Response to Anonymous Referee #1

This manuscript reports one-year continuous measurements of trace gases and particulate matters at a National Reference Climatological Station in Hangzhou, southern Yangtze River Delta region. The data were analyzed in terms of seasonal variations, interspecies correlations, and potential contributions from local emissions and regional transport. The measurement data of the present study are much valuable, and the analysis and interpretation of the data are fairly well. Thus, it is recommended that this manuscript can be considered for publication after the following comments being addressed.

Response: Thanks for your recognition and positive comments on our manuscript. According to your suggestions, we made the corresponding corrections in the revised manuscript. We expect this version would meet the requirement for publication in ACP.

Specific Comments:

1. Overall, the interpretation and analyses of the measurement results are fairly well, but there is lack of comparison with the other studies and importance or implications of the present study. To date there have been many measurement studies in the YRD region, such as at Lin'an, Shanghai and Nanjing. The authors should point to the new findings or difference between this new piece of work and the previous studies.

Response: Thank you very much for the suggestions about making comparison with the other studies. As depicted in the revised manuscript, we have elaborated more examples about the knowledge gap between this work and the previous studies and further pointed out our new finding.

2. The first paragraph of the Introduction section contains a lot of very basic information on the individual trace gases. I presume that the readership of the Journal should be expertise of this field, and suggest the authors to remove (or shorten) such general description and just focus on the key knowledge gaps and motivation of the study in the Introduction part.

Response: Yes, you are right. According to your comments, we removed some basic introduction related to the well-known trace gases including NO_x, CO, and SO_2 in the first paragraph in the revised manuscript.

3. Page 2, Line 52: intermediates/products

Response: As you suggested, we made corrections as "intermediates/products" in the revised manuscript.

4. Page 2, Line 55: and/or

Response: As you suggested, we replaced it with "and/or" in the revised manuscript.

5. Page 3, Lines 80-81: it is not clear what the "large knowledge gap and discrepancy" means. Please

elaborate more about the knowledge gap.

Response: Thanks for your valuable comments about "large knowledge gap and discrepancy". As mentioned in Response 2 above, we elaborated more examples about the knowledge gap in the revised manuscript.

6. Line 3, Line 89: Experiment and meteorological conditions

Response: Thanks for your valuable comments about "large knowledge gap and discrepancy". As mentioned in Response 2 above, we elaborated more examples about the knowledge gap in the revised manuscript.

6. Line 3, Line 89: Experiment and meteorological conditions

Response: In the revised manuscript, we changed it with "Experiment and meteorological conditions" as you suggested.

7. Line 3, Line 93: please provide the standard deviations for the average temperature, RH and rainfall.

Response: Thanks for your mention on this expression to improve our manuscript. As you suggested, we added the standard deviations for the average air temperature and RH. However, as shown in Table 1, we used the accumulated monthly value for rainfall (not the average) and thus it has no standard deviations.

8. Section 2.1: the authors need clearly state the type (e.g., urban, suburban or rural) of the study site. What are the major emission sources surrounding the site?

Response: We have stated the type of our study site (urban site) in the revised manuscript. With respect to the major emission sources surrounding this site, we have made the specific introduction as below in the revised manuscript.

"As a typical urban site, NRCS station is situated in the commercial and residential areas in the southern Hangzhou and thus it's characterized as a polluted receptor site as it receives local urban plumes and regional air masses from the YRD region when northwesterly wind prevails. In addition, as the right top map shown in Fig. 1, the site is adjacent to Prince Bay Park (area, 0.8 km^2) and situated in the northeastern part of West Lake famous scenic spot (area, 49 km^2). Therefore it can also capture the signature of vegetation emission in urban Hangzhou under southwesterly winds. Moreover, there are no local industrial pollution sources around the site. In brief, this site can be representative of urban Hangzhou."

9. Section 2.2, on the measurements of NO_2 and CO: what kind of converter was used for the conversion from NO_2 to NO? Is there auto-zero or auto-reference function for the CO analyzer, and what is the time frequency for the auto-zeroing during the campaign?

Response: Your suggestions are very important and valuable. In our study, we used internal and external MoO converter for the conversion from NO_2 and NOz to NO, respectively. For CO analyzer, there was auto-zero function and its time frequency was every 6 h during the campaign. The corresponding corrections were made in the revised manuscript, as shown below.

"NO and NO_x were detected by a chemiluminescence analyzer coupled with an internal MoO catalytic converter (TEI, 42i). Note that the differentiated value of NO_2 from NO_x and NO represents the upper limit concentration of atmospheric NO_2 due to the interference of other nitrogen-containing components (e.g., PAN, HNO_3, and HONO) in the conversion. Similar with NO_x, NO_y was also detected by a chemiluminescence analyzer (TEI 42i-Y) but equipped with an external MoO catalytic converter. CO was monitored with a gas filter correlation, infrared absorption analyser (TEI, 48i), with automatic zeroing every 6 hours."

14. Page 7, Lines 188-191: some studies have also investigated the seasonal variations of O_3 in Hong Kong and North China, and the authors should acknowledge these earlier studies.

Response: As you suggested, we have added the two earlier studies in the revised manuscript.

15. Page 7, Line 192: Xianlin?

Response: Yes, you are right. We are sorry for this mistake. This site is "Xianlin"

16. Page 8, Lines 227-228: revise this sentence

Response: According to your comment, we have changed this sentence with "In summer (Fig. 2f), an abnormally high level of O_x was found in winter with low O_3." in the revised manuscript.

17. Page 9, Line 257: color-coded

Response: According to your comment, we have replaced "coded" with "color-coded" in the revised manuscript.

18. Page 9, Line 258: led to

Response: According to your comment, we have replaced it with "led to" in the revised manuscript.

19. Page 9, Line 275: change "in addition to" to "in view of"

Response: According to your comment, we have changed "in addition to" with "in view of" in the revised manuscript.

20. Page 10, Line 297: pay attention to

Response: According to your comment, we have replaced it with "pay attention to" in the revised manuscript.

21. Page 11, Lines 329-330: why the air masses coming from open seas contained higher concentrations of NO_x and O_3? The authors need elaborate more about the reason for this interesting result.

Response: At first, we are so sorry for the incorrectly expression "long transports from Yellow Sea, East Sea, and South Sea were also important potential sources for NO_x and O_3" in the initial manuscript. After careful examination, we found that air masses with the seemed high WPSCF values for NO_x were not originating far from these open seas. They were just the broadening "tails" of high values contained in the areas with intensive anthropogenic NO_x emissions from inland well-industrialized cities. This phenomenon was also found in other studies by using trajectory statistical method (Riuttanen et al., 2013).

Similar with NO_x, air masses containing the high WPSCF values of O_3 also didn't come from the open seas. Indeed, as depicted in the manuscript, such air masses were mostly from the offshore area of East China Sea, Yellow China Sea, or South China, respectively on southeastern Zhejiang, Jiangsu, and Fujian Province. We speculated the recirculation of pollutants by sea- and land-breeze circulations around the cities along the YRD and Hangzhou Bay which has been confirmed by Li et al. (2015, 2016b), was largely responsible for the increased concentration of O_3 at NRCS site.

Also, such an increase in O_3 concentrations in urbanized coastal areas have been observed and modeled in a number of studies (Oh et al., 2006; Levy et al., 2008; Martins et al., 2012). Moreover, to further judge whether air masses came from open seas contained higher concentrations of NO_x and O_3, we used the results of MOZART-4/GEOS-5 simulation to draw the distribution maps of NO_x and O_3 concentrations within the identical domain (15-55 ^{o}N and 105-135 ^{o}E) with WPSCF analysis. As clearly seen from the Figure 1 below, high NO_x mainly distributed in terrestrial regions, especially in industrialized cities, but very low NO_x were found in open seas. In comparison, significantly high O_3 were elucidated covering the offshore regions of either East China Sea, Yellow China Sea, or South China Sea (Fig. 2). Then, along with the seasonal cluster analysis of back trajectories from NRCS site in Hangzhou, it's well confirmed that our speculation about the contribution of the recirculation of pollutants by sea- and land-breeze circulations in the offshore area to the observed O_3 at NRCS site.

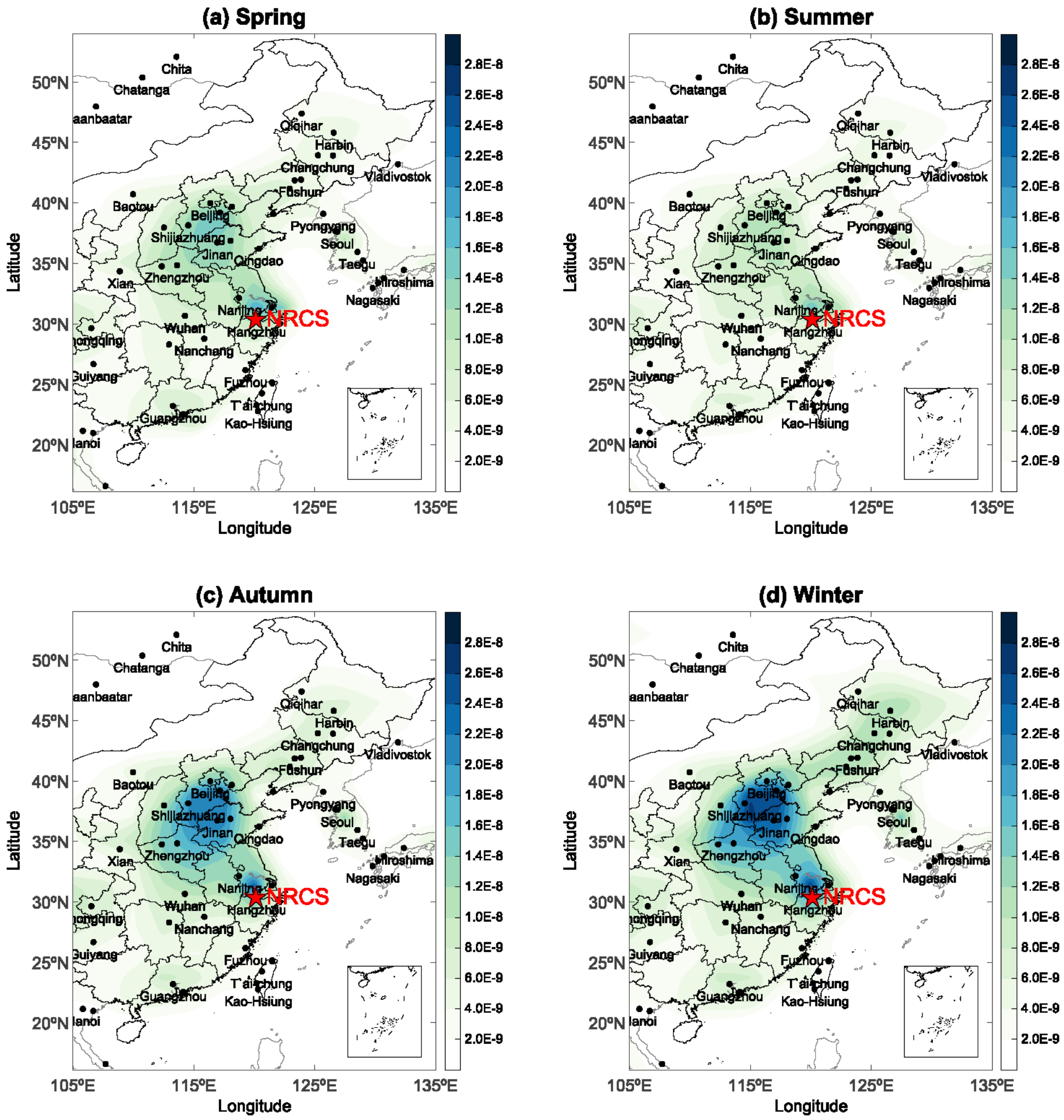

Figure 1 Seasonal and spatial distributions of NO_x volume mixing ratio (VMR) simulated by MOZART-4/GEOS-5. The sample site is marked in pentacle.

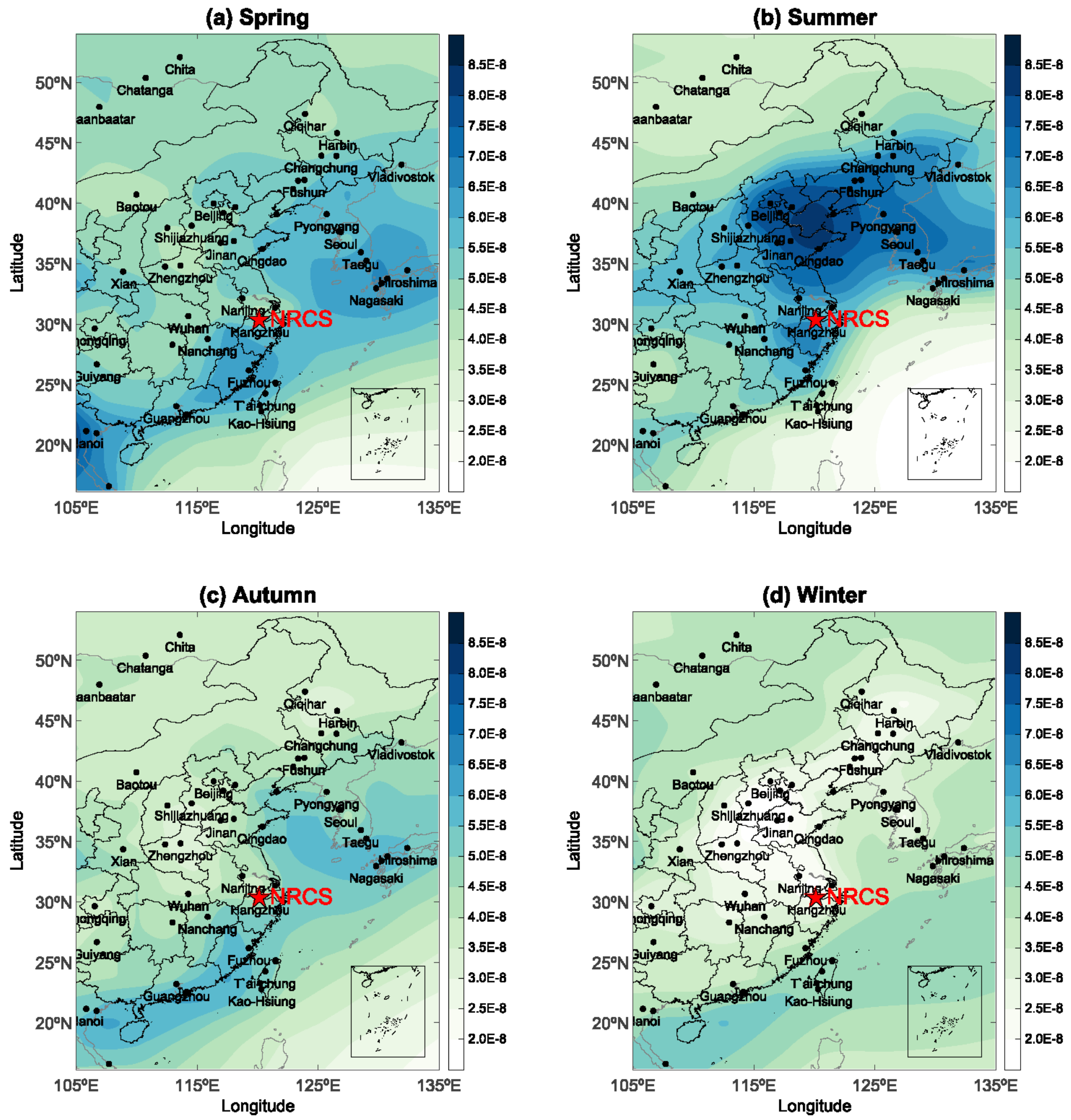

Figure 2 Seasonal and spatial distributions of O_3 volume mixing ratio (VMR) simulated by MOZART-4/GEOS-5. The sample site is marked in pentacle.

Reference

Levy, I., Dayan, U., and Mahrer, Y.: A five-year study of coastal recirculation and its effect on air pollutants over the east Mediterranean region, J. Geophys. Res., 113, D16121, 2008.

Li, M. M., Mao, Z. C., Song, Y., Liu, M. X., and Huang, X.: Impact of the decadal urbanization on thermally induced circulations in eastern China, J. Appl. Meteorol. Clim., 54, 259-282, 2015.

Li, M. M., Song, Y., Mao, Z. C., Liu, M. X., and Huang, X.: Impact of thermal circulations induced by urbanization on ozone formation in the Pearl River Delta, China, Atmos. Environ., 127, 382-392, 2016b.

Martins, D. K., Stauffer, R. M., Thompson, A. M., Knepp, T. N., and Pippin, M.: Surface ozone at a coastal suburban site in 2009 and 2010: relationships to chemical and meteorological processes, J. Geophys. Res., 117, D5, 5306, 2012.

Oh, I. B., Kim, Y. K., Lee, H. W., and Kim, C. H.: An observational and numerical study of the effects of the late sea breeze on ozone distributions in the Busan metropolitan area, Korea, Atmos. Environ., 40, 1284-1298, 2006.

Riuttanen, L., Hulkkonen, M., Dal Maso, M., Junninen, H., and Kulmala, M.: Trajectory analysis of atmospheric transport of fine particles, SO_2, NO_x and O_3 to the SMEAR II station in Finland in 1996-2008, Atmos. Chem. Phys., 13, 2153-2164, 2013.

22. Page 12, Line 359: long distance transport

Response: According to your comment, we have replaced it with "long distance transport" in the revised manuscript.

23. Figures 3-7: it would be better to combine these figures into one figure.

Response: According to your comment, we have combined the four figures, the scatter plots of NO_y-O_3 coded with air temperature (a), NO_y-$PM_{2.5}$ coded with relative humidity (b), NO_y-SO_2 coded with relative humidity (c), and O_3-$PM_{2.5}$ coded with air temperature (d), into one figure as Fig. 3 shown in the revised manuscript. In order to facilitate the clear view of the subpicture showing the scatter with O_3 mixing ratios above 80 ppbv, we keep the scatter plots of NO_y with CO in a single figure as Fig. 4 shown in the revised manuscript.

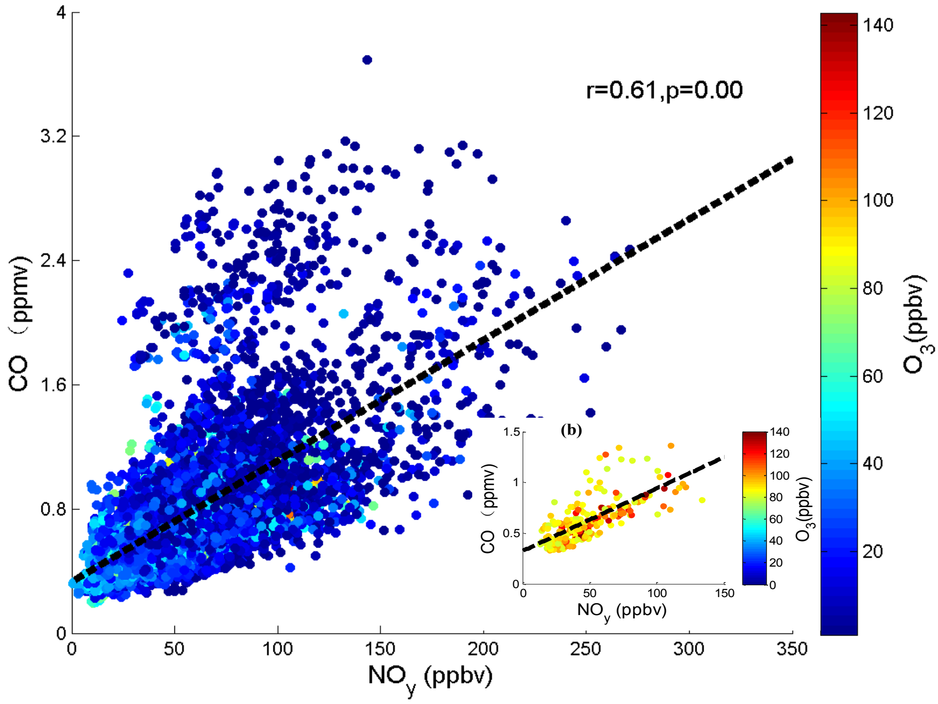

Fig. 4. Scatter plots of NO_y with CO coded with O_3 mixing ratios, along the subpicture (b) showing the scatter with O_3 mixing ratios above 80 ppbv.