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Atmospheric Chemistry and Physics Discussions

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.

Huimin Chen et al.

blzhuang@nju.edu.cn

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To Editors and Anonymous Referee #3:

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

in the supplement.

Anonymous Referee #3 Received and published: 21 December 2018

The paper written by Chen et al., performed the continuous measurements of particles and trace gases in Nanjing during cold seasons. Although the interaction of atmospheric components (e.g., trace gases, aerosols) and meteorological conditions has been analyzed, the originality should be addressed especially in abstract before publication. Besides, the paper still suffered from many minor flaws throughout the manuscript. Thus, I suggest this paper could be published after revising the minor errors.

The detailed suggestions are as follows:

1. It was well documented that the air pollutants were closely linked to the weather system and meteorological conditions. (Line 32) The author only revealed the important effects of weather system and human activities on the environment in the YRD region, which has been investigated by many previous studies. The originality was not addressed in the manuscript. In my opinion, the abstract should be rewritten to stress the new contribution of this paper to atmospheric chemistry rather than reporting the pollution level simply.

R:

We sincerely thanks for your comments. In the revised manuscript, the authors stress the originality of the study.

1. Indeed, some researches on the air pollutants related to weather system and human activities have been carried out in most sites of YRD recently. However, previous studies using observation data in Nanjing often concentrated on characteristics of one of the particles, such as BC and carbonaceous aerosols (e.g., Zhuang et al., 2014), or PMs (e.g., Deng et al., 2011; Shen et al., 2014), or ozone and its precursors (e.g., Tu et al., 2007; Wang et al., 2008; An et al., 2015). Thus, it is necessary to achieve

a relatively comprehensive understanding of the air pollution problem directly through analysis of various species. In addition, most of them described the temporal and spatial distributions of concentrations, and the influence of meteorological effects. In this study, discussion of aerosols characteristics, especially particles, is not limited to the concentrations but taking optical properties into consideration as well. Moreover, most of them lay less emphasis on the inter-species correlations and the combined effects of more than one pollutant, especially the possible underlying chemical progress, during severe pollution episodes except Ding et al. (2013b), who described the characteristics of O3 and PM2.5 with near-surface observation data in rural area of Nanjing. As implied in Zhang et al. (2012), aerosols are complicated in compositions and spatial distributions especially in fast developing regions with intense human activities (such as Nanjing). Thus, differences of the aerosol characteristics, for instance, concentrations as well as optical properties, might exist to degrees among the sites located in different parts of Nanjing with different land use. Additionally, a better understanding of spatial and temporal variations of pollutants can contribute to the adoption of effective measures to reduce air pollution on the urban scale. Therefore, it's necessary to investigate the characteristics of air pollutants in urban area of west YRD.

2. To make a better insight of the correlation and interaction between particles and ozone (the two main pollutants) through observation data, this study further identifies the influence of associated affecting factors, including UV radiation, temperature, and precursor's concentrations on the interaction (Section 3.3). Most of previous studies present the findings from 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.). However, only a few studies discussed the correlation based on observation. In Nanjing, Ding et al. (2013b) described a correlation between PM2.5 and O3. But only temperature is regarded as an affecting factor. Thus, it is believed that our study would contribute to a more comprehensive understanding of the underlying mechanisms from observation.

3. Back to the site, the site is located in the city center, one of the highly residential

areas of Nanjing, with concentrated human activities with residential areas, schools, institutions and business districts, and the main road of urban transportation around. Therefore, the results could suggest the characteristics and interactions of pollutants in the urban region very well. Also, the results could further imply the effects of the urban underlying surface and human activities to degrees. Besides, as a typical urban area, the results in this study would probably bring new knowledge of aerosol characteristics, like the pollution level variation in different years and different regions through comparison with previous studies based on observation and numerical simulations.

Overall, this manuscript presents more comprehensive, systematic and deeper analysis on main pollutants like particles and ozone in urban area of west YRD. Results further indicate the characteristics of the particles and trace gases and reveal the possible chemistry process and interactions among different species and meteorological variables in west YRD. And they are also advantageous to improve the understanding of the detailed variations (seasonal, monthly, and diurnal) and its effects in east regions of China.

According to your comments, questions and suggestions, not only the abstract, but the introduction, discussion and conclusion have also been rephrased carefully. The originality (listed above) and finding(s) of this study have been refined in better ways of expression. Details can be found in the revised manuscript.

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2. Line 71, the author said observation-based studies of particles were relatively limited. I think it was very subjective because there were hundreds of observation-based studies about the aerosol particles in the past decades. Meanwhile, in line 75, the author said there were only very limited studies of O3 in the urban of YRD. Actually, the O3 concentration has been widely monitored in YRD because it was one of the most important gaseous pollutants in YRD. I think the author should review a large amount of papers before writing this paper.

R:

Thanks for your suggestions. We have reviewed more papers and refined the expression in the introduction, making it more integrated. More details could be found in the revised version.

3. Line 108-112, the author should highlight the objective of the present study. In addition, the sentence between line 110 and line 112 should be replaced by the environmental implication of the research.

R:

Thank you for your advice. The sentences have been rewritten to highlight the objective of our study, and readers can find more details in the revised manuscript. Generally speaking, the emphasis of our objective is to improve the insight in the characteristics, interactions of main pollutants, and the influence of integrated meteorology variables based on the observation data at an urban site above ground, and further investigate the possible underlying reasons and mechanisms, which is also helpful to achieve a thorough understanding of particles and trace gases pollution in these polluted areas by just using the conventional observations

The detailed environmental implication of the research could be concluded as follows.

1. An in-depth discussion on particles variations is performed, not limited to the concentrations but taking optical properties into consideration as well, to quantify the polluted level in detail to receive an overview of the inter-annual variations in the urban region.

2. A detailed description of O3 variations can also be found in our study, including the analysis of the main precursors as trace gases (NOx, NOy and CO), to have a general and quantitative insight in O3 pollution situations. Both of the pollutants are analyzed considering the effects of meteorology variables including but not limited to precipitation and temperature.

3. Analysis of inter-species correlations gives a relatively thorough overview of the interactions among various species, for instance, O3 and particles (BC and PM2.5), O3 and precursors (NOx and CO), and particles (BC and PM2.5) and precursors (NOx and CO). For a better insight, this study further identifies the influence of associated affecting factors on the interaction, such as UV radiation and temperature. Deduction of the underlying chemical mechanisms and process based on the results and previous studies is also presented in our study.

4. Backward trajectories analysis is conducted for improving the knowledge of regional/sub-regional transport process in urban Nanjing. Discussion of pollutants in different clusters suggests main transport progress in each season and the effects of air masses coming from various regions.

5. A case study for high particles and O3 episode is implemented to emphasize the integrated influence of meteorology fields on regional air pollution.

4. Line 123, the instruments used to monitor the gaseous pollutants such as O3 should be added in the methods. Additionally, NOy generally consisted of a large of N-bearing gaseous pollutants. The detailed NOy species should be introduced in this part.

R:

Thank you for your suggestions. In the revised version, the detailed description of instruments measuring trace gases (CO, NOx, NOy and O3) and the measurement of NOy species (where NOy = NO + NO2 + PAN + HNO3 + NO3- + N2O5 + HONO + organic nitrates, etc.) has been added in the Section 2.1.

5. Line 263-264, the author did not show the variation trend of BC, PM10, and PM2.5. Furthermore, how do you know the sources of these pollutants shared the similar sources? The relevant references were also missing. Line 265, what does transport emission mean?

R:

Thanks for your questions. We agree with you that the statement of the similar pattern without the variation trend is not acceptable enough, thus, the expression has been rephrased to be more precise in the revised manuscript.

Chow et al. (2011) reported a wide range of EC and OC abundances (highly correlated with BC) in PM2.5 source profiles, which represented the same source type. Zhuang et al. (2014) also reported same emission sources for BC and PM2.5 in Nanjing based on the well-correlated relationship, which is also found in this study (R=0.75). Wang et al.

(2008) suggested that the major constituents of PM2.5 were black carbon (BC), cluster elements, nitrates, ammonium salts, and sulfates based on the long-term monitoring data, and PM2.5 and PM10 share various common sources, i.e., soil dust, coal combustion, industrial emission, and biomass burning in Beijing. Schlemicher et al. (2013) also reported quite similar main sources for both particle size classes. Moreover, it is found in Gong et al. (2013) that PM2.5 is one of the major contributors to PM10 with a good correlation in Wuhan, especially when the concentration of PM10 is not extremely high. Thus, we assume that these pollutants possibly shared the similar sources.

Relevant references have been added in the revised manuscript. As for the second question, the sentence has been rewritten to be clearer.

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sphere, 2015, 6(9): 1377-1387.

6. Line 272-274, the author said the high loadings of particulate matter in early October was mainly due to the increase in aerosol concentrations with high scatter coefficient (SC). I do not understand the association between PM concentration and the aerosol concentrations with high SC. Please explain the reasons in details.

R:

Thanks again for your advice. The high concentrations of PMs in early October are possibly resulted from the increase in scattering aerosols. First of all, scatter coefficient is high during that period. Besides, BC, as one of typical absorbing aerosols, does not show such peak in concentrations during that period. The statement has been rephrased and corresponding reasons have also been discussed in the revised manuscript.

7. Line 284-286, Nanjing is located in Southeast China. The combustion of fossil fuels for domestic heating is not common in the winter of Nanjing. I do not understand why the increased anthropogenic emission of fossil fuels in the winter of Nanjing contributed to the high aerosol loadings.

R:

Thanks for your question. Here in our study, anthropogenic particle emissions from fossil fuel are not limited to those for domestic heating. First, though the combustion of fossil fuels for domestic heating is not common in Nanjing to some degree, the emission rates indeed increase in winter in YRD are higher (Zhuang et al., 2018; Li et al., 2017). Besides, burning of crop residues during autumn harvest could also contribute to the high aerosol concentrations to some degrees (Qian et al., 2014). Although it has been strictly controlled in China, the influence of crop residues burning still exist and cannot be ignored (e.g., Wang and Zhang, 2008; Zhu et al., 2010; Ni et al., 2015; Mehmood et al., 2018, etc.). Additionally, sub-regional transport also plays an important role, for

example, in winter, air masses coming from North China Plain where emission rates from fossil fuels of domestic heating are high, account for 31% and have high particles concentrations (Section 3.4). More details have been added in the revised manuscript.

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straw and biomass burnings in China from 2002 to 2016. Environmental Chemistry Letters, 2018, 16(1):301-309.

8. Line 294, the diurnal variation of BC concentration was generally associated with the vehicle volume. I am very curious about the higher BC levels during 8-11 pm. I think Nanjing showed the higher vehicle volume during 5-8 pm. The author should explain the unusual characteristics.

R:

Thank you for your question and suggestion. Considering a higher vehicle volume during 5$\sim$ 8 pm, the peak occurs during 9$\sim$11 pm is probably related to a more stable atmosphere stratification and a lower planetary boundary layer (PBL) after around 4 pm when the temperature decreases (Qian et al., 2014, Chen et al., 2016), both of which would result the accumulation of BC levels, thus the lag of the peak.

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9. Line 336, the author inferred that the BC and CO in the atmosphere were mainly originated from biomass burning. The fire point data should be added to demonstrate the potential source of BC and CO.

R:

Thanks for your comments. A remarkable correlation between BC and CO was found in a number of studies (Jennings et al., 1996; Derwent et al., 2001; Badarinath et

al., 2007; Spackman et al., 2008; Zhuang et al., 2014). Additionally, BC is mostly produced by the incomplete combustion of carbonaceous material, and so is CO (Pan et al., 2011). Therefore, both BC and CO might come from the same sources.

BC in Nanjing might mostly come from combustions of domestic bio-fuel, industry-coal, and vehicle-gasoline (Zhuang et al., 2014; Cheng et al., 2017), instead of biomass burning only. And during the autumn harvest (September$\sim$ November), though not so much as that in summer, the crop burning emissions still make contribution to pollutants (e.g., Qian et al., 2014; Yang et al., 2008; Yin et al., 2016, etc.). Moreover, in Yin et al.2016, spatial distribution of crop residue burning spots number from September to December in 2015 deriving from MODIS data shows that autumn crop residue burning has started in October in YRD, and could cause a rise in pollutants. According to Huang et al. (2012) and Li et al. (2016), spatiotemporal distribution of agricultural fire occurrences in China during 2003$\sim$ 2010 as well as 2012 has been presented associated with the spatial distribution of CO emission from residue open burning. Both of them suggested the crop residue burning in autumn is noteworthy and Jiangsu as well as the surrounding provinces including Henan, Shandong, and Anhui are the regions with highest emissions. The problems of writings might mislead the readers, for example, the explanation is probably not comprehensive enough to degrees. The sentences have been rephrased in the current version.

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10. Line 495-496, what does the sentence mean? The author should point out the relationship between CO and ozone production.

R:

Thank you for your question and advice. The sentences have been rephrased. A more detailed and in-depth discussion about the relationship between CO and ozone production has been included in the revised manuscript.

11. The conclusion should be condensed and stress the new contribution to the atmospheric chemistry.

R:

Thanks for your suggestion. Conclusion has been rephrased significantly and shortened necessarily and it's believed that the revised version is much more readable. The readers would more easily grasp the useful information of the results. In the revised version, the authors highlight the contributions to the atmospheric chemistry, especially deduction of the underlying chemical mechanisms based on the results of our study and previous studies through inter-species correlations analysis.

12. There are many grammar and format errors throughout the paper. I suggest the author should revise all of these minor flaws from words to words carefully.

R:

Thanks a lot for figuring out the problems of the manuscript's writings. The manuscript has been rephrased to be clearer. And it's also corrected carefully by Professor J. Liu, who is from University of Toronto and also is a co-author of this study with great contributions.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-927/acp-2018-927-AC1-supplement.pdf

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