

Interactive comment on "Characteristics of ozone and particles in the near-surface atmosphere in urban area of the Yangtze River Delta, China" by Huimin Chen et al.

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To Editors and Anonymous Referee #2:

Dear editors and reviewers:

Thank you very much for dedicating time to reviewing the manuscript and providing us the important comments and suggestions on our study. We have learned a lot from your advice and made great efforts to improve the manuscript accordingly. A carefully point by point response to your comments has been listed below which we hope meet with approval. The revised details can be referred to the new version of the manuscript

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in the supplement.

Anonymous Referee #2 Received and published: 5 December 2018

A major revision of the MS must be made. Reconsideration of the MS is only possible pending the responses from the authors to the points listed below. The MS reports the observational data but barely digs enough into it, let alone a sufficient and reasonable discussion without conceptual mistakes. Moreover, the MS is not comfortably readable and lacks brevity. There are quite a few grammatical errors to be corrected. It would also be better if the language could be polished in the revision.

R:

We sincerely thanks for pointing out the problem of manuscript's analysis and writings. First, according to your suggestions, the authors conduct a more detailed analysis and discussion on the observational data. And we have also checked and corrected the all confusing statements in the manuscript. For example, we describe the similar role CO play in ozone production as volatile organic compounds (VOCs) and the criterion for VOC/NOx sensitive region in the revised manuscript after a comprehensive study of related work, making the deduction of the VOC-limited region through the CO-NOx-O3 correlation in this study more convincing. Also, for a sufficient use of observation data, like the aerosol optical properties, we have further analyzed the optical properties data to some extent for a better understanding of particle characteristics, such as its size and light extinction effects. It would contribute to the analysis of aerosols characteristics. Moreover, the manuscript has been rephrased significantly and shortened in necessarily throughout the whole text. Most parts of the manuscript have been shortened, especially for the Sections 2.2, 3.1, and 3.2. For example, the calculation of the aerosol optical properties and truncation correction of Aurora-3000 (Section 2.2), which have been stated clearly in previous studies (e.g., Zhuang et al., 2015, 2017; Anderson and Ogren, 1998; Müller et al., 2011, etc.) have been rephrased to a briefer but more legible version. More details could be found in the revised manuscript, and it's

believed that the revised version of the manuscript is much clearer and more readable.

With regard to your comments, questions and suggestions, the manuscript has been rephrased throughout the whole text. The finding(s) of this study have also been refined in better ways of expression, which could be found in most parts of the revised manuscript, including in the sections of Abstract, Introduction, Discussions, as well as Conclusion. Details can be found in the revised manuscript.

References:

Zhuang, B. L., Wang, T. J., Liu, J., Ma, Y., Yin, C. Q., Li, S., Xie, M., Han, Y., Zhu, J. L., Yang, X. Q., Fu, C. B.: Absorption coefficient of urban aerosol in Nanjing, west Yangtze River Delta, China, Atmos. Chem. Phys., 15, 13633–13646, doi:10.5194/acp-15-13633-2015, 2015.

Zhuang, B. L., Wang, T. J., Liu, J., Li, S., Xie, M., Han, Y., Chen, P. L., Hu, Q. D., Yang X.Q., Fu, C. B., Zhu, J. L.: The surface aerosol optical properties in the urban area of Nanjing, west GTH River Delta, China. Atmos. Chem. Phys., 17, 1143–1160, doi:10.5194/acp-17-1143-2017, 2017.

Anderson, T. L., Ogren, J. A.: Determining aerosol radiative properties using the TSI 3563 integrating nephelometer, Aerosol Sci. Tech., 29, 57–69, 1998.

Müller, T., Laborde, M., Kassell, G., Wiedensohler, A.: Design and performance of a three-wavelength LED-based total scatter and backscatter integrating nephelometer, Atmos. Meas. Tech., 4, 1291–1303, doi:10.5194/amt-4-1291-2011, 2011.

Main points:

1. The structure of the introduction apparently lacks logic organization. Even more, major scientific issues the MS to be addressed are not clearly stated.

R:

According to your suggestions, the structure of the introduction has been reorganized.

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It is believed to be more readable and easier for readers to grasp the major scientific issues of the study. Please refer to the revised manuscript for more details.

2. A detailed description of the environment where all instruments are installed should be given in section 2.1. How about the drying system upstream AE-31 and Aurora-3000? The instruments used to measure trace gases should be at least briefly described, instead of having not even a single word on that.

R:

Thank you for your suggestions and question. Section 2.1 has been extended to degrees. Description of the environment where all instruments are installed has been included in the revised manuscript. For the drying system, there is no heater for AE-31, which is similar to the settings in other sites (e.g., Wu et al., 2012; Wu et al., 2013; Gong et al., 2015, etc.). Both external and internal heaters are equipped for Aurora-3000. However, the internal heater has been turned off during the study period because RH in the tube is mostly lower than 50% in this period. Corresponding statements on settings of AE-31 and Aurora-3000 have also been included in the revised manuscript. For the instruments of the trace gases, more detailed description can be found in the revised manuscript.

References:

Wu, Y. F., Zhang, R. J., Pu, Y. F., Zhang, L. M., Ho, K. F., Fu, C.B.: Aerosol optical properties observed at a semi-arid rural site in northeastern China, Aerosol Air Qual. Res., 12, 503–514, 2012.

Wu, D., Wu, C., Liao, B., Chen, H., Wu, M., Li, F., Tan, H., Deng, T., Li, H., Jiang, D., Yu, J. Z.: Black carbon over the South China Sea and in various continental locations in South China, Atmos. Chem. Phys., 13, 12257–12270, doi:10.5194/acp-13-12257-2013, 2013.

Gong, W., Zhang, M., Han, G., Ma, X., Zhu, Z.: An investigation of aerosol scattering

and absorption properties in Wuhan, Central China, Atmosphere, 6, 503-520, 2015

3. The SC2006 is adopted in this study to correct the systematic biases inherent in the principle of AE-31. What are the parameters used in your procedure? How about the values of your correction factors? The description needs to be more specific.

R:

Thanks for your question. Previous investigation indicated that both Weingartner corrected (WC2003 for short, hereinafter) and Schmid corrected (SC2006 for short, hereinafter) absorptions show good agreements with the one from the Multi-Angle Absorption Photometer (Collaud Coen et al., 2010). Therefore, we have applied several correction algorithms to calculate the aerosol absorption coefficient according to SC2006, WC2003, and indirect calculation (IDC). And the aerosol optical properties and certain parameters used in the correction procedures are based on our observation data and previous work (Wu et al., 2009; Wu et al., 2013). Results showed that corrected σ a at 532 nm is consistent with each other among WC2003, SC2006 and IDC. However, the absorption Ångström exponent from SC2006 might be closer to the real ones than WC2003s as suggested in Zhuang et al. (2015). Therefore, the SC2006 is adopted in this study.

The parameters in the correction procedure are derived from the local optical properties (ω 0 and α s were set to 0.922 and 1.51, respectively). The values of correction factors C and R are as follows: R=1 when ATN \leq 10 and f=1.2, and C in Nanjing is 2.95, 3.37, 3.56, 3.79, 3.99, 4.51 and 4.64 at 370, 470, 520, 590, 660, 880 and 950 nm. Detailed procedures of the calculations could be referred to Zhuang et al. (2015). Relatively in-depth description has been added in Section 2.2 in the revised manuscript.

References:

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J. S., Jennings, S. G., Moerman, M., Petzold, A., Schmid, O., Bal-

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tensperger, U.: Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms, Atmos. Meas. Tech. 2010, 3, 457–474.

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4. The truncation correction of Aurora-3000 is based on Mie calculations. If I understand it correctly, Mie calculation is not performed in this study, since obviously there is no measurement of particle number size distribution. Instead, correction parameters are directly taken from the literature in this study. How much uncertainty might be introduced to scattering coefficients due to the choice of the correction factors?

R:

Thank you for your question. Mie calculation is not performed due to the lack of measurement of particle number size distribution at the site. However, in Müller et al. (2011), it is pointed out that the calculation performed for Aurora-3000 in the study is accurate for a wide range of atmospheric aerosols, and the correction parameters have been used for correction in previous studies (e.g., Virkkula et al., 2015; Gong et al., 2015; Perrone et al., 2014; Pandolfi et al., 2014, etc.). For the single scattering albedo larger than 0.8, the uncertainty of the correction is not expected to be larger than 3 % (Bond et al., 2009). Here in our study, over 99% of the single scattering albedo is larger than 0.8, thus the uncertainty is about $2\% \sim 3\%$, which could meet the precision requirements to degrees.

References:

Müller, T., Laborde, M., Kassell, G., Wiedensohler, A.: Design and performance of a three-wavelength LED-based total scatter and backscatter integrating nephelometer, Atmos. Meas. Tech., 4, 1291–1303, doi:10.5194/amt-4-1291-2011, 2011.

Virkkula, A., Chi, X., Ding, A.J., Shen, Y., Nie, W., Qi, X., Zheng, L., Huang, X., Xie, Y., Wang, J., Petäjä, T., Kulmala, M.: On the interpretation of the loading correction of the aethalometer. Atmospheric Measurement Techniques,8,10(2015-10-21), 2015, 8(8), 7373-7411.

Gong, W., Zhang, M., Han, G., Ma, X., Zhu, Z.: An Investigation of Aerosol Scattering and Absorption Properties in Wuhan, Central China. Atmosphere, 2015, 6, 503–520.

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Pandolfi, M., Ripoll, A., Querol, X., Alastuey, A.: Climatology of aerosol optical properties and black carbon mass absorption cross section at a remote high-altitude site in the western mediterranean basin. Atmospheric Chemistry and Physics, 2014, 14(12), 6443-6460.

Bond, T. C., Covert, D. S., Müller, T.: Truncation and Angular-Scattering Corrections for Absorbing Aerosol in the TSI 3563 Nephelometer, Aerosol Sci. Tech., 2009, 43, 866–871.

5. HYSPLIT model is driven by NCEP data with a temporal resolution of 6 hours and a spatial resolution of 2.5 degrees in this study. I doubt that the resolution is adequate

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for carrying a simulation of near surface transport process.

R:

Thanks for your comments. We have checked the data for driving HYS-PLIT model according to your comments. Instead of the NCAP data, HYSPLIT model is driven by GDAS (Ground Data Acquisition System, ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1) data with a temporal resolution of 6 hours and a spatial resolution of 1.0 degrees in our study, which is thought to be adequate enough for carrying a simulation of the transport process (e.g., Rolph et al., 2014; Su et al., 2015; Guo et al., 2015, etc.).

References:

Rolph, G.D., Ngan, F., Draxler, R.R.: Modeling the fallout from stabilized nuclear clouds using the HYSPLIT atmospheric dispersion model. Journal of Environmental Radioac-tivity, 2014, 136:41-55.

Su, L., Yuan, Z.B., Fung, J., Lau, A.: A comparison of HYSPLIT backward trajectories generated from two GDAS datasets. Sci. Total Environ, 2015, 506-507:527-537.

Guo, Z.B., Jiang, W.J., Chen, S.L., Sun, D.L., Shi, L., Zeng, G., Rui, M.L.: Stable isotopic compositions of elemental carbon in PM1.1 in north suburb of Nanjing Region, China. Atmospheric Research, 2015, 168:105-111.

6. Page 14, Line 283-284, particles especially sub-micron particles could hardly be removed from the atmosphere by rain droplets. It is strong wind before the rain that sweeps them out.

R:

Thank you for your comments. We agree with you that strong wind would also play a significant role in removing particles. With regard to your comment, the wind speed is further investigated during the study period. The meteorological data is downloaded

from National Climate Data Center (ftp://ftp.ncdc.noaa.gov/pub/data/noaa/) and Wunderground Global Weather Precision Forecast (www.wunderground.com), which has also been used in other researches (e.g., Bolling et al., 2005; Wang et al., 2006; Pokharel and Kaplan, 2017; Huang et al., 2018, etc.) . Results indicate that the monthly averages of daily wind speeds are relatively close to each other in fall and winter, ranging from 1.1 (February) to 1.8 m/s (November), whereas the aerosol concentrations are not. For example, the average of PM2.5 concentration in October was 35.8 μ g/m³ but 42.5 μ g/m³ in September, which increased roughly 20% in a month. The monthly average wind speed, however, increased only 0.1 m/s from September to October. And the mean precipitation increased from 2.3 mm/h in September to 3.1 mm/h in October correspondingly. Considering the emission rates of these two months are relatively close (emission in October is a little bit stronger according to Zhang et al., 2006), it is suggested that a higher precipitation in October thus with a larger scavenging efficiency might have larger contribution to small concentrations of particles.

References:

Bolling, B. G., Kennedy, J. H., and Zimmerman, E. G. (2005). Seasonal dynamics of four potential West Nile vector species in north-central Texas., Journal of Vector Ecology, 2005, 30(2), 186-194. Wang, Y., Zhuang, G.S., Zhang, X.Y., Huang, K., Xu, C., Tang, A.H., Chen, J.M., An, Z.S.: The ion chemistry, seasonal cycle, and sources of PM2.5 and TSP aerosol in Shanghai. Atmospheric Environment, 2006, 40(16):2935-2952.

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Zhang, Q., Streets, D.G., Carmichael, G.R., He, K.B., Huo, H., Kannari, A., Klimont, Z.,

Park, I.S., Reddy, S., Fu, J.S., Chen, D., Duan, L., Lei, Y., Wang, L.T., Yao, Z.L., 2009. Asian emissions in 2006 for the NASA INTEX-B mission. Atmos. Chem. Phys.2009, 9(14): 5131–5153.

7. Page 18, Line 385-386, the author states that the diurnal pattern of NOx is mainly governed by photochemical processes and meteorology. However, emission is a key factor that should not be ignored.

R:

Thanks again. Emission has been stated as a key factor in the current version. The sentence has been rephrased in the revised manuscript.

8. Page 22, Line 454-460, the concept about fog is completely wrong. Fog only occurs above 100% RH, though droplets can exist below 100% due to the hygroscopic growth of particles.

R:

Thank you for your advice. We agree with you that fog generally refers to a weather phenomenon created by the condensation of water vapor when the relative humidity approaches 100% (saturated) with the visibility less than 1 km, and we have rephrased the sentence in the revised manuscript.

9. Page 22, Line 467-471, the existence of aerosols might affect solar radiation to some extent and thus ozone photochemical production (not always to a measurable amount). However, the main reason for the observed variation of ozone should not be attributed to aerosols. The author tempts to build a relationship between aerosols and ozone, but I find the analysis of data and deduction not robust and even incorrect, just like here and discussions elsewhere in the MS, e.g, Page 24, Line 503-504.

R:

Thanks for the comments.

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Firstly, the existence of particulates could affect ozone photochemical production because particulates could inhibit the photolysis reactions near the surface in reducing the photolysis frequencies in the atmosphere, which would result in the decrease of O3 concentrations near the ground. In our study, the negative correlation between particulates and O3 coincides with the above assumption, which was also found in various numerical models (e.g., Li et al., 2005; Bian et al., 2007; Deng et al., 2010; Li et al., 2011; Li et al., 2018, etc.). Most of the simulated results above showed an obvious change in the amount of ozone concentration and production due to aerosols. For example, Bian et al. (2007) reported the ratio of Δ [O3]/ Δ [AOD] ranged from -4 \sim -16 ppb in Tianjin, and in Li et al. (2011), aerosols decreased the average O3 \rightarrow O(1D) photolysis frequency by 53%, 37% and 21% in the lower, middle and upper troposphere in central east China, and as implied in Li et al. (2017), high concentrations of aerosols result in a 0.1 \sim 5.0 ppb (12.0%) reduction of near-surface ozone in central Nanjing.

Besides, we agree with you that the main reasons for the observed variation of ozone might be attributed to the effects of radiation, concentrations of precursors, other weather conditions, etc. And we have taken the effects above into consideration when discussing the ozone variation. For example, in Section 3.2, the discussion of ozone temporal variation contains the influence of radiation, precursor concentrations, as well as the meteorology field. And to make a better insight of the correlation and interaction between particles and ozone through observation data, this study further identifies the influence of associated affecting factors, including UV radiation, temperature, and precursors (NOx, NOy, and CO) concentrations, on the interaction (Section 3.3). For a more comprehensive overview, we not only analyze the correlation between particulates and ozone but also the one between particulates and the precursor (NOx and CO). It is found that particles (PM2.5 and BC) are well-correlated with precursors (NOx and CO), which could be another possible reason for the negative correlation between aerosols and ozone. In our study, we have discussed the abovementioned possible reasons for the correlation thoroughly, instead of just laying emphasis on the impact of aerosols on the ozone photochemical production in the revised manuscript. Thus, the

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main points of our analysis and discussions is to propose the possible reasons for the effects of aerosols on ozone concentration (by influencing the radiation and the precursors concentrations) based on the observation data, rather than regard aerosols as a decisive factor of the observed ozone variation.

As for the analysis of data and deduction, according to your suggestion, Section 3.3 has been extended to degrees. More in-depth discussions on the aerosol classification and identification have been included in the current version. More details can be found in the revised manuscript.

References:

Li, G. H., Zhang, R. Y., Fan, J. W.: Impacts of black carbon aerosol on photolysis and ozone. Journal of Geophysical Research., 2005, 110(D23206).

Bian, H., Han, S., Tie, X., Sun, M., Liu, A.: Evidence of impact of aerosols on surface ozone concentration in Tianjin, China. Atmospheric Environment, 2007, 41, 4672-4681.

Deng, J., Wang, T., Liu, L., Jiang, F.: Modeling heterogeneous chemical processes on aerosol surface. Particuology, 2010, 8(4):308-318.

Li, J., Wang, Z., Wang, X., Yamaji, K., Takigawa, M., Kanaya, Y., Pochanart, P., Liu, Y., Irie, H., Hu, B., Tanimoto, H., Akimoto, H.: Impacts of aerosols on summertime tropospheric photolysis frequencies and photochemistry over Central Eastern China. Atmospheric Environment, 2011, 45(10):1817-1829.

Li, M., Wang, T., Xie, M., Li, S., Zhuang, B., Chen, P.: Agricultural fire impacts on ozone photochemistry over the Yangtze River Delta region, East China. Journal of Geophysical Research: Atmospheres, 2018.

Li, M., Wang, T., Xie, M., Li, S., Zhuang, B., Chen, P.: Impacts of aerosol-radiation feedback on local air quality during a severe haze episode in Nanjing megacity, eastern China, Tellus B: Chemical and Physical Meteorology, 2017, 69(1):1339548.

10. Page 23, Line 491-499, the author draws a conclusion that ozone photochemical production is VOC-limited by using CO/particle-O3-NOx relationship. I find it very unconvincing. R: Thank you for your comments.

In our study, we have no VOCs measurement, thus CO is chosen as the reference tracer, which is similar to other studies (e.g., Hsu et al., 2010; Shao et al., 2011; Yao et al., 2012, etc.). First of all, the measured mixing ratios of CO showed significant correlations with the measured levels of most anthropogenic VOCs, which has been verified in many previous studies (e.g., Baker et al., 2008; von Schneidemesser et al., 2010; Wang et al., 2014, etc.). In addition, as a significant precursor of ozone, CO also plays a similar role as VOCs. HO2 produced from oxidation of CO initiates photochemical reactions which result in the net formation of O3 (Novelli et al., 1998; Atkinson et al., 2000; Gao et al., 2005).

If O3 formation is under VOC-sensitive regime, the reduction of NOx will lead to increase in O3 concentrations, which is used for observation data to determine the ozone photochemical production in the region is VOC-limited or NOx-limited (Geng et al., 2008; Ding et al., 2013b). It is found that an increase of CO, as well as PM2.5 and BC, always results in higher O3 concentration for NOx lower than 40 ppb, while NOx reverses. To be specific, when NOx reduces for CO lower than 1500 ppb, O3 has a sharp increase. Also, an increase in the CO level would lead to an in increase in the O3 concentration, especially when NOx is lower than 40 ppb. Therefore, we suggest that the region is VOC-sensitive.

References:

Hsu, Y. K., VanCuren, T., Park, S., Jakober, C., Herner, J., FitzGibbon, M., Blake, D. R., Parrish, D. D.: Methane emissions inventory verification in southern California, Atmospheric Environment, 2010, 44(1):1-7.

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von Schneidemesser, E., Monks, P. S., and Plass-Duelmer, C.: Global comparison of VOC and CO observations in urban areas, Atmos. Environ., 2010, 44(39): 5053–5064.

Wang, M., Shao, M., Chen, W., Yuan, B., Lu, S., Zhang, Q., Zeng, L., Wang, Q.: A temporally and spatially resolved validation of emission inventories by measurements of ambient volatile organic compounds in Beijing, China. Atmos. Chem. Phys., 2014, 14(12): 5871–5891.

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western Yangtze River Delta: an overview of 1 yr data at the SORPEs station, Atmos. Chem. Phys., 2013, 13, 5813–5830.

Minor points:

1. The full form should be given for the abbreviation of BTH in the abstract.

R:

Thanks for your advice. The full form for the abbreviation of BTH has been given in the revised abstract.

2. The abbreviation of several aerosol optical parameters such as AAC and SC is not common. It would be better to follow the convention in the community.

R:

Thanks again for your suggestion. According to your suggestion, in the revised manuscript, aerosol optical parameters, including scattering (SC), back-scattering (Bsp), absorption (AAC), and extinction (EC) coefficient, scattering (SAE) and absorbing (AAE) Ångström exponent, asymmetry parameter (ASP), and single-scattering albedo (SSA), have been adapted to σ ts, σ bs, σ a, σ e, α ts, α a, g, and ω 0, respectively (e.g., Hess, 1998; Andreae et al., 2008; Müller et al., 2011, etc.).

References:

Hess, M.: Optical properties of aerosols and clouds: The software package OPAC. Bull. Am. Meteor. Soc. 1998, 79(5):831-844.

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Atmos. Meas. Tech., , 2011, 4, 1291–1303.

3. Page 13, Line 270-274, the sentence beginning with 'For example' is supposed to illustrate the point brought forward by the sentence before it. However, I find their connection rather confusing.

R:

Thank you for pointing out the problem. This part has been rephrased to be more intelligible in the revised manuscript.

Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2018-927/acp-2018-927-AC2supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-927, 2018.