in (a-1) Hangzhou, (a-2) Xiaoshan, (a-3) Fuyang, (a-4) LinAn, (a-5) Tonglu, (a-6) Jiande, (a-7) ChunAn and MODIS/Terra DT AOD at 550 nm with the CARSNET AOD at 10km in (b-1) Hangzhou, (b-2) Xiaoshan, (b-3) Fuyang, (b-4) LinAn, (b-5) Tonglu, (b-6) Jiande, (b-7) ChunAn. The red solid line represents the linear regression. The two black dotted lines represent the expected errors in the MODIS retrievals.*

Moreover, the monthly and diurnal AOD at 500nm has been analyzed for the entire manuscript in Table 1 and section 3.2. In this study, all sites have AOD measurements at 340, 380, 440, 500, 670,870, 1020, 1640nm, respectively. The direct measured AOD<sub>500nm</sub> is more accurate than AOD<sub>550nm</sub> transferred by the interpolated and extrapolated value from by AOD values at 440 and 675nm. To avoid uncertainty, the 550nm AOD were calculated for MODIS validation. The direct measured AOD<sub>500nm</sub> were used to analyze the aerosol optical characteristics and also is to consistent and comparable to previous studies We added the analysis of monthly and diurnal cycle of AOD at 500nm in section 3.2 to display a unique

aspect of the data in this region.

### Section 3.

15. L177: from Table 1, Xiaoshan and Fuyang have far smaller observation samples than other sites. Why? Does this affect the comparability between the sites? ChunAn AOD is only 10% less than other sites. I would not say it is a representative background site.

Response: Thanks for the suggestions. The problem of smaller observation samples in Xiaoshan and Fuyang has been explained in section 2 as follows in 246-251: “*Smaller observation sample has been found in Xiaoshan and Fuyang with 180 and 217 available observation days, respectively. The number of 180 observation days in Xiaoshan is less than half of the year may have less representative and need further observation accumulation, while the observation days of 217 in Fuyang was more than half of the year may not affect the comparability between the other sites.*”. According to the AOD level of ChunAn comparable to urban site, we changed the description of ChunAn as “rural site” in Table 1 and “clean site” as “*The rural site of ChunAn can be regarded as a representative clean site less affected by local and regional pollution.*” In section 2 line 145 and “*The site at LinAn is regarded as the clean suburban site in eastern China with an average AOD about  $0.73\pm0.44$ ,*” in section 4 on line 813-814 and “*e.g. particles from urban/industrial activities) over the clean rural sites than over urban or suburban sites.*” In Section 3.3 on lines 516-518 instead of the background site in this manuscript. And this has been response in question 11 in the above.

16. L180-188: this part has serious redundancy. Rewrite it to be more concise.

Response: According to the reviewer’s helpful suggestion, this part in line 180-188 has been rewritten concisely in section 3.2 line 251-265 as follows as “*The annual values of the AOD<sub>440nm</sub> at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn were about  $0.76\pm0.42$ ,  $0.76\pm0.43$ ,  $0.76\pm0.45$ ,  $0.73\pm0.44$ ,  $0.71\pm0.41$ ,  $0.73\pm0.40$  and  $0.68\pm0.38$ , respectively, which suggests that column aerosol loading is at a high level at all seven urban, suburban and rural sites in the YRD on the regional rather than the local scale. The AOD<sub>440nm</sub> decreased from the eastern coast to the inland areas towards the west (from*

*~0.76±0.42 at Hangzhou to ~0.68±0.38 at ChunAn) due to the high aerosol loading from economic development and anthropogenic influences. The annual AOD<sub>440nm</sub> shows that the aerosol loading has similar level in Hangzhou, Xiaoshan and Fuyang, and with the 4%-10% decrease in LinAn, Tonglu, Jiande and ChunAn, respectively. The AOD<sub>440nm</sub> at the urban site of Hangzhou was the highest as a result of the more industrial activity and high resident density in the eastern part metropolis region resulting in higher aerosol emissions compared with the other suburban and rural sites.”*

17. L188-206: It is dangerous to use AOD solely as a metric for pollution (PM) or emission level. Keep in mind that AOD is an optical quantity (at a specific wavelength) and measures the light extinction of a vertical atmospheric column from the ground to TOA. The AOD difference between sites may not reflect the ambient pollution severity if some sites are affected by transported events. The AOD difference between cities may not reflect the emission sources if the meteorological conditions are so different to affect the dispersion and lifetime...The author needs a more solid analysis here.

Response: The reviewer’s suggestions are important and constructive. In the revised paper, we used aerosol extinction ability instead of describing a metric for pollution (PM) or emission level based on the value of AOD. These modification has been modified in DISCUSSION as “The AOD in Hangzhou in urban eastern China was similar to that in Shenyang (0.75) in urban northeast China (Zhao et al., 2013), and in Beijing (0.76) and Tianjin (0.74) in urban north China (Che et al., 2015b), indicating that the aerosol extinction is both common and at a similar level throughout most urban areas of China. The AOD values at the urban and suburban sites of Hangzhou were slightly higher than at Pudong (0.70) and Hefei (0.69), other urban areas in eastern China, suggesting that higher aerosol extinction ability were observed here (He et al., 2012; Liu et al., 2017). However, the AOD at all seven sites was lower than that obtained at Wuhan (1.05), Nanjing (0.88), Dongtan (0.85), Taihu(0.77) and Xuzhou (0.92) in previous studies in eastern China (Wang et al., 2015; Li et al., 2015; Pan et al., 2010; Xia et al., 2007; Wu et al., 2016). This indicates that the aerosol loading caused by anthropogenic activities is very high in both urban and suburban areas in eastern China. The site at LinAn is regarded as the clean suburban site in eastern China with an average AOD about 0.73±0.44,

*which is higher than that at the other regional background stations of China, such as Longfengshan (0.35; northeastern China), Mt Waliguan (0.14, inland Asia), Xinglong (0.28, northern China), Akedala (0.20, northwestern China) and Shangri-La (0.11, southwestern China) (Wang et al., 2010; Che et al., 2011; Zhu et al., 2014; Che et al., 2015b). The aerosol loading in eastern China (especially in the YRD region) is at least twice as high as in other regions of China which indicate the strong aerosol extinction.”.*

18. L225-230: Another example of redundancy. Rewrite to be more concise.

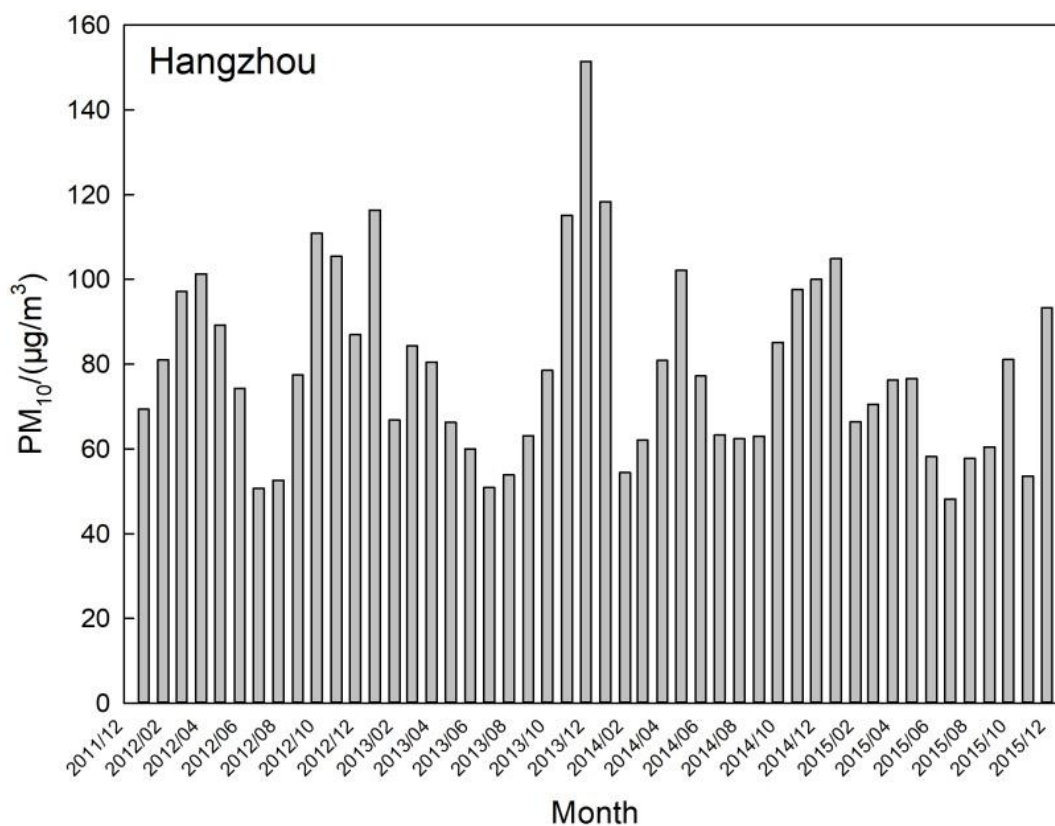
Response: Thanks for the helpful suggestion. A concise description has been re-written in the manuscript in section 3.2 line 314-342 as follows “*The annual fine-mode AOD values at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn were about  $0.68\pm0.42$ ,  $0.69\pm0.41$ ,  $0.69\pm0.44$ ,  $0.66\pm0.43$ ,  $0.64\pm0.41$ ,  $0.66\pm0.40$  and  $0.61\pm0.38$ , respectively (Fig.3). The seasonal variation in the AOD was similar to the total AOD at these urban, suburban and rural sites. The fine-mode fraction of AOD consistently exceeded 0.90 which indicates a major contribution of fine mode fraction to the total AOD in the YRD. Moreover, the figure 3 shows that the EAE at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn was about  $1.29\pm0.26$ ,  $1.37\pm0.24$ ,  $1.32\pm0.24$ ,  $1.29\pm0.27$ ,  $1.30\pm0.26$ ,  $1.32\pm0.28$  and  $1.22\pm0.25$ , respectively. Values of EAE >1.20 were found in all months throughout the year, indicating that small particle size distributions were favored in the YRD region. The annual coarse-mode AOD values at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn were between about 0.06 and 0.08 with the coarse mode fraction of AOD about 0.10 which indicates the 10% contribution of coarse mode fraction to the AOD in the YRD. The less coarse mode fraction indicated that there is no obvious effect of the coarse particles in the YRD region than that contributed to the higher aerosol loading in other north/northeast China (Zhang et al., 2012). Some dusts cases can be observed in YRD region that transported from north/northwest China during 2012-2015 reflect the effect of mineral dust aerosols (Gong et al., 2003). The fugitive dust from road traffic and construction activity is another more persistent and significant source for China’s cities as well as these eastern megacities.”*

19. L231-235: Another example of unconvincing analysis. Is the transported dust event verified?



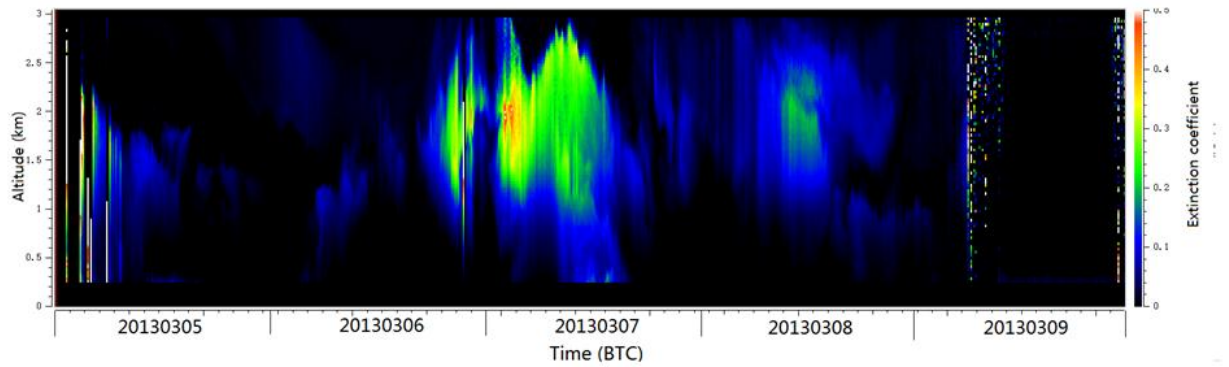
Please describe the specific events (2012-2015) rather than citing a climatological study. I suspect that fugitive dust from road traffic or construction activity is a more persistent and significant source for China's cities.

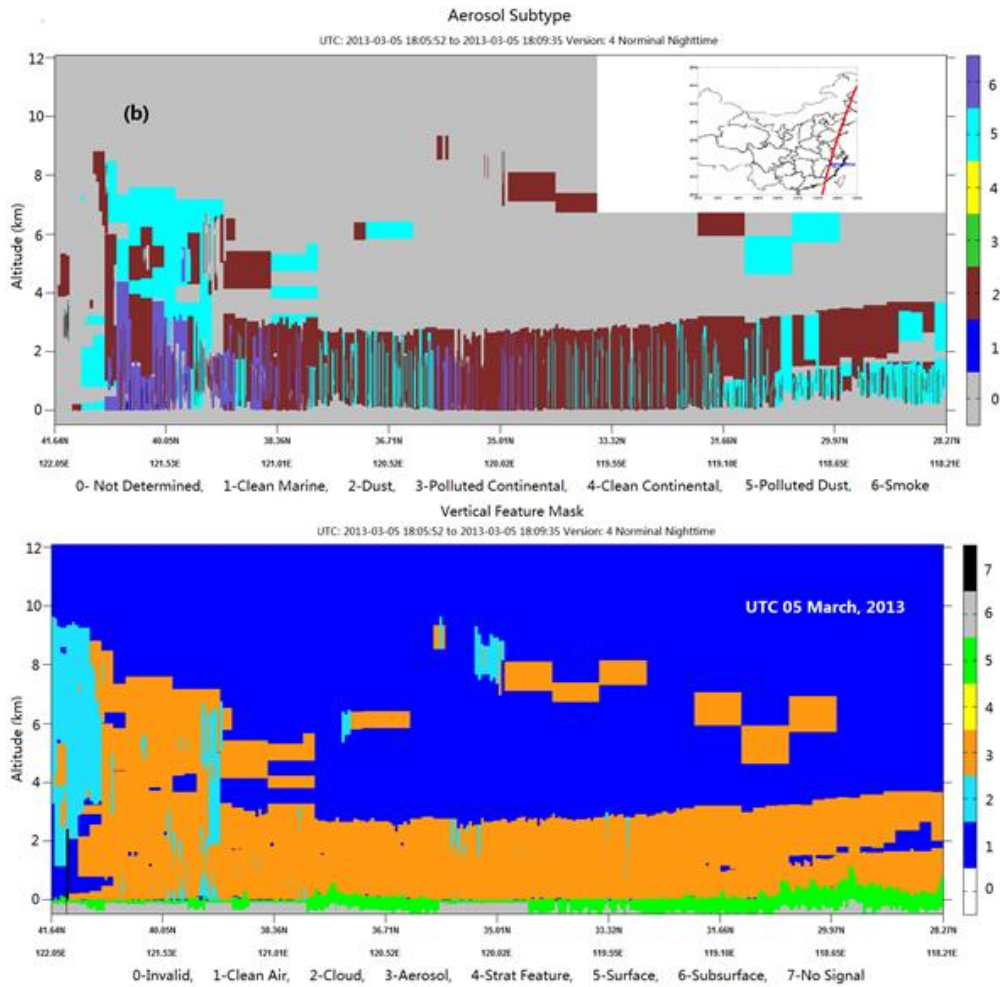
Response: Thanks for the important suggestions. The two reasons of fugitive dust from road traffic or construction activity has been added in line 340-342. According to the reviewer's suggestion, we rechecked the PM10 mass concentration observation data from 2012-2015 and describe a specific events on 5 March 2013. Moreover, the Lidar and CALIPSO data have also been investigated. As can be seen in the figure below, the PM10 mass concentration is higher in March to May than the value in February and June. This pattern clearly indicates the influence of dust in spring of Hangzhou. The Eastern China has been affected by a wide range of dust event in North China. These dust events across the northwest to the northeast area of China, then continue keep going south and east China. The higher PM10 mass concentration is obviously affected by the dust come from the northwest.



For example, there is a dust transportation event occurred in Hangzhou on 05-09 March, 2013. A series of transported aerosol masses over Hangzhou are monitored by MPL shown through

time-height cross section of extinction coefficient at 527nm. Referring to CALIPSO L2 retrieval results of vertical feature mask and aerosol subtypes illustrated in the Figure bellowing, the northwestern upwind areas of Hangzhou exists an aerosol layer mixed of “dust” and “polluted dust” about 3km thick from the surface in 5 March 2013, since when a thin external layer is detected concurrently. Therefore, the higher AOD in spring described in this study is significantly affected by dust process.





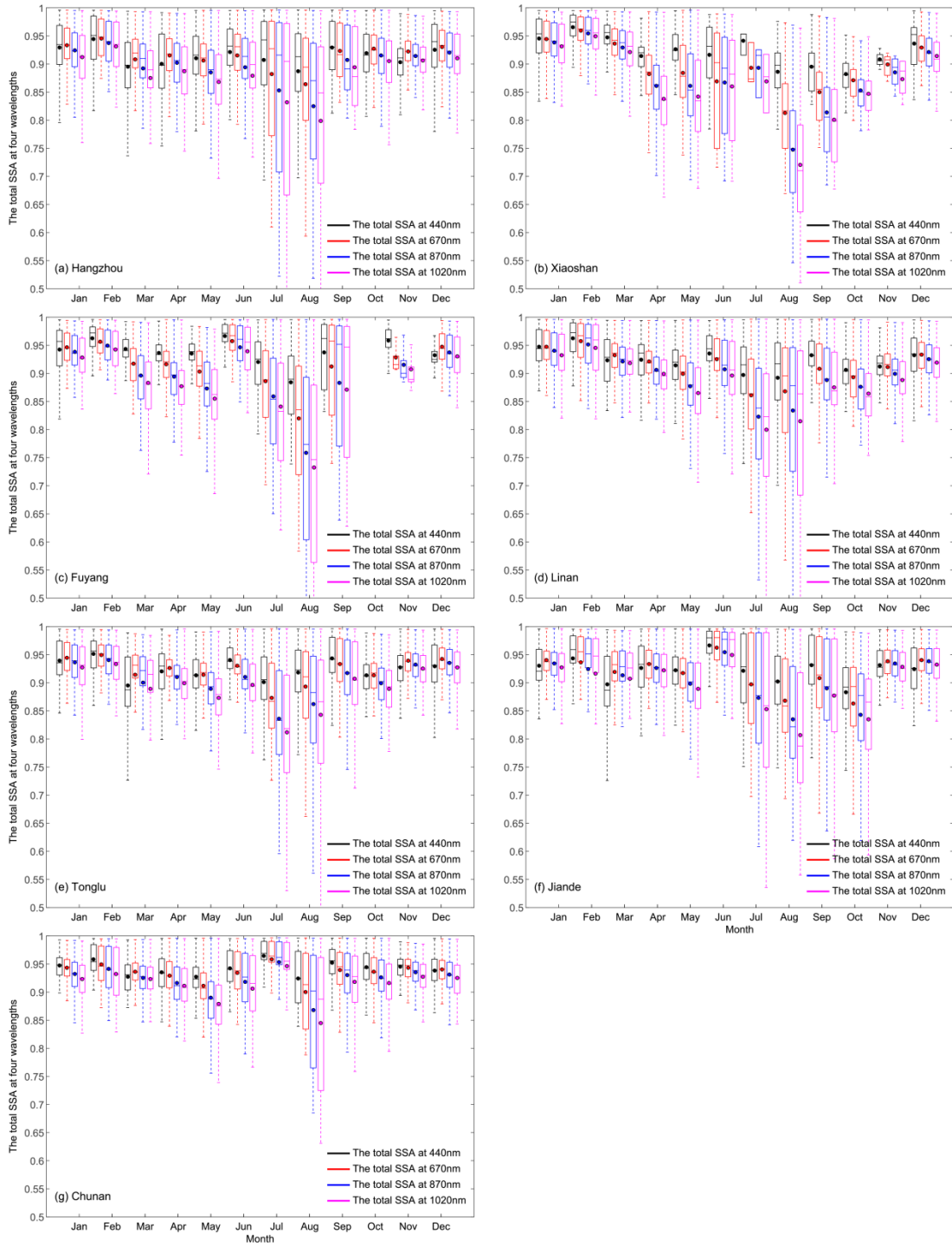
20. L241-250: The findings based on EAE are very similar to those from FMF (L222-235). Consider merging them to be more concise.

Response: According to the reviewer's helpful suggestion, the findings based on EAE in line 241-250 and those from FMF in Line 222-235 has been merged and modified to be more concise. The revised content is as following "The monthly and diurnal cycle of AOD at 500nm has also been discussed in Fig.4 and Fig.5. The annual values of AOD 500nm over the seven urban, suburban and rural sites in this study varied from 0.53 (ChunAn) to 0.68 (Hangzhou).The results show that two peaks of AOD at 500nm occurs in June and September with values of  $1.25 \pm 0.59$  and  $1.00 \pm 0.42$  in the urban site of Hangzhou, respectively which has the similar pattern as the other sites. The increase of AOD at 500nm in June is not corresponding to the same increase pattern of EAE (about 1.5) which indicates the aerosols types may be relatively constant in this region. The Fig.5 depicts the diurnal patterns of AOD at

500nm in this megacity area of eastern China. We can see that there are two types of diurnal patterns in this region. The daily AOD has been found increased in early morning (08:00 hr to 09:00 hr) and afternoon (12:00 hr to 14:00 hr) about the value of 0.60 to 0.70 in Hangzhou, Xiaoshan, Fuyang and Linan, while the decreasing of daily AOD has been observed from 0.70 to 0.50 during the daytime (from 07:00 hr to 16:00 hr) in Tonglu, Jiande and ChunAn. The high AOD during 07:00~09:00 in the urban area may be due to the anthropogenic activities and aerosol emissions from the morning rush hour. The decreased AOD with the value of  $0.37 \pm 0.36$  occurred in the suburban cities of Tonglu, Jiande and ChunAn may be due to the meteorological conditions more than anthropogenic effects. During the day, the aerosols in the near-surface may spread into vertical as a result of turbulence due to the more and more unstable atmosphere by the continuous strengthening of solar radiation.” in line 364-384.

21. L322-334: SSA depends on two factors - particle size and composition. It is expected that coarse mode SSA is less than fine mode SSA. I do not see the need for two SSAs for fine and coarse modes, separately. This paragraph presents no new findings and can be removed.

Response: Thanks for the important suggestions. Both reviewers mentioned this question. According to the two reviewer's suggestion, the section of coarse mode SSA and fine mode SSA has been removed in section 3.3 according to the suggestions of reviewer. Then we added the discussion of SSA depends on different wavelength in section 3.2 line 539-551 as “The wavelength dependence of SSA present specific absorption/scattering properties of different type aerosol (Sokolik and Toon, 1999; Eck et al., 2010). The SSA of dust in spring shown a dependence on the spectrum from 440nm to 1020nm in general (Cheng et al., 2006; Dubovik et al., 2002). Especially in March, the SSA at 440nm in Hangzhou, LinAn, Jiande and ChunAn was obviously lower at short wavelength than that in the longer wavelength. This result has shown a strong absorption of dust in the short wavelength in the YRD region over eastern China. It's worth noting that there is an obvious and strong decreasing of SSA in the longer wavelength of aerosol from biomass burning or industrial emissions in August. The wavelength dependence of SSA in YRD could be used to simply describe the aerosol types including dust or the biomass burning smoke.”.



*Fig.10. Variation in the SSA at 440nm, 670nm 870nm and 1020nm over (a) Hangzhou, (b) Xiaoshan, (c) Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn. The boxes represent the 25th to 75th percentile distribution, while the dots and solid lines within each box represent the mean and median, respectively.*

22. L349-351: The real part of refractive index is related to scattering, while the imaginary part

to absorption. Does the author mean to say spring dust absorption increases the imaginary part (not real part)?

Response: Yes. The sentences of *“High imaginary parts of the refractive index occurred in August at all urban, suburban and rural sites in the YRD, which may be due to the higher emission of absorptive particles by the post-harvest burning of crop residues with more spectral dependence. The burning of crop residues may cause a large deterioration in the regional air quality in the YRD region. A higher level of spring dust aerosols with absorption could contribute to a higher value of the imaginary part of the refractive index.”* has been modified in section 3.3 line 597-602.

23. L377-378: AOD values have very large uncertainties (standard deviation is comparable to mean). Why? Given these uncertain estimates, it would be difficult to make a robust comparison between sites.

Response: Thanks for the important suggestions. AOD values have very large uncertainties has been explained in line 630-634 as follows *“The AOD values may have very large an uncertainty because of the dataset is including all the values in one month. Nevertheless, there is also some varies in AOD according to the changes of the SSA in section 3.2. These differences in the AOD were mostly dependent on the type of aerosol and the ratio of absorbing and non-absorbing components in the aerosols.”*

24. L392, this paragraph seems redundant as the later part from L416 discusses EAE and AAE links to aerosol types.

Response: According to the reviewer’s suggestion, the paragraph in line 392 and the later part from L416 about the EAE and AAE links to aerosol types has been re-discussed in the revised manuscript as follows *“As fig.12 shown, the AAE was<1.00 in June and August at all urban, suburban and rural sites of the YRD, which suggested the presence aerosols coated with absorbing or non-absorbing material in summer season. This process is favored by high temperatures and high humidity under conditions of strong solar radiation (Shen et al., 2015, Zhang et al., 2015). The particles coagulate and grow rapidly in the presence of sufficient water vapor (Li et al., 2016). The AAE became increasingly close to, or larger than, 1.00 at all*

seven sites from September, which is consistent with decreasing amounts of precipitation. This increase in the AAE was related to the emission of black carbon from biomass burning (Soni et al., 2010; Russell et al., 2010). According to the corresponding annual mean values for the AAE at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn ( $1.13\pm 0.46$ ,  $0.88\pm 0.42$ ,  $0.85\pm 0.43$ ,  $0.98\pm 0.35$ ,  $1.11\pm 0.49$ ,  $1.16\pm 0.44$  and  $0.93\pm 0.31$ ) in Fig. 12, the seven sites has been attributed to three categories with AAE levels. The mean values of the AAE at Xiaoshan and Fuyang were  $<1.00$ , suggesting the presence of absorbing or non-absorbing materials coating black carbon at these suburban and rural sites (Bergstrom et al., 2007; Lack and Cappa et al., 2010; Gyawali et al., 2009). The AAE values were close to 1.00 at LinAn and ChunAn, indicating that the absorptive aerosols were dominated by particles of black carbon (Zhang et al., 2012; Li et al., 2016). By contrast, the AAE values at Hangzhou, Tonglu and Jiande were  $>1.00$ , indicating the presence of absorptive aerosols from the burning of biomass. This difference in the AAE distribution indicates the absorbing aerosols have different characteristics resulting from the different emission sources at urban, suburban and rural sites in the YRD.” in line 641-661.

25. L430-432: explain why the YRD region is “completely different from north/northeast China”

Response: The detailed explanation has been added according to the reviewer’s suggestion. The north/northeast China region is with lower precipitation and less vegetation coverage. And large dust particles from regional dust events transportation or local fugitive dust emission could have substantial contribution of aerosol loading in this region. While in the YRD region, there are large precipitation and dense vegetation coverage, resulting in less dust particle contribution to aerosol loading in YRD. The sentence has been changed as “*This indicates the YRD region is completely different from other north/northeast region in China where the dust particles could contribute to the aerosol loading substantially.*” In line 724-727.

26. Figure 10: really bad visualization. Consider other options, like histograms.

Response: The Figure 10 has been redrawn as Figure 14 in the manuscript.

27. L483: do you intend to say “large surface reflectance (or albedo)”

Response: Yes. The word has been changed as “large surface reflectance” in line 790.

28. Section 4 should be shortened. There is no need to repeat all results. There should be one or two paragraphs discussing the implications of the most important findings.

Response: Thank for the constructive suggestions. Section 4 Summary and discussion has been rewritten according to two reviewers' suggestions. The discussions part has been added and the summary part has been shortened and concise.



1 **Aerosol optical properties and ~~instantaneous~~ radiative forcing based on**  
2 **synchronous measurements of China Aerosol Remote Sensing Network**  
3 **(CARSNET) over eastern China**~~hightemporospatial spatiotemporal~~  
4 **resolution of China Aerosol Remote Sensing Network (CARSNET)**  
5 **ground-based measurements over eastern China**

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7 Victor Estelles<sup>6</sup>, Emilio Cuevas-Agulló<sup>7</sup>, Luc Blarel<sup>3</sup>, Yunfei Wu<sup>8</sup>, Jun Zhu<sup>9</sup>, Rongguang Du<sup>2</sup>,  
8 Yaqiang WANG<sup>1</sup>, Hong Wang<sup>1</sup>, Ke Gui<sup>1</sup>, Jie Yu<sup>1</sup>, Yu Zheng<sup>9</sup>, Tianze Sun<sup>1</sup>, Quanliang Chen<sup>10</sup>,  
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34

## Abstract

Variations in the optical properties of aerosols and their radiative forcing were investigated based on long-term synchronous observations made at three-minute intervals from 2011 to 2015 over seven adjacent CARSNET (China Aerosol Remote Sensing NETWORK) urban (Hangzhou), suburban (Xiaoshan, Fuyang, LinAn, Tonglu, Jiande) and rural (ChunAn) stations in the Yangtze River Delta region, eastern China. The fine-mode radii in the Yangtze River Delta region were  $\sim 0.2\text{--}0.3\ \mu\text{m}$  with a volume fraction of  $0.10\text{--}0.12\ \mu\text{m}^3$  and the coarse-mode radii were  $\sim 2.0\ \mu\text{m}$  with a volume fraction close to  $0.07\ \mu\text{m}^3$ . The radii of fine volume fraction in the Yangtze River Delta region were  $\sim 0.2\text{--}0.3\ \mu\text{m}$  with a volume of  $0.10\text{--}0.12\ \mu\text{m}^3$  and the radii of coarse volume fraction were  $\sim 2.0\ \mu\text{m}$  with a volume close to  $0.07\ \mu\text{m}^3$ . The fine-mode aerosols were obviously larger in June and September than in other months at almost the sites.

The aerosol optical depth (AOD at 440nm) varied from 0.68 to 0.76, with two peaks in June and September, and decreased from the eastern coast to western inland areas. The ratio of the AOD of fine-mode particles fine mode fraction to the total AOD was  $>0.90$  and the extinction Angström exponent was  $>1.20$  throughout the year at all seven sites. The AOD at 500nm has also been studied because of the wavelength dependent of optical properties to show the monthly and diurnal cycle. ~~—against~~ The Moderate Resolution Imaging Spectroradiometer (MODIS) C6 retrieval AOD was validated by comparison with ground-based observations. The correlation coefficients ( $R^2$ ) between the MODIS C6 AOD data and the values measured on the ground were  $0.73\text{--}0.89$ . The MODIS/Terra C6 retrieval AOD values was generally more stable in the YRD region compared with the MODIS/Aqua product with the two Deep Blue (10km) and Dark Target (3km and 10km) methods against ground-based observations. The single-scattering albedo varied from 0.91 to 0.94, indicating that scattering aerosol particles are dominant in this region. ~~The real parts of the refractive index were  $1.41\text{--}1.43$ , with no significant difference among the seven urban, suburban and rural sites.~~ Large imaginary parts of the refractive index were seen in August at all urban, suburban and rural sites. The fine-mode radii in the Yangtze River Delta region were  $\sim 0.2\text{--}0.3\ \mu\text{m}$  with a volume of  $0.10\text{--}0.12\ \mu\text{m}^3$  and the coarse-mode radii were  $\sim 2.0\ \mu\text{m}$  with a volume close to  $0.07\ \mu\text{m}^3$ . The fine-mode aerosols were obviously larger in June and September than in other months at almost the sites.

64 The absorption AOD was low in the winter. The absorption Angström exponent and the  
65 extinction Angström exponent ~~were used to classify the different types of aerosol and the~~  
66 ~~components of mixtures.~~ shows that the “mostly dust” category was very low in the suburban  
67 and rural sites (<0.01%) and also less in the urban site (~0.24%). The aerosols caused  
68 negative radiative forcing both at the Earth’s surface and at the top of the atmosphere all year  
69 round in the Yangtze River Delta region ~~of eastern China~~ with the lower surface albedo ~~–in a~~  
70 unique geographical climate condition of better vegetation in the YRD region than in  
71 north/northeast China.\_\_\_\_\_

## 72 1. Introduction

73 Aerosols have important effects on the Earth's climate at both global and regional scales,  
74 although there are still great uncertainties in assessing their impact (Hansen et al.2000;  
75 Solomon et al., 2007; Schwartz and Andreae, 1996). Aerosols affect not only the radiative  
76 balance of the Earth–atmosphere system by directly scattering and absorbing solar radiation  
77 (Charlson et al., 1992; Ackerman and Toon, 1981), but also indirectly affect the climate through  
78 aerosol – cloud interactions (Twomey et al., 1984; Albrecht et al., 1989; Li et al., 2016).

79 The optical properties of aerosols influence the aerosol radiative balance and can be  
80 used to predict and assess global and regional changes in the Earth's climate (Eck et al., 2005;  
81 Myhre et al., 2009; IPCC, 2013; Panicker et al., 2013). Long-term, ground-based observations  
82 are crucial to our understanding of the global and regional variations in the optical properties of  
83 aerosols and their effects on the Earth's climate (Holben et al., 2001; Kaufman et al., 2002;  
84 Sanap and Pandithurai, 2014; Li et al., 2016). Ground-based monitoring networks have been  
85 established worldwide—for instance, AERONET (Aerosol Robotic Network) (Holben et  
86 al.,1998; Goloub et al., 2007), SKYNET (SKYrad Network) (Takamura et al., 2004), EARLINET  
87 (European aerosol Lidar network) (Pappalardo et al.,2014) and the GAW-PFR Network  
88 (Global Atmosphere Watch Programmer-Precision Filter Radiometers) (Wehrli, 2002; Estelles  
89 et al., 2012),~~).~~The above networks exclude EARLINET~~which~~ includes several automated sites  
90 in China. CARSNET (the China Aerosol Remote Sensing NETwork) (Che et al., 2009a, 2015b)  
91 and CSHNET (the Chinese Sun Hazemeter Network) were established to obtain data on  
92 aerosol optical characteristics in China (Xin et al., 2007).

93 Most of the ground-based studies of the optical properties of aerosols in China have been  
94 concentrated in urban regions undergoing rapid economic development, which have high  
95 aerosol loadings and serious environmental problems (Cheng et al., 2015; Pan et al., 2010;  
96 Xia et al., 2013; Wang et al., 2015; Che et al., 2015a). Analyses of the aerosol optical depth  
97 (AOD), the types of aerosol ~~presents~~ and the classification of ambient aerosol populations  
98 based on their size and absorption properties(Giles et al., 2011) are needed to understand  
99 their effects on the Earth's climate and environment (Che et al., 2009b; Wang et al., 2010; Zhu

100 et al., 2014).

101 The Yangtze River Delta (YRD) region in eastern China has undergone rapid economic  
102 growth and has high emissions of aerosols (Fu et al., 2008; Zhang et al., 2009). There have  
103 been many studies of the optical properties of aerosols in eastern China and these are  
104 important in our understanding of both the local air quality and regional climate change (Duan  
105 and Mao, 2007; Pan et al., 2010; Ding et al., 2016). Basic investigations of the variation in the  
106 optical characteristics of aerosols over the YRD region have been carried out at Nanjing, Hefei,  
107 Shanghai, Shouxian and Taihu (Zhuang et al. 2014; Li et al., 2015; Wang et al., 2015; He et al.,  
108 | 2012; Lee et al., 2010; Cheng et al., 2015; Xia et al., 2007). These studies in the YRD region  
109 have mostly been single-site and/or short-period investigations. The study sites are ~100 km  
110 apart from each other, which makes high spatial resolution satellite and modeling validations  
111 difficult. Thus there is still a lack of long-term, continuous and synchronous observations of the  
112 optical characteristics of aerosols, especially over adjacent urban, suburban and rural areas in  
113 the YRD region.

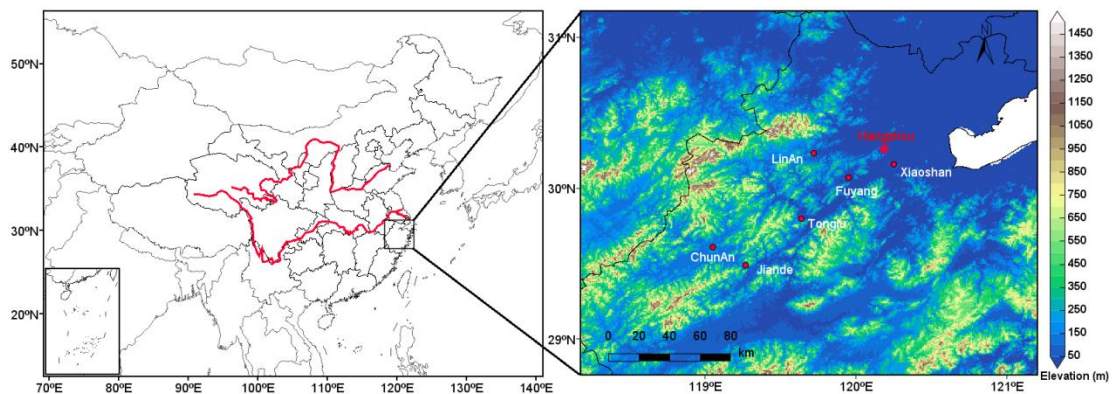
114 High-frequency ground-based observations of the variations in the optical characteristics  
115 of aerosols are necessary to our understanding of the processes involved in air pollution (e.g.  
116 the source, transport and diurnal variations of the pollution) and their effect on the regional  
117 climate. Ground-based observations are also important in the validation and improvement of  
118 satellite retrieval data (Holben et al., 2017; Xie et al., 2011). A high density of ground-based  
119 sun-and sky-scanning spectral radiometers within a local or meso-scale region is required to  
120 capture small-scale variations in aerosols for the accurate validation of satellite observations  
121 and to compare in situ versus remote sensing observations (Xiao et al., 2016; Holben et al.,  
122 2017). The MODIS (Moderate Resolution Imaging Spectroradiometer) retrieval AOD has a  
123 high accuracy with a wide spectral coverage (Tanré et al., 1997; Kaufman, et al. 1997) and the  
124 algorithm has been validated and improved based on AERONET data (Chu et al., 2002;  
125 | Ichoku et al., 2002; Remer et al., 2005; Levy et al., 2010;). Levy et al. (2013) refined the  
126 | MODIS Collection 6 (C6) aerosol retrieval process to provide better-more accurate AOD  
127 retrievals. Some validations of satellite aerosol retrievals have been carried out in China with

128 ground-based observations from CSHNET (Li, et al., 2007; Wang, et al., 2007; Xin, et al., 2007)  
129 and CARSNET (Che et al., 2009a, ~~Che et al., 2011a~~; Tao et al., 2015).

130 We investigated the variation in the optical properties of aerosols and aerosol radiative  
131 forcing (ARF) using three-minute intervals of sunphotometer measurements from 2011 to 2015  
132 at seven adjacent CARSNET (~10–40 km) urban, suburban and rural sites over eastern China.  
133 The aims of this study were: (1) to investigate the synchronous variations and differences in  
134 the optical properties of aerosols over urban, suburban and rural areas of the YRD megacity,  
135 eastern China; (2) to analyze the type and dominant distribution pattern of aerosols in the YRD  
136 via the extinction and absorption properties of aerosols; (3) to understand the difference in the  
137 ARF calculated from ground-based measurements of the optical properties of aerosols over  
138 urban, suburban and rural areas in eastern China; and (4) to evaluate the MODIS AOD  
139 retrieval data using the CARSNET AOD for the YRD. The results of this study will help the  
140 satellite and modeling communities to improve future aerosol retrieval data and simulations.

## 141 2. Site descriptions, measurements and data

142 Fig. 1 shows the geographical locations of the seven CARSNET sites in the YRD; these  
143 locations are described in Table 1.



145 Fig.-1. Geographical location and elevation map for the seven CARSNET sites in the YRD.

146 The rural site of ChunAn can be regarded as a representative ~~background location~~  
147 ~~site less un~~affected by local and regional pollution. The site has a small population and a good

148 ecological environment, although there is some agricultural activity and burning of biomass  
149 from crop residues. Hangzhou is a densely populated urban site with a large volume of  
150 vehicular traffic and is therefore more affected by anthropogenic activity. LinAn, Fuyang,  
151 Jiande, Xiaoshan, ~~and Tonglu and Xiaoshan~~ are suburban sites and are all affected by both  
152 anthropogenic activity and pollution from industrial and agricultural production.

153 CE-318 sun photometers (Cimel Electronique, Paris, France) were installed at these  
154 seven sites in the YRD from 2011 to 2015. The instruments were standardized and calibrated  
155 annually according to the protocols reported by Che et al. (2009a). The instruments in this  
156 study were made inter-comparison calibration by the CARSNET reference instruments, which  
157 were periodically calibrated at Izaña in Spain. The cloud-screened AOD at different  
158 wavelengths was obtained using ASTPwin software (Cimel Electronique) (Smirnov et al., 2000).  
159 Instantaneous ~~direct AOD measurements observation data for the AOD were selected at~~  
160 ~~least more than~~ ten times ~~at~~ each day ~~were selected at a temporal resolution of about three~~  
161 ~~minutes and this can eliminate about 20% data according to for daily average calculation and~~  
162 ~~statistical analysis to increase the representability of the aerosol optical characteristics (Che et~~  
163 ~~al., (2015). The large AOD were checked by MODerate-resolution Imaging Spectroradiometer~~  
164 ~~(MODIS) images (<http://modis-atmos.gsfc.nasa.gov/IMAGES/>) to further determine the cloud~~  
165 ~~contamination. and t~~he corresponding values of Angström exponent ( $\alpha$ ) were calculated by  
166 instantaneous AOD values at 440 and 870 nm.

167 The aerosol ~~microphysical properties of the volume size distribution and aerosol~~ optical  
168 properties—including the single-scattering albedo (SSA), the complex refractive index, ~~the~~  
169 ~~volume size distribution,~~ the absorption AOD (AAOD), the absorption Angström exponent  
170 (AAE) and the fraction of spherical particles—were retrieved from the almucantar irradiance  
171 measurements according to the methods of Dubovik and King (2000) and Dubovik et al. (2002,  
172 2006). ~~The inversion algorithm is under an assumption of homogeneous nonsphericitye~~  
173 ~~aerosol particles distribution according to Dubovik and King (20006) and has been applied in~~  
174 ~~many different types of areas world widely. The accuracies of SSA is ~0.03, and the errors are~~  
175 ~~about 30%–50%/0.04 for the imaginary/real part of the complex refractive index under the~~

176 conditions of AOD at 440nm larger than 0.4 with the solar zenith angle more than 50°. The  
 177 SSA was retrieved using only AOD<sub>440nm</sub>>0.40 measurements to avoid the large uncertainties  
 178 inherent in a low AOD (Dubovik et al. 2002, 2006). Real and imaginary parts of refractive index  
 179 at 4 wavelengths (440, 675, 870, and 1020 nm) were retrieved from sky radiance and were  
 180 confined in the range of ~~The complex refractive index was also retrieved by sky irradiance~~  
 181 ~~measurements in the range 1.33–1.60 and 0.0005–0.50, respectively for the real part and in~~  
 182 ~~the range 0.0005–0.50 for the imaginary part~~ (Dubovik\_ and King, 2000; Che\_ et al., 2015b).  
 183 Also retrieved were aerosol volumes of 22 size bins within the 0.05 - 15 um radius range.~~In the~~  
 184 ~~volume size distribution, the radius range is selected from 0.05–15µm.~~ The EAOD in this study  
 185 has been defined as extinction aerosol optical depth, and ~~t~~The AAOD and the AAE were  
 186 calculated as described in equations (1) and (2):

$$187 \quad \text{AAOD}(\lambda) = [1 - \text{SSA}(\lambda)] \times \text{EAOD}(\lambda) \quad (1)$$

$$188 \quad \text{AAE} = -\text{dln}[\text{AAOD}(\lambda)]/\text{dln}(\lambda) \quad (2)$$

189 The ARF (aerosol aerosol radiative forcing) data were calculated by the radiative transfer  
 190 module used by the AERONET inversion (García et al., 2012) under the assumption of  
 191 cloud-free consideration. In this code, the aerosol vertical properties have been considered  
 192 into a homogeneous atmosphere layers because of the weak dependent of ground radiances  
 193 on the whole atmospheric column with minor uncertainties (Dubovik et al., 2000). The  
 194 ~~broadband~~ fluxes from 0.20 to 4.0µm were calculated according to the radiative transfer model  
 195 GAME (Global Atmospheric ModEI) (Dubuisson et al., 1996, 2006; Roger et al., 2006). While  
 196 the broadband radiation was calculated based on the aerosol optical depth, single scattering  
 197 albedo and asymmetry factor based on those properties at four distinct wavelengths (440, 670,  
 198 870, 1020) which were linearly interpolated and extrapolated from the retrieval of the  
 199 sun/sky-radiometer measurements. The uncertainties have been found to about 30% including  
 200 the influence of spectral and solar zenith angle in the aerosol radiative effect (Myhre et al.,  
 201 2003; Zhou et al., 2005). The size distribution, complex refractive index, and spherical  
 202 particles fraction has been retrieved from the almucantar plane in the measurements. The SA  
 203 (surface albedo) is obtained from the MODIS albedo product (MCD43C3) with the interpolation



204 value of 440, 670, 870, and 1020 nm. The water vapor at 940 nm data has been retrieved in  
205 the 940 nm channel of by the sun photometer. The ozone content was fixed using the monthly  
206 climatological values of the total ozone content obtained from NASA Total Ozone Mapping  
207 Spectrometer measurements from 1978 to 2004. And oOther atmospheric gaseous  
208 profiles data were came obtained from the US standard 1976 atmosphere model. In this study,  
209 we used the two parameters of ARF at the surface (ARF-BOA) and at the top of the  
210 atmosphere (ARF-TOA) have been calculated to describes the aerosol direct radiation effect to  
211 account for the changes of the solar radiation by calculating the difference energy between the  
212 aerosols presentation and absentation.-

213 The MODIS C6 aerosol optical thickness products refined by Levy et al. (2013) were used  
214 to compare the MODIS AOD retrievals with evaluated against our ground-based observations.  
215 The MODIS C6 AOD retrievals were formed into a merged dataset combining by the Deep  
216 Blue (at 10km) and Dark Target methods (at 3km and 10km) methods separately. This version  
217 of MODIS includes some important changes from earlier versions—such as the central  
218 wavelength assumptions, Rayleigh scattering and the gas absorption performance (Levy et al.,  
219 2013)—and improvements in the radiometric calibration (Lyapustin et al., 2014). All cloud- and  
220 snow-free land surfaces have been expanded in the MODIS C6 aerosol products (Hsu et al.,  
221 2013). The AOD averaged data from Terra-MODIS and Aqua-MODIS were validated by  
222 matching the averaged CARSNET AODs within 30 minutes of the MODIS overpass within the  
223 35x5 3 pixels surrounding the CARSNET site (Tao et al., 2015). The AOD at 550 nm was  
224 interpolated between two wavelengths of the ground-based AOD measurements at 440 and  
225 675 nm.-

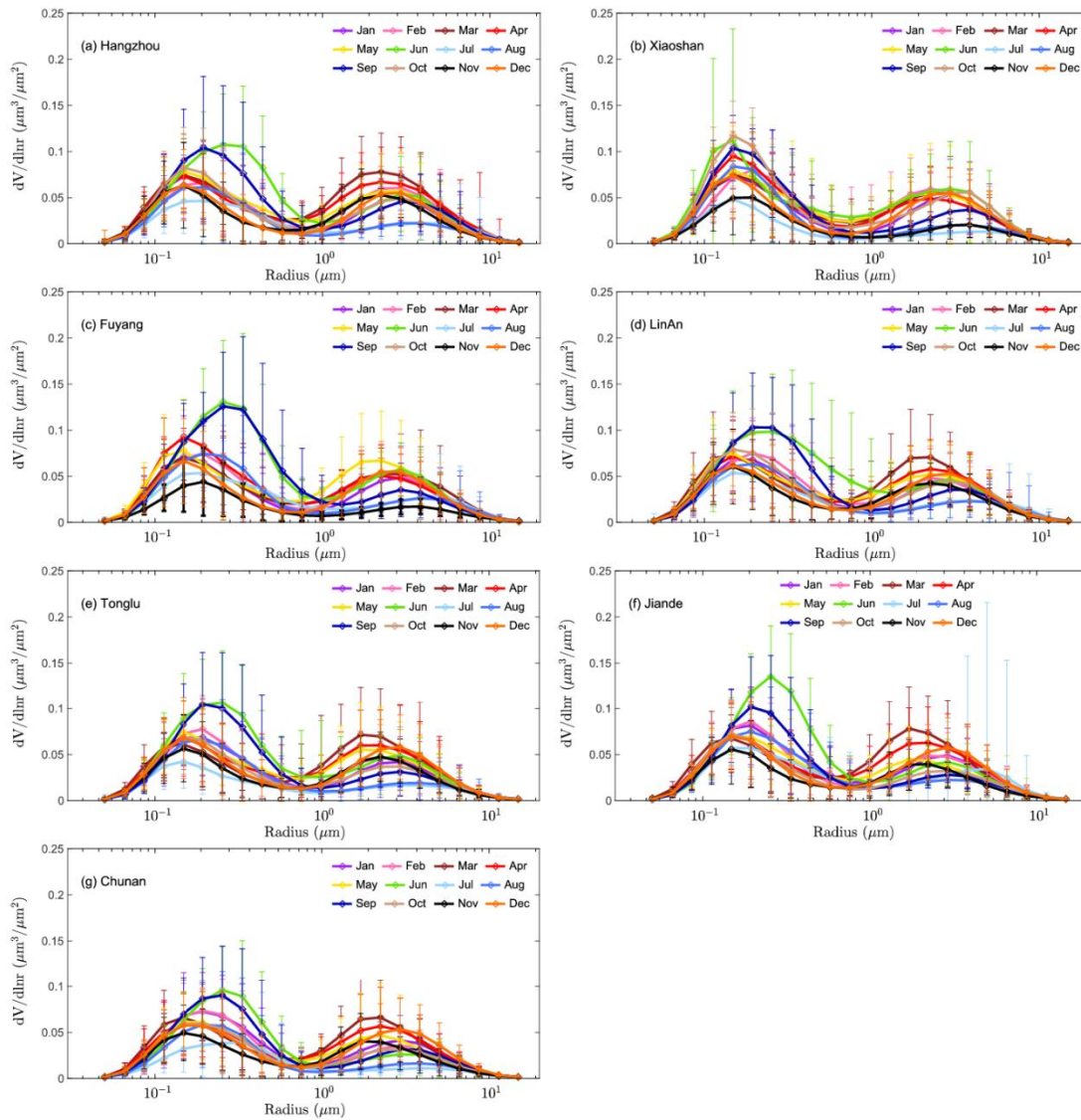
### 226 **3. Results and discussion**

#### 227 **3.1 Aerosol microphysical properties of radius and volume size distributions**

228 Fig.2 shows the monthly aerosol size distribution (dV/dlnr) in the YRD for all sites. The  
229 volumes of fine-mode aerosols were obviously higher than those of coarse-mode aerosols  
230 over all sites. The radii of fine volume fraction fine-mode radii were ~0.2–0.3 μm in the YRD

231 with a volume fraction of 0.10–0.12  $\mu\text{m}^3$  and the coarse-mode radii were  $\sim 2.0 \mu\text{m}$  with a  
232 volume fraction close to 0.07  $\mu\text{m}^3$ . The amount of fine-mode aerosols was higher in June and  
233 September than in other months at almost sites, except for Xiaoshan. This could be caused by  
234 aerosol humidification (Eck et al., 2012; Li et al., 2010, 2014; Huang et al., 2016). This  
235 phenomenon is also found over Beijing and Shenyang in north/northeast China, suggesting  
236 that hygroscopic growth occurs over many regions of China (Li et al., 2011; Che et al., 2015c).

237 The coarse-mode radius in spring at all sites was smaller than in other cities in north and  
238 northeast China affected by frequent dust transport events in spring (Kong et al., 2011; Zhao et  
239 al., 2015). The coarse-mode particles showed a larger effective radius at all seven urban,  
240 suburban and rural sites in the summer, which may due to the adhesion of new particles onto  
241 larger particles (such as fly ash).



242

243 Fig.2.Variation in the annual volume size distribution over (a) Hangzhou, (b) Xiaoshan, (c)  
 244 Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn.

245 **3.4-2 Aerosol optical properties of Aerosol optical depth and Angström exponent**

246 The annual mean of AOD at 440nm over the seven urban, suburban and rural sites in this  
 247 study varied ranges from 0.68 to 0.76 (Table 1). Smaller observation samples hasSmaller  
 248 observation samples hasve been found in Xiaoshan and Fuyang with 180 and 217 available  
 249 observation days, respectively. The number of 180 observation days in Xiaoshan is less than  
 250 half of the year may have less representative and need further data accumulation, while the  
 251 the observation days of 217 in Fuyang was more than half of the year may not affect the

252 [comparability between the other sites.](#) The annual values of the AOD<sub>440nm</sub> at Hangzhou,  
253 Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn were about 0.76±0.42, 0.76±0.43,  
254 0.76±0.45, 0.73±0.44, 0.71±0.41, 0.73±0.40 and 0.68±0.38, respectively, which suggests that  
255 [column](#) aerosol loading is at a high level at all seven urban, suburban and rural sites in the  
256 YRD. ~~This suggests that aerosol pollution is~~ on the regional rather than the local scale ~~in the~~  
257 ~~YRD region.~~ [The AOD<sub>440nm</sub> decreased from the eastern coast to the inland areas towards the](#)  
258 [west \(from ~0.76±0.42 at Hangzhou to ~0.68±0.38 at ChunAn\) due to the high aerosol loading](#)  
259 [from economic development and anthropogenic influences.](#) ~~The annual annual AOD<sub>440nm</sub>~~  
260 [shows that the aerosol loading has similar level in Hangzhou, Xiaoshan and Fuyang, and with](#)  
261 [the 4%-10% decrease in LinAn, Tonglu, Jiande and ChunAn, respectively.](#) The AOD<sub>440nm</sub> at the  
262 urban site of Hangzhou was ~~the highest of all the study sites~~ as a result of ~~high local~~  
263 ~~anthropogenic activity~~ in this urban area ~~compared with the other suburban and rural sites.~~ [the](#)  
264 [more industrial activity and high resident density in the eastern part of the Hangzhou](#)  
265 [metropolis region resulting in larger aerosol emissions compared with the other suburban and](#)  
266 [rural sites.](#) ~~The AOD at the rural site of ChunAn was lower than at the urban and suburban sites~~  
267 ~~due to lower levels of anthropogenic activity.~~ ~~The AOD decreased from the eastern coast to the~~  
268 ~~inland areas towards the west (from ~0.76±0.42 at Hangzhou to ~0.68±0.38 at ChunAn).~~ ~~This is~~  
269 ~~due to the high aerosol loading from economic development and anthropogenic~~  
270 ~~influences.~~ ~~There is more industrial activity and high resident density in the eastern part of the~~  
271 ~~Hangzhou metropolis region, resulting in higher aerosol emissions.~~

272 ~~The AOD in Hangzhou in urban eastern China was similar to that in Shenyang (0.75) in~~  
273 ~~urban northeast China (Zhao et al., 2013), and in Beijing (0.76) and Tianjin (0.74) in urban~~  
274 ~~north China (Che et al., 2015b), indicating that the aerosol extinction pollution is both common~~  
275 ~~and at a similar level throughout most urban areas of China.~~ ~~The AOD values at the urban and~~  
276 ~~suburban sites of Hangzhou were slightly higher than at Pudong (0.70) and Hefei (0.69), other~~  
277 ~~urban areas in eastern China, suggesting that higher aerosol extinction ability loadings were~~  
278 ~~emitted here observed here (He et al., 2012; Liu et al., 2017).~~ ~~However, the AOD at all seven~~  
279 ~~sites was lower than that obtained at Wuhan (1.05), Nanjing (0.88), Dongtan (0.85), Taihu (0.77)~~  
280 ~~and Xuzhou (0.92) in previous studies in eastern China (Wang et al., 2015; Li et al., 2015; Pan~~

281 ~~et al., 2010; Xia et al., 2007; Wu et al., 2016). This indicates that the aerosol loading caused by~~  
 282 ~~anthropogenic activities is very high in both urban and suburban areas in eastern China. The~~  
 283 ~~site at LinAn is regarded as the regional background clean site in eastern China and is~~  
 284 ~~representative of the background atmospheric characteristics of this region (Che et al., 2009c).~~  
 285 ~~The with an average AOD at LinAn was about 0.73±0.44, which is higher than that at the other~~  
 286 ~~regional background stations of China, such as Longfengshan (0.35; northeastern China), Mt~~  
 287 ~~Waliguan (0.14, inland Asia), Xinglong (0.28, northern China), Akedala (0.20, northwestern~~  
 288 ~~China) and Shangri-La (0.11, southwestern China) (Wang et al., 2010; Che et al., 2011; Zhu et~~  
 289 ~~al., 2014; Che et al., 2015b). The aerosol loading in eastern China (especially in the YRD~~  
 290 ~~region) is at least twice as high as in other regions of China which indicate the strong aerosol~~  
 291 ~~extinction.~~

292 Table1. Geographical location and annual mean optical parameters of aerosols at the seven  
 293 observation sites in the YRD.

	Hangzhou	Xiaoshan	Fuyang	LinAn	Tonglu	Jiande	
Site type	Urban	Suburban	Suburban	Suburban	Suburban	Suburban	
Longitude (°E)	120.19	120.25	119.95	119.72	119.64	119.27	
Latitude (°N)	30.26	30.16	30.07	30.23	29.80	29.49	
Altitude (m)	41.9	14.0	17.0	139	46.1	88.9	
<sup>a</sup> N <sub>day</sub>	485	180	217	562	498	480	
<sup>b</sup> N <sub>inst.</sub>	2052	752	906	2410	2255	1952	
<sup>c</sup> AOD <sub>500nm</sub>	<u>0.68±0.46</u>	<u>0.67±0.43</u>	<u>0.66±0.43</u>	<u>0.60±0.42</u>	<u>0.60±0.41</u>	<u>0.63±0.38</u>	
<sup>e</sup> AOD <sub>440nm</sub>	0.76±0.42	0.76±0.43	0.76±0.45	0.73±0.44	0.71±0.41	0.73±0.40	
<sup>e</sup> AOD <sub>fine</sub> <sup>d</sup> AOD <sub>fine(440nm)</sub>	0.68±0.42	0.69±0.41	0.69±0.44	0.66±0.43	0.64±0.41	0.66±0.40	
<sup>e</sup> AOD <sub>coarse</sub> <sup>d</sup> AOD <sub>coarse(440nm)</sub>	0.08±0.06	0.07±0.06	0.07±0.06	0.07±0.07	0.07±0.06	0.07±0.07	
<sup>d</sup> EAE <sup>e</sup> EAE	1.29±0.26	1.37±0.24	1.32±0.24	1.29±0.27	1.30±0.26	1.32±0.28	
<sup>e</sup> SSA <sub>440nm</sub> <sup>d</sup> SSA <sub>440nm</sub>	0.91±0.06	0.93±0.04	0.94±0.04	0.93±0.05	0.92±0.04	0.92±0.05	
<sup>e</sup> SSA <sub>fine</sub> <sup>ef</sup> SSA <sub>670nm</sub> <sup>fine</sup>	0.923±0.065	0.915±0.064	0.935±0.064	0.924±0.054	0.934±0.054	0.924±0.075	
<sup>e</sup> SSA <sub>coarse</sub> <sup>dg</sup> SSA <sub>870nm</sub> <sup>coarse</sup>	0.9082±0.079	0.9083±0.078	0.9184±0.08	0.981±0.068	0.981±0.068	0.9082±0.089	
<sup>h</sup> SSA <sub>1020nm</sub>	<u>0.89±0.08</u>	<u>0.89±0.08</u>	<u>0.89±0.09</u>	<u>0.90±0.07</u>	<u>0.90±0.07</u>	<u>0.90±0.09</u>	
<sup>e</sup> Real <sup>d</sup> Real	1.43±0.07	1.41±0.06	1.41±0.06	1.42±0.06	1.43±0.06	1.41±0.05	
<sup>e</sup> Imaginary <sup>d</sup> Imaginary	0.011±0.010	0.008±0.006	0.007±0.006	0.009±0.007	0.009±0.007	0.010±0.009	
<sup>e</sup> AAOD <sup>d</sup> AAOD	0.06±0.05	0.05±0.04	0.04±0.04	0.05±0.04	0.05±0.04	0.06±0.04	
<sup>d</sup> AAE <sup>e</sup> AAE	1.13±0.46	0.88±0.42	0.85±0.43	0.98±0.35	1.11±0.49	1.16±0.44	
<sup>e</sup> R <sub>meas</sub> <sup>d</sup> R <sub>meas</sub> (μm)	0.70±0.34	0.65±0.31	0.66±0.33	0.66±0.33	0.65±0.33	0.62±0.24	
<sup>e</sup> R <sub>meas</sub> <sub>fine</sub> <sup>d</sup> R <sub>meas</sub> <sub>fine</sub> (μm)	0.18±0.05	0.18±0.04	0.19±0.05	0.19±0.05	0.19±0.05	0.19±0.05	
<sup>e</sup> R <sub>meas</sub> <sub>coarse</sub> <sup>d</sup> R <sub>meas</sub> <sub>coarse</sub> (μm)	2.67±0.47	2.73±0.42	2.75±0.45	2.71±0.52	2.66±0.48	2.63±0.47	
<sup>e</sup> Reff <sup>d</sup> Reff(μm)	0.30±0.10	0.29±0.09	0.30±0.09	0.29±0.10	0.29±0.10	0.29±0.09	

<sup>e</sup> Reff <sub>fine</sub> <sup>d</sup> Reff <sub>fine</sub> (μm)	0.16±0.04	0.16±0.03	0.17±0.04	0.16±0.04	0.16±0.04	0.17±0.04
<sup>e</sup> Reff <sub>coarse</sub> <sup>d</sup> Reff <sub>coarse</sub> (μm)	2.21±0.40	2.26±0.35	2.30±0.39	2.24±0.44	2.19±0.41	2.16±0.39
<sup>e</sup> Volume <sup>d</sup> Volume (μm <sup>3</sup> )	0.19±0.09	0.19±0.09	0.19±0.09	0.18±0.09	0.17±0.09	0.18±0.09
<sup>e</sup> Volume <sub>fine</sub> <sup>d</sup> Volume <sub>fine</sub> (μm <sup>3</sup> )	0.10±0.06	0.11±0.06	0.11±0.07	0.10±0.06	0.10±0.06	0.10±0.06
<sup>e</sup> Volume <sub>coarse</sub> <sup>d</sup> Volume <sub>coarse</sub> (μm <sup>3</sup> )	0.09±0.06	0.08±0.05	0.08±0.06	0.08±0.05	0.08±0.06	0.08±0.07
<sup>e</sup> ARF <sup>d</sup> ARF-BOT (W/m <sup>2</sup> )	-93±44	-84±41	-80±40	-81±39	-79±39	-82±40
<sup>e</sup> ARF <sup>d</sup> ARF-TOA (W/m <sup>2</sup> )	-35±20	-36±21	-37±21	-36±21	-35±20	-35±21

294 <sup>a</sup> Number of available observation days.

295 <sup>b</sup> Number of instantaneous observations.

296 <sup>c</sup> Optical parameters at a wavelength of 440-500nm.

297 <sup>d</sup> [Optical parameters at a wavelength of 440 nm.](#)

298 <sup>d</sup> [Angström](#)-<sup>e</sup> [Angström](#) exponents between 440 and 870 nm.

299 <sup>f</sup> [Optical parameters at a wavelength of 670 nm.](#)

300 <sup>g</sup> [Optical parameters at a wavelength of 870 nm.](#)

301 <sup>h</sup> [Optical parameters at a wavelength of 1020 nm.](#)

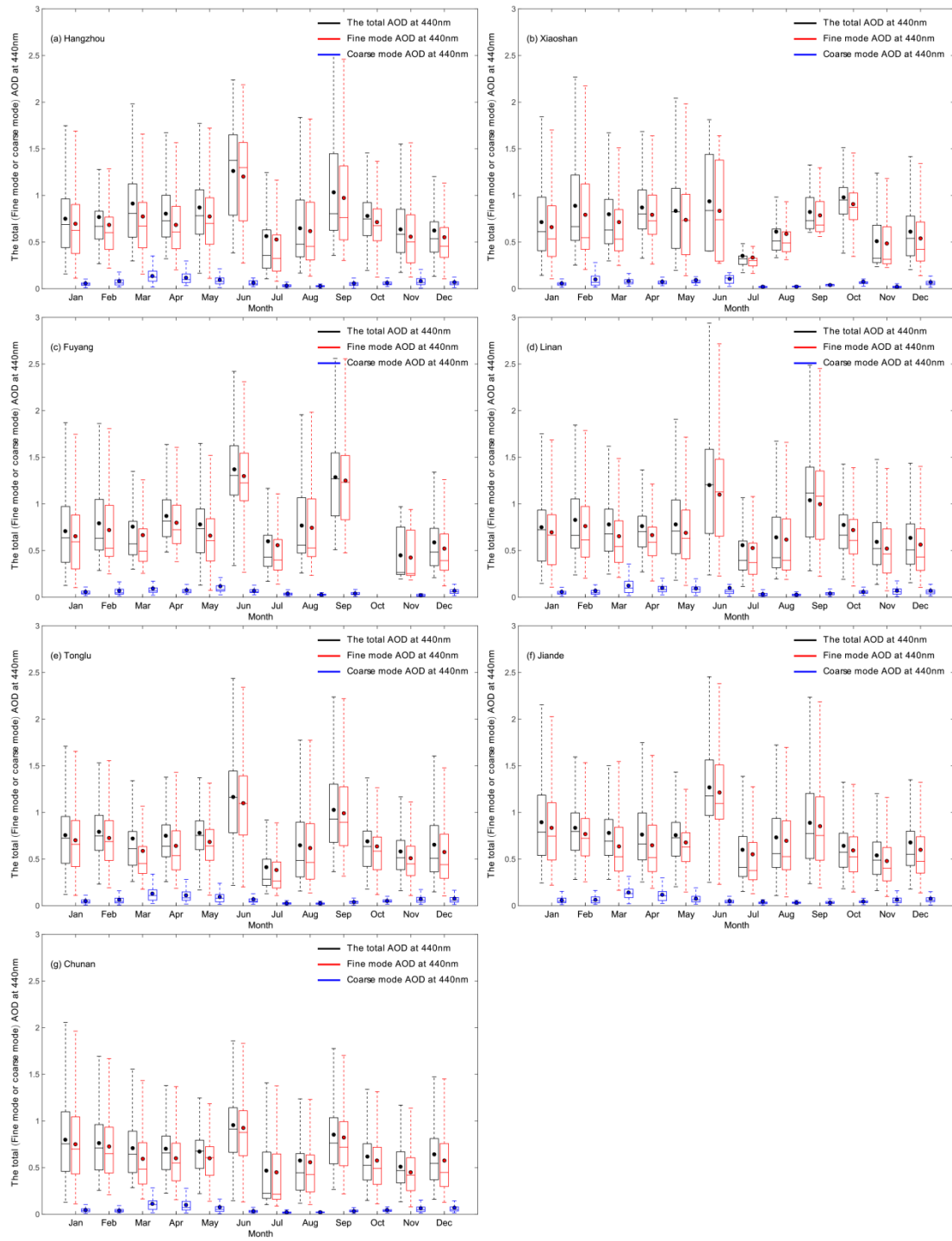
302

303 Ding et al. (2013a,b) showed that plumes from agricultural burning in June may  
304 significantly and seriously affect the radiation balance and air quality of the YRD region. In this  
305 study, the monthly averaged AODs at most sites showed two peaks in June and September  
306 (Fig.-23) with values of ~1.26±0.50 and ~1.03±0.57, respectively. This may be attributed to the  
307 accumulation of fine-mode particles via hygroscopic growth in the summer season and the  
308 burning of crop residue biomass under a continental high-pressure system with good  
309 atmospheric stability and frequent temperature inversions. These conditions lead to the poor  
310 diffusion of pollutants (Xia et al., 2007). [As Ffig.3 shown, tThe monthly average value of the](#)  
311 [extinction Angström exponent \(EAE, -dln\[EAOD\(λ\)\]/dln\(λ\)\) EAE in Hangzhou was higher in](#)  
312 [January \(~1.40±0.23\) and September \(~1.43±0.24\). This conclusion is also indicated the](#)  
313 [dominance of small particles from anthropogenic emissions and agricultural activity in autumn](#)  
314 [and winter \(Tan et al., 2009\).](#)

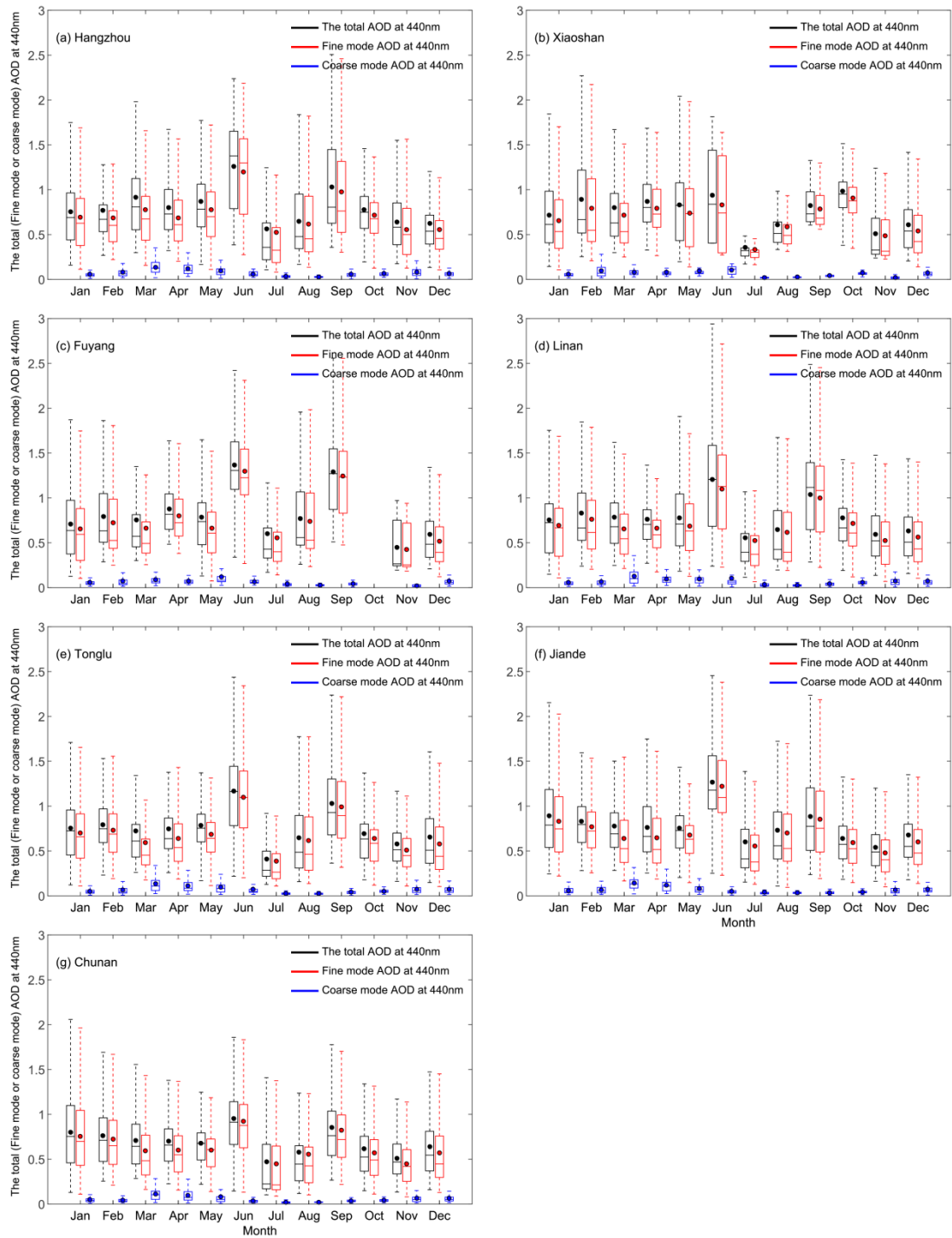
315 The annual fine-mode AOD values at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu,  
316 Jiande and ChunAn were about 0.68±0.42, 0.69±0.41, 0.69±0.44, 0.66±0.43, 0.64±0.41,  
317 0.66±0.40 and 0.61±0.38, respectively (Fig.-23). The seasonal variation in the AOD was  
318 similar to the total AOD at these urban, suburban and rural sites. The [fine-mode fraction of](#)  
319 [AODratio AOD<sub>f</sub>/AOD<sub>t</sub>](#) consistently exceeded 0.90 [\\_at all sites,](#) which indicates [that fine-mode](#)  
320 [particles make a major contribution of fine mode fraction](#) to the total AOD in the YRD.

321 ~~Moreover, the f~~Figure 3 shows that the annual extinction Angström exponent (EAE) ~~EAE at~~  
322 ~~Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn was about 1.29±0.26,~~  
323 ~~1.37±0.24, 1.32±0.24, 1.29±0.27, 1.30±0.26, 1.32±0.28 and 1.22±0.25, respectively. Values of~~  
324 ~~EAE >1.20 were found in all months throughout the year, indicating that small particle size~~  
325 ~~distributions were favored in the YRD region.~~ The annual coarse-mode AOD values at  
326 Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn were between about 0.06  
327 and 0.08. ~~The with the ratio coarse mode fraction of  $AOD_{coarse}/AOD_t$  was~~ about 0.10, which  
328 indicates ~~that about 10% of the 10% contribution of coarse mode fraction~~ to the AOD in the  
329 YRD ~~region is from coarse particles.~~ The variation in the coarse-mode AOD (Fig. 2) also  
330 ~~showed a significant increase in March at all seven sites of about 0.14±0.08, 0.08±0.04,~~  
331 ~~0.09±0.09, 0.13±0.11, 0.13±0.11, 0.14±0.08 and 0.11±0.07 at Hangzhou, Xiaoshan, Fuyang,~~  
332 ~~LinAn, Tonglu, Jiande and ChunAn, respectively.~~ The monthly average value of the EAE in  
333 ~~Hangzhou was higher in January (-1.40±0.23) and September (-1.43±0.24). This indicated~~  
334 ~~the dominance of small particles from anthropogenic emissions and agricultural activity in~~  
335 ~~autumn and winter (Tan et al., 2009). The lower EAE was lower in March (-1.16±0.24) and~~  
336 ~~April (-1.13±0.22).~~ ~~Though~~ The less coarse mode fraction indicated that there is no obvious  
337 ~~effect of the coarse particles in the YRD region than that contributed to the higher aerosol~~  
338 ~~loading in other north/northeast China that contributed to the higher aerosol loading (Zhang et~~  
339 ~~al., 2012).~~ Some ~~dust~~ dust cases ~~has~~ also can be observed ~~found~~ in YRD region that  
340 ~~transported from north/northwest China during 2012-2015 reflect the effect of mineral dust~~  
341 ~~aerosols (Gong et al., 2003).~~ ~~I suspect that~~ The fugitive dust from road traffic ~~or~~ and  
342 ~~construction activity is another more persistent and significant source for China's cities as well~~  
343 ~~as these eastern megacities,~~ which reflects the effect of mineral dust aerosols (Gong et al.,  
344 ~~2003). However, this effect is not as obvious in the YRD region as other regions in north or~~  
345 ~~northeast China which contributed to the optical properties of aerosols in this region (Zhang et al.,~~  
346 ~~2012).~~

347 This was mainly caused by dust episodes from north/northwest China,  
348 which contributed to the optical properties of aerosols in this region (Zhang et al., 2012).







350

351 Fig. 23. Variation in the total, fine- and coarse-mode AOD<sub>440 nm</sub> over (a) Hangzhou, (b)  
 352 Xiaoshan, (c) Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn. The boxes represent  
 353 the 25th to 75th percentile distribution, while the dots and solid lines within each box represent  
 354 the mean and median, respectively.

355 ~~Figure 3 shows that the annual extinction Angström exponent (EAE) at Hangzhou,~~

356 ~~Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn was about  $1.29 \pm 0.26$ ,  $1.37 \pm 0.24$ ,~~  
357  ~~$1.32 \pm 0.24$ ,  $1.29 \pm 0.27$ ,  $1.30 \pm 0.26$ ,  $1.32 \pm 0.28$  and  $1.22 \pm 0.25$ , respectively. Values of~~  
358 ~~EAE  $> 1.20$  were found in all months throughout the year, indicating that small particle size~~  
359 ~~distributions were favored in the YRD region. The monthly average value of the EAE in~~  
360 ~~Hangzhou was higher in January ( $-1.40 \pm 0.23$ ) and September ( $-1.43 \pm 0.24$ ). This indicated the~~  
361 ~~dominance of small particles from anthropogenic emissions and agricultural activity in autumn~~  
362 ~~and winter (Tan et al., 2009). The EAE was lower in March ( $-1.16 \pm 0.24$ ) and April ( $-1.13 \pm 0.22$ ),~~  
363 ~~which reflect the effect of mineral dust aerosols (Gong et al., 2003). However, this effect is not as~~  
364 ~~obvious in the YRD region as other regions in north or northeast China.~~

365 Moreover, we also discuss the monthly and diurnal cycle of AOD at 500nm has also  
366 been discussed in Fig.4 and Fig.5. The annual values of AOD<sub>500nm</sub> over the seven urban,  
367 suburban and rural sites in this study varied from 0.5368 (ChunAn) to 0.7668 (Hangzhou). The  
368 results show that two peaks of AOD at 500nm occurs in June and September in the seven  
369 megacity of eastern China. The higher AOD<sub>500nm</sub> occurs in June and September with values of  
370  $0.491.25 \pm 0.5919$  and  $0.231.00 \pm 0.3442$  in the urban site of Hangzhou, respectively which  
371 has the similar pattern as the other sites. The increase of AOD at 500nm in June is not  
372 corresponding to the same increase pattern of EAE (about 1.5) which indicates the aerosols  
373 types may be relatively constant in this region. The Fig.452 depicts the diurnal patterns of AOD  
374 at 500nm in this megacity area of eastern China. We can see that there are two types of  
375 diurnal patterns in this region. The daily AOD has been found increased in early morning  
376 (08:00 hr to 09:00 hr) about and afternoon (12:00 hr to 14:00 hr) about the value of 0.60 to  
377 0.70 has been found in Hangzhou, Xiaoshan, Fuyang and Linan, while the decreasing of daily  
378 AOD has been observed from 0.70 to 0.50 during the daytime (from 07:00 hr to 16:00 hr) in  
379 Tonglu, Jiande and ChunAn. The high AOD during 07:00~09:00 in the urban area may be due  
380 to the anthropogenic activities and aerosol emissions from the morning rush hour. The  
381 decreased AOD with the value of  $0.37 \pm 0.36$  occurred in the suburban cities of Tonglu, Jiande  
382 and ChunAn may be due to the meteorological conditions more than anthropogenic effects.  
383 During the day, the aerosols in the near-surface may spread into vertical as a result of  
384 turbulence due to the more and more unstable atmosphere by the continuous strengthening of

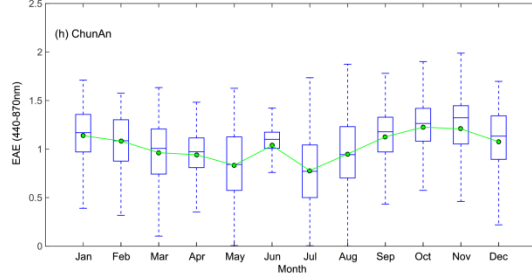
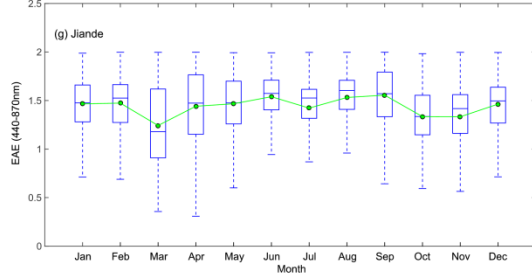
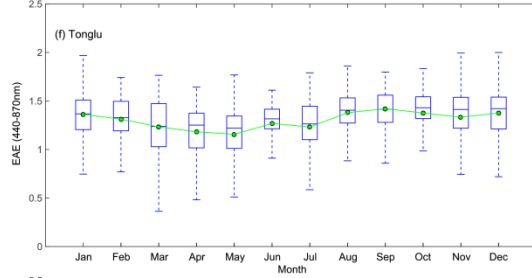
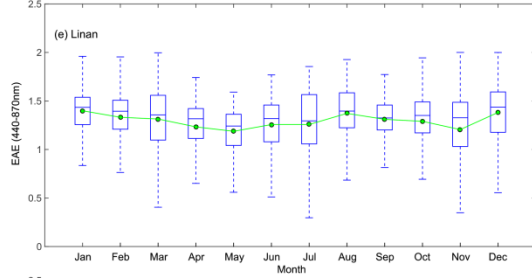
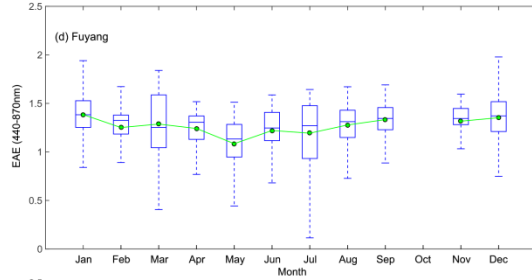
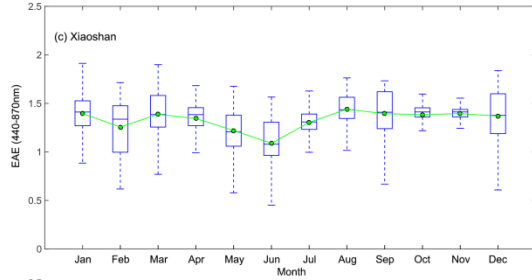
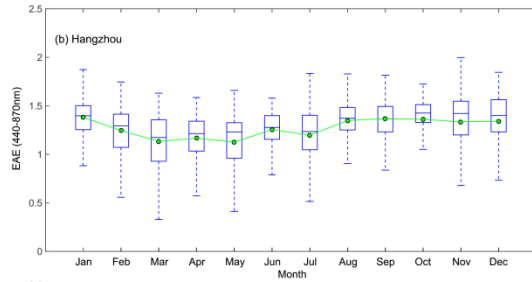
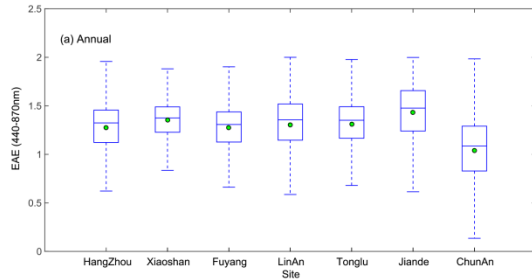
385

solar radiation.

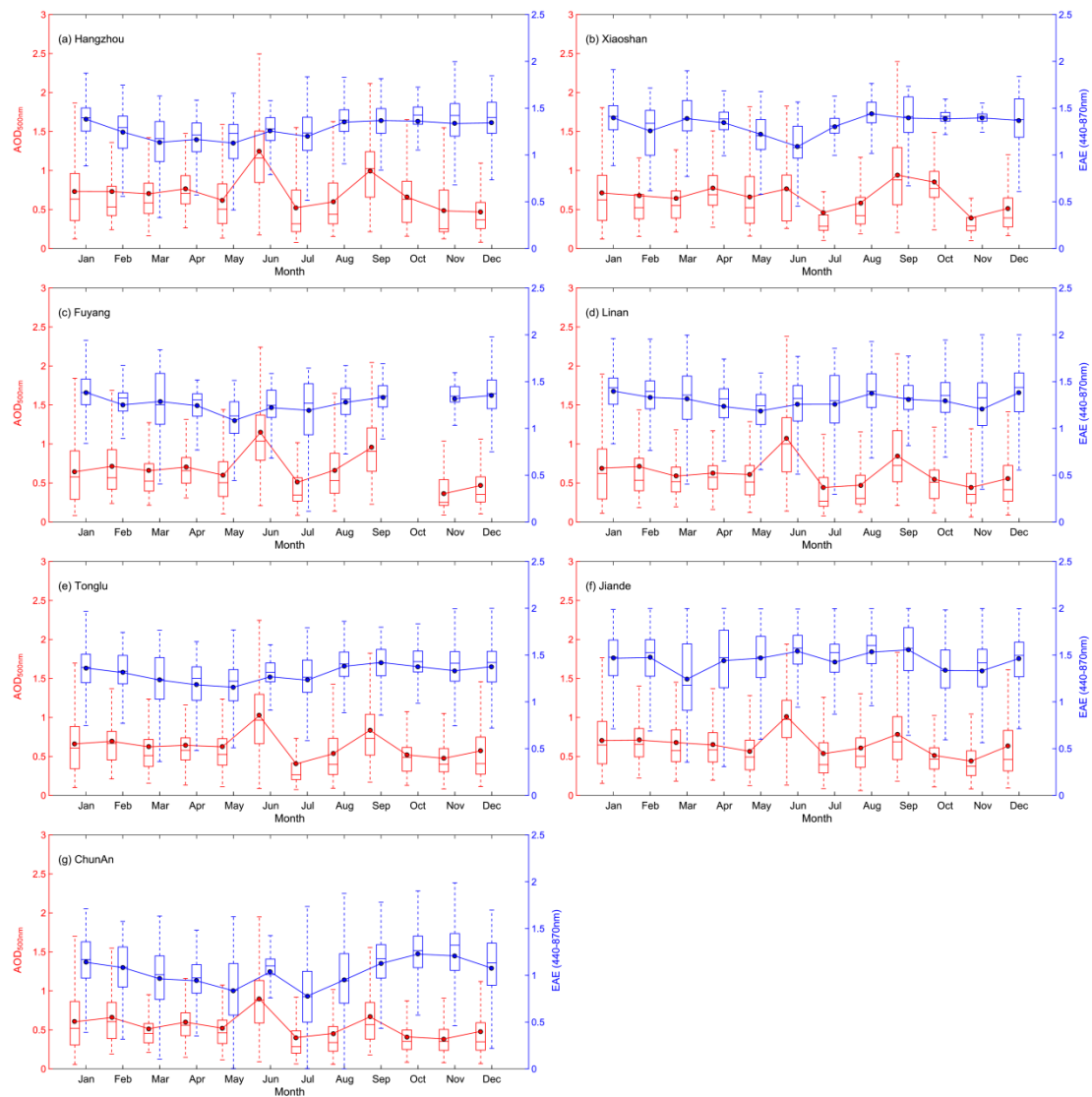
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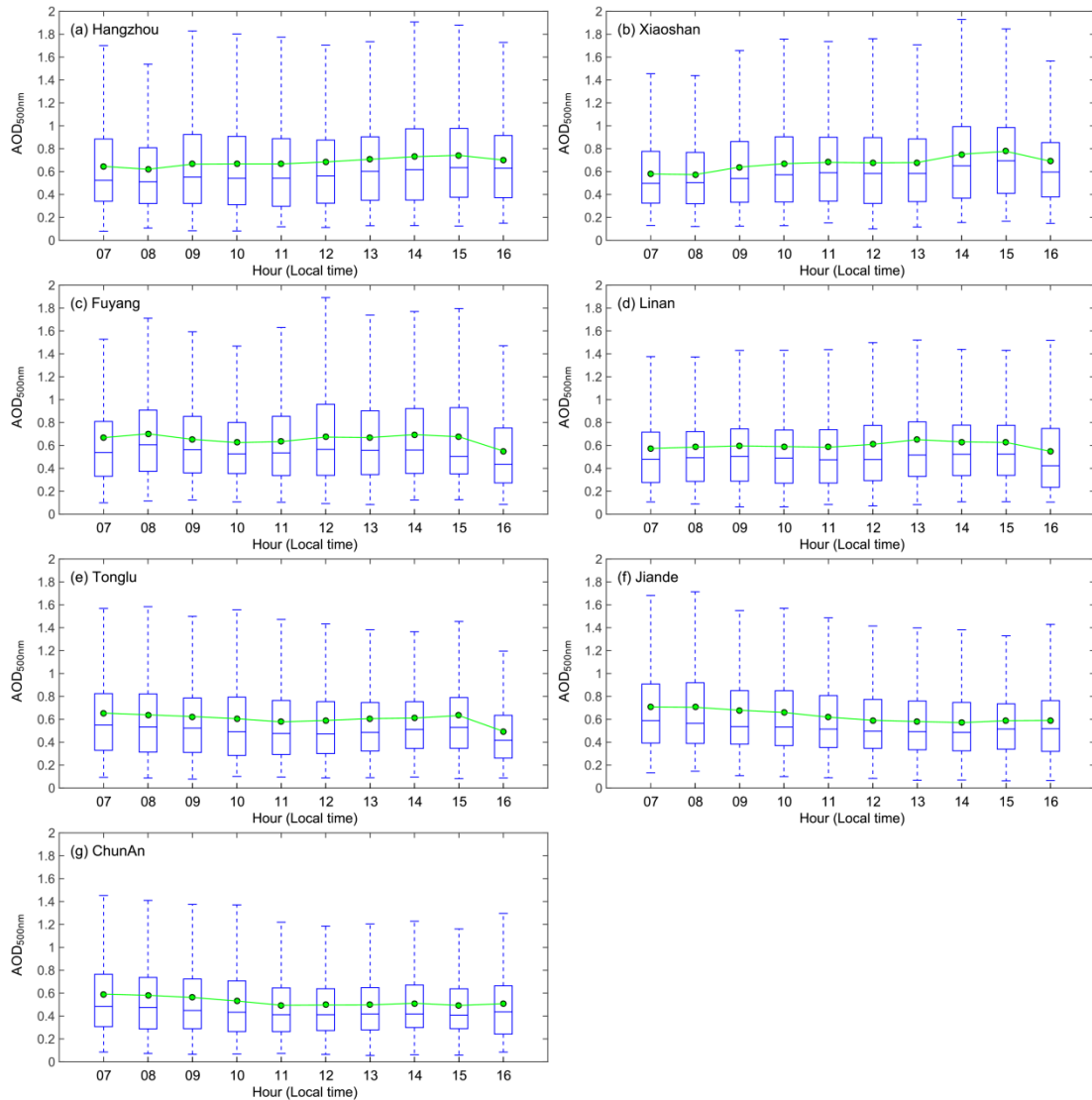
389



390

391 Fig.34. (a) Annual variation in the EAE at 440–870 nm. Variation in the AOD at 500nm & EAE  
 392 at 440–870 nm over (ba) Hangzhou, (eb) Xiaoshan, (ec) Fuyang, (ed) LinAn, (fe) Tonglu, (gf)  
 393 Jiande and (hg) ChunAn. The boxes represent the 25th to 75th percentile distribution, while  
 394 the dots and solid lines within each box represent the mean and median, respectively.

395



396

397 Fig.5. Variation of diurnal cycle in the AOD at 500 nm over (a) Hangzhou, (b) Xiaoshan, (c)  
 398 Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn. The boxes represent the 25th to 75th  
 399 percentile distribution, while the dots and solid lines within each box represent the mean and  
 400 median, respectively.

401 Validation of the MODIS C6 retrieval AOD values was carried out by comparison with  
 402 ground-based observations (Figure 4).The product of Terra-MODIS/Terra and  
 403 Aqua-MODIS/Aqua with Deep Blue (at 10km) and Dark Target (at 3km and 10km) methods at  
 404 3km and 10km has been evaluated against by ground-based observations separately in Figure.  
 405 654-802. We use the better estimated data of Quality flag = 3 and Quality flag=2, 3 for DT and  
 406 TB methods, respectively. The systematic performance of the Terra-MODIS/TerraMODIS C6

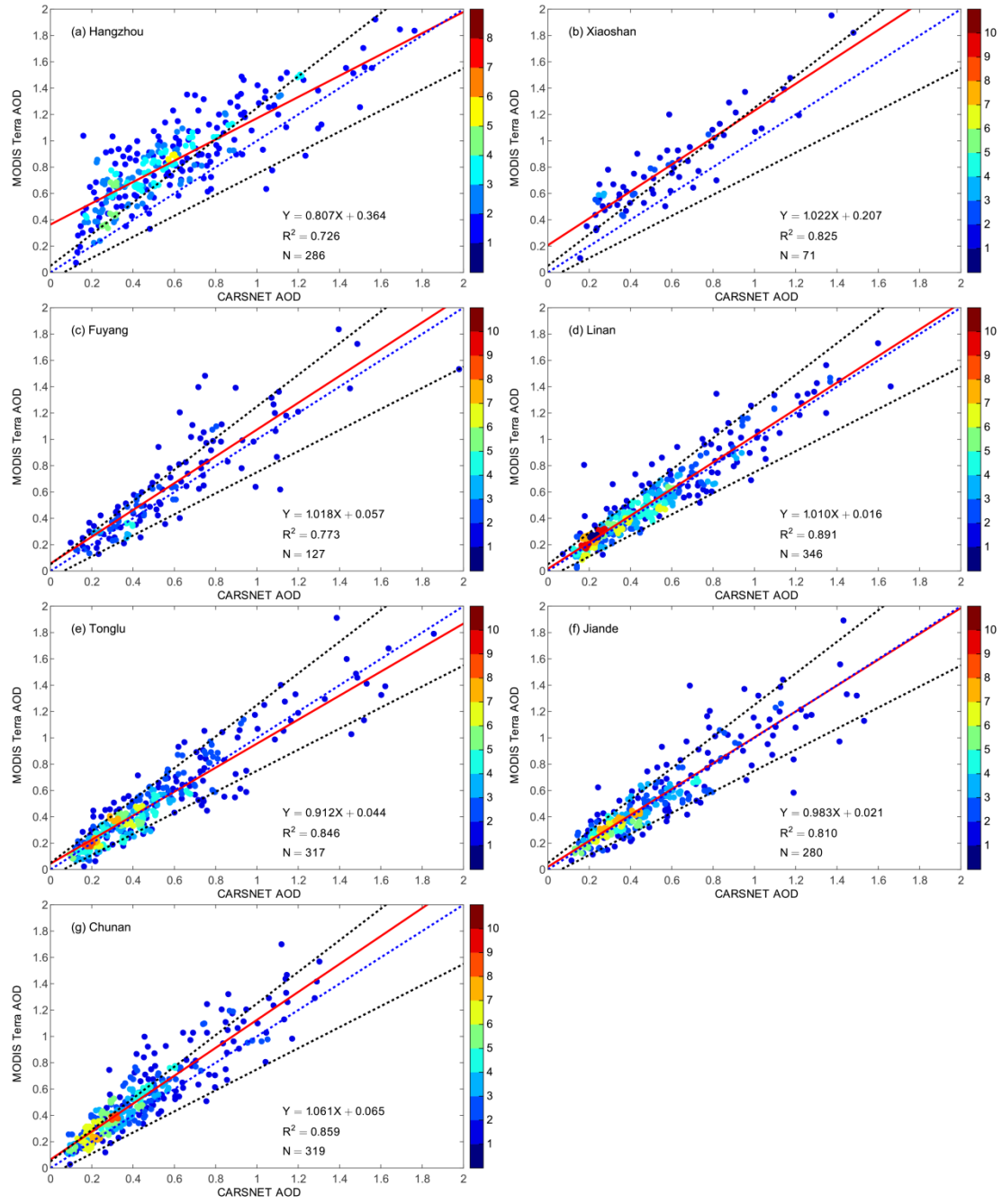
407 retrieval AOD values was generally more stable in the YRD region compared with the  
408 Aqua-MODIS/Aqua product with the two Deep Blue and Dark Target methods, with which most  
409 of the plots scattered around the 1:1 regression line. The correlation coefficients ( $R^2$ ) fitting  
410 relations between the Terra-MODIS and sun photometer AOD (550 nm) values by the Deep  
411 Blue methods at 10km were better than that of by the Dark Target methods.

412 about 0.73, 0.83, 0.77, 0.89, 0.85, 0.81 and 0.86 at Hangzhou, Xiaoshan, Fuyang, LinAn,  
413 Tonglu, Jiande and ChunAn, respectively. The correlation coefficients (R) of between the  
414 Aqua-MODIS/Aqua and Terra-MODIS/Terra between by the Dark Target methods at 3km and  
415 sun photometer AOD (550 nm) values by the Dark Target methods at 3km were about 0.7084  
416 to 0.8492 and 0.7385 to 0.8994 in the YRD region, respectively. The linear regression fitting  
417 performed better at the suburban sites of LinAn and Jiande according to the product of  
418 MODIS/TerraTerra-MODIS by the Dark Target methods at 3km. The fitting curve was almost  
419 consistent with the 1:1 reference line, which suggests that the aerosol properties were well  
420 defined for the MODIS C6 products. A large part of the MODIS retrieval AOD value was  
421 outside the expected error envelope of  $\pm (0.05 + 20\%T_{CARSNET})$ , especially for AOD  
422 values  $< 0.80$  in Hangzhou and Xiaoshan. This indicates that the MODIS retrieval algorithm  
423 could still be improved, especially in urban areas. The MODIS retrieval AOD performed better  
424 at the other five sites (Fuyang, LinAn, Tonglu, Jiande and ChunAn) in the YRD; most of the  
425 retrieved AOD values for these sites fell within the expected error envelope. The  
426 MODIS/AquaMODIS retrievals with Dark Target methods at 3km were overestimated  
427 underestimated while the MODIS/Terra retrievals with Dark Target methods at 3km were  
428 overestimated except at Hangzhou, Xiaoshan-Tonglu and ChunAnJiande. This could be  
429 because the MODIS SSA was underestimated at and near to urban sites (Tao et al., 2015).  
430 The small deviation at the suburban sites suggested that the MODIS C6 retrieval using the DT  
431 method was suitable for capturing the optical properties of aerosols in suburban areas with  
432 dense vegetation coverage of the YRD. However, this method may have larger difference in  
433 the urban areas with less vegetation such as Hangzhou. The correlation coefficients (R) of the  
434 MODIS/Aqua and MODIS/TerraAqua-MODIS and Terra-MODIS between sun photometer AOD  
435 (550 nm) values by the Deep Blue and Dark Target methods at 10km were about 0.6581 to

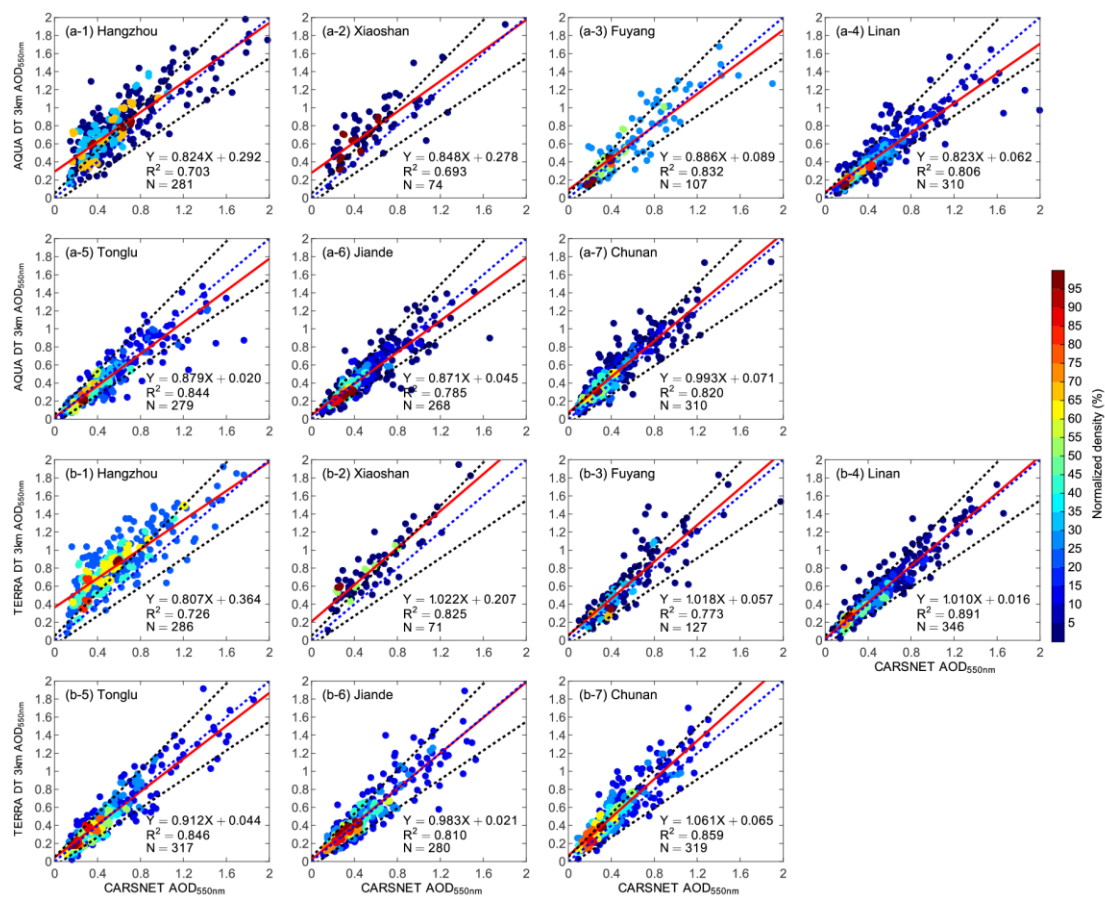
436 0.9084, 0.8573 to 0.9084, 0.6948 to 0.9182 and 0.8572 to 0.9386 in the YRD region,  
437 respectively. The MODIS/Aqua and MODIS/Terra retrievals with Deep Blue and Dark Target  
438 methods at 10km were underestimated except Hangzhou and Xiaoshan. In particular, the  
439 biases of the correlation coefficients (R) occurred in LinAn and Jiande –has decreased from  
440 0.94 and 0.90 to 0.87 and 0.88. The validation results correlation indicates is not as better as  
441 the MODIS product at 3km which indicate a good MODIS/TerraMODIS matching with better  
442 fitting correlation at 3km rather than 10km products.

443

444 The AOD overestimation retrieved using Dark Target (DT) and Deep Blue (DB) methods  
445 are more influenced by the SSA and the phase function of aerosol in eastern China with  
446 AOD >0.4 (Tao et al. 2015). Therefore, the detailed ground-based observation in this work is  
447 more helpful to the calibration of MODIS retrievals in eastern China.







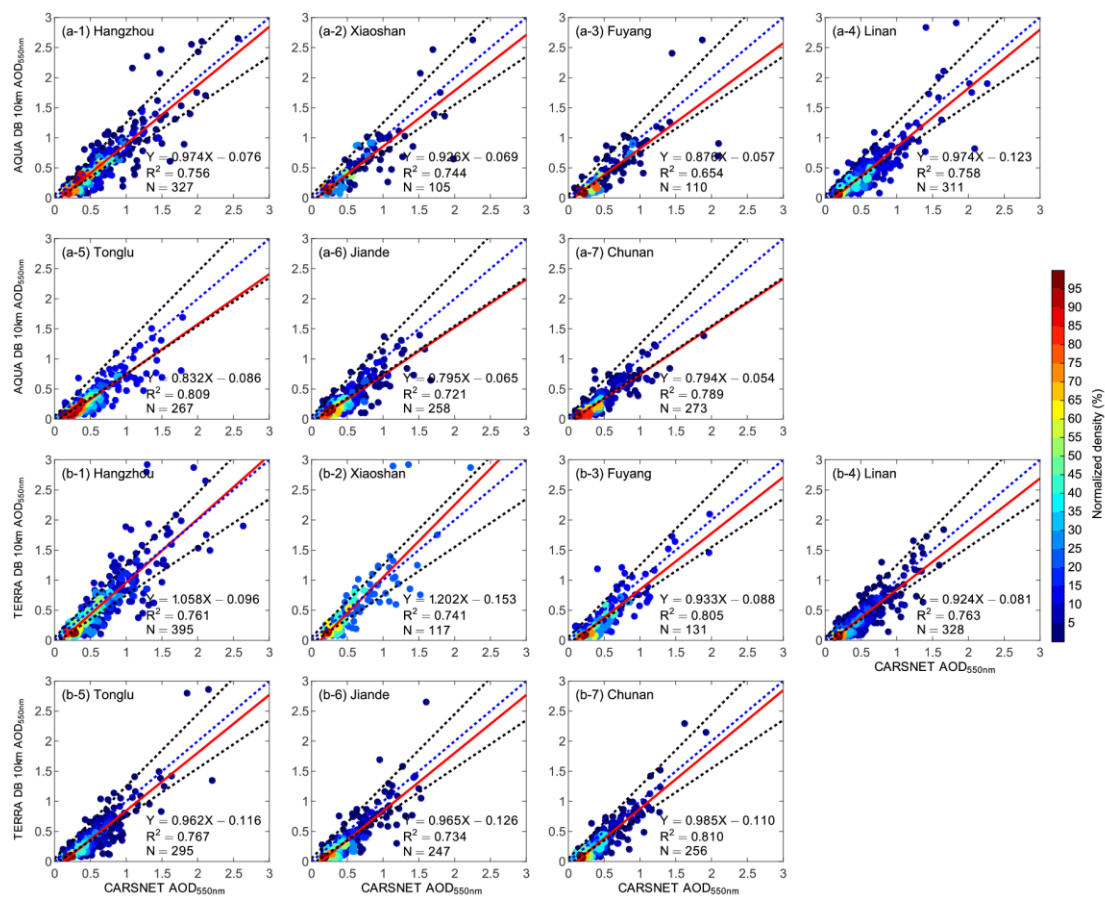
449

450 Fig.564. Comparison of  $\text{C6-MODIS/Aqua Dark Target (DT)MODIS AOD at 550 nm}$  with  
 451 the CARSNET AOD by the Dark Target methods at 3km in (a-1) Hangzhou, (a-2) Xiaoshan,  
 452 (a-3) Fuyang, (a-4) LinAn, (a-5) Tonglu, (a-6) Jiande, (a-7) ChunAn and  
 453 MODIS/TerraTerra-MODIS DT AOD at 550 nm with the CARSNET AOD by the Dark Target  
 454 methods at 3km in (b-1) Hangzhou, (b-2) Xiaoshan, (b-3) Fuyang, (b-4) LinAn, (b-5) Tonglu,  
 455 (b-6) Jiande, (b-7) ChunAn. The red solid line represents the linear regression. The two black  
 456 dotted lines represent the expected errors in the MODIS retrievals.

457 6Terra-at 3km

458

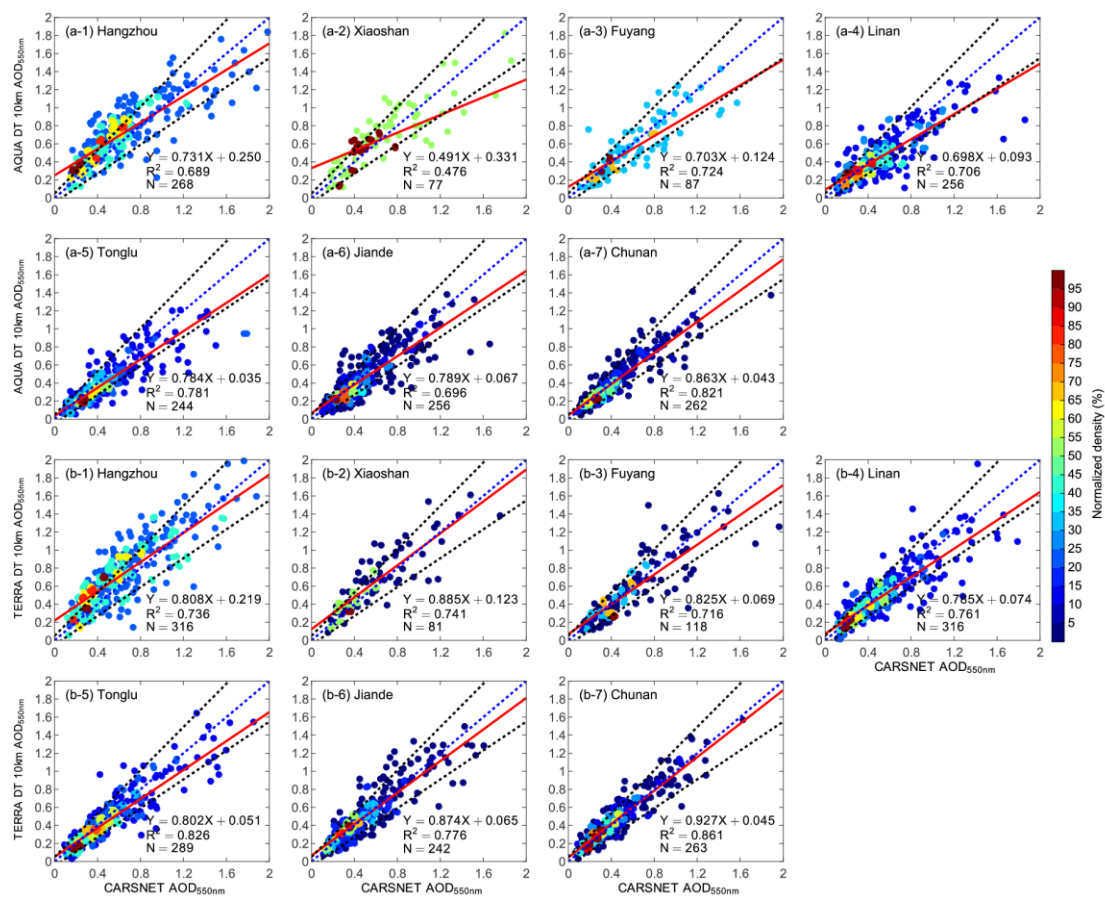
459



460

461 Fig. 477. Comparison of C6-MODIS/AquaS Deep Blue (DB) AOD at 550 nm with the  
 462 CARSNET AOD by the Deep Blue methods at 10km in (a-1) Hangzhou, (a-2b) Xiaoshan, (a-3e)  
 463 Fuyang, (a-4d) LinAn, (a-5e) Tonglu, (a-6f) Jiande—and, (a-7g) ChunAn- and  
 464 Terra-MODIS/Terra AOD DB at 550 nm with the CARSNET AOD by the Deep Blue methods at  
 465 10km in (b-1) Hangzhou, (b-2) Xiaoshan, (b-3) Fuyang, (b-4) LinAn, (b-5) Tonglu, (b-6) Jiande,  
 466 (b-7) ChunAn. The red solid line represents the linear regression. The two black dotted lines  
 467 represent the expected errors in the MODIS retrievals.

468 8Terra \_\_\_\_\_ at \_\_\_\_\_ 10km



469

470 Fig.8. Comparison of Aqua-MODIS/Aqua AOD DT at 550 nm with the CARSNET AOD by  
 471 the Dark Target methods at 10km in (a-1) Hangzhou, (a-2) Xiaoshan, (a-3) Fuyang, (a-4)  
 472 LinAn, (a-5) Tonglu, (a-6) Jiande, (a-7) ChunAn and Terra-MODIS/Terra DT AOD at 550 nm  
 473 with the CARSNET AOD by the Dark Target methods at 10km in (b-1) Hangzhou, (b-2)  
 474 Xiaoshan, (b-3) Fuyang, (b-4) LinAn, (b-5) Tonglu, (b-6) Jiande, (b-7) ChunAn. The red solid  
 475 line represents the linear regression. The two black dotted lines represent the expected errors  
 476 in the MODIS retrievals.

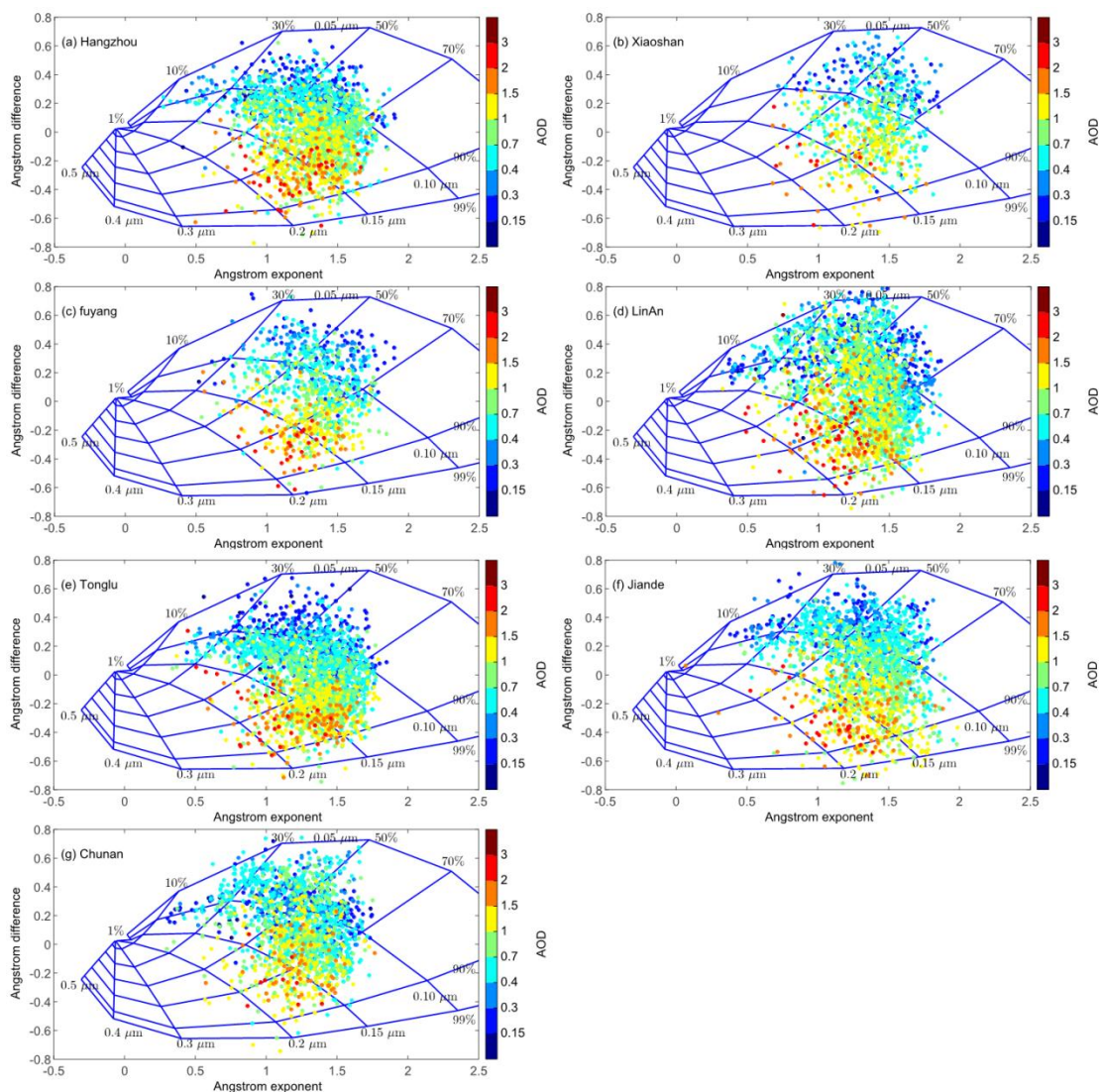
477 9Aqua- at 10km 10Terra- at 10km

478 The relationship between the EAE and the spectral difference in the EAE  
 479 ( $\delta EAE = EAE_{440-675nm} - EAE_{675-870nm}$ ) was analyzed to investigate the contribution of fine  
 480 particles ( $R_f$ ) and their fraction ( $\eta$ ) to the total extinction (EAOD) at 440 nm (Gobbi et al., 2007).  
 481 In this framework, values of  $AOD > 0.15$  are represented by different colors to avoid errors in  
 482 the  $\delta EAE$ . The lines indicate contribution of the fixed radius ( $R_f$ ) and fraction ( $\eta$ ) of the

483 fine-mode particles to the total extinction. Gobbi et al. (2007) used the difference in the EAE  
484 and AOD data to determine the growth of fine-mode particles or contamination by  
485 coarse-mode particles at eight AERONET stations: Beijing (China), Rome (Italy), Kanpur  
486 (India), Ispra (Italy), Mexico City (Mexico), NASA Goddard Space Flight Center (GSFC, USA),  
487 Mongu (Zambia) and Alta Floresta (Brazil).

488 | Fig. 5 shows 914 shows that the high EAOD values ( $>1.00$ ) cluster in the plots for all  
489 seven urban, suburban and rural sites, which is attributed to fine-mode particles with  $\delta EAE < 0$   
490 and  $\eta \sim 50\text{--}90\%$ . This variation in the fine-mode particles is similar to the results from Beijing  
491 and Kanpur ( $\eta \sim 70\text{--}90\%$ ). However, there were very few coarse-mode particles ( $\delta EAE \sim 0$ ,  
492  $\eta \sim 0\text{--}10\%$ ) in this study, suggesting that the dominance of dust is not significant in eastern  
493 China. These results showed a different pattern from that of other regions in north/northeast  
494 China (Wang et al., 2010; Zhu et al., 2014). For  $\delta EAE \sim 0$  and  $10\% < \eta < 30\%$ , high extinction was  
495 associated with a mixture dominated by fine-mode particles and less persistent coarse-mode  
496 particles. Clustering concentrated in the region  $\alpha \sim 1.5$ ,  $\delta \alpha \sim -0.5$  with high AOD values at all  
497 sites, which may be linked to an increase in size of the fine-mode particles by coagulation as  
498 the aged and hygroscopic events, as seen at other locations (e.g. Ispra, Italy; Mexico City,  
499 Mexico; GSFC, USA).

500



501

502 Fig. 5.449. Angström exponent difference as a function of  $\alpha_{440-870 \text{ nm}}$  and the  $\text{AOD}_{440 \text{ nm}}$  over (a)  
 503 Hangzhou, (b) Xiaoshan, (c) Fuyang, (d), LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn.

504 **3.2-3 Aerosol optical properties of Single-scattering albedo and aerosol complex**  
 505 **refractive index**

506 The distribution of the ~~total, fine and coarse mode~~ SSAs at the wavelengths of 440nm,  
 507 670nm, 870nm and 1020nm over the seven sites in the YRD are shown in Fig. 6.4210-. The  
 508 ~~total~~ SSA varied from 0.91 to 0.94, which is similar to the range seen in other regions of China,  
 509 such as Wuhan (0.92), Beijing (0.89) and Xinglong (0.92) (Wang et al., 2015; Xin et al., 2014;  
 510 Zhu et al., 2014). This indicated that scattering aerosol particles in eastern China resulting  
 511 from high levels of industrial and anthropogenic activity were dominant. The characteristics of

512 the SSA at these seven sites gradually increased from the east coast ( $0.91\pm 0.06$  at Hangzhou)  
513 inland toward the west ( $0.94\pm 0.03$  at ChunAn). The seven observation sites may always  
514 controlled by the same weather system that indicates a weak effect of meteorological  
515 elements in each site to the change of aerosol optical characteristics. These results indicate  
516 the emissions caused by human activity affect the absorption of aerosols in urban areas. The  
517 SSA was higher at LinAn and ChunAn than at the other sites, which may reflect the presence  
518 of a larger number of scattering aerosols (e.g. particles from urban/industrial activities) over  
519 the clean rural sites than over urban or suburban sites.

520 ~~The range of variation in the SSA of fine particles ( $SSA_f$ ) was 0.93–0.95, whereas the SSA~~  
521 ~~for coarse-mode particles ( $SSA_c$ ) was 0.81–0.84 at the seven sites (Fig. 6). The~~  
522 ~~absorption/scattering properties of fine- and coarse-mode particles determine the total SSA in~~  
523 ~~the YRD. The SSA was higher at LinAn and ChunAn than at the other sites, which may reflect~~  
524 ~~the presence of a larger number of scattering aerosols (e.g. particles from urban/industrial~~  
525 ~~activities) over the regional background/rural sites than over urban or suburban sites. The SSA~~  
526 ~~over urban and suburban sites showed the largest monthly variation. The monthly average~~  
527 ~~values of  $SSA_{T_i}$  were high in February ( $\sim 0.94\pm 0.05$ ) and June ( $\sim 0.92\pm 0.06$ ), but low in March~~  
528 ~~( $\sim 0.90\pm 0.06$ ) and August ( $\sim 0.89\pm 0.09$ ) in Hangzhou. However, the monthly SSA values at the~~  
529 ~~rural site of ChunAn only varied from 0.92 to 0.95. We concluded that the type of aerosol at~~  
530 ~~urban/suburban sites was more complex than at rural sites. The increased level of scattering~~  
531 ~~aerosols with higher SSA in June may be influenced by the hygroscopic growth in favor of the~~  
532 ~~interaction between aerosol aerosols from different emissions sources (Xia et al., 2007). The~~  
533 ~~existence of light-absorbing dust aerosols may contribute to the weaker lower SSA in spring~~  
534 ~~while the aerosols from biomass burning were probably due to the strong decreased in SSA~~  
535 ~~values in August (Yang et al., 2009).~~

536 ~~The lower SSA of coarse-mode particles in spring has been found in March/April~~  
537 ~~( $\sim 0.79\pm 0.08$ / $\sim 0.81\pm 0.07$ ) which may reflect the existence of light-absorbing dust aerosols in~~  
538 ~~the dominance, and the lower fine-mode SSA values in August ( $\sim 0.90\pm 0.08$ ) were probably a~~  
539 ~~result of aerosols from biomass burning in Hangzhou which has a larger contribution to the total~~

540 ~~SSA (Yang et al., 2009). The wavelength dependence of SSA present specific~~  
541 ~~absorption/scattering properties of different type aerosol seasons (Sokolik and Toon, 1999;~~  
542 ~~Eck et al., 2010). The SSA of dust in spring shown a weak dependence on the spectrum from~~  
543 ~~440nm to 1020nm in general (Cheng et al., 2006; Dubovik et al., 2002). Especially in the~~  
544 ~~March, the SSA at 440nm in Hangzhou, LinAn, Jiande and ChunAn was obviously lower at~~  
545 ~~short wavelength than that in the longer wavelength. This results This result has shown~~  
546 ~~a strong absorption of dust in the short wavelength in the YRD region over eastern China. It's~~  
547 ~~worth noting that there is an obvious and strongly decreasing of SSA in the longer wavelength~~  
548 ~~wavelength dependence of SSA by the type of aerosol from biomass burning or industrial~~  
549 ~~emissions in August (Alam et al., 2011; Janjai et al., 2012). The wavelength dependence of~~  
550 ~~SSA in YRD could be used to simply describe included to examine the aerosol types absorbing~~  
551 ~~aerosol type, as different absorbing particles (including dust or the and biomass burning smoke)~~  
552 ~~appear different spectral contrast of SSA.~~

553 -

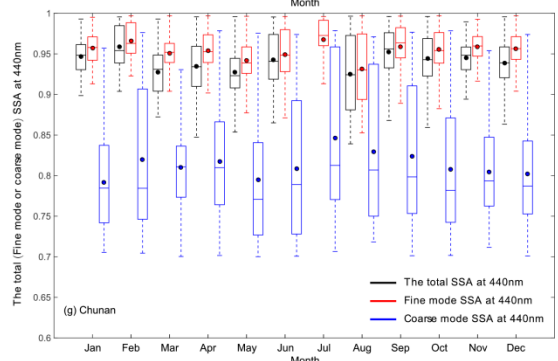
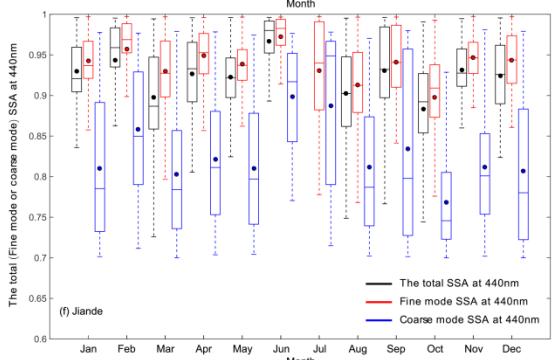
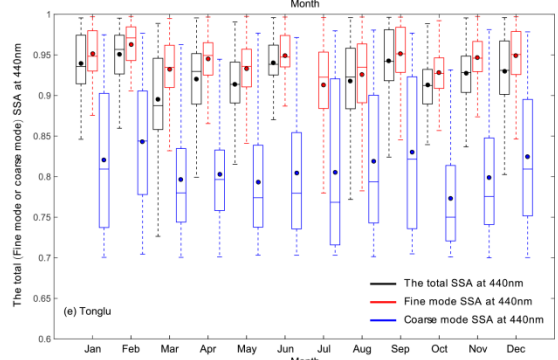
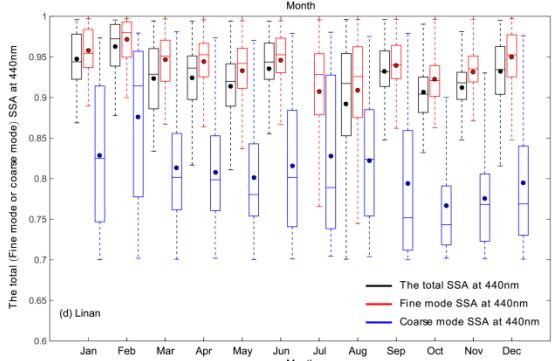
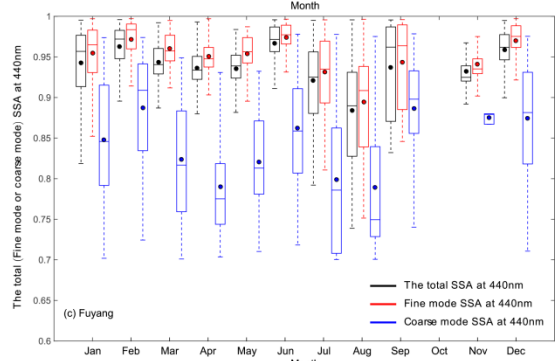
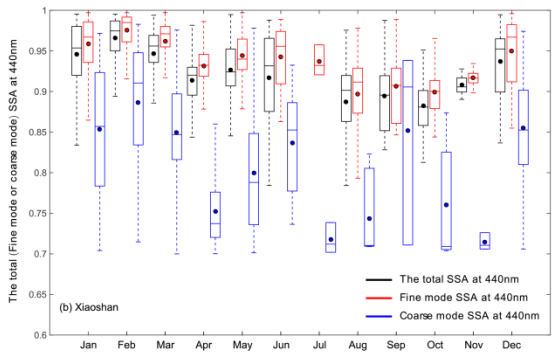
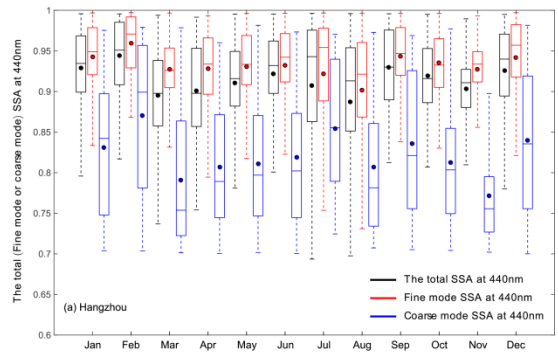
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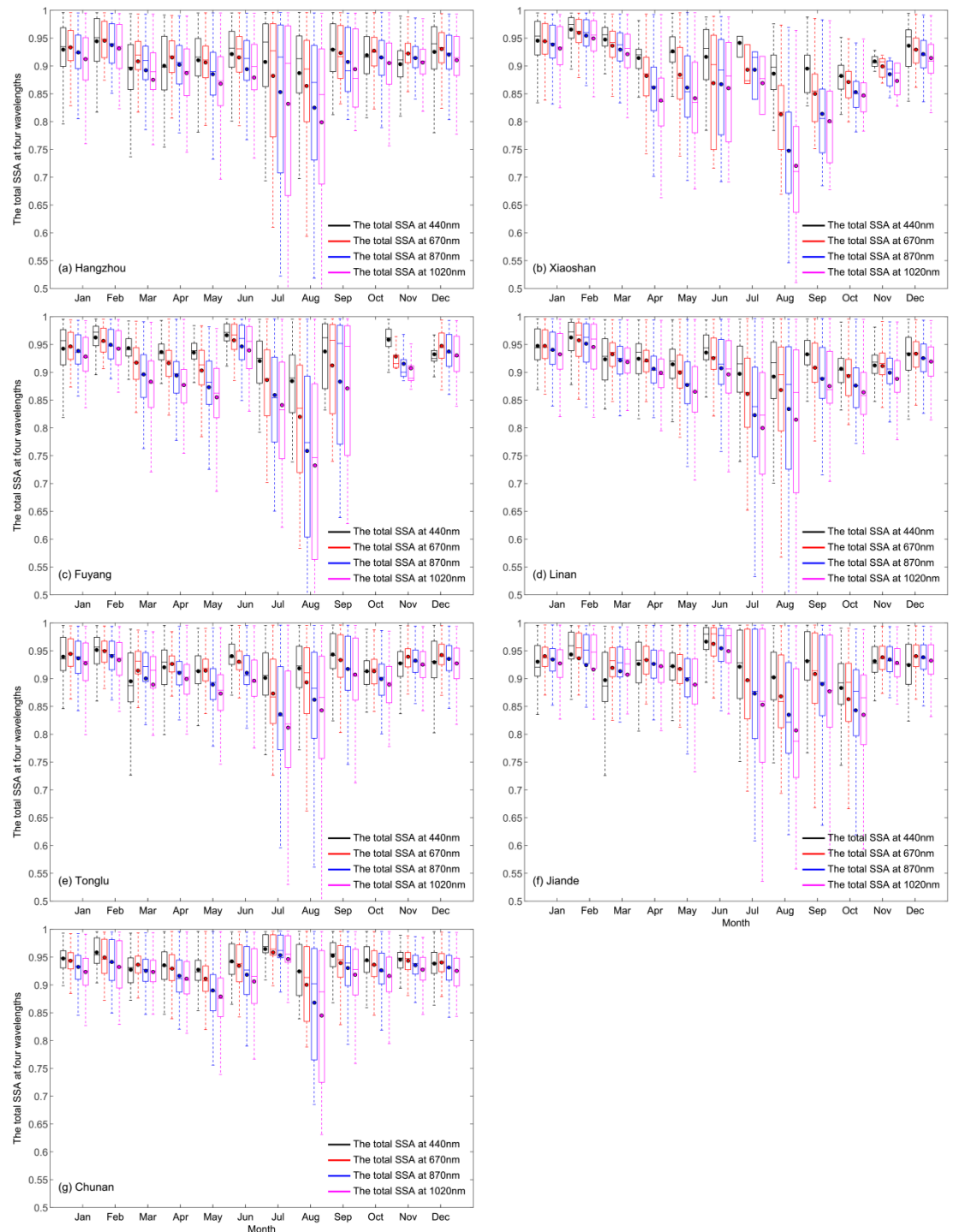
555 ~~However, the monthly SSA values at the rural site of ChunAn only varied from 0.92 to 0.95. We~~  
556 ~~concluded that the type of aerosol at urban/suburban sites was more complex than at rural~~  
557 ~~sites. Fig. 6 shows a significant decrease in the fine-mode SSA in July/August and in the~~  
558 ~~coarse-mode SSA in March/April. At Hangzhou, the lower fine-mode SSA values in~~  
559 ~~July/August ( $-0.92 \pm 0.08 / -0.90 \pm 0.08$ ) were probably a result of aerosols from biomass burning~~  
560 ~~and the lower coarse-mode SSA values in March/April ( $-0.79 \pm 0.08 / -0.81 \pm 0.07$ ) may reflect the~~  
561 ~~existence of light-absorbing dust aerosols (Yang et al., 2009). The SSA depends on the~~  
562 ~~wavelength and dust particles absorb strongly at short wavelengths, resulting in a lower SSA at~~  
563 ~~440nm (Eck et al., 2010).~~

564 ~~The range of variation in the SSA of fine particles ( $SSA_f$ ) was 0.93–0.95, whereas the SSA~~  
565 ~~for coarse-mode particles ( $SSA_c$ ) was 0.81–0.84 at the seven sites (Fig. 6). The fine- and~~  
566 ~~coarse-mode particles displayed significant scattering and absorption abilities in the urban,~~

567 suburban and rural areas of the YRD region. Fig. 6 shows a significant decrease in the  
568 fine mode SSA in July/August and in the coarse mode SSA in March/April. At Hangzhou, the  
569 lower fine mode SSA values in July/August ( $0.92 \pm 0.08 / 0.90 \pm 0.08$ ) were probably a result of  
570 aerosols from biomass burning and the lower coarse mode SSA values in March/April  
571 ( $0.79 \pm 0.08 / 0.81 \pm 0.07$ ) may reflect the existence of light absorbing dust aerosols (Yang et al.,  
572 2009). The SSA depends on the wavelength and dust particles absorb strongly at short  
573 wavelengths, resulting in a lower SSA at 440nm (Eck et al., 2010). The absorption/scattering  
574 properties of fine and coarse mode particles determine the total SSA in the YRD. These  
575 differences in the SSA were mostly dependent on the type of aerosol and the ratio of absorbing  
576 and non-absorbing components in the aerosols.







578

579 Fig.61210. Variation in the total, fine and coarse mode SSA at 440nm, 670nm 870nm and  
 580 1020nm<sub>440-nm</sub> over (a) Hangzhou, (b) Xiaoshan, (c) Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande  
 581 and (g) ChunAn. The boxes represent the 25th to 75th percentile distribution, while the dots  
 582 and solid lines within each box represent the mean and median, respectively.

583 The real and imaginary parts of the refractive index represent the scattering and

584 absorption capacity of particles, respectively. The refractive index is determined by the  
585 hygroscopic conditions and the chemical composition of the aerosols (Dubovik and King, 2000).  
586 There was no significant difference between the real parts of the refractive index among the  
587 seven urban, suburban and rural sites in this study (range 1.41–1.43). The real parts of the  
588 refractive index in this study were smaller than the real parts of ammonium sulfate and  
589 ammonium nitrate (1.55), which may be due to the hygroscopic conditions or the mixture of  
590 dust particles. The real part of the refractive index was highest in March ( $\sim 1.46 \pm 0.06$ ) and  
591 November ( $\sim 1.45 \pm 0.06$ ) and lowest in July ( $\sim 1.42 \pm 0.06$ ) and August ( $\sim 1.41 \pm 0.07$ ) at the urban  
592 sites. ~~A higher level of dust aerosols with weak scattering in spring and autumn could contribute~~  
593 ~~to a higher value of the real part of the refractive index; this was reduced or eliminated by~~  
594 ~~rainfall during the summer months.~~

595  
596 The imaginary part of the refractive index was higher at the urban site of Hangzhou ( $\sim 0.0112 \pm$   
597  $0.0104$ ) as a result of the high loading of absorption aerosols in this region and was consistent  
598 with the lower SSA. High imaginary parts of the refractive index occurred in August at all urban,  
599 suburban and rural sites in the YRD, which may be due to the higher emission of absorptive  
600 particles by the post-harvest burning of crop residues— with more spectral dependence. The  
601 burning of crop residues may cause a large deterioration in the regional air quality in the YRD  
602 region. A higher level of spring dust aerosols with absorption could contribute to a higher value  
603 of the imaginary part of the refractive index.

604

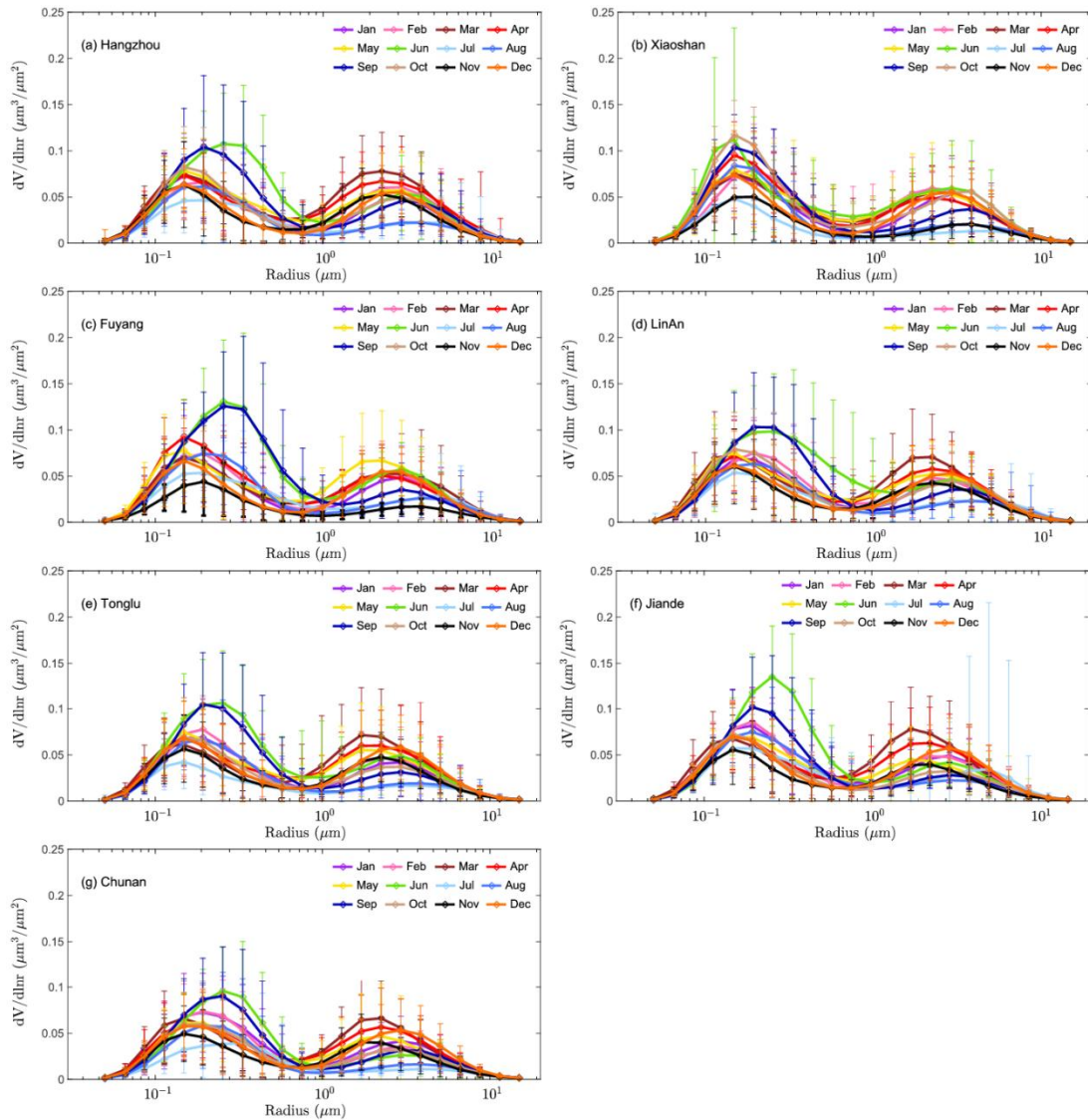
605

### 606 ~~3.3 Radius and aerosol volume size distributions~~

607 ~~Fig.7 shows 13 shows the monthly aerosol size distribution ( $dV/dlnr$ ) in the YRD for all~~  
608 ~~sites. The volumes of fine-mode aerosols were obviously higher than those of coarse-mode~~  
609 ~~aerosols over all sites. The fine-mode radii were  $\sim 0.2$ – $0.3 \mu\text{m}$  in the YRD with a volume of~~

610 ~~0.10–0.12  $\mu\text{m}^3$  and the coarse-mode radii were  $\sim 2.0 \mu\text{m}$  with a volume close to  $0.07 \mu\text{m}^3$ . The~~  
611 ~~amount of fine-mode aerosols was higher in June and September than in other months at~~  
612 ~~almost sites, except for Xiaoshan. This could be caused by aerosol humidification (Eck et al.,~~  
613 ~~2012; Li et al., 2010, 2014; Huang et al., 2016). This phenomenon is also found over Beijing~~  
614 ~~and Shenyang in north/northeast China, suggesting that hygroscopic growth occurs over many~~  
615 ~~regions of China (Li et al., 2011; Che et al., 2015c).~~

616 ~~The coarse-mode radius in spring at all sites was smaller than in other cities in north and~~  
617 ~~northeast China affected by frequent dust transport events in spring (Kong et al., 2011; Zhao et~~  
618 ~~al., 2015). The coarse-mode particles showed a larger effective radius at all seven urban,~~  
619 ~~suburban and rural sites in the summer, which may due to the adhesion of new particles onto~~  
620 ~~larger particles (such as fly ash).~~



621

622 ~~Fig.137. Variation in the annual volume size distribution over (a) Hangzhou, (b) Xiaoshan, (c)~~

623 ~~Fuyang, (d) LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn.~~

624 **3.4 Aerosol optical properties of Absorption aerosol optical depth and**  
 625 **absorption Angström exponent**

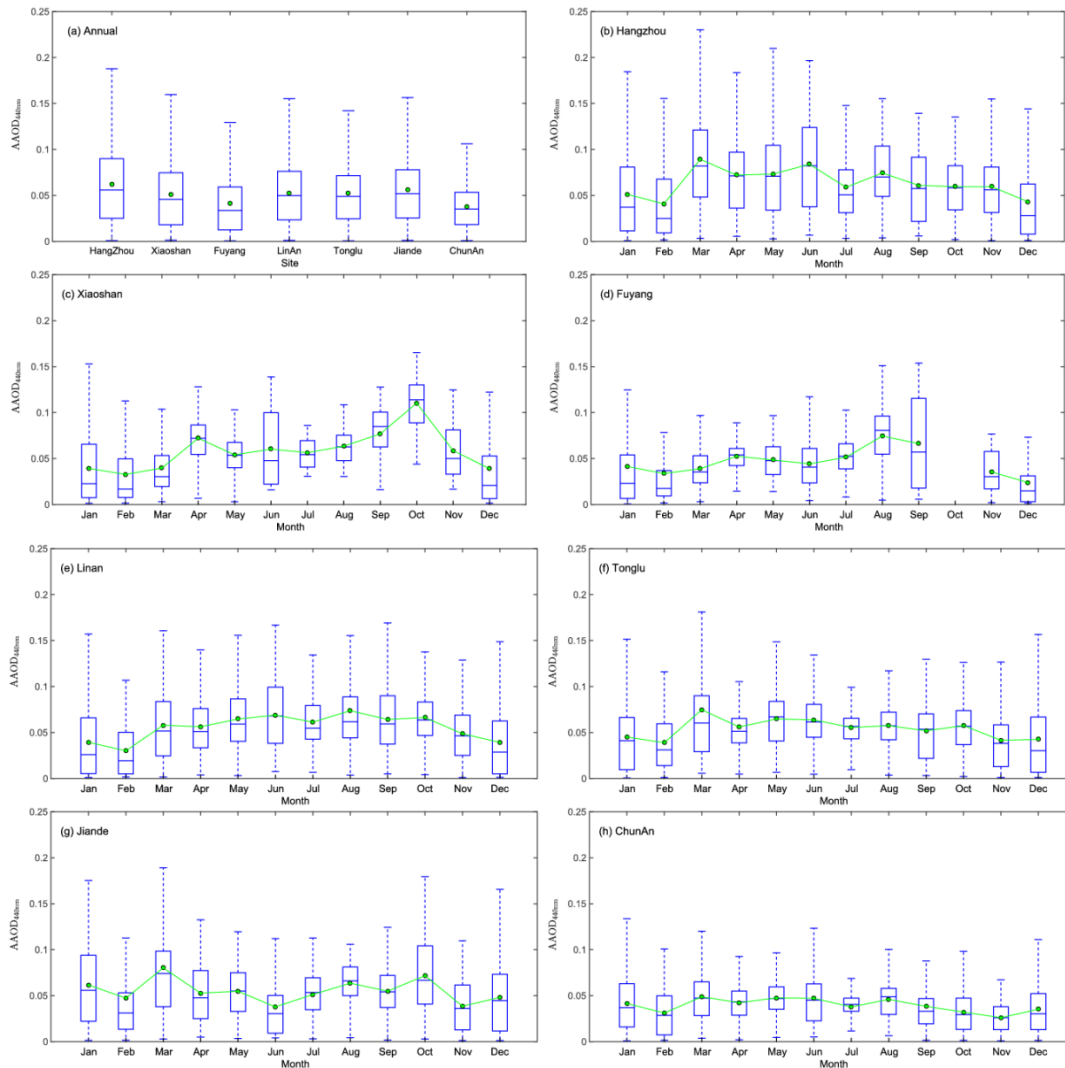
626 The annual AAODs at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and ChunAn  
 627 were about  $0.06 \pm 0.05$ ,  $0.05 \pm 0.04$ ,  $0.04 \pm 0.04$ ,  $0.05 \pm 0.04$ ,  $0.05 \pm 0.04$ ,  $0.06 \pm 0.04$  and  $0.04 \pm 0.03$ ,  
 628 respectively (Fig. 81411). ~~The higher annual values of the AAOD in Hangzhou and~~  
 629 ~~Jiande indicate that there are more absorbing aerosol particles at these sites.~~ The similar AAOD  
 630 level at the seven sites (0.04-0.06) suggests that absorbing aerosols are distributed

631 homogeneously in the YRD region. The AAOD values may have very large uncertainties and  
632 uncertainty because of the dataset is including all the values in one month-. Nevertheless,  
633 there is also some varies in AAOD according to the changes of the SSA in section 3.3. These  
634 differences in the AAOD were mostly dependent on the type of aerosol and the ratio of  
635 absorbing and non-absorbing components in the aerosols.

636 These differences in the SSA were mostly dependent on the type of aerosol and the ratio-  
637 of absorbing and non-absorbing components in the aerosols.

638 The monthly AAOD at the urban site of Hangzhou was  $0.09 \pm 0.06$  in March as a result of  
639 the presence of absorbing dust particles. The AAOD of about  $0.07 \pm 0.04$  in August is related to  
640 the burning of crop residues. The AAODs in the winter season at all the sites in the YRD region  
641 were  $< 0.05$ , which suggests that absorbing aerosol emissions did not frequently occur at these  
642 sites, unlike in the northern regions of China. As fig.152 shown, tThe AAE was  $< 1.00$  in June  
643 and August at all urban, suburban and rural sites of the YRD, which suggested the presence  
644 aerosols coated with absorbing or non-absorbing material in summer season. This process is  
645 favored by high temperatures and high humidity under conditions of strong solar radiation  
646 (Shen et al., 2015, Zhang et al., 2015). The particles coagulate and grow rapidly in the  
647 presence of sufficient water vapor (Li et al., 2016). The AAE became increasingly close to, or  
648 larger than, 1.00 at all seven sites from September, which is consistent with decreasing  
649 amounts of precipitation. This increase in the AAE was related to the emission of black carbon  
650 from biomass burning (Soni et al., 2010; Russell et al., 2010). According to the corresponding  
651 annual mean values for the AAE at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and  
652 ChunAn ( $1.13 \pm 0.46$ ,  $0.88 \pm 0.42$ ,  $0.85 \pm 0.43$ ,  $0.98 \pm 0.35$ ,  $1.11 \pm 0.49$ ,  $1.16 \pm 0.44$  and  $0.93 \pm 0.31$ ) in  
653 Fig. 12, the seven sites has been attributed to three categories with AAE levels. The mean  
654 values of the AAE at Xiaoshan and Fuyang were  $< 1.00$ , suggesting the presence of absorbing  
655 or non-absorbing materials coating black carbon at these suburban and rural sites (Bergstrom  
656 et al., 2007; Lack and Cappa et al., 2010; Gyawali et al., 2009). The AAE values were close to  
657 1.00 at LinAn and ChunAn, indicating that the absorptive aerosols were dominated by particles  
658 of black carbon (Zhang et al., 2012; Li et al., 2016). By contrast, the AAE values at Hangzhou,

659 Tonglu and Jiande were >1.00, indicating the presence of absorptive aerosols from the burning  
660 of biomass. This difference in the AAE distribution indicates the absorbing aerosols have  
661 different characteristics resulting from the different emission sources at urban, suburban and  
662 rural sites in the YRD. The AAE was <1.00 in June – August at all urban, suburban and rural sites  
663 of the YRD, which suggested the presence of aerosols coated with absorbing or non-absorbing  
664 material in summer season. This process is favored by high temperatures and high humidity  
665 under conditions of strong solar radiation (Shen et al., 2015; Zhang et al., 2015). The particles  
666 coagulate and grow rapidly in the presence of sufficient water vapor (Li et al., 2016). The  
667 AAE became increasingly close to, or larger than, 1.00 at all seven sites from September,  
668 which is consistent with decreasing amounts of precipitation. This increase in the AAE was  
669 related to the emission of black carbon from biomass burning (Soni et al., 2010; Russell et al.,  
670 2010).



671

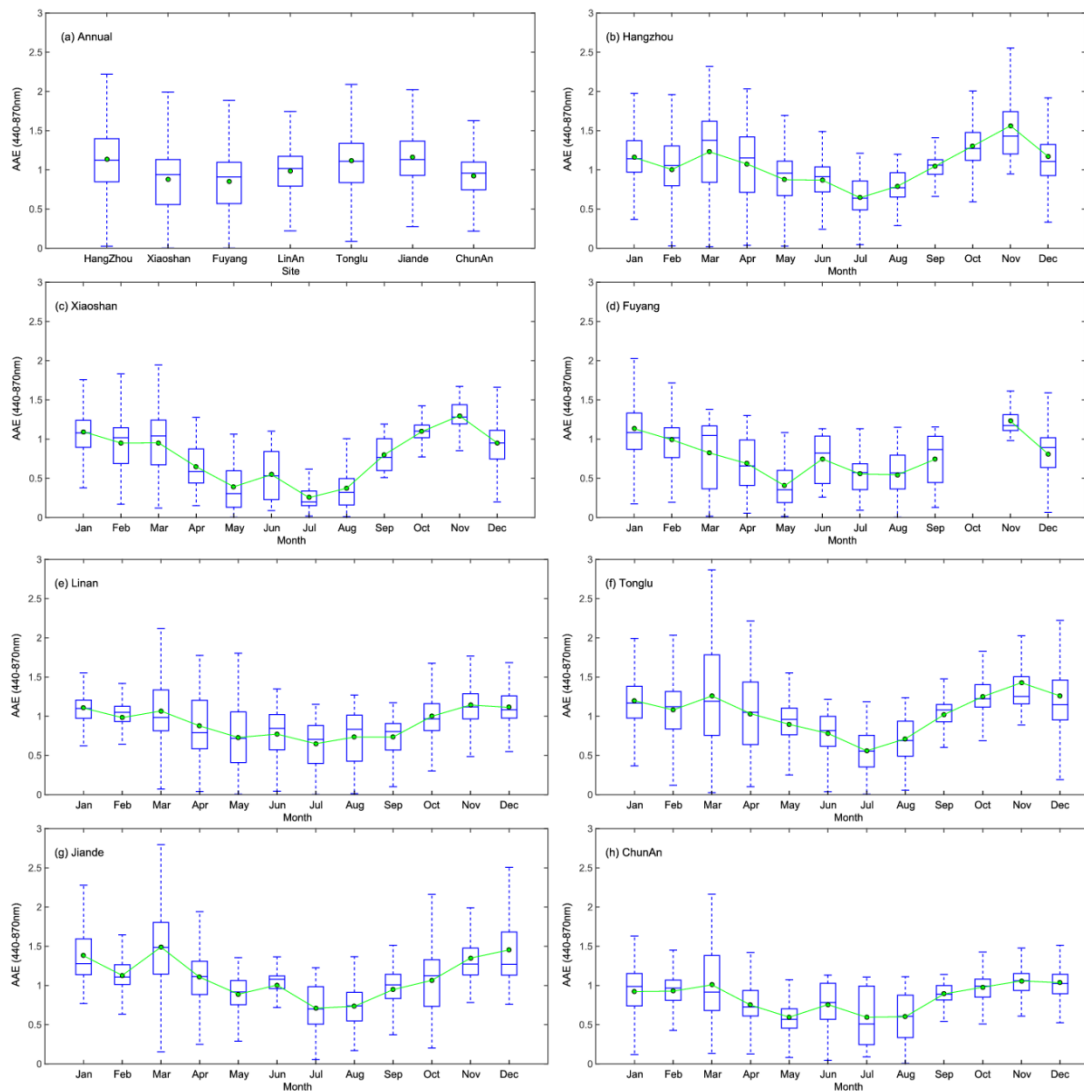
672 Fig.8.141. (a) Annual variation in the absorption aerosol optical depth at 440 nm ( $AAOD_{440\text{ nm}}$ )  
 673 over (b) Hangzhou, (c) Xiaoshan, (d) Fuyang, (e) LinAn, (f) Tonglu, (g) Jiande and (h) ChunAn.  
 674 The boxes represent the 25th to 75th percentile distribution, while the dots and solid lines  
 675 within each box represent the mean and median, respectively.

676 ~~The annual mean values for the AAE at Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu,~~  
 677 ~~Jiande and ChunAn were about  $1.13 \pm 0.46$ ,  $0.88 \pm 0.42$ ,  $0.85 \pm 0.43$ ,  $0.98 \pm 0.35$ ,  $1.11 \pm 0.49$ ,~~  
 678  ~~$1.16 \pm 0.44$  and  $0.93 \pm 0.31$ , respectively(Fig. 9). The meanvalues of the AAE at Xiaoshan and~~  
 679 ~~Fuyangwere  $< 1.00$ , suggesting the presence of absorbing or non-absorbing materialscoating~~  
 680 ~~black carbonat these suburban and rural sites(Bergstrom et al., 2007; Lack and Cappa et al.,~~  
 681 ~~2010; Gyawali et al., 2009).The AAE valueswere close to 1.00 at LinAn and ChunAn,~~



682 ~~indicating that the absorptive aerosols were dominated by particles of black carbon (Zhang et al.,~~  
683 ~~2012; Li et al., 2016). By contrast, the AAE values at Hangzhou, Tonglu and Jiande were >1.00,~~  
684 ~~indicating the presence of absorptive aerosols from the burning of biomass. This difference in~~  
685 ~~the AAE distribution indicates the absorbing aerosols have different characteristics resulting~~  
686 ~~from the different emission sources at urban, suburban and rural sites in the YRD. The~~  
687 ~~AAE was <1.00 in June – August at all urban, suburban and rural sites of the YRD, which~~  
688 ~~suggested the presence of aerosols coated with absorbing or non-absorbing material in summer~~  
689 ~~season. This process is favored by high temperatures and high humidity under conditions of~~  
690 ~~strong solar radiation (Shou et al., 2015; Zhang et al., 2015). The particles coagulate and grow~~  
691 ~~rapidly in the presence of sufficient water vapor (Li et al., 2016). The~~  
692 ~~AAE became increasingly close to, or larger than, 1.00 at all seven sites from September, which~~  
693 ~~is consistent with decreasing amounts of precipitation. This increase in the AAE was related~~  
694 ~~to the emission of black carbon from biomass burning (Soni et al., 2010; Russell et al.,~~  
695 ~~2010). The AAE can be used to indicate the major types (urban/industrial, biomass burning,~~  
696 ~~dust/mixed dust) or optical mixtures of absorbing aerosol particles (Schnaiter et al., 2006;~~  
697 ~~Russell et al., 2010; Giles et al., 2011; 2012; Mishra and Shibata, 2012). Giles et al., (2011)~~  
698 ~~examined AAE/EAE data from Kanpur to classify the categories of absorbing aerosols. The~~  
699 ~~“mostly dust” category has been defined as having an EAE value  $\leq 0.50$  and sphericity fraction~~  
700  ~~$< 0.20$  with an AAE value  $> 2.00$ . The “mostly black carbon” category has been defined as~~  
701 ~~having an EAE value  $> 0.80$  and a sphericity fraction  $\geq 0.20$  with  $1.00 < \text{AAE} \leq 2.00$ . Values of~~  
702 ~~EAE  $> 0.80$  and AAE  $> 2.00$  indicate a concentration of organic carbon (Arola et al., 2011). The~~  
703 ~~“mixed black carbon and dust” category was centered at EAE  $\sim 0.50$  with AAE  $\sim 1.50$  and used~~  
704 ~~to represent an optical mixture with black carbon and mineral dust particles as the dominant~~  
705 ~~absorbers.~~

706



707

708 Fig. 9152. (a) Annual variation in the absorption Angström exponent at 440 nm ( $AAE_{440\text{nm}}$ ) over  
 709 (b) Hangzhou, (c) Xiaoshan, (d) Fuyang, (e) LinAn, (f) Tonglu, (g) Jiande and (h) ChunAn. The  
 710 boxes represent the 25th to 75th percentile distribution, while the dots and solid lines within  
 711 each box represent the mean and median, respectively.

712 ~~The AAE can be used to indicate the major types (urban/industrial, biomass burning,~~  
 713 ~~dust/mixed dust) or optical mixtures of absorbing aerosol particles (Schnaiter et al., 2006;~~  
 714 ~~Russell et al., 2010; Giles et al., 2011, 2012; Mishra and Shibata, 2012). Giles et al., (2011)~~  
 715 ~~examined AAE/EAE data from Kanpur to classify the categories of absorbing aerosols. The~~  
 716 ~~“mostly dust” category has been defined as having an EAE value  $\leq 0.50$  and sphericity fraction~~  
 717  ~~$\leq 0.20$  with an AAE value  $> 2.00$ . The “mostly black carbon” category has been defined as having~~

~~an EAE value > 0.80 and a sphericity fraction  $\geq 0.20$  with  $1.00 < AAE \leq 2.00$ . Values of EAE > 0.80 and AAE > 2.00 indicate a concentration of organic carbon (Arola et al., 2011). The “mixed black carbon and dust” category was centered at EAE ~ 0.50 with AAE ~ 1.50 and used to represent an optical mixture with black carbon and mineral dust particles as the dominant absorbers.~~

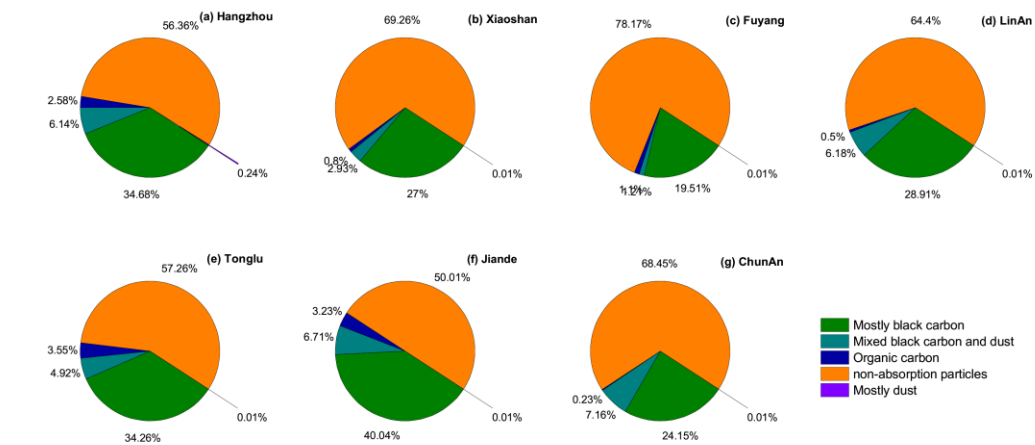
We used the instantaneous AAE and EAE values to classify the dominant absorbing aerosol types in urban, suburban and rural areas of the YRD (Fig. 40163; Table 2). Fig. 13 Table 2 shows that the “mostly dust” category was very low at both suburban and rural sites (<0.01%) and just ~0.24% at the urban site of Hangzhou. This indicates ~~that dust does not dominate the absorbing aerosol particles in the YRD region of eastern China, which is completely different from other regions of north/northeast region in China where the dust particles could contribute to the aerosol loading substantially.~~ The “mostly black carbon” category dominates the absorbing aerosols in the urban, suburban and rural areas in the YRD region. The percentage “mostly black carbon” varied from ~20 to 40% depending on each site, indicating the mixing of black carbon as well as brown and soot carbon species from biomass burning and urban/industrial activities. Because of the long-distance transportation and local fugitive dust effect, the “mixed black carbon and dust” category contributed ~5% of the absorbing aerosol particles in the YRD region. There was also ~1-4% of the “organic carbon” category identified as absorbing aerosol particles in this region.

The non-absorption particles account for ~50 to 80% in the YRD region. There is higher contribution of non-absorption particles about 78.17% in Fuyang and less non-absorption particles about 50.01% in Jiande. The result is consistent with the level of total SSA at 440nm of Fuyang (0.94) with more scattering particles and than Jiande (0.92).

Particles with EAE values of ~0.40 and ~1.25 could be regarded as “mixed large particles” greater than microns in size and submicron “mixed small particles”, respectively (Giles et al. 2012). The frequency of “mixed large particles” was <0.5% at the urban, suburban and rural sites (Table 2). By contrast, the frequency of “mixed small particles” was ~18-36%.

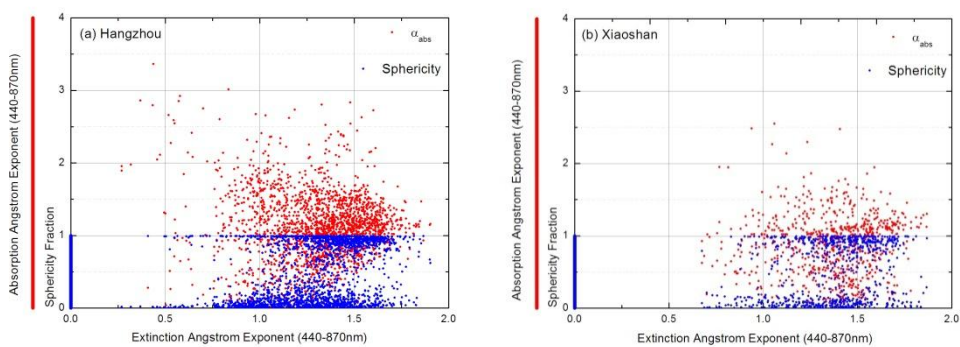
The EAE ( $\alpha_{ext}$ ) and AAE ( $\alpha_{abs}$ ) values at all the urban, suburban and rural sites were

745 distributed mainly around 1.25 and 1.00–1.50 (Fig.1016), respectively (Fig.14). In contrast with  
 746 the results of Giles et al. (2011), the sphericity fraction did not show an obvious transition from  
 747 non-spherical to spherical particles from the urban, suburban and rural sites in YRD. The  
 748 sphericity fraction showed a dispersed distribution of spherical particles, indicating a mixture of  
 749 fine-mode particles derived from anthropogenic sources and coarse-mode particles, such as  
 750 dust events transported from north/northwest China or local fugitive dust emissions.

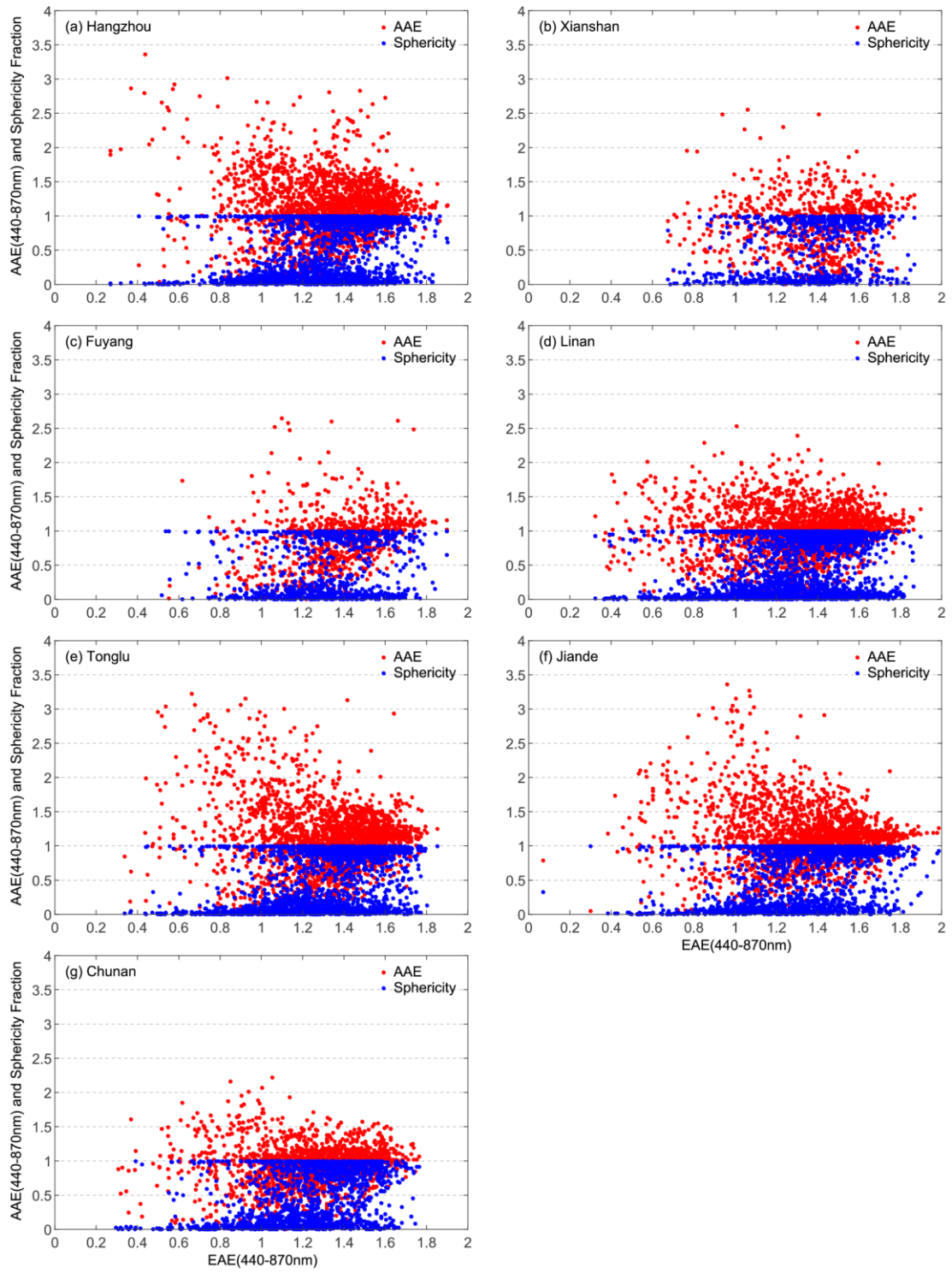


751

752 Fig.13.Types of aerosol over (a) Hangzhou, (b) Xiaoshan, (c) Fuyang, (d), LinAn, (e)  
 753 Tonglu, (f) Jiande and (g) ChunAn.



754



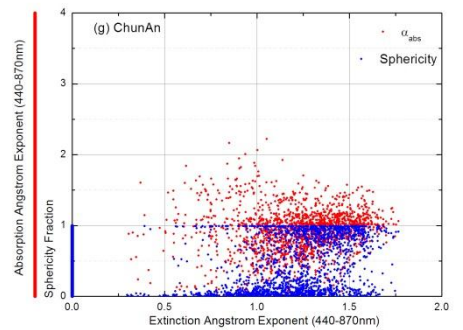
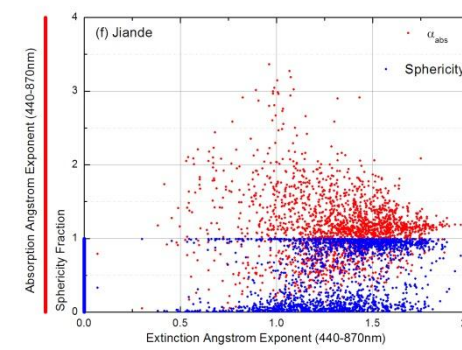
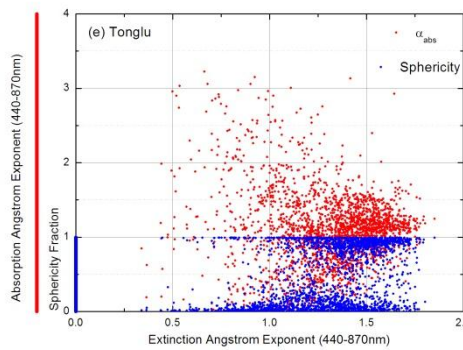
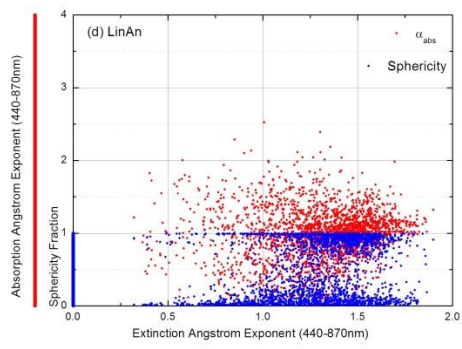
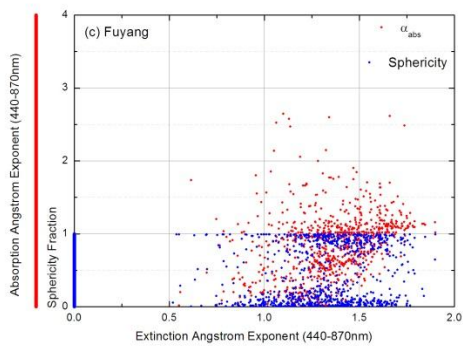


Fig. 40164. The AAE (red dot) and the sphericity fraction (blue dot) as a function of the EAE at 440–870 nm over (a) Hangzhou, (b) Xiaoshan, (c) Fuyang, (d), LinAn, (e) Tonglu, (f) Jiande and (g) ChunAn.

~~Table 2. Types of aerosol at the seven sites in the Yangtze River Delta.~~

Mostly	Mixed	Mostly	Organic	Mixed—large	Mixed—small
--------	-------	--------	---------	-------------	-------------

	dust(%)	black-carbon and dust(%)	black-carbon(%)	carbon(%)	particles(%)	particles(%)
Hangzhou	0.24	6.14	34.68	2.58	0.19	36.34
Xiaoshan	<0.01	2.93	27.00	0.80	<0.01	23.40
Fuyang	<0.01	1.21	19.51	1.10	<0.01	18.63
LinAn	<0.01	6.18	28.91	0.50	0.37	28.04
Tonglu	<0.01	4.92	34.26	3.55	0.18	33.33
Jiande	<0.01	6.71	40.04	3.23	0.26	35.28
ChunAn	<0.01	7.16	24.15	0.23	0.12	26.75

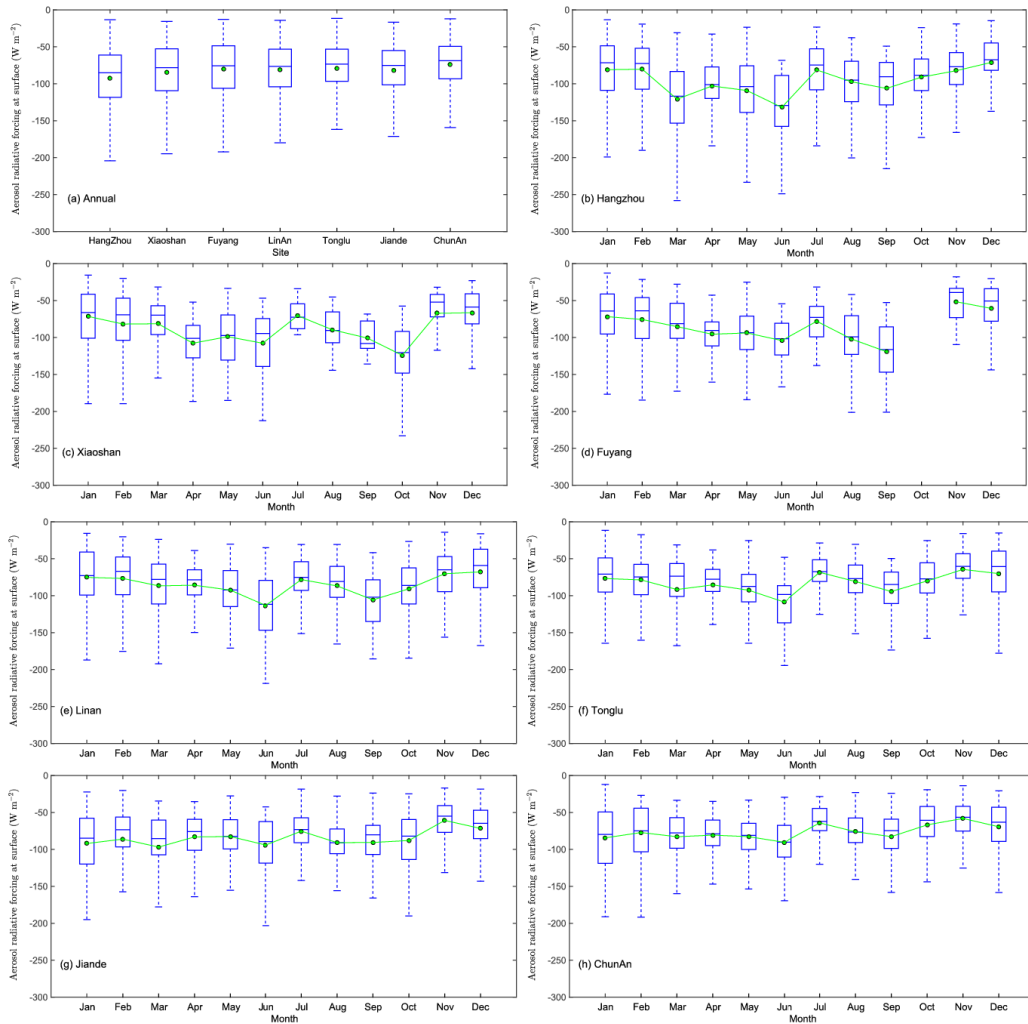
764

765 **3.5 Aerosol optical properties of Aerosol-aerosol radiative forcing at the Earth's surface**

766 **and top of the atmosphere**

767 [Figures 11](#) and [Figures 17, 15, and 18, 6, 12](#) show the variations in ARF at the surface  
768 (ARF-BOA) and at the top of the atmosphere (ARF-TOA) at the urban, suburban and rural  
769 sites in the YRD region.

770 The annual ARF-BOA values for Hangzhou, Xiaoshan, Fuyang, LinAn, Tonglu, Jiande and  
771 ChunAn were about  $-93\pm 44$ ,  $-84\pm 40$ ,  $-80\pm 40$ ,  $-81\pm 39$ ,  $-79\pm 39$ ,  $-82\pm 40$  and  $-74\pm 34$  W/m<sup>2</sup>,  
772 respectively. The higher ARF-BOA values in Hangzhou indicate that there was high aerosol  
773 loading at this site, which scattered and absorbed more radiation and caused a significant  
774 cooling effect at the surface. The monthly value of the ARF-BOA in Hangzhou was higher in  
775 June (about  $-132\pm 48$  W/m<sup>2</sup>) and September (about  $-106\pm 48$  W/m<sup>2</sup>), which is consistent with  
776 the timing of burning biomass from crop residues. Ding et al. (2016) found that black carbon  
777 emitted from biomass burning can modify the meteorology of the planetary boundary layer and  
778 substantially decrease the surface heat flux. Hygroscopic growth at the same time enhances  
779 the aerosol optical extinction (Yan et al., 2009; Zhang et al., 2015); this was also an important  
780 factor in the large ARF-BOA values in June and September at the urban, suburban and rural  
781 sites in the YRD.



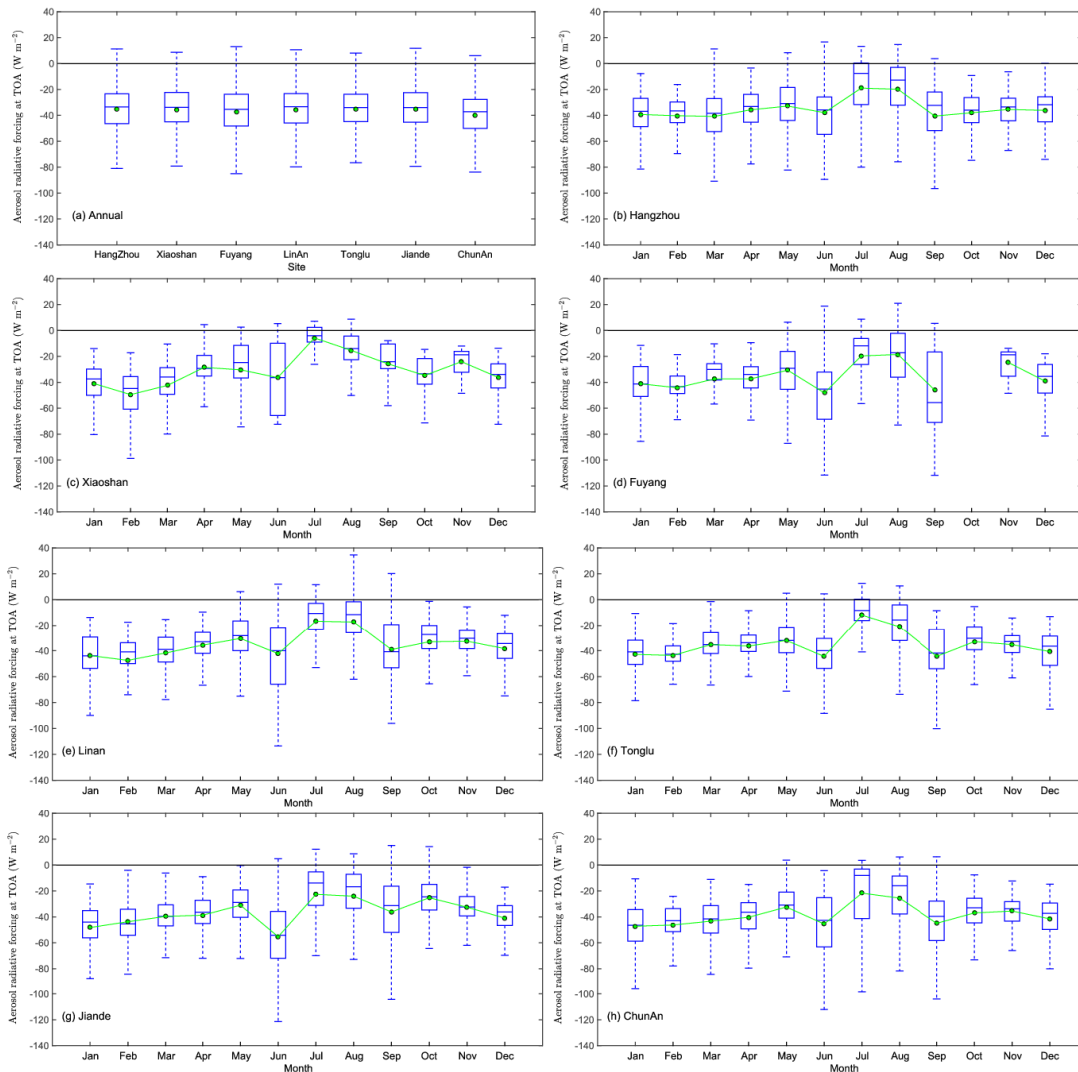
782

783 Fig. 17. (a) Annual variation of the ARF at the surface over (b) Hangzhou, (c) Xiaoshan, (d)  
 784 Fuyang, (e) LinAn, (f) Tonglu, (g) Jiande and (h) ChunAn. The boxes represent the 25th to 75th  
 785 percentile distribution, while the dots and solid lines within each box represent the mean and  
 786 median, respectively.

787 The ARF-TOA values were less than  $-40 \text{ W/m}^2$  at the urban, suburban and rural sites in  
 788 the YRD. The AFR-TOA values were negative all year, which suggests that the aerosols  
 789 caused a cooling effect at the TOA as well as at surface in the YRD. This is different from the  
 790 north/northeast regions of China, where the instantaneous AFR-TOA value can be positive in  
 791 the winter season as a result of the large surface reflectance area reflecting on short  
 792 wavelength radiation and heating caused by absorbing aerosols (Che et al., 2014). The  
 793 surface albedo in the YRD region is lower than in north/northeast China as a result of better



794 vegetation. At the same time, there is also a low level of absorbing aerosol emissions in winter.  
 795 This caused obvious negative AFR at the TOA at the urban, suburban and rural sites in the  
 796 YRD.



797  
 798 Fig. 18. (a) Annual variation in the aerosol radiative forcing at the top of the atmosphere  
 799 (TOA) in (b) Hangzhou, (c) Xiaoshan, (d) Fuyang, (e) LinAn, (f) Tonglu, (g) Jiande and (h)  
 800 ChunAn. The boxes represent the 25th to 75th percentile distribution, while the dots and solid  
 801 lines within each box represent the mean and median, respectively.

802 **4. Discussion and Summary**

803 In this paper, the aerosol optical properties, including the AOD, EAE, SSA, complex  
 804 refractive index, volume size distribution, and the absorption properties of the AAOD and AAE

805 were retrieved from ground-based measurements data over the YRD in eastern China for the  
806 period 2011–2015. The AOD in Hangzhou in urban eastern China was similar to that in  
807 Shenyang (0.75) in urban northeast China (Zhao et al., 2013), and in Beijing (0.76) and Tianjin  
808 (0.74) in urban north China (Che et al., 2015b), indicating that the aerosol extinction is both  
809 common and at a similar level throughout most urban areas of China. The AOD values at the  
810 urban and suburban sites of Hangzhou were a little bit higher slightly higher than at Pudong  
811 (0.70) and Hefei (0.69), other urban areas in eastern China, suggesting that higher aerosol  
812 extinction ability were observed here (He et al., 2012; Liu et al., 2017). However, the AOD at all  
813 seven sites was lower than that obtained at Wuhan (1.05), Nanjing (0.88), Dongtan (0.85),  
814 Taihu(0.77) and Xuzhou (0.92) in previous studies in eastern China (Wang et al., 2015; Li et al.,  
815 2015; Pan sphericity et al., 2010; Xia et al., 2007; Wu et al., 2016). This indicates that the  
816 aerosol loading caused by anthropogenic activities is very high in both urban and suburban  
817 areas in eastern China. The site at LinAn is regarded as the clean suburban site in eastern  
818 China with an average AOD about  $0.73\pm 0.44$ , which is higher than that at the other regional  
819 background stations of China, such as Longfengshan (0.35; northeastern China), Mt Waliguan  
820 (0.14, inland Asia), Xinglong (0.28, northern China), Akedala (0.20, northwestern China) and  
821 Shangri-La (0.11, southwestern China) (Wang et al., 2010; Che et al., 2011; Zhu et al., 2014;  
822 Che et al., 2015b). The aerosol loading in eastern China (especially in the YRD region) is at  
823 least twice as high as in other regions of China which indicate the strong aerosol extinction.  
824 Moreover, aerosol extinction loading was at a high level over both urban and suburban sites  
825 and even over the rural sites in the YRD which suggests large regional scale aerosol loading  
826 extinction over eastern China in recent years. In this paper, tThe aerosol optical properties,  
827 including the AOD, EAE, SSA, complex refractive index, volume size distribution, and the  
828 absorption properties of the AAOD and AAE were retrieved from ground-based  
829 measurementsatellite data over the YRD in eastern China for the period 2011–2015.

830 Aerosol loading was at a high level over both urban and suburban sites and even over the  
831 rural sites in the YRD, which suggests that pollution from aerosols is not just local, but has  
832 occurred at a regional scale aerosol extinction over eastern China in recent years. The AOD  
833 showed a decreasing trend from the east coast inland to the west as a result of contributions

834 ~~from anthropogenic activity. Hygroscopic growth and the burning of biomass from crop~~  
835 ~~residues in the summer season could cause this obvious increase in the AOD. The ratios of~~  
836 ~~AOD<sub>fine</sub>/AOD<sub>total</sub>, fine mode fraction of AOD was (>0.90) and coarse mode fraction of AOD (~0.10)~~  
837 ~~consistently >0.90, indicating that fine-mode particles made a major contribution to the total~~  
838 ~~AOD in the YRD. The as well as the~~ relationship between the EAE and the spectral difference in  
839 the EAE suggested ~~that~~ the dominance of fine mode fraction to the AOD and the subordinate  
840 position of coarse mode fraction in the YRD. dust is not important in eastern China. The  
841 validation results indicates a good Terra-MODIS matching with better fitting correlation at 3km  
842 rather than 10km products with the ~~The MODIS C6 AOD~~ retrievals performed better in  
843 suburban than in urban and rural areas, but were systematically over estimated in rural and  
844 urban areas and their immediate surroundings. ~~A large part of the MODIS retrieval AOD was~~  
845 ~~outside the expected error, especially at AOD values <0.80 in urban areas and their immediate~~  
846 ~~surroundings.~~

847 The range of ~~variation of the total, fine- and coarse-mode SSA at 440nm values~~ was about  
848 0.91–0.94, 0.93–0.95 and 0.81–0.84, respectively, in the YRD region which, suggesting the  
849 presence of mainly scattering aerosol particles in eastern China as a result of high industrial  
850 and anthropogenic activity. ~~The fine- and coarse-mode particles showed significant scattering~~  
851 ~~and absorption in the urban, suburban and rural areas of the YRD region. The SSA of dust was~~  
852 weakly lower at short wavelength while the SSA of aerosol from biomass burning has the  
853 strong wavelength dependence in the longer wavelength. The imaginary part of the refractive  
854 index was larger at urban sites as a result of the high loading of absorption aerosols. The large  
855 imaginary parts occurring in August may be due to the higher emission of absorptive particles  
856 from the post-harvest burning of biomass.

857 The similar AAOD levels at the seven sites indicated that absorbing aerosols were  
858 homogeneously distributed in the YRD region. ~~The low AAODs in the winter season suggest~~  
859 ~~fewer absorbing aerosol emissions at the urban, suburban and rural sites.~~ The difference in the  
860 distribution of the AAE suggests that the absorbing aerosols have different characteristics  
861 depending on the emission source. ~~Hygroscopic growth not only contributed to the high aerosol~~

862 ~~extinction values, but also increased the size of the fine-mode particles in the summer in~~  
863 ~~the YRD region.~~ The “mostly black carbon” category was the dominant contributor of absorbing  
864 aerosols at the urban, suburban and rural sites in the YRD region. The submicron “mixed small  
865 particle” category had a significant effect on the aerosol optical properties over the YRD region.  
866 The sphericity fraction showed a dispersed distribution of spherical particles, indicating a  
867 mixture of both fine- and coarse-mode particles from anthropogenic and natural sources.

868 The large ARF-BOA indicated a high aerosol loading that scattered and absorbed more  
869 radiation. ~~It also showed that with the stronger the aerosol~~ cooling effect ~~of the aerosols~~ at the  
870 surface ~~was stronger~~ in the YRD region. Both the burning of biomass from crop residues and  
871 the hygroscopic growth of particles could make important contributions to the ARF-BOA in  
872 summer over the YRD region. The AFR-TOA values were negative all year, ~~suggesting that~~  
873 ~~the aerosols had with an aerosol~~ cooling effect at the TOA, ~~while the instantaneous positive in~~  
874 AFR-TOA value in the winter by the large surface reflectance of better vegetation has been  
875 found different from the north/northeast China.

876  
877 The column aerosol optical properties over urban, suburban and rural areas of YRD  
878 region of China were investigated and the results will increase our understanding of the  
879 characteristics and sources of aerosol emissions over eastern China. Future research should  
880 consider the vertical distribution of aerosols by Lidar, the validation of the aerosol optical  
881 results of other satellite products such as VIIRS and GOCI, and a comprehensive analysis of  
882 the physical and chemical properties of aerosols and meteorological factors.

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891 Framework Programme (FP7/2007-2013) under grant agreement no. 262254.

892 **Reference**

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