Reviewer 1

General comments:

This study makes an extensive and comprehensive national distribution of the aerosol optical properties and direct radiative effect during 2008-2017 in China over decade change. The aerosol key optical parameter obtained from CARSNET, and this ground-based observation net was established independently with Chinese characteristics. The instrument calibration and inversion algorithm from CARSNET has been recognized by the international community, and the results have also been compared by the global ground based observational organizations such as AERONET, etc. Generally, five regions including 50 ground stations nationwide were defined in this study, covers almost the whole region of China, which is of great research value contributed to the regional aerosol optical properties in China, East Asia or the whole world. An emphasis on the estimation of the aerosol optical properties over China vast and varied terrain under different background and aerosol sources driven by the meteorological factors and climatology changes have been employed. The objective of the paper is challenging to use the National scale, ground-based measurements of aerosol microphysical and its optical properties as well as direct radiative effect obtained from the sunphotometer with good quality and large databases. The paper is well written with most importance to complement and support the climatology for aerosol microphysical and optical properties of China and provide better understanding of the aerosols' climate effects over the different types of sites covering a broad expanse of China. Thus, I would suggest a minor revision before it is considered for publication as following.

Response: Thanks for the reviewer's important comments; some important revisions and the grammar have been corrected according to the reviewer's suggestions.

Special comments:

1. Line 40, the time period for the data at the observation site need a brief description in the Abstract.

Response: Thanks for the suggestions. The time period for the data at the observation site has been briefly descripted as "Multi-year observations of …" in the Abstract.

2. Line 60-61, the word “useful” could be changed as “important” to avoid repetition.

Response: The word “useful” has been changed as “important” to avoid repetition in the manuscript.

3. Line 95-96, some references could be added there.

Response: Following the suggestion of reviewer, some references have been added in the revised paper as “…in many regions of China (Che et al., 2009c, 2018; Zhao et al., 2018)”.

4. Line 126-127, “aerosol size distribution” could be changed as “aerosol size distribution (volume and aerosol effective radii)”.

Response: The words “aerosol size distribution” has been changed as “aerosol size distribution (volume and aerosol effective radii)” in the revised manuscript.

5. Line 196, “Angström” should be changed as “Ångström exponent” to make consistency in the text.

Response: Thank for the suggestions of reviewers. The words “Angström” has been changed as “Ångström” and this change applied to the rest of the manuscript.

6. Line 271, “…was found to be substantially…” should be better revised as “…was found substantially…”.

Response: According the reviewer’s suggestions, the words “…was found to be substantially…” has been modified as “*…was found substantially…”*.

7. Line 297, “…also was high, 0.30 μm3/μm2.” should be better revised as “…also was high to 0.30 μm3/μm2.”.

Response: Thanks for the suggestion. The words “…also was high, 0.30 μm3/μm2.” has been modified in the revised manuscript as “…also was high to 0.30 μm3/μm2.”.

8. Line 331-332, please specify the results of Zhao et al. (2018) in detail.

Response: Follow up on the reviewer’s suggestion, the sentences has been changed as “Zhao

et al. (2018) also reported the effect of sea salt aerosol on the aerosol absorption and radiative effects in the coastal region over northeastern China." in the revised manuscript.

9. Line 375, "…in and around…" should be better revised as "…in or around…".

Response: Done. The words of "…in and around…" was changed as "…in or around…" in the revised paper.

10. Line 432, "…these particles originate from a multitude of sources…" should be better revised as "…these particles originate from multitude sources…".

Response: The sentence "…these particles originate from a multitude of sources…" has been changed as "…these particles originate from multitude sources…".

11. Line 437, "were more than likely" should be better revised as "were more likely".

Response: The words "were more than likely" have been changed as "were more likely" in section 3.3.

12. Line 442, "(EAE 0.25)" should be better revised as "(EAE = 0.25)".

Response: The words "(EAE 0.25)" has been changed as "(EAE = 0.25)" in the revised version.

13. Line 474, "…the aerosol was more absorbing in fall, …" should be better revised as "…the aerosol was more absorbing in autumn, …".

Response: The words "…the aerosol was more absorbing in fall, …" has been changed as "…the aerosol was more absorbing in autumn, …"， and this change applied to the rest of the manuscript.

14. Line 482-483, "Therefore, the different SSA440nm distributions in the two regions may be attributed by the special aerosol composition." Is the special aerosol composition because of the industrial structure of different regions Like the Northeastern China was once the significant heavy industries base in China.

Response: According to the suggestion, the sentences “Therefore, the different SSA440nm distributions in the two regions may be attributed by the special aerosol composition” has been changed as “Therefore, the different SSA440nm distributions in the two regions may be attributed by the special aerosol composition related to the urban-industrial background of northeastern China (lower SSA440nm) and more anthropogenic sources in the eastern China environmental (higher SSA440nm).” in the revised manuscript.

15. Line 542, “aerosol direct radiative effect” in the section title should be better revised as “direct aerosol radiative effect”.

Response: Thanks for the suggestions. The title of the section 3.5 has been changed as “direct aerosol radiative effect” to consistent with the rest of revised manuscript.

16. Line 554, “-22.13 and -17.43” should be better revised as “-22.13 and -17.43 W/m2”.

Response: According to the suggestion, the “-22.13 and -17.43” has been changed as “-22.13 and -17.43 W/m^2” in the revised manuscript.

17. Line 583, “A notably small” should be better revised as “A notably small positive”.

Response: Thanks for the reviewer’s suggestions. We checked the value of DARE-TOA in Akedala carefully, and found that it should be corrected as -0.42 W/m2 in the text by a typing mistake. Moreover, the value was correct in the relevant descriptive sentences, Table and charts through the text.

18. Line 588, “(SSA 0.92)” should be better revised as “(SSA = 0.92)”.

Response: Done. The words “(SSA 0.92)” has been changed as “(SSA = 0.92)”.

19. Line 607, “as” should be deleted.

Response: The word “as” have been deleted in the revised paper.

Reviewer 2

General comments

This paper characterizes the climatology of aerosol microphysical and optical properties in China using ground-based remote sensing from the CARSNET network. This is one of the most systematic dataset of aerosol optical properties reported in the literature, and is valuable for improving the estimate of aerosol radiative effects and for evaluation of satellite data and climate models. The paper is generally well written. I think it can be considered for publication after the author addresses the following minor comments and suggestions. Besides the following comments, however, there are many outstanding grammar errors in the paper. I strongly suggest that the author ask a native speaker to carefully edit and improve the language.

Response: Thanks for the reviewer's important and constructive comments and suggestions. Some minor comments and suggestions have been revised carefully in the manuscript. Moreover, the grammar has been carefully checked in the paper and the language of this manuscript has been improved by a native speaker.

Special comments:

(1) Line 72-77: The descriptions of the roles of AOD, absorptivity, and SSA are very similar. Please revise a bit to reflect their respective roles.

Response: According the reviewer's suggestions, the descriptions of the roles of AOD, absorptivity, and SSA has been modified to reflect their roles, respectively. The text is as follows:

"Aerosol optical depth (AOD) is one of the key measures of the total aerosol extinction effects on climate (Breon et al., 2002), and the extinction Ångström exponent (EAE) with spectral dependence can be used to obtain the information about aerosol size distributions (Gobbi et al., 2007; Eck et al., 1999; Zheng et al., 2017). The aerosols' absorptivity depends on particle composition is a key determinant to calculate the direct aerosol radiative effect (Haywood and Shine, 1995; Li et al., 2016; Zheng et al., 2018), and the single scattering albedo (SSA) is the parameter has spectral dependence to distinguish major aerosol particle types (Jacobson et al., 2000; Dubovik et al., 2002; Gelencser et al., 2004; Russell et al., 2010;

Giles et al., 2012).".

(2) Line 241-243: Which radiative transfer model is used to calculate the direct aerosol radiative effect?

Response: Following the reviewer's suggestions, the radiative transfer model of Discrete Ordinates (DISORT) approach was used to calculate the direct aerosol radiative effect which has been descript in the manuscript.

(3) Line 258-259: The assumption of single fixed aerosol vertical distribution (exponential to 1 km) may deviate from the real-world situation significantly. What's the potential impact on calculated aerosol radiative effect?

Response: Thank for the reviewer's comments. Firstly, according to the Zhao et al. (2019, AE) pointed out that the distribution of annual MLH in the most land regions of China were less than 1km. Aerosols are mainly distributed within the mixing layer in the atmosphere and usually decrease with the increase of mixing layer height. Therefore, this paper assumes 1 km to calculate the radiation flux is reliable and reasonable. In addition, the single fixed aerosol vertical distribution (exponential to 1 km) assumed in this study were to be consistent with other previous researches in the world, which also adopted this standard.

(4) Line 260-262: What does this error refer to and how is it quantified?

Response: Thank for the suggestions of reviewer. "The error for the observed solar radiation at the surface in global was +2.1 ± 3.0% for an overestimation of about +9 ± 12 Wm^{-2}." was refer to García et al. (2008). The text has been modified as "García et al. (2008) pointed out that the error for the observed solar radiation at the surface in global was +2.1 ± 3.0% for an overestimation of about +9 ± 12 Wm^{-2}.".

Moreover, García et al. (2008) found that a small overestimation of 9 ± 12 Wm^{-2} for measured radiation in global terms within the uncertainty of solar measurements by BSRN (Baseline Surface Radiation) and SolRad-Net (Solar Radiation Networks) observed data.

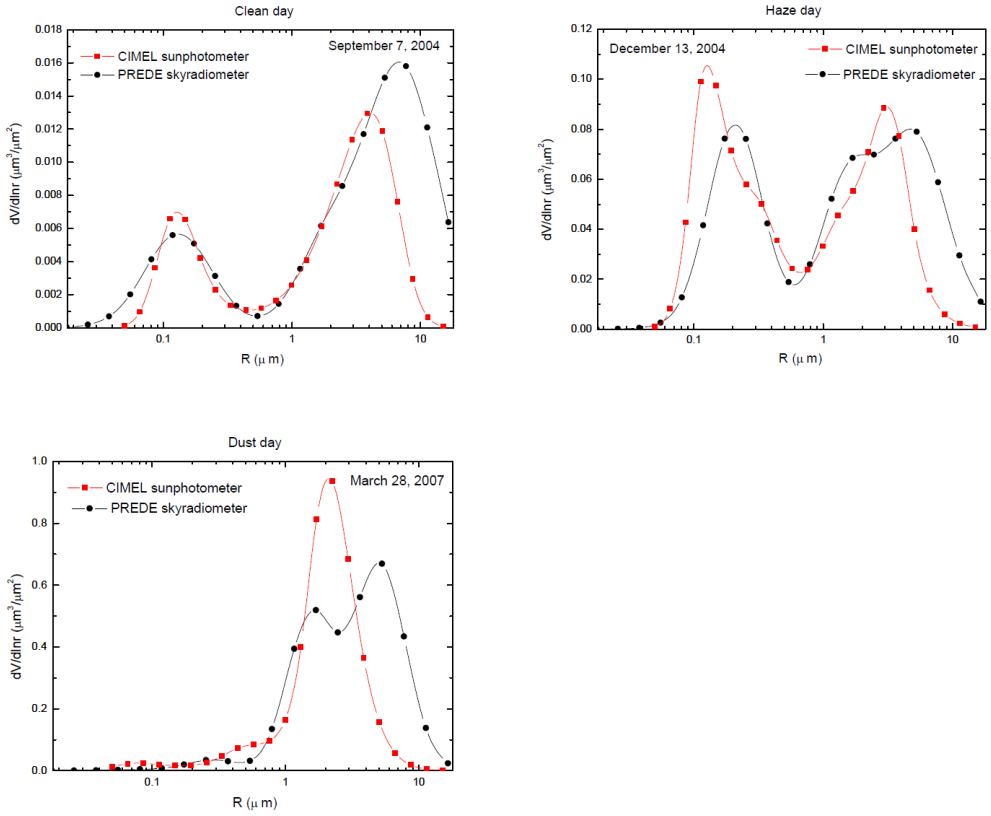
(5) Line 285-287: PVF and PVC have been defined before (Line 216) and the full names used

in these two places are different. Please define only once and use consistent terms.

Response: Thanks for the suggestions. The “PVF” and “PVC” have been defined as “PV_F” and “PV_C” to make consistent throughout the text in the revised paper.

(6) Line 314-316: In urban sites, the volume concentration of coarse particles is higher than fine particles, which sounds counterintuitive. Does this agree with previous studies?

Response: The authors agree with the reviewer’s opinion. Firstly, the volume concentration of fine particles is higher than that of the coarse particles at most of the pollution period; but during the non-pollution period, the volume concentration of coarse particles is higher than fine particles. Che et al. (2008, ACP) has investigated that the volume concentration of coarse particles is higher than fine particles in the clean day as can be seen in the figure below. The results of this manuscript are a multi-year average and agree with previous studies. Moreover, Zhao et al. (2018, JGR) has also found the larger volume concentration of coarse mode particles than fine mode particles in the industrial-urban site of Fushun in Northeastern China. Finally, in these 23 urban sites of this study, there are almost 15 cities in Northern China, where have less precipitation and higher fugitive dust contribution level. These climatic conditions could lead to the higher volume concentration of coarse particles in such urban sites.



(7) Line 334-335: The several studies listed here did not support the hygroscopic growth of fine-mode particles.

Response: Thanks for the reviewer's comments. More references have been added to supplement the hygroscopic growth of fine-mode particles in the revised version. The text is as follows:

"Cheng et al. (2015) found different aerosol volume size distributions for dust and sea salt at Shanghai in the eastern China, and they showed that their relative abundances varied with season and in response to local or long-range transport. Zhao et al. (2018) also reported the effect of sea salt aerosol on the aerosol absorption and radiative effects in the coastal region over northeastern China. Especially the particles hygroscopic growth with different composition observed in special climatic conditions could affect aerosol microphysical properties by geographically variable effects (Zhang et al., 2015; Sun et al., 2010). Like in the YRD region, hygroscopic growth of fine-mode particles could lead to larger AOD and scattering enhancing reported by Sun et al. (2018) and Che et al. (2018). Xia et al. (2019) observed the aerosol hygroscopic growth on fine particle scattering coefficient in Beijing.".

Moreover, some references has been added as following:

Sun, J. Y., Zhang, Q., Canagaratna, M. R., Zhang, Y. M., Ng, N. L., Sun, Y. L., Jayne, J. T., Zhang, X. C., Zhang, X. Y., and Worsnop, D. R.: Highly time- and size-resolved characterization of submicron aerosol particles in Beijing using an Aerodyne Aerosol Mass Spectrometer, Atmos. Environ., 44, 131-140, 2010.

Sun, T., Che, H., Qi, B., Wang, Y., Dong, Y., Xia, X., Wang, H., Gui, K., Zheng, Y., Zhao, H., Ma, Q., Du, R., and Zhang, X.: Aerosol optical characteristics and their vertical distributions under enhanced haze pollution events: effect of the regional transport of different aerosol types over eastern China, Atmos. Chem. Phys., 18, 2949–2971, https://doi.org/10.5194/acp-18-2949-2018, 2018.

Xia, C., Sun, J. Y., Qi, X. F., Shen, X. J., Zhong, J. T., Zhang, X. Y., Wang, Y. Q., Zhang, Y. M., and Hu, X. Y.: Observational study of aerosol hygroscopic growth on scattering coefficient in Beijing: A case study in March of 2018, Sci. Total Environ., 685, 239-247, 2019.

Zhang, L., Sun, J. Y., Shen, X. J., Zhang, Y. M., Che, H., Ma, Q. L., Zhang, Y. W., Zhang, X. Y., and Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China, Atmos. Chem. Phys., 15, 8439-8454, https://doi.org/10.5194/acp-15-8439-2015, 2015.

(8) Line 369, Line 577: Wuhan is not located in the YRD region.

Response: According to the reviewer's suggestion, the location of Wuhan has been checked and modified to central China all through the text.

(9) Line 583-585: Why is the DARE-TOA positive in Akedala? Due to a strong absorption?

Response: Thanks for the important suggestions. We checked the value of DARE-TOA in Akedala, it should be corrected as -0.42 W/m2 in the text by a typing mistake. Moreover, the value was correct in the relevant descriptive sentences, Table and charts through the text.

(10) Line 610-612: I think the strong cooling is not due to strong absorption.

Response: According to the reviewer's helpful suggestion, the sentences "The high DARE-TOA values at these urban sites imply relatively strong cooling effects due to moderate to strong light absorption by the particles." has been revised as "The high DARE-TOA values at these urban sites imply relatively strong cooling effects due to higher aerosol loadings in the atmosphere.

(11) Fig. 2, 3, 8: The scales of the legend should be modified to differentiate large and small values more clearly. For example, in Fig. 8, most values fall between -40 and 0 and hence show the same color.

Response: Thanks for the suggestions. The legend scales of Figure 2 - 8 have been changed in the revised manuscript to differentiate large and small values more clearly as follows:

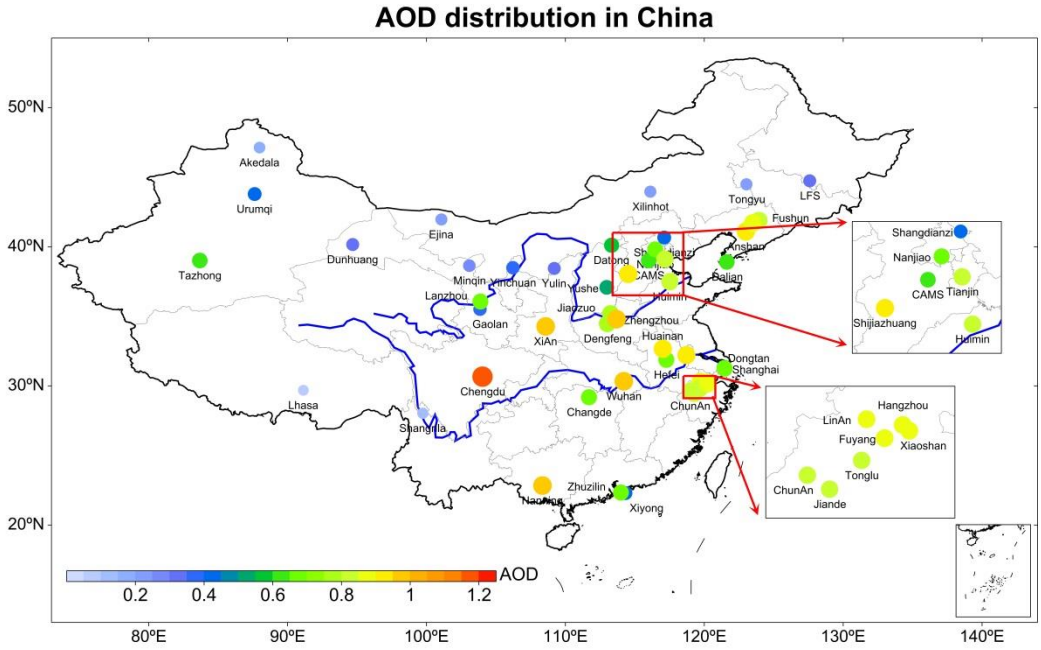


Figure 2. Annual spatial distribution of aerosol optical depth (AOD) at 440 nm at the CARSNET sites

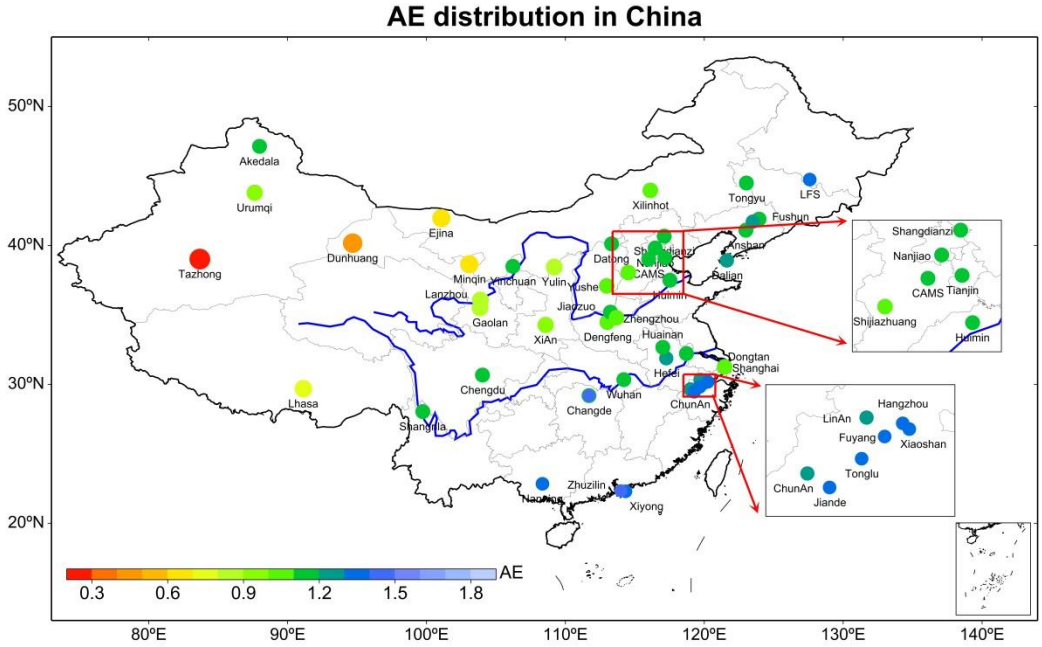


Figure 3. Annual spatial distribution of extinction Ångström exponent (AE) 440-870 nm at the CARSNET sites

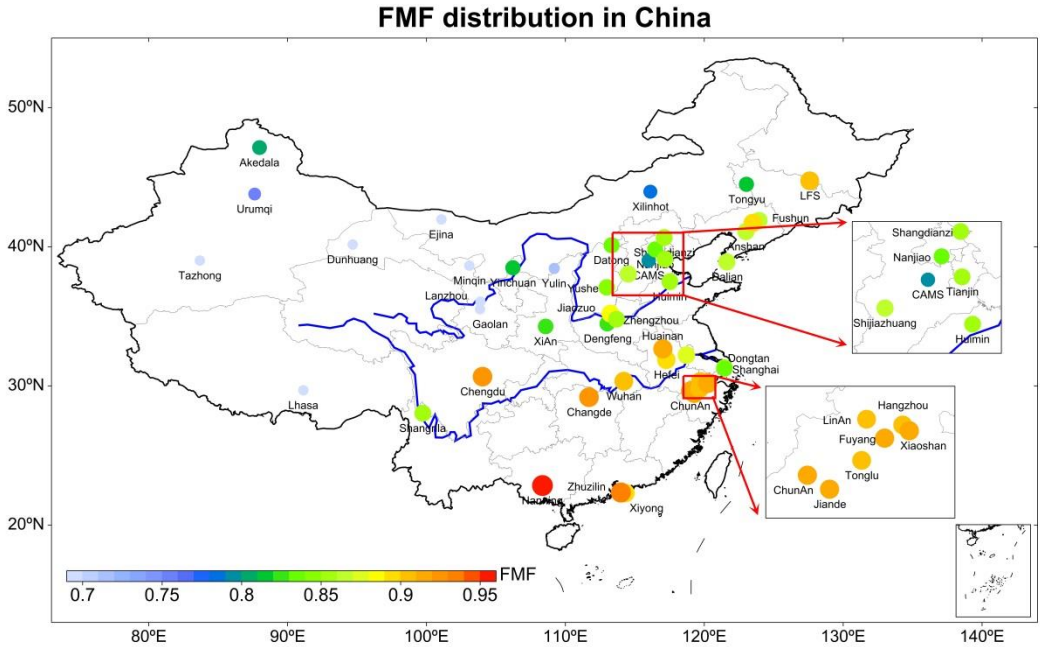


Figure 4. Annual spatial distribution of fine mode fraction at the CARSNET sites

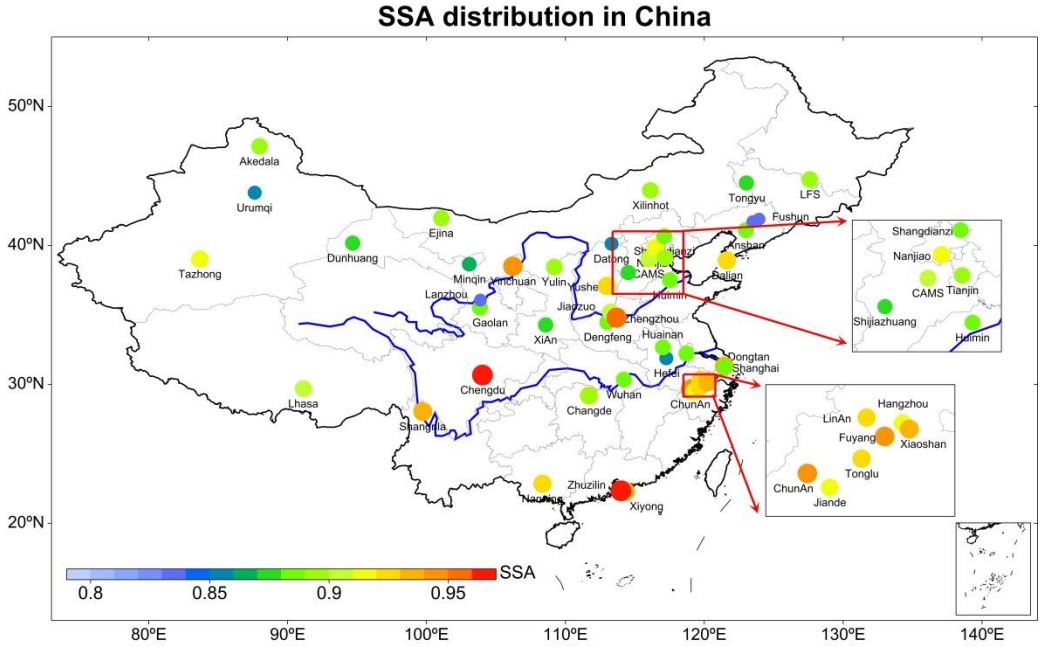


Figure 5. Annual spatial distribution of the single scattering albedo (SSA) at 440 nm at the CARSNET sites

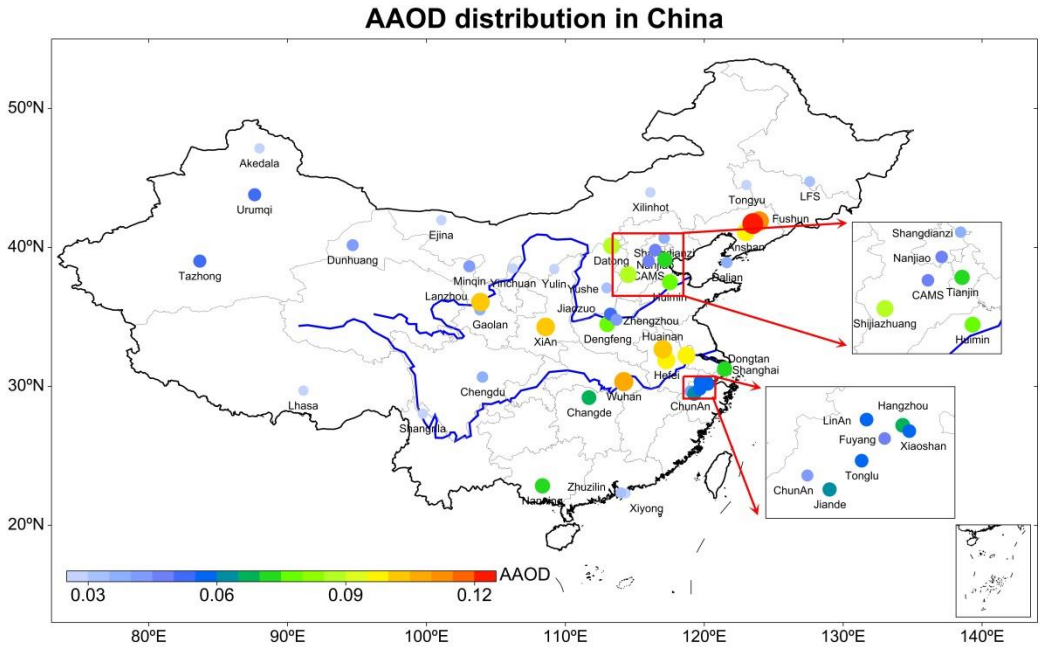


Figure 6. Annual spatial distribution of absorption aerosol optical depth (AAOD) at 440 nm at the CARSNET sites

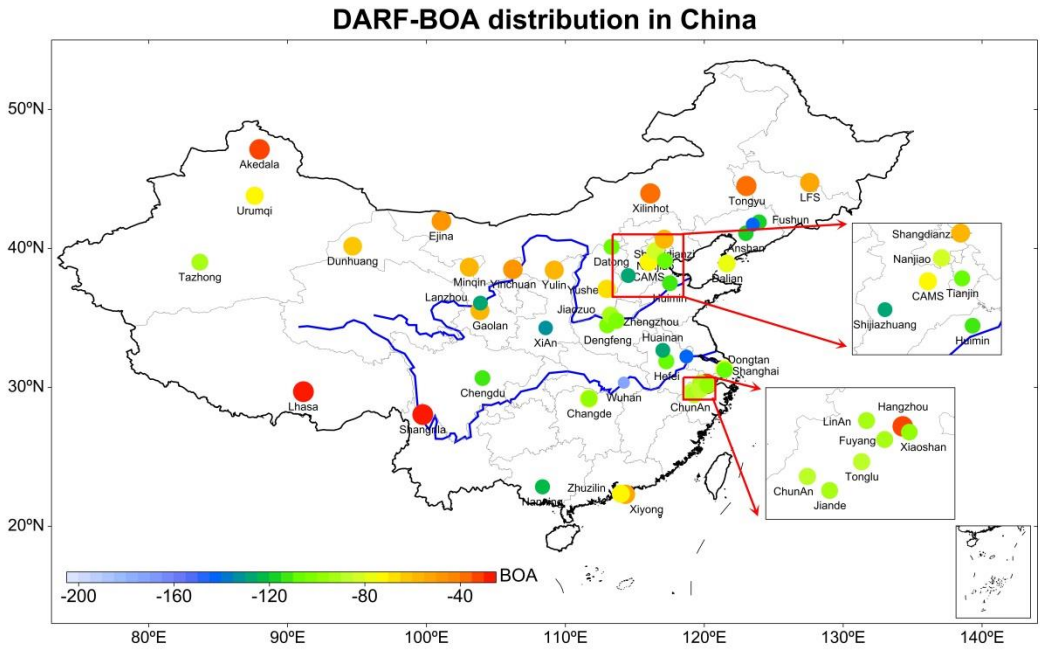


Figure 7. Annual spatial distribution of direct aerosol radiative effect at the bottom of the atmosphere at the CARSNET sites

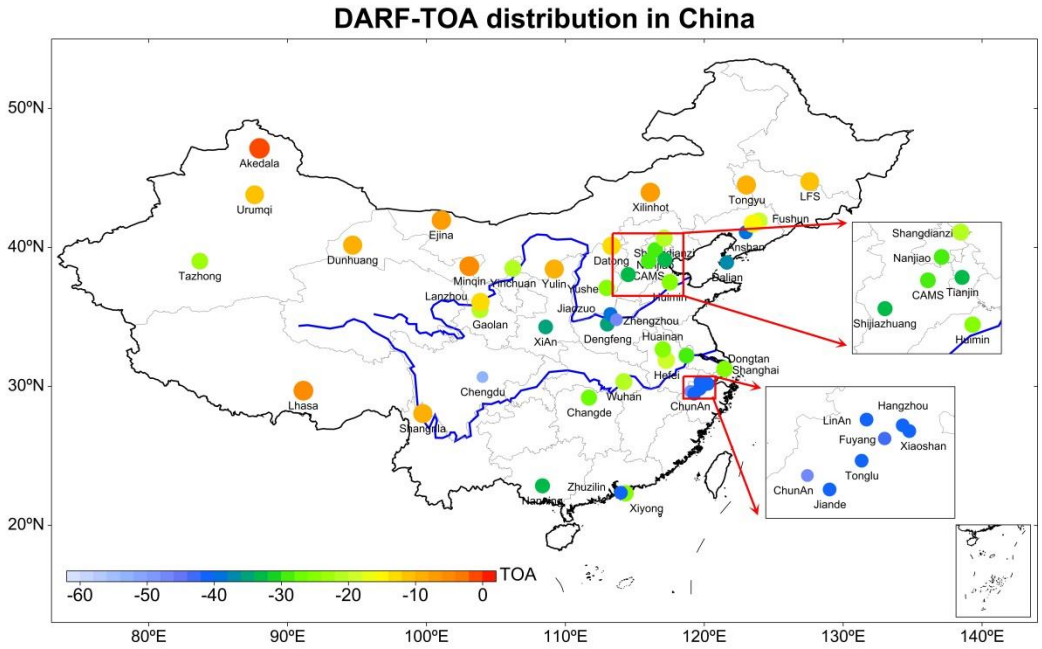


Figure 8. Annual spatial distribution of direct aerosol radiative effect at the top of the atmosphere at the CARSNET sites

Reviewer 3

General comments:

Che et al. “Spatial distribution of aerosol microphysical and optical properties and direct radiative effect from the China Aerosol Remote Sensing Network” This study takes the advantage of the multi-band original observation data of the sunphotometer for many years from the China Aerosol Remote Sensing Network, and applies ground inversion method to calculate the atmospheric aerosols optical characteristic parameters. The authors sorted and processed the data results in aerosol optical properties in China. The results of this study established a ground aerosol remote sensing optical parameters dataset which is more complete, accurate and reliable in China. This work will help to improve China’s climate change research and even for the East Asia. This research enhanced the understanding of aerosol optical properties in different regions of China. I would suggest its publication after minor revision.

Response: We thank for the reviewer’s comments and suggestions. The manuscript has been revised in some minor changes.

Special comments:

1. Lines 67-69. It is well known that the aerosol-cloud interaction plays very important roles to the radiation balance and remains as the largest uncertainty in climate model prediction, even more important than the aerosol direct radiative effect. Before diving into aerosol direct effect, one or two sentences of description about the aerosol indirect effects might be necessary. For example, Garrett and Zhao (2006, DOI: 10.1038/nature04636) and Zhao and Garrett (2015, doi:10.1002/2014GL062015) have shown the warming effect in longwave radiation by aerosol indirect effect in the Arctic, Xie et al. (2013, 10.1175/JCLI-D-12-00517.1) show the cooling effect of aerosol indirect effect. For aerosol direct radiative effect, Yang et al. (2016, doi:10.1002/2016JD024938) particularly indicate (also) the optical properties of aerosols determine the particle direct radiation effect.

Response: Thanks for the reviewer’s suggestion. The authors agree with the reviewer’s opinion that the aerosol indirect effects could be same important as the aerosol direct radiative effect such as the aerosol-cloud interaction which play very important roles to the radiation

balance. Some description about the aerosol indirect effects has been added in the revised paper. The text is as follows:

"Atmospheric aerosols have important direct effects on climate because they can scatter and absorb radiant energy and in so doing affect the Earth's energy balance (Charlson et al., 1992; Yang et al., 2016). Meanwhile, the aerosols can be served as cloud condensation nuclei or ice nuclei to affect the climate indirectly through aerosol–cloud interactions (Twomey et al., 1984; Garrett et al., 2006; Zhao et al., 2015; Xie et al., 2013).".

Moreover, the following references were now cited:

Garrett, T. J., and Zhao, C.: Increased Arctic cloud longwave emissivity associated with pollution from mid-latitudes, Nature, 440, 787–789, doi:10.1038/nature04636, 2006.

Xie, S., Liu, X., Zhao, C., and Zhang, Y.: Sensitivity of CAM5-Simulated Arctic Clouds and Radiation to Ice Nucleation Parameterization, J. Climate., 26, 5981–5999, doi:10.1175/jcli-d-12-00517.1, 2013.

Yang, X., Zhao, C., Zhou, L., Wang, Y., and Liu, X.: Distinct impact of different types of aerosols on surface solar radiation in China, J. Geophys. Res.-Atmospheres, 121, 6459–6471. DOI: https://doi.org/10.1002/2016jd024938, 2016.

Zhao, C., and Garrett, T. J.: Effects of Arctic haze on surface cloud radiative forcing, Geophys. Res. Lett., 42, 557–564. doi:10.1002/2014gl062015, 2015.

2. Lines 69-75, In addition to the climate impacts, aerosols also play important impacts on weather. For example, Zhao et al. (2018, doi: 10.1029/2018GL079427) showed that aerosol can enlarge the rainfall area of tropical cyclone, causing severe flooding damage.

Response: According to the helpful suggestions of reviewer, the text has been rewritten in the revised manuscript as follows:

"The optical properties of the aerosol determine the particles' direct effects on the Earth's radiative balance and weather-climate change (Ramanathan et al., 2001; Eck et al., 2005;

Myhre, 2009; Zhao et al., 2018).”

And the following reference was now cited:

Zhao, C., Lin, Y., Wu, F., Wang, Y., Li, Z., Rosenfeld, D., and Wang, Y.: Enlarging Rainfall Area of Tropical Cyclones by Atmospheric Aerosols, Geophys. Res. Lett., doi:10.1029/2018gl079427, 2018.

3. Lines 73-75, Zheng et al. (2017, doi: 10.5194/acp-17-13473-2017) have done a comprehensive study about the aerosols based on AOD, size, angstrom exponent, PM2.5, and so on, which is worthy to mention here.

Response: Thanks for the suggestions of the reviewer. The comprehensive study of Zheng et al. (2017) about the AOD, size, angstrom exponent, PM2.5 has been added in the revised manuscript. The following reference was now cited:

Zheng, C., Zhao, C., Zhu, Y., Wang, Y., Shi, X., Wu, X., Chen, T.,Wu, F., and Qiu, Y.: Analysis of influential factors for the relationship between PM2.5 and AOD in Beijing, Atmos. Chem. Phys., 17, 13473–13489, doi: 10.5194/acp-17-13473-2017, 2017.

4. Lines 75-76, This is true. Actually, aerosol direct radiative effect can also be used to derive the absorptivity of aerosols. Some recent references might be worthy to be cited.

Response: According the reviewer’s suggestions, some recent references have been added in the revised paper and the text is as follows:

“The aerosols’ absorptivity depends on particle composition is a key determinant to calculate the direct aerosol radiative effect (Haywood and Shine, 1995; Li et al., 2016; Zheng et al., 2018), …”

The following reference was now cited:

Zheng, Y., Che, H., Xia, X., Wang, Y., Wang, H., Wu, Y., Tao, J., Zhao, H., An, L., Li, L., Gui, K., Sun, T., Li, X., Sheng, Z., Liu, C., Yang, X., Liang, Y., Zhang, L., Kuang, X., Luo, S., and You, Y.: Five-year observation of aerosol optical properties and its radiative effects to planetary boundary layer during air pollution episodes in North China: Intercomparison of a plain site and

a mountainous site in Beijing, Sci. Total. Environ., 674, 140-158, 2019.

5. Lines 98-100, I would suggest changing “due to” to “associated with”, since I do not think the economy development is the direct reason for aerosol emissions. Moreover, a few more citation could be added here, such as Yang et al. (2018, DOI: 10.1016/j.atmosres.2018.04.029), Zhao et al. (2019, doi: 10.1029/2018JD028888), Yang et al. (2019, DOI: 10.1016/j.atmosres.2019.01.027), and Li et al. (2016, doi:10.1002/2015RG000500).

Response: Thanks for the important suggestions. The words “due to” has been changed as “associated with” in the revised paper. A few more references were now added and cited as following:

Li, Z., Lau, W. K.-M., Ramanathan, V., Wu, G., Ding, Y., Manoj, M. G., Liu, J., Qian, Y., Li, J., Zhou, T., Fan, J., Rosenfeld, D., Ming, Y., Wang, Y., Huang, J., Wang, B., Xu, X., Lee, S., Cribb, M., Zhang, F., Yang, X., Takemura, T., Wang, K., Xia, X., Yin, Y., Zhang, H., Guo, J., Zhai, P., Sugimoto, N., Babu, S., Brasseur, G. P., and Zhao, C.: Aerosol and monsoon climate interactions over Asia. Rev. Geophys., 54, 866–929. doi:10.1002/2015rg000500, 2016.

Yang, X., Zhao, C., Zhou, L., Li, Z., Cribb, M., and Yang, S.: Wintertime cooling and a potential connection with transported aerosols in Hong Kong during recent decades, Atmos. Res., 211, 52–61, doi:10.1016/j.atmosres.2018.04.029, 2018.

Yang, Y., Zhao, C., Dong, X., Fan, G., Zhou, Y., Wang, Y., Zhao, L., Lv, F., and Yan, F.: Toward understanding the process-level impacts of aerosols on microphysical properties of shallow cumulus cloud using aircraft observations, Atmos. Res., doi:10.1016/j.atmosres.2019.01.027, 2019.

Zhao, C., Wang, Y., Shi, X., Zhang, D., Wang, C., Jiang, J. H., Zhang, Q., and Fan, H.: Estimating the contribution of local primary emissions to particulate pollution using high-density station observations, J. Geophys. Res.-Atmospheres, doi:10.1029/2018jd028888, 2019.

6. Line 103, “pay” -> “paid”.

Response: Done. The word “pay” has been revised as “paid”.

7. Line 103-106, There are also studies about the effect of aerosol optical properties on the sea-land breeze by changing the surface radiation, such as Shen et al. (2019, doi: 10.1016/j.atmosres.2019.05.007).

Response: Thanks for the suggestions. The reference of Shen et al. (2019) about the effect of aerosol optical properties on the sea-land breeze by changing the surface radiation has been added in the revised paper.

Shen, L., Zhao, C., Ma, Z., Li, Z., Li, J., and Wang, K.: Observed decrease of summer sea-land breeze in Shanghai from 1994 to 2014 and its association with urbanization, Atmos. Res., doi:10.1016/j.atmosres.2019.05.007, 2019.

8. Line 107-109, “conducted” -> “were conducted”, and some more recent studies should be cited, such as Zhang et al. (2019, doi: 10.1007/s13143-019-00125-w), and Yang et al. (2019, doi: 10.1029/2019EA000574).

Response: According to the reviewer’s suggestion, the word “conducted” has been changed as “were conducted”. Some more recent studies have been cited as follows:

Zhang, K., Zhao, C., Fan, H., Yang, Y., and Sun, Y.: Toward Understanding the Differences of PM2.5 Characteristics Among Five China Urban Cities, Asia-Pac. J. Atmos. Sci., doi:10.1007/s13143-019-00125-w, 2019.

Yang, Y., Zhao, C., Sun, L., and Wei, J.: Improved aerosol retrievals over complex regions using NPP Visible Infrared Imaging Radiometer Suite observations, Earth. Space. Sci., doi:10.1029/2019ea000574, 2019.

9. Lines 109-112, I think the aerosol optical properties should be studied over much more locations than that listed here, you may use “et al.” or other words to be more accurate. For

example, Zhao et al. (2018, doi: 10.1007/s00376-017-7069-3) have studied the aerosol properties over Xianghe, Hebei province.

Response: Thanks for the comments and suggestions. The sentences has been revised as "Many studies of aerosol optical properties were conducted in northern China with high aerosol loadings, such as the Beijing-Tianjin-Hebei region (Che et al. 2014; Xia et al., 2013; Fan et al., 2006; Xie et al., 2008; Zhang et al., 2019; Yang et al., 2019; Zhao et al., 2018). Aerosol optical properties also have been investigated at Hefei, Shouxian, Nanjing, Taihu, Shanghai and other sites in eastern China (Lee et al., 2010; He et al., 2012; Zhuang et al., 2014; Wang Z et al., 2015; Che et al., 2018)." in the revised paper to make the text more accurately.

10. Lines 126-128, I think the purpose of these aerosol properties measured are not only for evaluation of aerosol first indirect effect. We may change the description as "... can at least be used ...".

Response: Thanks for the suggestions. The description has been revised as "The measurements of greatest interest include aerosol size distributions (volume and aerosol effective radii), optical properties (AOD, AE, SSA, absorption AOD) because those data can at least be used to evaluate direct radiative effect."

11. Lines 135-136, "can be provided".

Response: Done. The words have been changed as "can be provided".

12. Line 151, delete "multi-year" since there is already "from 2010 to 2017".

Response: Done. The words "in multi-year" have been deleted in the revises paper.

13. Line 157, there are two "five sites", please delete one to avoid redundant.

Response: Done. The words "(five sites)" have been deleted to to avoid redundant in the revised manuscript.

14. Lines 169-171, "There are several ... that have been used at the 50 sites in this network as follows".

Response: Thanks for the suggestions. The sentence has been modified as “There are several different types of the Cimel instruments that have been used at the 50 sites in this network as follows:” in the revised paper.

Spatial distribution of aerosol microphysical and optical properties and direct radiative effect from the China Aerosol Remote Sensing Network

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Abstract

Multi-yearLong-term observations of aerosol microphysical and optical properties obtained through ground-based remote sensing at 50 China Aerosol Remote Sensing Network (CARSNET) sites were used to characterize the aerosol climatology for representative remote, rural, and urban areas over China to assess effects on climate. The annual mean effective radii for total particles (R_{efft}) decreased from north to south and from rural to urban sites, and high total particle volumes were found at the urban sites. The aerosol optical depth at 440 nm (AOD_{440nm}) increased from remote/rural sites (0.12) to urban sites (0.79), and the extinction Ångström exponent ($EAE_{440\text{-}870nm}$) increased from 0.71 at the arid/semi-arid sites to 1.15 at the urban sites, presumably due to anthropogenic emissions. Single scattering albedos ($SSA_{440\ nm}$) ranged from 0.88 to 0.92 indicating slightly to strongly absorbing aerosols. Absorption AOD_{440nm}'s were 0.01 at the remote sites versus 0.07 at the urban sites. The average direct aerosol radiative effect (DARE) at the bottom of atmosphere increased from the sites in the remote (-24.40 W/m^2) to the urban area (-103.28 W/m^2) indicating increased cooling at the latter. The DARE for the top of the atmosphere increased from -4.79 W/m^2 at the remote sites to -30.05 W/m^2 at the urban sites, indicating overall cooling effects for the earth-atmosphere system. A classification method based on $SSA_{440\ nm}$, fine mode fraction (FMF), and $EAE_{440\text{-}870\ nm}$ showed that coarse mode particles (mainly dust) were dominant at the rural sites near the northwestern deserts, while light-absorbing, fine-mode particles were important at most urban sites. This study will be ~~useful~~ important for understanding aerosol climate effects and regional environmental pollution, and the results will provide useful information for satellite validation and the improvement of climate modelings.

Keywords: aerosol optical properties; direct aerosol radiative effect; aerosol type; climatology; China Aerosol Remote Sensing Network

1. Introduction

Atmospheric aerosols have important direct effects on climate because they can scatter and absorb radiant energy and in so doing affect the Earth's energy balance (Charlson et al., 1992; Yang et al., 2016). Meanwhile, the aerosols can be served as cloud condensation nuclei or ice nuclei to affect the climate indirectly through aerosol–cloud interactions (Twomey et al., 1984; Garrett et al., 2006; Zhao et al., 2015; Xie et al., 2013). The optical properties of the aerosol determine the particles' direct effects on the Earth's radiative balance and weather-climate change (Ramanathan et al., 2001; Eck et al., 2005; Myhre, 2009; Zhao et al., 2018). Aerosol optical depth (AOD) is one of the key measures of the total aerosol extinction~~aerosols'~~ effects on climate (Breon et al., 2002), and the extinction Ångström exponent (EAE) with spectral dependence can be used to obtain the information about aerosol size distributions~~together with AOD to study aerosol sizes and types~~ (Gobbi et al., 2007; Eck et al., 1999; Zheng et al., 2017). The aerosols' absorptivity depends ~~is a key determinant of absorption~~ on particle composition ~~—~~is a key determinant ~~–~~to calculate the direct aerosol radiative effect (Haywood and Shine, 1995; Li et al., 2016; Zheng et al., 2018~~ STE~~), and the single scattering albedo (SSA) is the parameter has spectral dependence to distinguish major aerosol particle types~~needed to calculate the direct aerosol radiative effect~~ (Jacobson et al., 2000; Dubovik et al., 2002; Gelencser et al., 2004; Russell et al., 2010; Giles et al., 2012).

With the recognition of the importance for climate, the aerosol optical properties have been obtained from ground-based monitoring networks worldwide; some of the major networks include AERONET-Aerosol Robotic Network) (Holben et al., 1998) and its sub-networks of PHOTONS-PHOtométrie pour le Traitement Opérationnel de Normalisation Satellitaire, AEROCAN-Canadian Sun-Photometer Network, and RIMA-Iberian Network for aerosol measurements ((Goloub et al., 2007; Bokoye et al., 2001; Prats et al., 2011), SKYNET-SKYrad Network (Takamura and Nakajima, 2004;

Che et al., 2008), EARLINET-European aerosol Lidar network (Pappalardo et al., 2014), the GAW-PFR Network-Global Atmosphere Watch Programmer-Precision Filter Radiometers (Wehrli, 2002; Estellés et al., 2012),. The CARSNET-China Aerosol Remote Sensing NETwork, the CSHNET-Chinese Sun Hazemeter Network and the SONET-Sun–Sky Radiometer Observation Network have been established to measure aerosol optical properties in China (Che et al., 2009a, 2015; Xin et al., 2007; Li et al., 2018). Furthermore, the aerosol optical properties have also been used in comprehensive studies of aerosol physical characteristics and chemical composition in many regions of China (Che et al., 2009c, 2018; Zhao et al., 2018).

China has become one of the largest aerosol sources in the world associated with~~due to~~ its rapid economic development, and this has caused significant effects on local environments and regional climate (Che et al., 2005; Xia, 2010; Li et al., 2016; Yang et al., 2018, 2019; Zhao et al., 2019). There have been numerous studies that have focused on aerosol optical properties obtained though ground-based remote sensing methods in China (Luo et al., 2002; Li et al., 2003; Duan and Mao, 2007). A few researches have ~~pay~~ paid more attention to the aerosol optical properties and its radiative effects over the urban-industrial areas as well as at coastal sites in northeastern and eastern China (Wang et al., 2010; Xin et al., 2011; Xia et al., 2007; Zhao et al., 2016; Wu et al. 2012; Shen et al., 2019). Many studies of aerosol optical properties were conducted in northern China with high aerosol loadings, such as the Beijing-Tianjin-Hebei region (Che et al. 2014; Xia et al., 2013; Fan et al., 2006; Xie et al., 2008; Zhang et al., 2019; Yang et al., 2019; Zhao et al., 2018). Aerosol optical properties also have been investigated at Hefei, Shouxian, Nanjing, Taihu ~~and,~~ Shanghai and other sites in eastern China (Lee et al., 2010; He et al., 2012; Zhuang et al., 2014; Wang Z et al., 2015; Che et al., 2018). Some studies of aerosol optical properties have been made in southern China (Wang et al., 2015; Tao et al., 2014), and those at remote and rural sites in

China provide information on regional background conditions (Che et al., 2009b; Wang et al., 2010; Xue et al., 2011; Zhu et al., 2014; Yuan et al., 2014).

China's vast size, varied terrain, and heterogeneity of aerosol sources, has led to strong temporal and spatial variability in aerosol optical and physical properties. The mixtures of aerosol types at most sites are complex, and aerosol populations' size and composition are affected by their sources, transformations that occurring during transportation and removing processes (Cao et al., 2007; Wang et al., 2007; Zhang et al., 2013; Wan et al., 2015). National scale, ground-based measurements of aerosol microphysical and its optical properties obtained from the sunphotometer provide for a better understanding of the aerosols' climate effects over the different regions of China. The measurements of greatest interest include aerosol size distributions (volume and aerosol effective radii), optical properties (AOD, AE, SSA, absorption AOD) because those data can at least be used~~can be used~~ to evaluate direct radiative effect.

The aim of this study was focused on the investigation of the climatological spatial distribution of aerosol microphysical and optical properties over regional-scales using spatial distribution data from the national CARSNET network. The data were collected at CARSNET sites, which include sites in the remote, rural and urban area, with the same calibration procedures and calculation algorithms were used at all sites. As a result, the data are directly comparable among sites (Che et al., 2009a), and the results can be provided to characterize the regional distribution and temporal variation of aerosol optical properties. This research focused on aerosol climate effects and regional environmental pollution, and the results should be useful for satellite validations and for the improvement of models in the future. The remainder of this paper is organized as following: Section 2 firstly describes the sites in detail, and then introduced the methods in data processing of the aerosol optical properties as well as the direct aerosol radiative effect calculation through the retrieved aerosol optical parameters. Section 3

illustrates the aerosol microphysical and optical properties, as well as its direct aerosol radiative effect. An aerosol type classification method is proposed according to the aerosol optical parameters. Section 4 presents the conclusions of the study.

2 Site description, instruments, and data

2.1 Site description

Sunphotometers (CE-318, Cimel Electronique, Paris, France, see Appendix A,) were installed at 50 CARSNET sites (Fig. 1) ~~in multi-year~~ from 2010 to 2017. The stations were classified as remote, rural, or urban sites based on administrative division (Appendix Table 1). Three of the remote stations were about more than 3000 m above the sea level on the Tibetan Plateau far from the anthropogenic influences, and one of them was a northwestern regional background site in China. The 23 rural sites represent (a) five sites of desert regions ~~(five sites)~~ affected by most of dust aerosols rather than anthropogenic particles, (b) two sites affected by both dust and anthropogenic activities on the Loess Plateau, and (c) 16 sites located near or surrounding the large cities relatively strong to the impacts of anthropogenic activities in the central and eastern China. The last category is 24 urban sites located in provincial capitals or heavily populated cities.

2.2 Instruments and calibration

The CE-318 sunphotometers used in this study were calibrated annually, using the CARSNET calibration protocol, to verify the accuracy and reliability of the sky irradiance measurements (Holben et al., 1998; Che et al., 2009; Tao et al., 2014). The reference instruments for CARSNET were periodically calibrated at Izaña, Tenerife, Spain located at 28.31°N, 16.50°W (2391.0 m a.s.l.) in conjunction with the AERONET program. There ~~is~~ are several different types of the Cimel instruments that have been used ~~of~~ at the 50 sites in this network as follows: (1) logical type CE-318 sunphotometers (440 nm, 675 nm, 870 nm, 940 nm, 1020 nm and three 870 nm at the polarization band), (2) numerical type CE-318 sunphotometers (440 nm, 675 nm, 870 nm, 940 nm,

1020 nm and three polarization bands at 870 nm), (3) numerical type CE-318 sunphotometers at eight wavelengths (340 nm, 380 nm, 440 nm, 500 nm, 675 nm, 870 nm, 940 nm, and 1020 nm), (4) and numerical type CE-318 sunphotometers at nine wavelengths (340 nm, 380 nm, 440 nm, 500 nm, 675 nm, 870 nm, 940 nm, 1020 nm and 1640 nm).

Measurements used to retrieve AODs were at 340 nm, 380 nm, 440 nm, 500 nm, 675 nm, 870 nm, 1020 nm, and 1640 nm, while the total precipitable water content was obtained by using those measurements at 940 nm (Holben et al., 1998; Dubovik and King, 2000). The cloud-screened AOD data were calculated by using the ASTPwin software, and extinction Ångström exponents (EAE) were calculated from the instantaneous AODs for wavelengths of 440 nm and 870 nm (Che et al., 2009, 2015). Sites with more than three daily AOD observations and more than 10 monthly AOD observation days were used to calculate the daily and monthly mean AODs and extinction Ångström exponents. The FMF is described as the fraction of fine mode particles of total AOD_{440nm} ($AOD_{fine440\ nm}/AOD_{440\ nm}$).

2.3. Data processing

The aerosol microphysical and optical properties, including volume size distributions (dV(r)/dln(r), the total, fine, and coarse mode aerosol effective radii (R_{effT}, R_{effF}, and R_{effC}, respectively) single-scattering albedo (SSA), complex refractive indices, absorption AODs (AAODs), and absorption Ångström~~Angström~~ exponents (AAEs), were retrieved from the observational data from the sky scattering channel of the sunphotometers at 440 nm, 670 nm, 870 nm, 1020 nm using the algorithms of Dubovik et al. (2002, 2006). In the process of retrieval, the data of surface albedo (SA) was interpolated or extrapolated to 440 nm, 670 nm, 870 nm, and 1020 nm based on the daily MCD43C3 data, a product from the MODIS-Moderate Resolution Imaging Spectroradiometer surface reflectance (https://ladsweb.modaps.eosdis.nasa.gov/). The algorithm used to calculate

aerosol volume size distributions ($dV/\ln r$) was under the assumption of a homogeneous distribution of non-spherical particles following the approach of Dubovik (2006). The sphericity fraction retrieved from the inversions is defined as: spherical particles/(spheroidal particles + spherical particles) (Giles et al., 2011).

As Dubovik et al. (2002, 2006) defined that all the particles with effective radii < 0.992 μm were considered as fine mode particles; and those > 0.992 μm were considered as coarse mode particles. For the total (R_{effT}), fine (R_{effF}) and coarse (R_{effC}) mode aerosols, the effective radii are calculated by the equation as follows:

$$R_{eff} = \frac{\int_{r_{min}}^{r_{max}} r^3 \frac{dN(r)}{dlnr} dlnr}{\int_{r_{min}}^{r_{max}} r^2 \frac{dN(r)}{dlnr} dlnr} \quad (1)$$

Where r_{min} denotes 0.05, 0.05, 0.992 μm and r_{max} denotes 15, 0.992, 15 μm of the total, fine and coarse mode particles, respectively.

The coarse (PV_C) and fine aerosol particle volumes distributions (PV_F) are calculated according to a bimodal lognormal function descript by Whitby (1978), Shettle and Fenn (1979) and Remer and Kaufman (1998):

$$\frac{dV(r)}{d\ln r} = \sum_{i=1}^{2} \frac{C_{v,i}}{\sqrt{2\pi}\sigma_i} exp\left[-\frac{(lnr - lnr_{V,i})^2}{2\sigma_i^2}\right] \quad (2)$$

where $C_{v,i}$ means for the volume concentration; $r_{V,i}$ means the median radius, and σ_i means the standard deviation.

The volume median radius is computed by fine and coarse modes particles as follows:

$$\ln r_V = \frac{\int_{r_{min}}^{r_{max}} \ln r \frac{dV(r)}{dlnr} dlnr}{\int_{r_{min}}^{r_{max}} \frac{dV(r)}{dlnr} dlnr} \quad (3)$$

Then the standard deviation is calculated from the volume median radius:

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$$\sigma_V = \sqrt{\frac{\int_{r_{min}}^{r_{max}} (lnr - lnr_V)^2 \frac{dV(r)}{d\ lnr} d\ lnr}{\int_{r_{min}}^{r_{max}} \frac{dV(r)}{d\ lnr} d\ lnr}} \quad (4)$$

The volume concentration ($\mu m^3/\mu m^2$) is speculated by the following equation:

$$C_V = \int_{r_{min}}^{r_{max}} \frac{dV(r)}{d\ lnr} d\ lnr \quad (5)$$

The SSA was retrieved only for AOD_{440nm} > 0.40; this was done to avoid the larger uncertainty inherent in the lower AOD retrieval according to Dubovik et al. (2002, 2006). The AAOD and AAE for wavelength λ were calculated as follows:

$$\mathrm{AAOD}(\lambda) = [1 - SSA(\lambda)] \times \mathrm{AOD}(\lambda) \quad (6)$$

$$\mathrm{AAE} = \frac{-\mathrm{dln}[\mathrm{AAOD}(\lambda)]}{\mathrm{dln}(\lambda)} \quad (7)$$

The total AODs' uncertainty was 0.01 to 0.02 according to Eck et al. (1999). The accuracy of SSA retrieved from AOD_{440nm} > 0.50 with solar zenith angles > 50 was 0.03 (Dubovik et al., 2002). The accuracy of the particle volume size distribution was 15–25% between 0.1 μm ≤ r ≤ 7.0 μm and 25–100% in conditions of r < 0.1 μm and r >7μm.

Direct aerosol radiative effect (DARE in W/m^2) was calculated by the radiative transfer module under cloud-free conditions, which is similar to the inversion of AERONET (García et al., 2008; 2012). The DARE at the bottom of the atmosphere (BOA) and the top of the atmosphere (TOA) was defined as the difference in the shortwave radiative fluxes with and without aerosol effects as follows:

$$\mathrm{DARE}_{TOA} = F_{TOA}^{\uparrow 0} - F_{TOA}^{\uparrow} \quad (8)$$

$$\mathrm{DARE}_{BOA} = F_{BOA}^{\downarrow} - F_{BOA}^{\downarrow 0} \quad (9)$$

Where F and F^0 denoted the broadband fluxes including and excluding aerosols, respectively at the BOA and TOA. The "↑" and "↓"mean the upward~~downward~~ fluxes and ~~upward~~downward fluxes, respectively.

In the radiative transfer module, the absorption and multiple scattering effects are taken into account during flux calculations by the Discrete Ordinates (DISORT) approach (Nakajima and Tanaka, 1988; Stamnes et al., 1988). The gaseous distributions and single fixed aerosol vertical distribution (exponential to 1 km) from the multi-layered US standard 1976 atmosphere were used in the radiative flux calculations (García et al., 2008). García et al. (2008) pointed out that t~~T~~he error for the observed solar radiation at the surface in global was +2.1 ± 3.0% for an overestimation of about +9 ± 12 Wm^{-2}. The data used in preparing the figures for the present paper have been made available as an Appendix.

3. Results and discussion

3.1 Spatial distribution of aerosol microphysical properties

A map showing the 50 CARSNET sampling sites and plots of the aerosol volume size distributions (dV/dlnr) at each of the sites is shown in Fig. 1. Generally, the annual mean effective radius of total particles (R_{effT}) decreased from the inland northwest to the southeastern coastal areas. Furthermore, the volume concentration of total particles was found ~~to be~~ substantially higher at the urban sites. The volume of the coarse mode particles was considerably larger than that of the fine mode particles at the remotes, arid/semi-arid sites and at those sites on the CLP-Chinese Loess Plateau or nearby, indicating that those areas were most strongly affected by larger particles, most likely mineral dust as discussed below.

The average (arithmetic mean) R_{effT} at the remote sites was about 0.47 μm with the volume about 0.05 $μm^3/μm^2$ (Table 1). A large R_{effT} (0.64 μm) was found at Lhasa, and the total aerosol volume there was 0.05 $μm^3/μm^2$. These

results are consistent with those reports by Li et al. (2018) who found high levels of coarse mode particles at Lhasa due to the presence of mineral dust. The two other remote sites, Akedala and Shangri-La, had smaller average R_{effT} values than Lhasa (0.36 and 0.39 μm, respectively), and corresponding volumes were 0.06 and 0.03 $\mu m^3/\mu m^2$. The average fine-mode effective radius (R_{effF}) was 0.14 μm at the remote sites, and fine-mode particle fractional volume (PV_F) was 0.01 $\mu m^3/\mu m^2$, while the average coarse-mode effective radii (R_{effC}) was 2.35 μm and the coarse-mode fractional volume (PV_C) was 0.03 $\mu m^3/\mu m^2$. These findings indicated that the contribution of coarse-mode particles to the total volume of aerosol was larger at the remote sites. A study by Cong et al. (2009) at the remote Nam Co site on the Tibetan Plateau showed that dust particles mainly affected the site in spring, while anthropogenic aerosols were prevalent in the summer.

The average R_{effT} at the arid and semi-arid sites (0.55 μm) was larger than at the remote sites, and the total volume of aerosols at the arid/semi-arid sites also was large (0.14 $\mu m^3/\mu m^2$), nearly three times that at the remote sites. Large R_{effT} values (0.71 μm) were found at Tazhong, which is near the northwestern deserts, and the aerosol volume there also was high~~,~~ to 0.30 $\mu m^3/\mu m^2$. Large PV_C's were found at the arid/semi-arid sites (0.05–0.27 $\mu m^3/\mu m^2$). The arithmetic mean R_{effT} (0.49 μm) at the rural sites on or near the CLP had total aerosol volumes (0.15 $\mu m^3/\mu m^2$) similar to those at the arid/semi-arid sites. These results also show a major contribution to the aerosol volumes by coarse-mode particles at the sites in or near the mineral dust source regions. Bi et al. (2011) similarly found that coarse particles dominated the volume-size distribution at the Semi-Arid Climate and Environment Observatory of Lanzhou University (SACOL) on the CLP.

Small R_{effT} values (0.33 μm) were found at the rural sites in eastern China, and relatively high aerosol volumes were observed there (0.18 $\mu m^3/\mu m^2$). In the Yangtze River Delta (YRD) region, the R_{effF} was large range for 0.16–0.17 μm, and the PV_F's were 0.12–0.13 $\mu m^3/\mu m^2$. At the Mt. Longfeng background

site in northeastern China, the total particle volume was low (0.08 $\mu m^3/\mu m^2$), which is consistent with minimal anthropogenic influences and low aerosol loadings. Compared with the other sites, the urban areas had relatively low coarse mode aerosol concentrations, but small particles were plentiful—the average R_{effT} was 0.37 μm and total volume was high at 0.21 $\mu m^3/\mu m^2$. The average R_{effF} of fine-mode particles at the urban sites was 0.16 μm with a PV_F of 0.10 $\mu m^3/\mu m^2$ while the R_{effC} was 2.22 μm and PV_C was 0.11 $\mu m^3/\mu m^2$.

The effective radii and PV_F values showed strong relationships with population density and vehicle emissions at the urban sites. High volumes of fine mode particles occurred at the northeastern urban site of Shenyang (R_{effT} = 0.16 μm, PV_F = 0.12 $\mu m^3/\mu m^2$); at major cities in northern China, including Shijiazhuang (R_{effT} = 0.16 μm, PV_F = 0.12 $\mu m^3/\mu m^2$) and Zhengzhou (R_{effT} = 0.18 μm, PV_F = 0.12 $\mu m^3/\mu m^2$); at Chengdu, a city in the Sichuan Basin of (R_{effT} = 0.21 μm, PV_F = 0.16 $\mu m^3/\mu m^2$); and at the urban regions of Nanning (R_{effT} = 0.18 μm, PV_F = 0.13 $\mu m^3/\mu m^2$) and Panyu (R_{effT} = 0.16 μm, PV_F = 0.10 $\mu m^3/\mu m^2$) in southern China. Overall, these results show that the volumes of fine-mode particles increased at the urban sites where anthropogenic influences were most apparent.

Cheng et al. (2015) found different aerosol volume size distributions for dust and sea salt at Shanghai in the eastern China, and they showed that their relative abundances varied with season and in response to local or long-range transport. Zhao et al. (2018) also reported the effect~~influence~~ of sea salt aerosol on the aerosol absorption and radiative effects in the coastal region over ~~of~~ northeastern China. ~~Cho et al. (2018) found that aerosol hygroscopicity affected the aerosol microphysical properties in the YRD region.~~ Especially the particles hygroscopic growth with different composition observed in special climatic conditions could affect aerosol microphysical properties by geographically variable effects (Zhang et al., 2015; Sun et al., 2010). Like in the YRD region, hygroscopic growth of fine-mode particles could lead to larger AOD and scattering enhancing reported by Sun et al. (2018) and Che et al.

(2018). Xia et al. (2019) observed the aerosol hygroscopic growth on fine particle scattering coefficient in Beijing.

~~Che et al. (2018) found that aerosol hygroscopicity affected the aerosol microphysical properties in the YRD region.~~ ~~These findings suggest that the hygroscopic growth of fine-mode particles can affect aerosol microphysical properties and that differences in climatic conditions could lead to geographically variable effects.~~

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3.2 Spatial distributions of AOD and EAE

The spatial distributions of $AOD_{440\ nm}$ and $EAE_{440\text{-}870\ nm}$ are shown in Fig. 2. The AOD_{440nm} increased from the remote/rural sites to the urban sites, and as one might expect, the remote sites were the least affected by particle emissions and had the lowest aerosol loadings. For example, the AOD_{440nm} at the remote stations was low and had an average value of 0.12. The Lhasa and Shangri-La sites on the Tibetan Plateau had similar average AOD_{440nm} values of 0.10. These phenomenons are similar to the study of Li et al. (2018), who showed clean air conditions at Lhasa with AOD < 0.1. Cong et al. (2007, 2009) also found a low AOD (0.05) at Nam Co, which was comparable to the background levels at other remote sites.

The AOD_{440nm}'s at the arid/semi-arid sites and those on or near the Loess Plateau ranged from 0.32–0.42, which is higher than at the remote sites. The high AOD_{440nm} at Tazhong (0.60), which is near the deserts in northwestern China was likely due to the large aerosol volume of 0.30 $\mu m^3/\mu m^2$ (section 3.1) caused by mineral dust. Indeed, arid and semi-arid regions in northwestern China are important sources of aeolian dust on a global scale (Bi et al., 2011). Li et al. (2012) showed that the contribution of dust to the average AOD at SACOL near Lanzhou was 28.4%. Other sites that showed large AOD_{440nm} include regions with strong anthropogenic influences, such as Dengfeng (0.79) on the North China Plain, Huimin (0.83) in the YRD (0.83 to 0.87) and Huainan (0.91) in the Guanzhong Plain.

Compared with the sites just discussed, lower AOD_{440nm}'s were found at the Mt. Longfeng background station of the Northeast China Plain (0.34), the semi-arid rural site as Tongyu in northeastern China (0.23), and the clean Xiyong site in southern China (0.41). Zhu et al. (2014) found a low AOD of 0.28 at the North China Plain regional background site. Che et al. (2009c) have pointed out that the large AOD at Lin'an was likely affected by the high aerosol loadings in YRD Region. Among the urban sites in China, large $AOD4_{40nm}$'s were found in the cities with strong influences of anthropogenic activities, such as the Northeastern Plain (Shenyang 0.89), North China Plain (Zhengzhou 0.99), Central China~~YRD region~~ (Wuhan 1.00) and Sichuan Basin (Chengdu 1.17); the average value for these sites was 0.79. Lower AOD_{440nm}'s, that is < 0.50, occurred at remote sites in northwestern China, including Urumqi (0.42), Yinchuan (0.37); those sites are affected less by industrial activities and the population densities are lower compared with the sites in northern or eastern China.

It is worth noting that the particle emissions in ~~and~~ or around the urban sites could lead to large optical extinctions due to hygroscopic aerosol growth, especially in summer when the relative humidity is often high. In a related study, Zhang et al. (2018) found a large AOD of 1.10 at Wuhan in central China and that was linked to secondary aerosol formation under the high summertime temperatures. Li et al. (2015) similarly concluded that high temperature and humidity promoted the formation of fine particles and led to hygroscopic aerosol growth at Nanjing. Qin et al. (2017) observed a high $AOD_{500\ nm}$ of 1.04 at Shijiazhuang and related this to the hygroscopic growth of aerosol fine-mode particles during polluted days.

Clear spatial variability in EAE values over China is evident in Fig. 3, and at the remote sites, the average EAEs were 1.03. The EAE at Lhasa (0.77) was lower than at Akedala (EAE = 1.13), which is in an arid region of central Asia, or at Shangri-La (EAE = 1.19) in Tibet. The average coarse-mode average effective radii (R_{effC}) at Lhasa was 2.26 μm and the fractional volume

was 0.04 $\mu m^3/\mu m^2$, this result suggests the major components of the large mineral dust particles in aerosol populations over that region. The smaller sphericity fraction (~42.70) and lower FMF (0.66) at Lhasa indications the presence of non-spherical aerosol coarse particles compared with the spherical fine particles in the urban sites.

At the arid and semi-arid sites in China, the average EAE value (0.71) was relatively low and the FMF also was low (0.58). The EAE was extremely low at Tazhong (0.25), which is in the Takliman Desert in the Xinjiang Uygur Autonomous Region of northwestern China and the sphericity fraction (12.87) and FMF (0.35) there were lower compared with most of the other sites. This finding indicates a strong contribution of large particles in this desert region consistent with large volume of the coarse-mode particles (0.27 $\mu m^3/\mu m^2$) noted in section 3.1. The average EAE reached 0.93 at the rural sites near the CLP, and the average value of FMF for those sites was 0.73. Eck et al. (2005) found especially low EAE values in March and April (0.3 and 0.4, respectively) at Yulin, China, where the dust aerosol dominated the optical column.

Large EAEs (1.23) were found at the sites in eastern China, and the FMFs also were large (0.89) at those sites. This result can be attributed to the strong impacts of anthropogenic in the more urbanized eastern part of the country. On the other hand, large EAE values also occurred at the clean sites in northeastern China, including Mt. Longfeng (1.38), where the sphericity fraction was 58.5 and the FMF 0.90. This shows that small particles can have stronger effects in these areas relative to some other regions of China. The EAE at Lin'an was larger than that at Shangdianzi in the Northern Plain or Longfengshan in Northeastern China for most months according to data from Che et al. (2009c). At the urban sites, large EAEs were found at sites in southern China, including Nanning (EAE = 1.36, sphericity fraction = 70.12, FMF = 0.95), Panyu (EAE = 1.43, sphericity fraction = 75.55, FMF = 0.93) and Zhuzilin (EAE = 1.45, sphericity fraction = 55.51, FMF = 0.94). This is likely because the large populations and widespread vehicle ownership in those

cities led to the dominance of fine-mode particles throughout the year. Cheng et al. (2015) found a uni-modal distribution of EAE centered in 1.1–1.6 with the occurrence frequency about 72%, which indicated an abundance of fine primary particles at Shanghai in eastern China. At the urban Nanjing site, which is in east-central China, small particles were dominant, and the annual average EAE was 1.21 ± 0.28 (Li et al., 2015).

3.3 Spatial distribution of aerosol single-scattering albedo

The spatial distribution of SSA at 440 nm of the 50 CARSNET stations is shown in Fig. 4. As a frame of reference, Eck et al. (2005) reported that that SSA_{440nm} from the AERONET retrievals were 0.82 to 0.98 globally. We note that SSA_{440nm} values in this range reflect slightly to strongly absorbing aerosols, and these particles originate from ~~a~~ multitude ~~of~~ sources (Che et al., 2018). The SSA_{440nm}'s decreased from remote/rural to the urban sites and from west to east, which means that there were higher percentages of absorbing particles at the urban and eastern stations. The average SSA_{440nm} at the remote sites was about 0.91, which is indicative of particles with moderate absorption. The absorbing aerosols at the remote sites were more ~~than~~ likely mineral dust particles because those sites are less likely to be affected by carbonaceous particles, which also are absorbing, but mainly produced by anthropogenic activities. The SSA_{440nm}'s for the arid and semi-arid sites were 0.89. The relatively high SSA at Tazhong (0.92) was probably due to slightly absorbing, coarse mode dust particles (~~EAE~~ EAE = 0.25).

A study by Bi et al. (2011) showed that SSAs increased slightly with wavelength when dust was present at the SACOL site. Moderately absorbing particles were found in our study on or near the Chinese Loess Plateau where the SSA_{440nm}'s were typically 0.88 to 0.89. Eck et al. (2005) concluded that the spectral SSA demonstrated effects of dust at Yulin because the SSA increased for wavelengths from 440 to 675 nm. At the rural sites in eastern China, large SSA_{440nm}'s mainly occurred at sites in the YRD affected anthropogenic

474 influences; these include Tonglu (0.93), Xiaoshan (0.93), Xiyong (0.94). Che et
475 al. (2018) found the slightly absorbing particles came from industrial activity
476 and anthropogenic sources at YRD region with the $SSA_{440\ nm}$ between 0.91 to
477 0.94.

478 The average value of SSA_{440nm} at the urban sites was 0.90, which
479 indicates that particles with moderate absorption dominated the aerosol
480 populations. Cheng et al. (2015) reported a seasonal range of SSA from 0.88
481 to 0.91 at Shanghai, with higher values in autumn and winter compared with
482 spring and summer. Lower SSA_{440nm}'s occurred at the urban sites and
483 industrial regions in northeastern China, such as Shenyang (0.84), Anshan
484 (0.89), Fuhsun (0.84), which indicates that the particles were more strongly
485 absorbing in that region. On the other hand, higher SSA_{440nm}'s were found at
486 urban sites in southern China, including Nanning (0.92), Panyu (0.90) and
487 Zhuzilin (0.96), and this indicates that the particles at those sites were slightly
488 or weakly absorbing.

489 Moreover, we found that the SSA_{440nm} spatial distribution reflected the
490 percentages of absorbing aerosols at the urban sites both in northern and
491 eastern China. The reports of Dubovik et al. (2000, 2002, 2006) showed that
492 SSA values vary with both particle size and composition, and Su et al. (2017)
493 used the variations in SSA with wavelength to indicate the presence of brown
494 carbon aerosols at Tianjin, a coastal megacity in China. Qin et al. (2017)
495 suggested that the small SSAs found at Shijiazhuang indicated the presence
496 of fine-mode absorbing particles, such as brown carbon. Zhuang et al. (2014)
497 reported that the SSA at the Nanjing urban site ranged from 0.90 to 0.95, and
498 the aerosol was more absorbing in ~~fall~~autumn, possibly due to the biomass
499 burning emission in the YRD. As evident in the results presented in section 3.1,
500 one can see that the R_{effT}, R_{effF} and R_{effC} between northeastern and southern
501 China was very similar. For example, at Shenyang, a megacity in northeastern
502 China, the effective radii of total, fine- and coarse-mode particles were 0.31,
503 0.16, 2.23 μm and the corresponding volumes were 0.22, 0.12, 0.10 $\mu m^3/\mu m^2$,

504 respectively. At Hangzhou in the YRD region, the R_{effT}, R_{effF} and R_{effC} were 0.30,
505 0.17, 2.21 μm with the volumes about 0.22, 0.12, 0.10 $\mu m^3/\mu m^2$, respectively.
506 Therefore, the different SSA_{440nm} distributions in the two regions may be
507 attributed by the special aerosol composition related to the urban-industrial
508 background of northeastern China (lower SSA440nm) and more
509 anthropogenic sources in the eastern China environmental (higher
510 SSA440nm).
511 Dust aerosols with light-absorbing occur more frequently in spring in
512 northeastern China than in more southern regions (Zhao et al., 2018).
513 Anthropogenic emissions from seasonal biomass burning and residential
514 heating are two other main factors that affect aerosol composition between the
515 two regions (Che et al., 2018). Especially in winter, there was high percentage
516 of absorbing aerosols at the northeastern sites, and that was more than likely
517 caused by emissions of carbonaceous aerosol from residential heating (Zhao
518 et al., 2015). Climatic conditions are also the main factors affecting the
519 absorption characteristics of aerosols in different regions of north and south
520 China. The increased light scattering could well be due to the particles
521 hygroscopic growth demonstrated in other studies. For example, Mai et al.
522 (2018) found that AODs and SSAs both increased with relative humidity at
523 Guangdong in the PRD region, which suggests that condensational growth
524 can affect the aerosol optical properties.
525
526 **3.4 Spatial distributions of absorption aerosol optical depth (AAOD)**
527 The spatial distribution of AAOD at 440 nm shown as Fig. 5 indicates that
528 overall, the $AAOD_{440nm}$'s increased from north to south and from remote/rural
529 to urban sites. Lower $AAOD_{440nm}$'s were found at the remote stations, where
530 the average value was 0.01. The $AAOD_{440nm}$ at Akedala, a remote site in
531 northwestern China, was 0.02, and that was higher than at Shangri-La or
532 Lhasa (0.01), both of which are on the Tibetan Plateau. The low $AAOD_{440nm}$'s
533 throughout that region indicates that the aerosol population was not strongly

absorbing. Compared with these three sites, the average $AAOD_{440nm}$'s at the arid and semi-arid sites were higher (0.03); for example, an $AAOD_{440nm}$ of 0.05was found at Tazhong, which is adjacent to the desert, and that indicates that the aerosol particles were more absorbing. As discussed in sections 3.2 and 3.3, dust aerosols likely make a significant contribution to aerosol light absorption in the areas impacted by the deserts.

The low $AAOD_{440nm}$ found at Xilinhot (0.02) was probably due to the low aerosol loadings (AOD_{440nm} = 0.21) in this region. The $AAOD_{440nm}$'s at the Mt. Gaolan and Yulin rural sites which on or around the CLP were about 0.04 and 0.03, respectively, and the particles were moderately absorbing (SSA = 0.89). The large $AAOD_{440nm}$ at Datong (0.09) can be explained by the high AOD_{440nm} (0.58) there. Indeed, large $AAOD_{440nm}$'s were found at rural sites in eastern China, where there were high AODs and low SSAs as noted in sections 3.2 and 3.3. Of these sites, Dengfeng (AOD_{440nm} = 0.08) and Huimin (AOD_{440nm} = 0.08) are located on the North China Plain, while Huainan (AOD_{440nm} = 0.10) is on the Guanzhong Plain. Lower $AAOD_{440nm}$'s, from 0.02–0.03, occurred at Tongyu (0.03), which is at a semi-arid region in northeastern China, at the Mt. Longfeng (0.03) regional background site on the Northeast China Plain, at the Yushe rural site in northern China (0.03), and at the clean Xiyong site in the PRD (0.02).

Several urban sites showed $AAOD_{440nm}$ values greater than 0.10; these include Fushun (0.11) and Shenyang (0.14) in the northeastern China, Lanzhou (0.10) in the northwestern China, and Nanjing (0.10) and Wuhan (0.11) in the eastern and central China. Lower $AAOD_{440nm}$'s occurred in other urban areas, such as Yinchuan ($AAOD_{440nm}$ = 0.02, AOD_{440nm} = 0.37) in the northwest and Zhuzilin ($AAOD_{440nm}$ = 0.03, AOD_{440nm} = 0.66) in the PRD; both of these sites had relatively low AOD_{440}'s indicating weaker anthropogenic influences compared with metropolitan regions of some other areas. We note that there are significant uncertainties in relating aerosol absorbing properties to particle types, such as black carbon, organic matter, as well as mineral dust

(Russell et al., 2010; Giles et al., 2012). Nonetheless, the information presented here on the spatial distribution of AAODs over China may be useful for the further investigations into the relationships between light absorption and particle type (Liu et al., 2017; Schuster et al., 2016a, 2016b).

3.5 Spatial distribution of direct aerosol ~~direct~~ radiative effect at the Earth's surface and top of the atmosphere

The spatial distributions of the DAREs calculated for both the bottom and top of the atmosphere are shown in Fig. 6. Overall, the DARE-BOAs increased from northwest to southeast and from rural to urban sites, consistent with impacts from the densely populated regions around the sites. The average DARE-BOA at the remote sites was -24.40 W/m^2, and in comparison, a higher DARE-BOA (-33.65 W/m^2) occurred at Akedala, which occurred on a remote region of northwestern China. The AOD_{440nm} at Akedala was relatively low (0.17) and the SSA moderate (0.90). The moderate absorption of aerosol could lead to more strong surface cooling effects with little higher DARE-BOA than the other remote sites. The DARE-BOAs for Lhasa and Shangri-La were -22.13 and -17.43 W/m^2, respectively. These results indicate weaker surface cooling effects at the remote sites relative to other regions because the aerosol loadings were relatively low, as indicated by AOD_{440nm}'s < 0.20.

The average DARE-BOTs at the arid and semi-arid sites of China were about -56.43 W/m^2, and those high DARE-BOAs can be explained by the moderately absorbing particles (SSA = 0.89) and large AOD_{440nm}'s (0.32) compared with the remote sites. A large DARE-BOA (-91.20 W/m^2) occurred at the Tazhong site near the northwestern deserts, and there, the high AOD (0.60) and the slight absorption of mineral dust (SSA = 0.92) imply substantial surface cooling. The average DARE-BOA for rural sites on the Chinese Loess Plateau or surrounding was -74.67 W/m^2, and that also implies cooling at the surface.

Several rural sites in northern and eastern China had large DARE-BOA values; these include Huimin (-111.58 W/m^2), Dengfeng (-104.78 W/m^2) and

Huainan (-129.17 W/m^2), and at those sites the AODs were high, from 0.80–0.90, and the SSAs were ~0.89. These results show stronger surface cooling effects at sites influenced by anthropogenic emissions compared with the remote sites or those near the deserts. The large negative DARE-BOA values (-103.28 W/m^2) at the urban sites indicate that the combination of high AOD_{440nm}'s (0.79) and moderate SSAs (0.90) can cause significant surface cooling. Indeed, anthropogenic emissions presumably led to the high DARE-BOAs at urban sites, including Shenyang (-144.88 W/m^2) and Fushun (-116.91 W/m^2) in the Northeastern Plain, Xian in the Guanzhong Plain (-132.55 W/m^2), Chengdu in the Sichuan Basin (-110.42 W/m^2), Lanzhou in the western region (-126.17 W/m^2), and Nanjing (-143.38 W/m^2) and Wuhan (-171.80 W/m^2) in the Central China~~YRD~~. These results indicate that anthropogenic aerosols can cause significant direct radiative effects at urban sites.

The DARE-TOAs increased from north to south and from rural to urban sites, and the average DARE-TOA for the remote stations was low, about -4.79 W/m^2 (Fig. 7). The DARE-TOAs at Lhasa and Shangri-La were -5.04 W/m^2 and -8.93 W/m^2, respectively. A notably small DARE-TOA was found at Akedala (-0.42 W/m^2), indicating that the effects of the aerosol on the temperature of earth-atmosphere system there would be weak. The average DARE-TOA at the arid and semi-arid sites was -10.17 W/m^2. The large DARE-TOA found at Tazhong (-23.49 W/m^2) could represent the larger contribution of slightly absorbing mineral aerosols (SSA = 0.92) and a large AOD (0.60); this indicates more cooling at surface through the absorption and scattering solar radiation compared with the less impacted sites. This is consistent with the results for Tazhong discussed in section 3.1 which showed high volumes of coarse mode particles with large radii.

The average DARE-TOA at rural sites on the Chinese Loess Plateau or nearby was about -14.56 W/m^2. Although the SSA_{440nm} were close to Gaolan and Yulin about 0.89, the TOAs were quite different (Mt. Gaolan -20.87 W/m^2;

Yulin -9.09 W/m^2) which could be due to the different AOD_{440nm} about 0.36 and 0.32, respectively. In rural eastern China, the DARE-TOA was about -32.40 W/m^2, and to put this in context, Che et al. (2018) found that DARE-TOAs of −40 W/m^2 at rural sites in the YRD region, which is indicative of a relatively strong cooling effect. Low DARE-TOAs were found at the Mt. Longfeng rural site in northeastern China (DARE-TOA = -11.34, AOD_{440nm} = 0.34, SSA = 0.89) and at the Tongyu semi-arid site in northeastern China as (DARE-TOA = -8.87, AOD_{440nm} = 0.23, SSA = 0.88) where the aerosol loadings were relatively low and the absorption was moderate.

In the urban sites at central and eastern China, the average DARE-TOA values were about -30.05 W/m^2. Higher DARE-TOAs occurred at Anshan in the Northeastern Plain (-39.66 W/m^2), Chengdu in the Sichuan Basin ~~as~~ (-52.21 W/m^2), Hangzhou in the YRD (-40.16 W/m^2), Jiaozuo (-39.35 W/m^2) and Zhengzhou (-46.18 W/m^2) in the North China Plain, and Zhuzilin (-40.15 W/m^2) in the PRD region. The high DARE-TOA values at these urban sites imply relatively strong cooling effects due to higher aerosol loadings in the atmosphere~~moderate to strong light absorption by the particles~~.

3.6 Spatial distributions of aerosol mixing properties

The spatial distribution of aerosol mixing properties (Figure 8) was obtained by using the SSA_{440nm}, FMF, and EAE results to classify the particles based on size and absorbing properties. In previous studies by Zheng et al. (2017) and Che et al. (2018), the particles in this study were grouped into eight types as Table 2 show. Moreover, the FMF has been provided to give the particle size information in the group of the particles.

At the remote Akedala and Lhasa sites (FMF = 0.70–0.78 and SSA_{440nm} = 0.85), the percentages of mixed absorbing particles (Type V) were 35-40%, while at Shangri-la (FMF = 0.76, SSA_{440nm} = 0.84), the percentage was slightly lower, 24.62%. The characteristics of the particles at these remote, high-altitude sites were probably affected by the rugged topography which

would promote particle mixing. The proportion of coarse mode, mainly dust, particles with moderate to strong absorption (Group VII) was highest at the arid and semi-arid sites. The percent abundances of Group VII particles were 57.90% at Dunhuang (AE = 0.26, SSA_{440nm} = 0.85, FMF = 0.43) and 58.52% at Tazhong (AE = 0.20, SSA_{440nm}= 0.87, FMF = 0.37), respectively. Mixed absorbing particles (Type V) and strongly absorbing dust particles (Group VII) accounted for 30 to 70% of the aerosol in the rural sites on or near the CLP. The percentages of mixed absorbing particles (Type V) at Gaolan, Yulin, and Datong were 31.98%, 45.22% and 29.04%, respectively, and the average FMFs at those sites ranged from 0.70–0.76.

The proportions of the coarse-mode aerosols with strongly absorbing in Group VII were about 35.23% at Gaolan and 21.21% at Yulin, which was mainly dust particles with the FMFs at those sites were 0.43 and 0.48, respectively. The proportion of coarse-mode particles with strongly absorbing in Group VII and coarse-mode particles with weakly-absorbing in Group VIII at the rural sites in eastern China were < 11%. These patterns indicated that the differences in the eastern region from northwestern China because in the east, coarse-mode particles have only a minor contribution to aerosol absorption. The percentage of fine-mode particles with weakly-absorbing in Type IV and mixed absorbing particles in Type V combined about ~50% at the eastern sites. This result suggests that mixed aerosols originated from a variety of sources and that many of the sites were affected by anthropogenic emissions from megacities upwind.

The fine-mode particles with absorbing in Types I, II, III and V accounted for 50 to 90% at most of the urban sites. The percentages of these four particle types combined were especially large in eastern China; for example, at Panyu, particle Types I—IV composed 90.83% of the total, and the FMF there was 0.90–0.94, while at Zhuzilin, the percentage of Types I–IV was 92.55%, and the FMF was 0.92–0.94. These results are another indication that fine-mode particles are important for light absorption in urban areas. In contrast, the

Lanzhou and Urumqi urban sites were less affected by absorbing fine-particles because the percentages of Type I–IV particles were only 19.73% and 18.36%, respectively. The mixed absorbing Type V particles accounted for large percentages of the total at Lanzhou (48.80%, EAE = 0.88, SSA = 0.82, FMF = 0.73) and at Urumqi (59.39%, EAE = 0.94, SSA = 0.84, FMF = 0.75). Different from the other urban sites, these patterns show that larger particles had significant contributions to the aerosol absorption at these two northwestern sites.

4. Conclusions

Aerosol microphysical and its optical properties obtained from the ground-based sunphotometer deployed at 50 CARSNET stations were used to begin the development of their climatology characteristics and to investigate potential aerosol-climate effects over vast area of China. Direct aerosol radiative effects (DAREs) at the bottom and at the top of the atmosphere were calculated, and eight types of aerosols were classified based on the particle size and absorbing properties. The annual mean values of the ReffT decreased from the arid and semi-arid sites (0.55 μm) to the urban sites (0.37 μm). The aerosol volumes increased from the remote sites (0.05 $\mu m^3/\mu m^2$) to the urban sites (0.21 $\mu m^3/\mu m^2$). The volumes of coarse-mode particle were larger than those for the fine mode at the remote and arid/semi-arid sites—this can be explained by the greater relative abundances of mineral dust compared with pollution-derived particles at those sites. At the urban sites, where anthropogenic influences were relatively strong, the proportion of fine mode particles increased gradually with aerosol volume.

The AOD_{440nm} progressively increased from the remote sites (0.12) to the arid and semi-arid sites (0.32) to rural sites in eastern China (0.70) and finally to the urban sites (0.79), which were the ones most strongly affected by anthropogenic activities. The average $EAE_{440\text{-}870\ nm}$'s at the arid and semi-arid sites were relatively low (0.71), which indicates an important contribution of

larger particles to the aerosol extinction in those regions. The consistently large $EAE_{440\text{-}870\ nm}$'s at the urban sites (> 1.20) and the high FMFs that those site (0.88) are the evidence that fine mode particles are prevalent throughout year. The average SSA_{440nm}'s at the remote, rural, and urban sites were relatively similar, averaging about 0.89, and this indicates the particles were moderately absorbing.

Overall, dust aerosols with light-absorbing in spring and emissions came from biomass burning and residential heating during the colder months were the main factors that led to spatial differences in the percentages of absorbing aerosols over China. The $AAOD_{440nm}$'s increased from the remote sites (0.01) to the arid and semi-arid sites (0.03) to the rural sites of eastern China (0.05) and finally to the urban sites (0.07). High $AAOD_{440nm}$'s were caused by light-absorbing dust aerosols at the rural sites and by the strong anthropogenic emissions in the metropolitan areas. The spatial patterns in the absorbing aerosols were not only affected by the chemical composition of aerosol, but also by physical effects imposed by topography, weather, and climate.

The average DARE-BOA values were -24.40 W/m^2 at the remote sites; -56.43 W/m^2 at the arid and semi-arid sites; -74.67 W/m^2 at the sites on the CLP or nearby; -85.25 W/m^2 at the rural sites in eastern China; and -103.28 W/m^2 at the urban sites. The larger DARE-BOA values at the urban sites imply stronger cooling effects from anthropogenic emissions compared with those from mineral dust at the remote sites or those near the desert. Moreover, larger DARE-TOA's also occurred at the urban sites (-30.05 W/m^2), which indicates strong cooling effects due to the large aerosol extinctions between the earth-atmosphere system displayed the moderate to strong light absorption. Mixed-absorbing particles were the most abundant aerosol type in the remote and rural sites on or near the Chinese Loess Plateau and in eastern China. Mineral dust particles with moderate to strong absorbing were dominant in the arid and semi-arid sites while absorbing fine-mode particles accounted for 50 to 90% of the aerosol at the most urban sites.

744 The results of the study have considerable value for ground truthing
745 satellite observations and for validating aerosol models. Moreover, the results
746 also have provided significant information on aerosol optical and radiative
747 properties for different types of sites covering a broad expanse of China.
748 These results also are a major step towards developing a climatology for
749 aerosol microphysical and optical properties for China and even East Asia.

750 **Data availability:**

751 The detailed CARSNET AOD dataset used in the study can be requested
752 by contacting the corresponding author.

753 **Competing interests.**

754 The authors declare that they have no conflict of interest.

755 **Author contribution:**

756 All authors contributed to shaping up the ideas and reviewing the paper.
757 HC, XX and XZ designed and implemented the research, as well as prepared
758 the manuscript; HC, HZ,YW and HW contributed to analysis of the CARSNET
759 dataset; HC, XX, JZ, OD, BNH, PG, and ECA contributed to the CARSNET
760 data retrieval; HC, BQ, WG, HY, RZ, LY, JC, YZ, KG, and XZ carried out the
761 CARSNET observations; OD, BNH, PG, and ECA provided constructive
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Figure captions

Figure 1. Annual spatial distribution of aerosol volume-size distributions at the CARSNET sites

Figure 2. Annual spatial distribution of aerosol optical depth (AOD) at 440 nm at the CARSNET sites

Figure 3. Annual spatial distribution of extinction Ångström exponent (AE) 440-870 nm at the CARSNET sites

Figure 4. Annual spatial distribution of fine mode fraction at the CARSNET sites

Figure 5. Annual spatial distribution of the single scattering albedo (SSA) at 440 nm at the CARSNET sites

Figure 6. Annual spatial distribution of absorption aerosol optical depth (AAOD) at 440 nm at the CARSNET sites

Figure 7. Annual spatial distribution of direct aerosol radiative effect at the bottom of the atmosphere at the CARSNET sites

Figure 8. Annual spatial distribution of direct aerosol radiative effect at the top of the atmosphere at the CARSNET sites

Figure 9. Annual spatial distribution of the aerosol type classification of types I–VII at the CARSNET sites

Table 1 The aerosol type classification based on the optical properties

Figure 1. Annual spatial distribution of aerosol volume-size distributions at the CARSNET sites

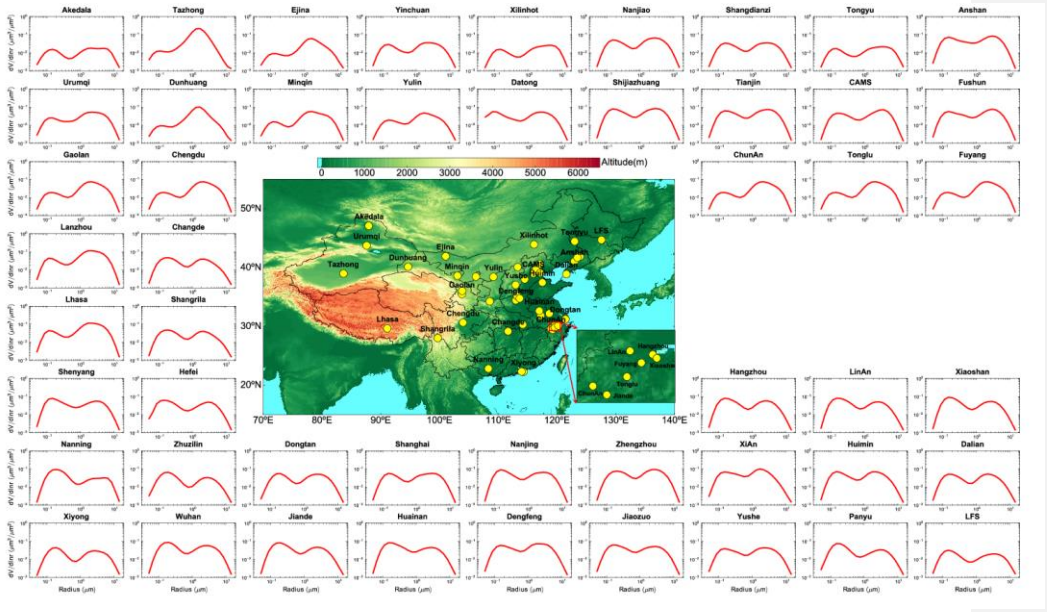


Figure 2. Annual spatial distribution of aerosol optical depth (AOD) at 440 nm at the CARSNET sites

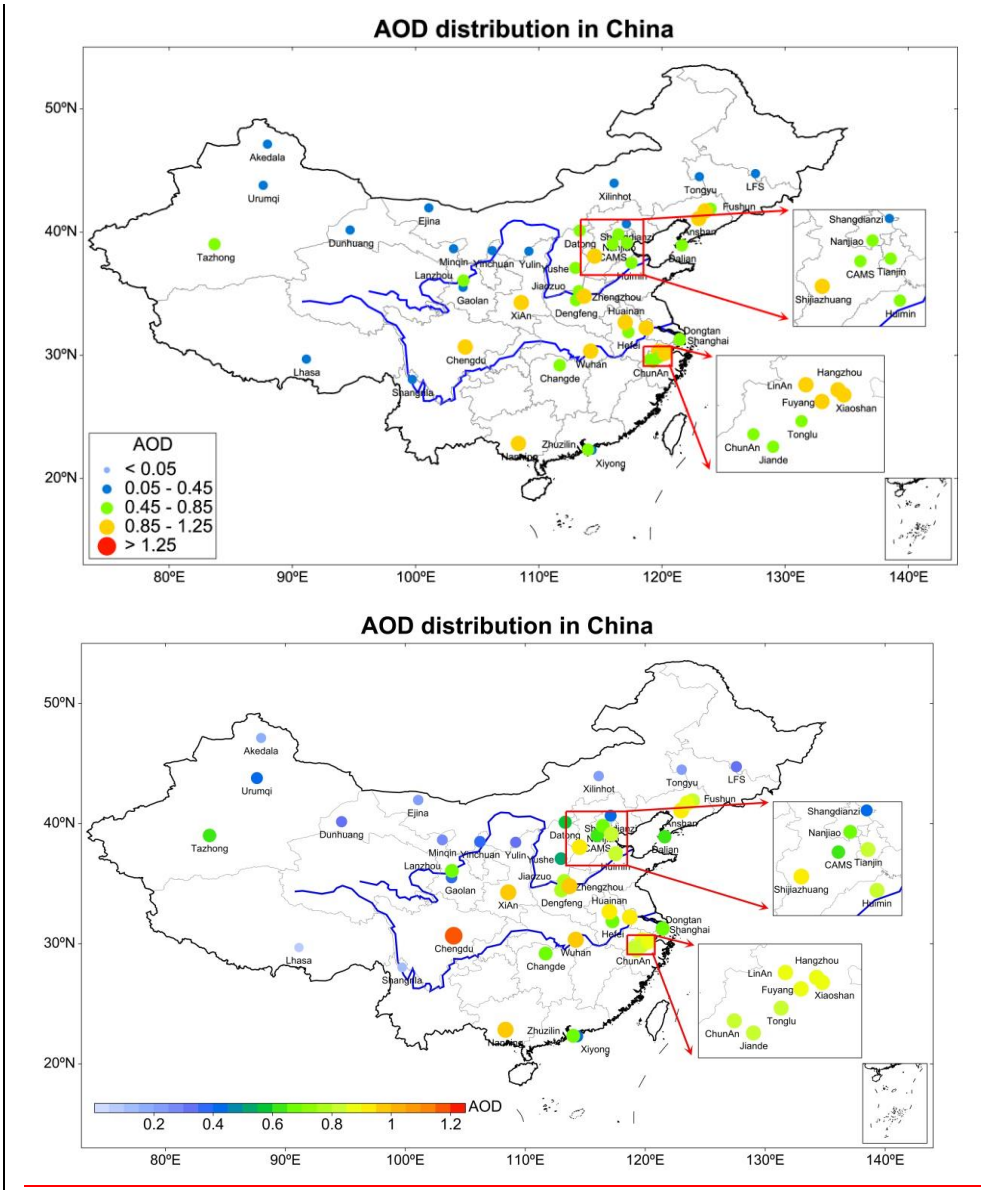


Figure 3. Annual spatial distribution of extinction Ångström exponent (AE) 440-870 nm at the CARSNET sites

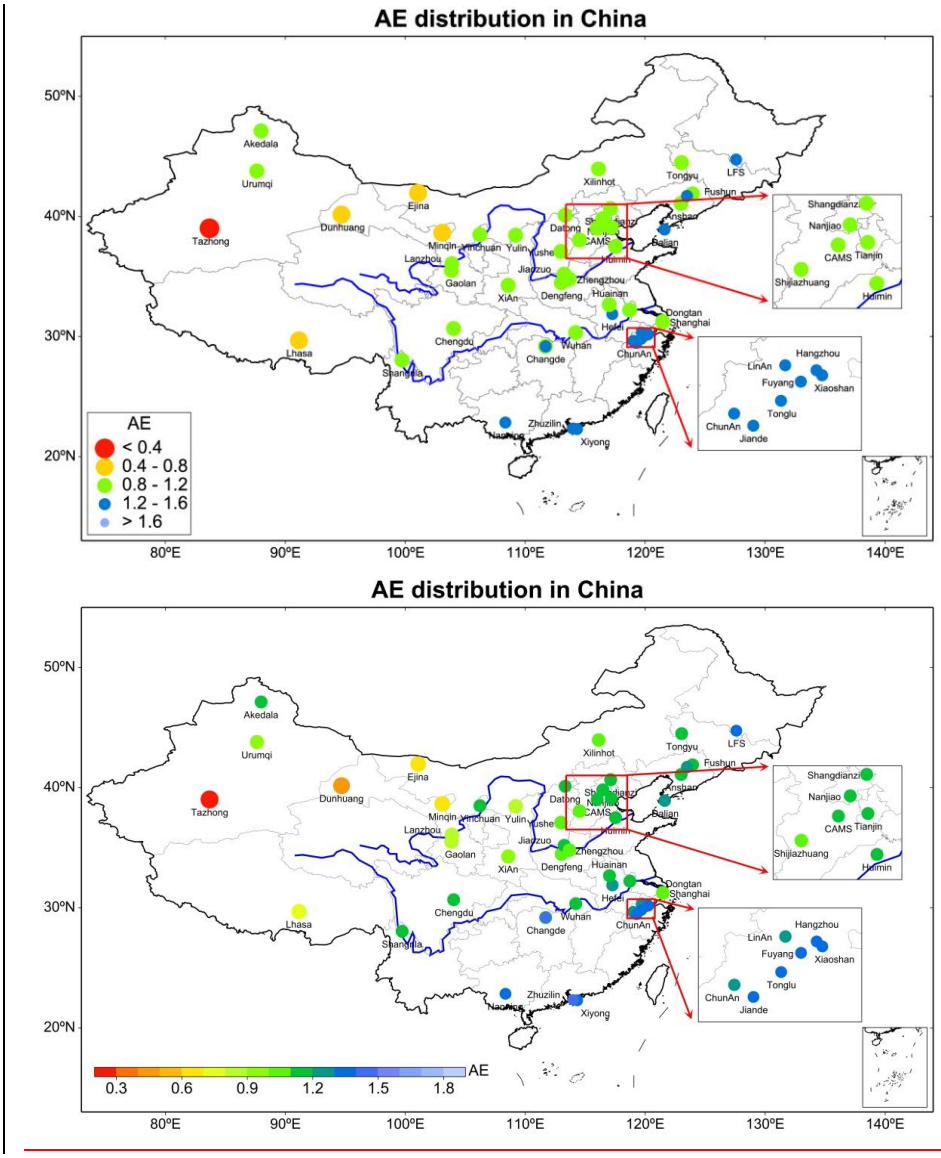


Figure 4. Annual spatial distribution of fine mode fraction at the CARSNET sites

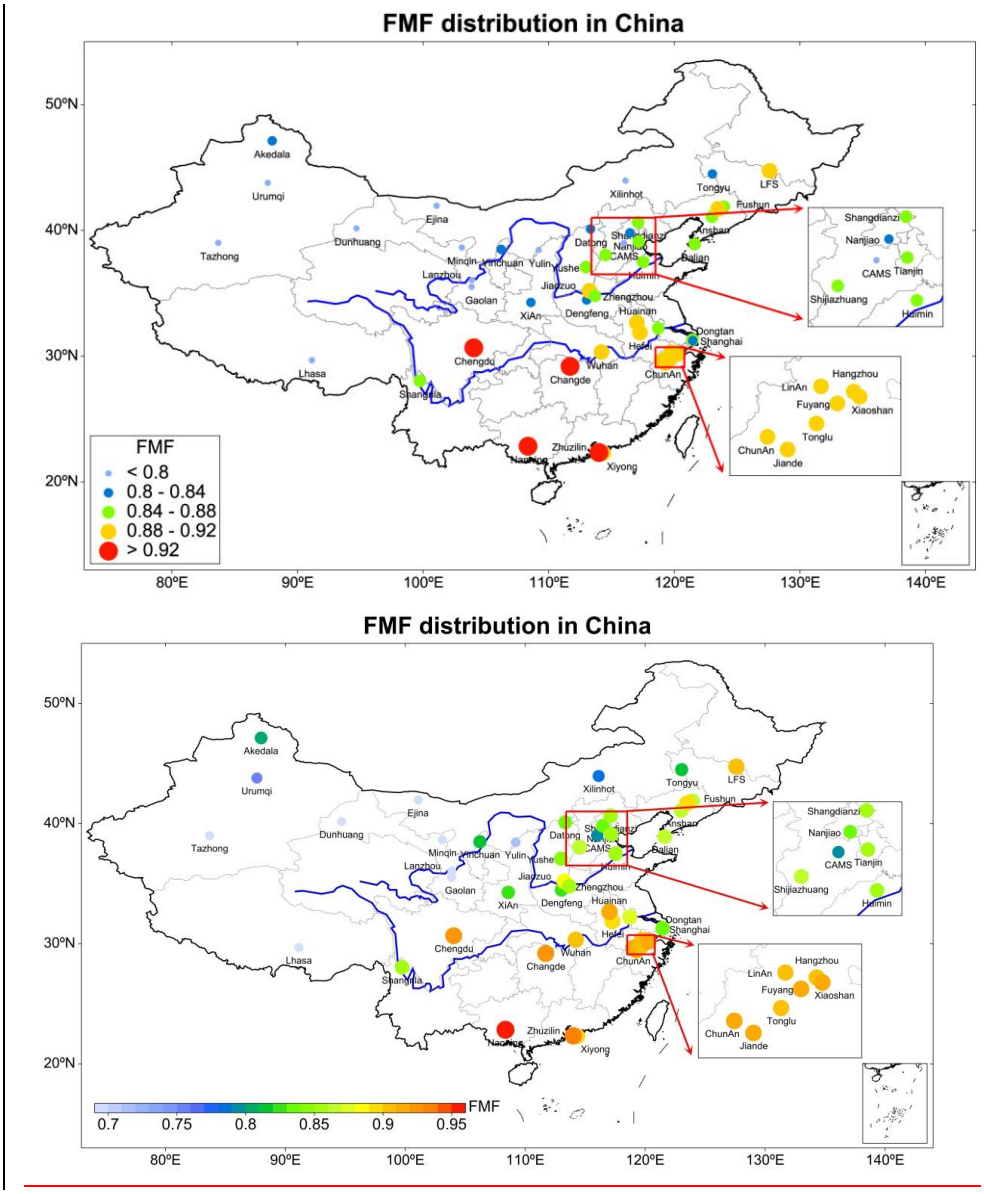


Figure 5. Annual spatial distribution of the single scattering albedo (SSA) at 440 nm at the CARSNET sites

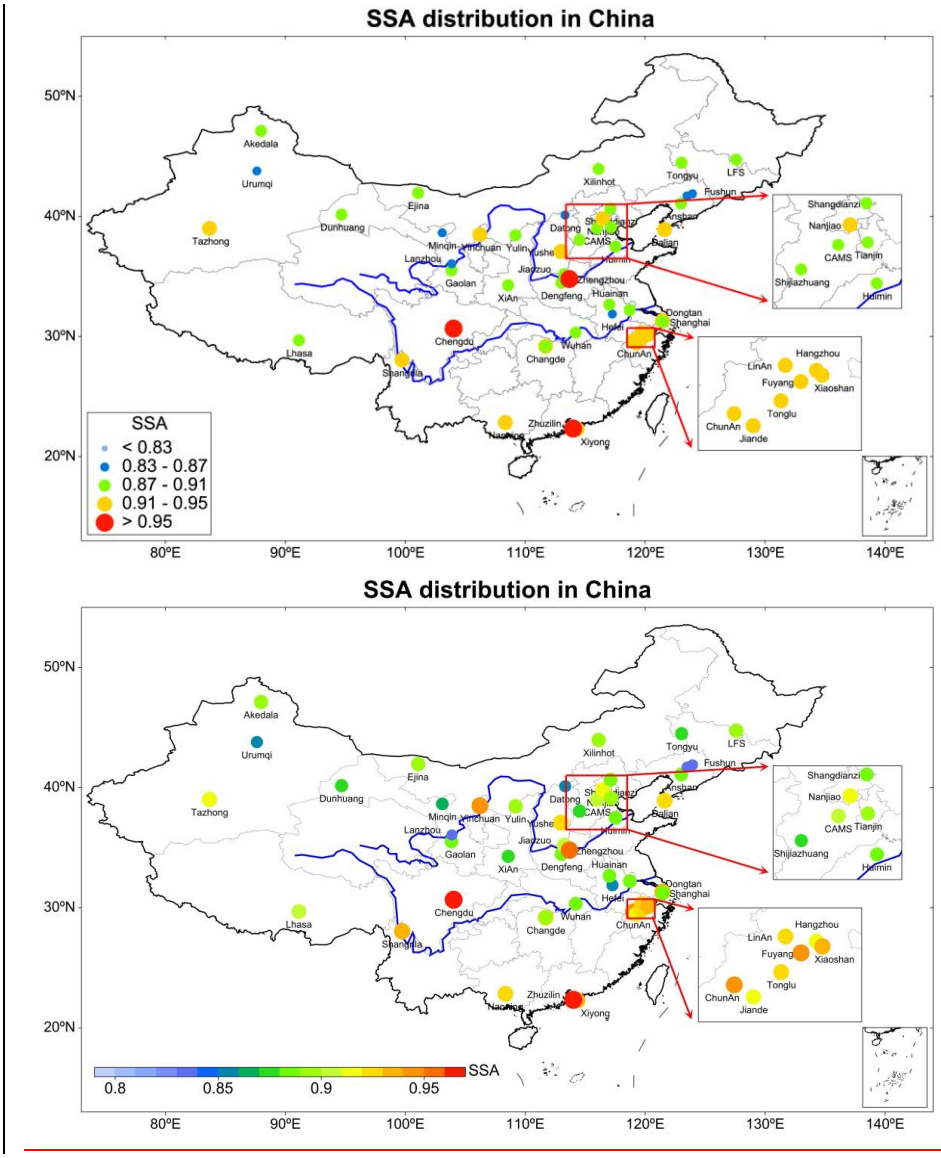


Figure 6. Annual spatial distribution of absorption aerosol optical depth (AAOD) at 440 nm at the CARSNET sites

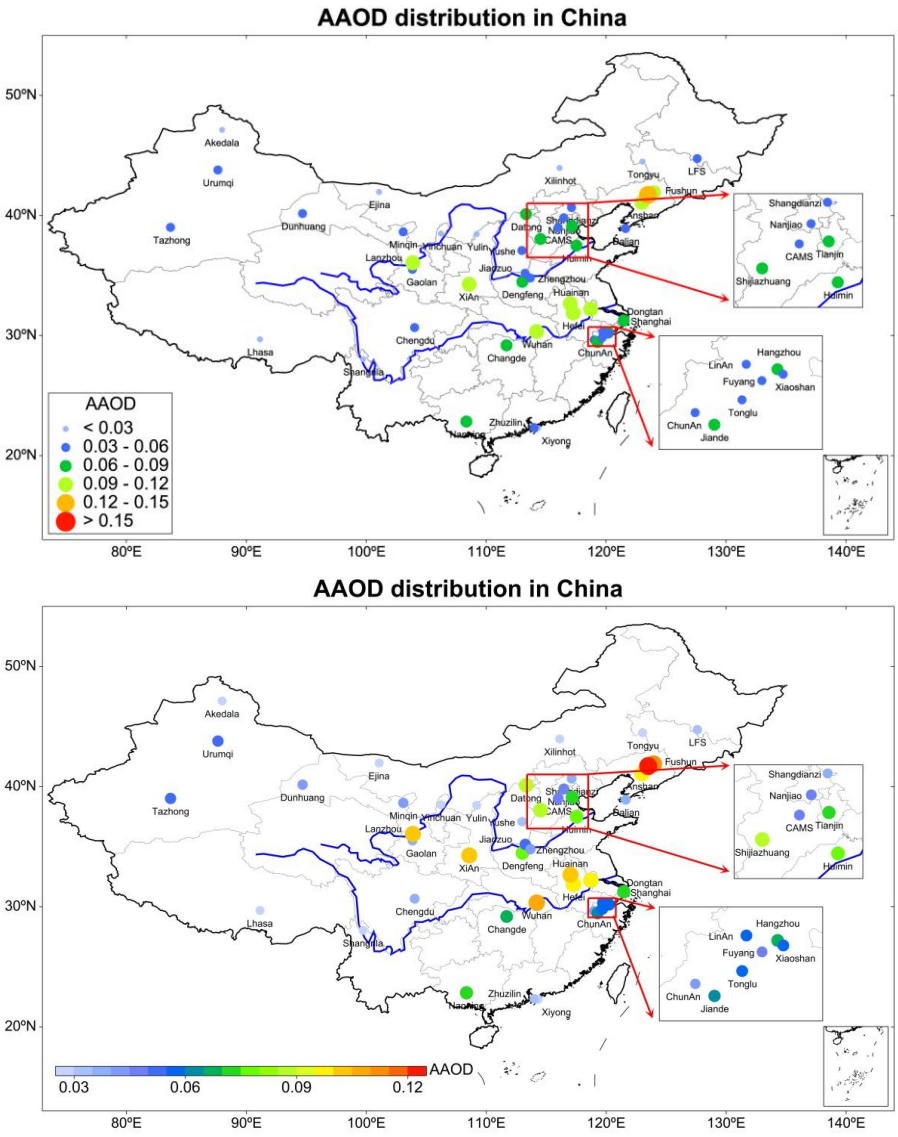
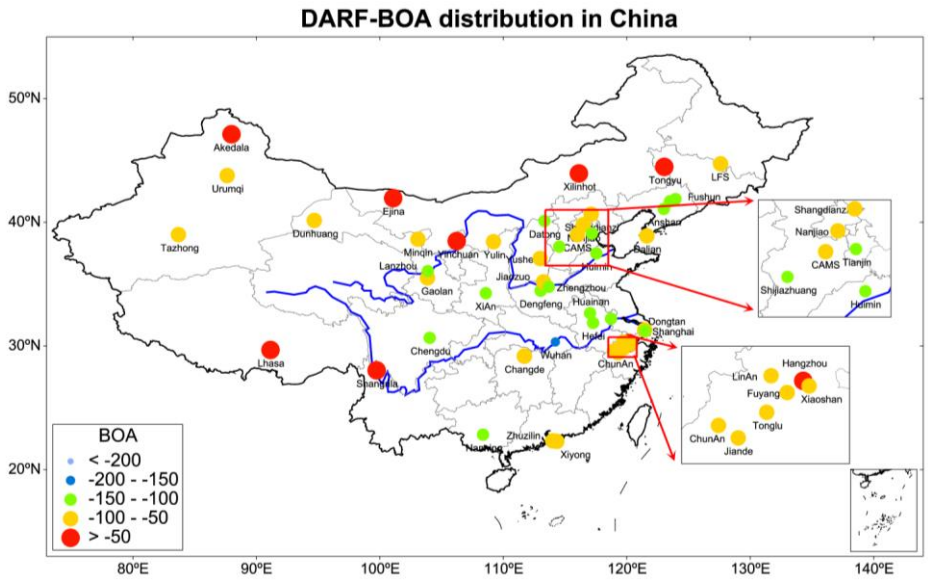


Figure 7. Annual spatial distribution of direct aerosol radiative effect at the bottom of the atmosphere at the CARSNET sites



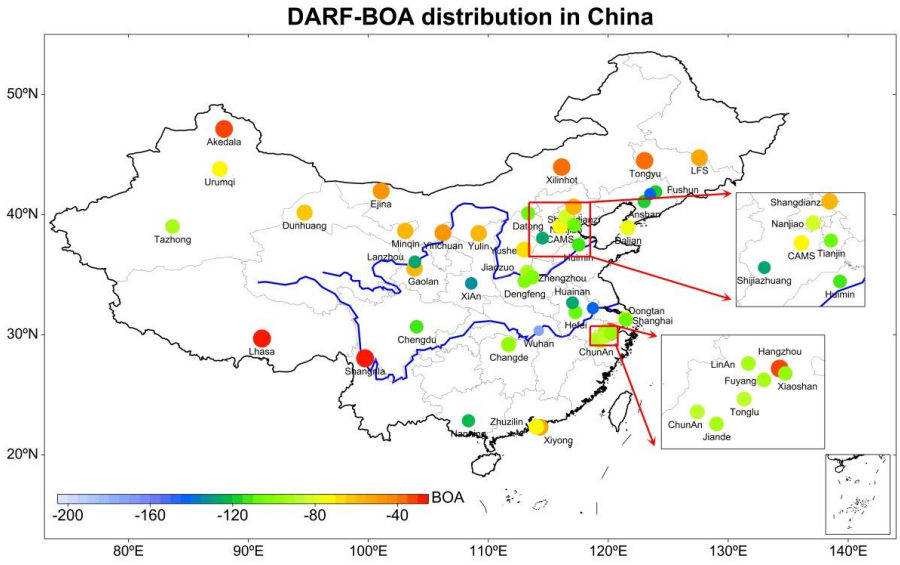
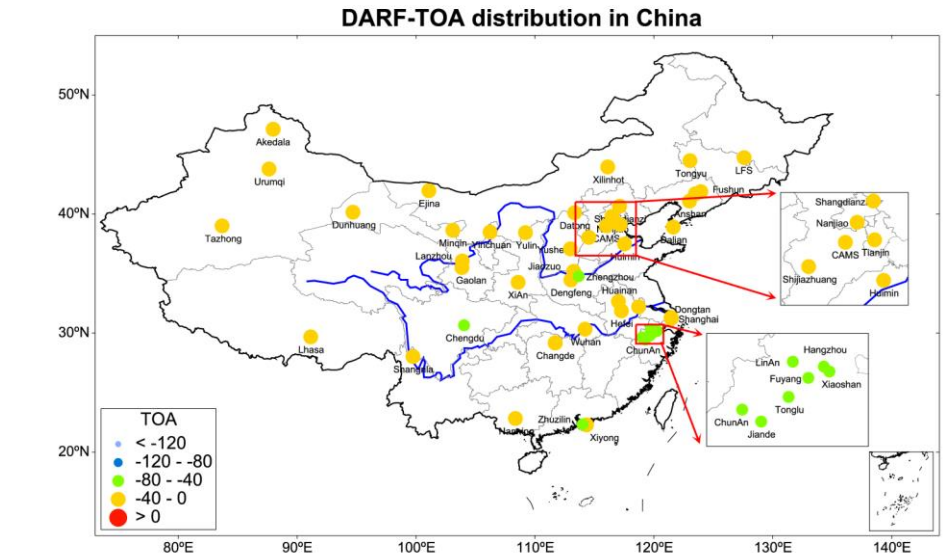


Figure 8. Annual spatial distribution of direct aerosol radiative effect at the top of the atmosphere at the CARSNET sites



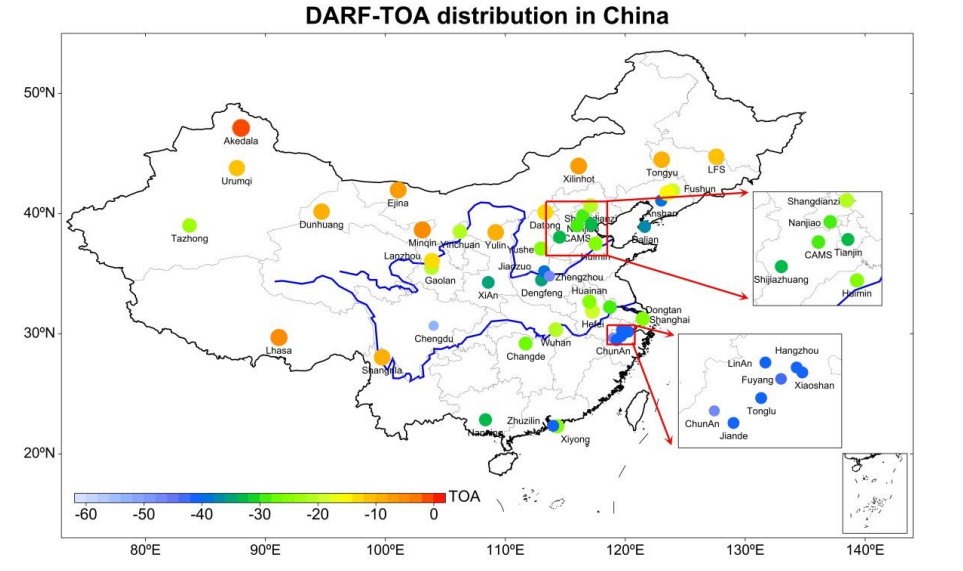


Figure 9. Annual spatial distribution of the aerosol type classification of types I–VII at the CARSNET sites

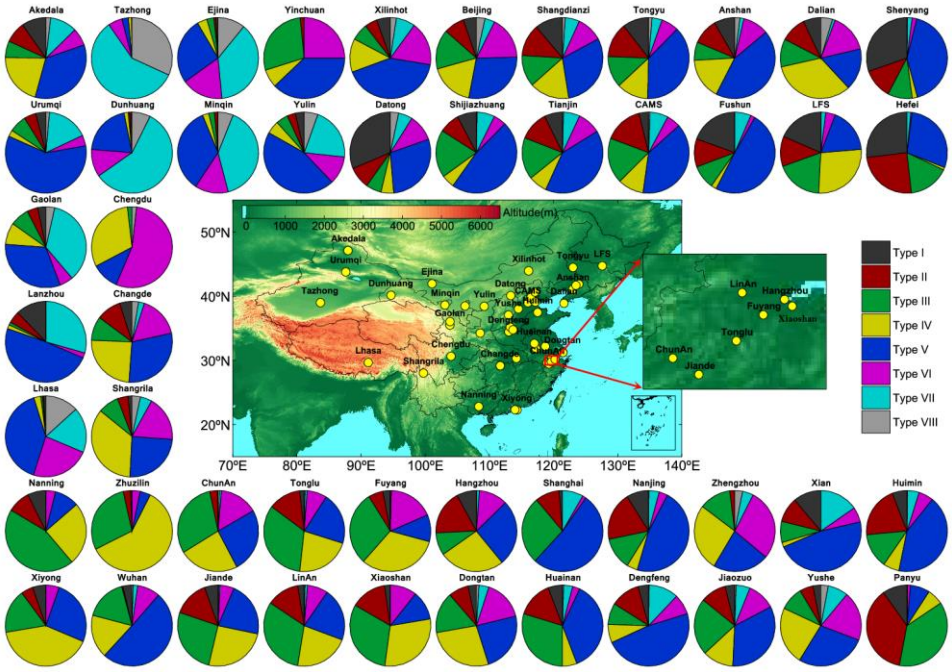


Table 1. The aerosol type classification based on the optical properties.

Type	EAE	SSA	absorbing properties and particle size
I	EAE > 1.20	$SSA_{440nm} \leqslant 0.85$	fine-mode particles with highly-absorbing
II	EAE > 1.20	$0.85 \leqslant SSA_{440nm} < 0.90$	fine-mode particles with moderately-absorbing
III	EAE > 1.20	$0.90 \leq SSA_{440nm} < 0.95$	fine-mode particles with slightly-absorbing
IV	EAE > 1.20	$SSA_{440nm} > 0.95$	fine-mode particles with weakly-absorbing
V	$0.60 \leq EAE < 1.20$	$SSA_{440nm} \leq 0.95$	mixed-absorbing particles
VI	$0.60 \leq EAE < 1.20$	$SSA_{440nm} > 0.95$	mixed-slightly absorbing particles
VII	$EAE \leq 0.60$	$SSA_{440nm} \leq 0.95$	coarse mode particles with strongly absorbing (mainly dust)
VIII	$AE \leq 0.60$	$SSA_{440nm} > 0.95$	coarse-mode particles with weakly-absorbing

Appendix

Table 1. Site information for the 50 CARSNET sites used in this study

No.	Site Name	Long.	Lat.	Alt.	Site information	Obs. Num	Period
					Remote sites (three sites)		
1	Akedala	47.12	87.97	562.0	55 km west of Fuhai county, Xinjiang province, and 250–300 km southeast of Kazakestan.	947	2010-2017
2	Lhasa	29.67	91.13	3663.0	In the center of Lhasa city, Qinghai-Xizang Plateau.	437	2012-2017
3	Shangri-La	28.02	99.73	3583.0	12 km northeast of Shangri-La county, Diqing area, Yunnan province	325	2013-2017
					Arid and semi-arid sites (six sites)		
4	Dunhuang	40.15	94.68	1139.0	1.5 km northeast of Dunhuang city, Gansu province; near Kumutage Desert of China	2030	2012-2017
5	Ejina	41.95	101.07	940.5	West ofIner-Mongolia Province, near Mongolia and Badanjilin desert	1970	2013-2017
6	Minqin	38.63	103.08	1367.0	In Minqin county, east to Tenggeli desert and north to Badanjilin Desert, Gansu Province	481	2013-2017
7	Tazhong	39.00	83.67	1099.4	In the middle of Takilamakan Desert, Xinjiang Province	1279	2013-2017
8	Xilinhot	43.95	116.12	1003.0	5 km southeast of Xilinhot City, near Hunshandake sand-land, Inner-Mongolia Province,	1464	2013-2017
9	Tongyu	44.42	122.87	151.0	In Tonyu city, west of Jilin Province	817	2010-2011
					Rural sites on the Chinese Loess Plateau or nearby (three sites)		
10	Mt.Gaolan	36.00	103.85	2161.6	5 km north of Lanzhou city in Gansu province	769	2015-2016
11	Yulin	38.43	109.20	1135.0	10 km north of Yulin city in Shaanxi province	716	2010-2016
12	Datong	40.10	113.33	1067.3	9 km of Datong City, but within area of rapid urbanization, Shanxi Province	914	2014-2017
					Rural sites in eastern China (15 sites)		
13	Changde	29.17	111.70	565.0	18 km northwest from Changde city, Hunan province.	344	2013-2016
14	Dongtan	31.52	121.96	10.0	In the Chongmin Island, 30km east of Shanghai city	986	2012-2016
15	ChunAn	29.61	119.05	171.4	151 km southwest from Hangzhou city, Zhejiang province.	1286	2011-2015
16	Huimin	37.48	117.53	11.7	100 km Northeast of Jinan City, Shandong Province	2243	2009-2017
17	Lin'an	30.30	119.73	138.6	150 km northeast of Shanghai, and 50 km west of Hangzhou city, Zhejiang province	1834	2011-2015
18	Mt.Longfeng	44.73	127.60	330.5	In Wuchang county, 175 km northeast of Harbin city, Heilongjiang Province	1515	2012-2016
19	Fuyang	30.07	119.95	17.0	44.1 km southwest from Hangzhou city, Zhejiang province.	710	2014-2015
20	Shangdianzi	40.65	117.12	293.0	In Miyun county, 150 km northeast to Beijing city.	1520	2014-2017
21	Yushe	37.07	112.98	1041.5	1.5 km east of Yushe city in Shanxi Province	1479	2013-2017
22	Dengfeng	34.46	113.02	350.0	75 km Southwest of Zhengzhou City, Henan Province	712	2013
23	Huainan	32.65	117.02	52.0	In the central of Hefei City, Anhui Province	794	2014-2015
24	Jiande	29.45	119.28	89.0	In the southwest from Hangzhou city, Zhejiang province.	1550	2011-2015
25	Tonglu	29.80	119.64	46.1	100 km northwest from Hangzhou city, Zhejiang province.	1717	2011-2015
26	Xiaoshan	30.16	120.25	14.0	In the south of Hangzhou city, Zhejiang province.	600	2014-2015
27	Xiyong	22.28	114.33	155.2	In the eastern of Shenzhen city, Guangdong province.	189	2016
					Urban sites (23 sites)		
28	Anshan	41.08	123.00	23.0	In Anshan city, central Liaoning province	193	2009-2013
29	Beijing-Nanjiao	39.80	116.47	31.3	In the southeast Beijing at city margin	1732	2014-2017
30	Beijing-CAMS	39.93	116.32	106.0	Chinese Academy of Meteorological Sciences in Beijing	1113	2012-2018
31	Chengdu	30.65	104.03	496.0	In Chengdu city, Sichuan province.	55	2014-2015
32	Dalian	38.90	121.63	91.5	Southeast coastal city in Liaoning Province	736	2012-2015
33	Fushun	41.88	123.95	80.0	In Fushun city, central Liaoning province.	231	2009-2013
34	Hangzhou	30.23	120.17	42.0	In Hangzhou city, Zhengjiang province.	1663	2011-2015

35	Hefei	31.98	116.38	92.0	In Hefei city, Anhui province.	197	2016
36	Jiaozuo	35.18	113.25	113.0	In the center of Jiaozuo city, Henan province.	981	2016-2017
37	Lanzhou	36.05	103.88	1517.3	In Lanzhou city, Gansu province.	1493	2013-2017
38	Nanjing	32.05	118.77	99.3	In Nanjing city, Jiangsu province	1258	2007-2015
39	Nanning	22.82	108.35	172.0	In Nanning city, Guangxi province	286	2013-2017
40	Panyu	23	113.35	145.0	In district of Guangzhou city, Guangdong Province	436	2012-2016
41	Shanghai	31.22	121.55	14.0	In Pudong district of Shanghai city	144	2016
42	Shenyang	41.77	123.50	60.0	In Shenyang city, central Liaoning province.	541	2009-2013
43	Tianjin	39.10	117.17	3.3	Northern coastal city in North China Plain	1705	2013-2017
44	Urumqi	43.78	87.62	935.0	In Urumuqi city, Xijiang province	1411	2012-2017
45	Xi'an	34.43	108.97	363.0	20 km north of center of Xian city, but within Jing RiverIndustrial District, Shaanxi province	652	2012-2016
46	Yinchuan	38.48	106.22	1111.5	In Yinchuan city, Ningxia province.	124	2017
47	Zhengzhou	34.78	113.68	99.0	In Zhengzhou city, Henan province.	1485	2013-2017
48	Shijiazhuang	38.03	114.53	75.0	In the center of Shijiazhuang city, Hebei province.	1178	2015-2017
49	Wuhan	30.32	114.21	30	In the center of Wuhan city, Hubei province	220	2008
50	Zhuzilin	22.32	114.00	63.0	In the central of Shenzhen city, Guangdong province.	915	2010-2017

Table 2. Annual data for aerosol microphysical properties, optical and direct radiative parameters

No.	Site	[a]ReffT	[a]ReffF	[a]ReffC	[a]VolT	[a]VolF	[a]VolC	[a]AODt	[b]Alpha	[a]FMF	[a]SSAT	[a]Image	[a]Real	[a]AAOD	[a]BOA	[a]TOA
Remote sites (3 sites)																
1	Akedala	0.36	0.14	2.45	0.06	0.02	0.04	0.17	1.13	0.81	0.90	0.0117	1.4540	0.02	-33.65	-0.42
2	Lhasa	0.64	0.13	2.26	0.05	0.01	0.04	0.10	0.77	0.66	0.90	0.0106	1.5541	0.01	-22.13	-5.04
3	Shangri-La	0.39	0.14	2.33	0.03	0.01	0.02	0.10	1.19	0.85	0.93	0.0086	1.4626	0.01	-17.43	-8.93
	Average	0.47	0.14	2.35	0.05	0.01	0.03	0.12	1.03	0.77	0.91	0.0103	1.4902	0.01	-24.40	-4.79
Arid and semi-arid sites (6 sites)																
4	Dunhuang	0.62	0.14	1.52	0.15	0.02	0.13	0.33	0.48	0.44	0.88	0.0103	1.5491	0.04	-63.61	-8.96
5	Ejina	0.56	0.14	1.78	0.11	0.02	0.09	0.24	0.64	0.52	0.89	0.0116	1.5265	0.03	-47.66	-7.20
6	Minqin	0.56	0.13	1.87	0.13	0.02	0.11	0.30	0.68	0.59	0.86	0.0145	1.5430	0.04	-59.83	-5.01
7	Tazhong	0.71	0.14	1.38	0.30	0.03	0.27	0.60	0.25	0.35	0.92	0.0054	1.5257	0.05	-91.20	-23.49
8	Xilinhot	0.48	0.13	2.45	0.08	0.02	0.05	0.21	1.03	0.78	0.89	0.0139	1.5183	0.02	-37.14	-7.47
9	Tongyu	0.39	0.13	2.36	0.07	0.02	0.05	0.23	1.16	0.82	0.88	0.0179	1.5377	0.03	-39.13	-8.87
	Average	0.55	0.14	1.89	0.14	0.02	0.12	0.32	0.71	0.58	0.89	0.0123	1.5334	0.03	-56.43	-10.17
Rural sites on the Chinese Loess Plateau or nearby (3 sites)																
10	Mt.Gaolan	0.58	0.14	2.03	0.16	0.03	0.13	0.36	0.81	0.64	0.89	0.0108	1.5154	0.04	-59.36	-20.87
11	Yulin	0.53	0.15	2.05	0.11	0.03	0.08	0.32	0.84	0.72	0.89	0.0122	1.5070	0.03	-56.81	-9.09
12	Datong	0.35	0.13	2.15	0.19	0.09	0.10	0.58	1.15	0.83	0.86	0.0171	1.4905	0.09	-107.86	-13.71
	Average	0.49	0.14	2.08	0.15	0.05	0.10	0.42	0.93	0.73	0.88	0.0134	1.5043	0.05	-74.67	-14.56
Rural sites in eastern China (15 sites)																
13	Changde	0.32	0.16	2.18	0.14	0.07	0.07	0.58	1.15	0.88	0.93	0.0101	1.4619	0.04	-75.33	-31.44
14	Dongtan	0.37	0.16	2.12	0.17	0.08	0.09	0.62	1.21	0.86	0.93	0.0080	1.4624	0.04	-79.41	-33.18
15	ChunAn	0.30	0.18	2.30	0.19	0.12	0.08	0.81	1.22	0.92	0.94	0.0066	1.4095	0.04	-86.49	-46.48
16	Huimin	0.36	0.15	2.07	0.22	0.10	0.12	0.83	1.14	0.86	0.89	0.0147	1.4852	0.08	-111.58	-25.49

17	Lin'an	0.29	0.17	2.24	0.21	0.12	0.09	0.87	1.29	0.91	0.93	0.0089	1.4172	0.06	-93.09	-41.73
18	Mt.Longfeng	0.28	0.15	2.44	0.08	0.04	0.04	0.34	1.38	0.90	0.89	0.0165	1.4647	0.03	-51.17	-11.34
19	Fuyang	0.29	0.17	2.28	0.21	0.13	0.09	0.89	1.31	0.92	0.94	0.0070	1.4147	0.05	-91.69	-42.29
20	Shangdianzi	0.40	0.15	2.33	0.12	0.05	0.07	0.43	1.17	0.86	0.89	0.0148	1.4840	0.04	-59.99	-20.58
21	Yushe	0.41	0.15	2.18	0.14	0.06	0.08	0.50	1.07	0.84	0.92	0.0090	1.4878	0.03	-66.72	-25.99
22	Dengfeng	0.39	0.15	2.03	0.23	0.09	0.13	0.79	1.02	0.83	0.89	0.0131	1.4782	0.08	-104.78	-35.84
23	Huainan	0.30	0.17	2.25	0.21	0.13	0.08	0.91	1.17	0.92	0.88	0.0166	1.4308	0.10	-129.17	-24.44
24	Jiande	0.29	0.17	2.18	0.20	0.12	0.08	0.84	1.34	0.91	0.92	0.0099	1.4085	0.06	-91.06	-40.07
25	Tonglu	0.29	0.17	2.20	0.20	0.12	0.08	0.83	1.31	0.91	0.93	0.0091	1.4269	0.06	-89.82	-41.28
26	Xiaoshan	0.28	0.17	2.24	0.22	0.13	0.09	0.87	1.35	0.91	0.93	0.0082	1.4134	0.06	-95.23	-40.39
27	Xiyong	0.33	0.16	2.43	0.11	0.06	0.05	0.41	1.32	0.89	0.94	0.0074	1.4072	0.02	-53.18	-25.45
	Average	0.33	0.16	2.23	0.18	0.09	0.08	0.70	1.23	0.89	0.92	0.0107	1.4435	0.05	-85.25	-32.40
							Urban sites (23 sites)									
28	Anshan	0.36	0.17	2.24	0.26	0.12	0.14	0.94	1.12	0.86	0.89	0.0158	1.4759	0.10	-117.99	-39.66
29	Beijing-Nanjiao	0.45	0.15	2.33	0.19	0.07	0.12	0.65	1.12	0.84	0.92	0.0100	1.4939	0.05	-82.06	-29.43
30	Beijing-CAMS	0.50	0.16	2.37	0.19	0.07	0.12	0.65	1.12	0.79	0.90	0.0115	1.5108	0.05	-72.66	-29.10
31	Chengdu	0.34	0.21	2.26	0.26	0.16	0.10	1.17	1.12	0.92	0.97	0.0033	1.4116	0.04	-110.42	-52.21
32	Dalian	0.35	0.16	2.24	0.16	0.08	0.09	0.62	1.22	0.87	0.93	0.0095	1.4584	0.04	-75.50	-37.42
33	Fushun	0.38	0.17	2.34	0.22	0.09	0.12	0.80	1.12	0.87	0.84	0.0244	1.4954	0.11	-116.91	-19.59
34	Hangzhou	0.30	0.17	2.21	0.22	0.12	0.10	0.87	1.30	0.90	0.91	0.0109	1.4337	0.07	-31.57	-40.16
35	Hefei	0.29	0.15	2.37	0.18	0.10	0.08	0.69	1.28	0.90	0.85	0.0195	1.4253	0.10	-105.83	-19.22
36	Jiaozuo	0.35	0.16	2.17	0.20	0.10	0.10	0.76	1.14	0.88	0.91	0.0105	1.4722	0.05	-92.29	-39.35
37	Lanzhou	0.54	0.14	2.04	0.28	0.06	0.22	0.66	0.81	0.66	0.83	0.0197	1.5193	0.10	-126.17	-13.81
38	Nanjing	0.33	0.16	2.16	0.25	0.12	0.12	0.94	1.13	0.88	0.88	0.0154	1.4446	0.10	-143.38	-28.29
39	Nanning	0.30	0.18	2.53	0.20	0.13	0.06	0.97	1.36	0.95	0.92	0.0107	1.4272	0.07	-121.92	-33.35
40	Panyu	0.26	0.16	2.29	0.16	0.10	0.06	0.69	1.43	0.93	0.90	0.0137	1.4155	0.07	-96.03	-26.56

41	Shanghai	0.40	0.15	1.93	0.19	0.08	0.11	0.68	1.10	0.84	0.88	0.0142	1.4814	0.07	-106.89	-24.34
42	Shenyang	0.31	0.16	2.23	0.22	0.12	0.10	0.89	1.20	0.90	0.84	0.0253	1.4589	0.14	-144.88	-15.02
43	Tianjin	0.42	0.16	2.26	0.23	0.10	0.13	0.83	1.11	0.86	0.89	0.0134	1.4957	0.07	-108.09	-33.26
44	Urumqi	0.48	0.14	2.14	0.15	0.04	0.10	0.42	0.93	0.75	0.85	0.0192	1.5371	0.05	-70.55	-11.74
45	Xi'an	0.37	0.16	1.85	0.26	0.11	0.15	0.98	0.98	0.82	0.88	0.0150	1.4888	0.10	-132.55	-35.93
46	Yinchuan	0.38	0.14	2.02	0.11	0.04	0.07	0.37	1.12	0.81	0.94	0.0054	1.4930	0.02	-48.67	-21.89
47	Zhengzhou	0.43	0.18	2.22	0.28	0.12	0.16	0.99	1.10	0.86	0.95	0.0045	1.4626	0.04	-101.10	-46.18
48	Shijiazhuang	0.40	0.16	2.28	0.26	0.12	0.14	0.95	1.09	0.87	0.88	0.0154	1.4754	0.09	-125.05	-33.66
49	Wuhan	0.34	0.17	2.22	0.22	0.12	0.10	1.00	1.16	0.91	0.88	0.0196	1.4779	0.11	-171.80	-20.40
50	Zhuzilin	0.27	0.17	2.45	0.15	0.09	0.05	0.66	1.45	0.94	0.96	0.0049	1.4438	0.03	-73.16	-40.65
	Average	0.37	0.16	2.22	0.21	0.10	0.11	0.79	1.15	0.86	0.90	0.0136	1.4695	0.07	-103.28	-30.05

Table 1 (Continued)

[a] Optical parameters at a wavelength of 440 nm.

[b] Angström exponents between 440 and 870 nm.