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Response to Referee #1

The manuscript studied the characteristics and changes of baseline ozone in oceanic air in East China based on 6-year continuous measurements conducted at an island site. Corresponding ozone changes under various transport conditions were detailed presented and the impacts of offshore ozone on ozone air quality in Shanghai were quantified using the WRF-Chem model. Since increasing ozone pollution has become an urgent environmental problem in coastal urban agglomerations in East China, the results of this study provide valuable insight into what needs to be considered in dealing with ozone pollution in coastal megacities like Shanghai.

We thank the reviewer for all the insightful comments. Please see our point-by-point response (in blue) to the general and specific comments below. The changes that have been made to the manuscript are also listed.

General comments: Results and discussion-Sect.3 presented the overall changes of ozone in oceanic air at SSI. However, the key point was not prominent enough in current version. I suggest focusing more on the novelties of the study (the changes of baseline O₃ in oceanic air). Questions of why O₃ changes in September and October were analyzed, and what could be the driver of the detected changes need to be deeply reformulated.

Response: Thanks very much for pointing out that. We have rewritten Sect. 3.3 in the revised manuscript as suggested by the reviewer. Please see below:

“As discussed in Sect. 3.1, the prevailing winds carried different levels of pollutants to the SSI, resulting in different impacts on the O₃ levels in different months. In September and October, the frequencies of SW and W winds that carried high levels of pollutants were lowest (Table 1–2), exerting least influence on the atmospheric composition at SSI. Therefore, the variations of surface O₃ concentrations in September and October at SSI were examined to further assess the changes of least contaminated O₃ in the oceanic air. Figure 6b presents the overall changes of daily mean surface O₃ concentrations in September and October at SSI and XJH, respectively during the six-year period. The

corresponding mean O₃ mixing ratios during the two months were 60.9 and 31.3 ppbv, respectively at SSI and XJH. Compared to the significant elevated O₃ concentrations at XJH (0.59 ppbv yr⁻¹, $\alpha < 0.10$) in September and October, observed O₃ at SSI during same months exhibited insignificant decreasing changes from 2012–2017. The changes (-0.72 ppbv yr⁻¹, $\alpha > 0.10$) were somewhat different from the overall O₃ changes (+1.12 ppbv yr⁻¹, $\alpha > 0.10$) at SSI, suggesting different causes of the observed O₃ changes in the oceanic air during September and October.

To investigate possible drivers of the observed changes in the least contaminated O₃ in September and October at SSI, Table 3 displays the statistical results of the MK test and Theil-Sen trend estimate for NO_x and CO mixing ratios, temperature, and wind speed during the 2012–2017 period. Statistically significant upward trends were detected in wind speed, with estimated increasing rates of 0.21 m s⁻¹ yr⁻¹ during the observation period ($\alpha < 0.05$). The significantly enhanced surface wind speeds were conducive to the diffusion of O₃, which might be an important meteorological driver of the observed decreasing changes in O₃ levels at SSI from 2012 to 2017. Observed NO_x and CO levels exhibited increases of 0.48 ppbv yr⁻¹ ($\alpha < 0.05$) and 2.67 ppbv yr⁻¹ ($\alpha > 0.10$), respectively in September and October during the six-year period, indicating enhanced transport of pollutants to the oceanic area. Tie et al. (2013) suggested that the VOC-limited regime of O₃ formation was not only confined in urban Shanghai, but also extended to a broader regional area surrounding Shanghai. Thus, the elevated NO_x concentrations might not only retard daytime O₃ production but also enhance nighttime O₃ depression at SSI. Figure 6c further presents corresponding variations of daytime (10:00-16:00 LST) and nighttime (23:00-04:00 LST) mean O₃ concentrations at SSI. Both daytime and nighttime O₃ concentrations exhibited downward changes, reflecting the O₃ response to the enhanced O₃ diffusion and depression in September and October. Therefore, the enhanced diffusion and depression of O₃ induced by the elevated wind speed and NO_x concentrations might be important causes of the observed O₃ changes in September and October at SSI. It should be noted that the influence of radiation cannot be analyzed since observations of solar radiation were not available during the study period. Therefore, more

measurements are still needed to further understand the O₃ changes and corresponding drivers in the oceanic air.”

Specific comments:

1. Line 59: The location where the increased ozone concentrations were observed should be specified.

Response: We have revised the sentence as “Based on 14-year observations at a coastal site in Hong Kong, Wang et al. (2009) pointed out that enhanced pollution flow from the upwind coastal regions contributed to most of the observed O₃ increases in the background atmosphere of South China during 1994–2007. And the increase in background O₃, in turn, made a strong contribution of 81% to the increasing rate of O₃ in urban Hong Kong.”.

2. Line 69: Change “the three” to “those”

Response: Changed.

3. Line 73: Remove “surface”

Response: Removed.

4. Line 74: Change “atmospheric oxidation capacity response to” to “atmospheric oxidation capacity of continental air responding to”

Response: Changed.

5. Line 76: Change “in” to “at”

Response: Changed.

6. Line 91: Change “covering...area” to “covering an area of...”

Response: Changed.

7. Line 93: Change “magnitude” to “levels”

Response: Changed.

8. Line 120-121: Remove “In addition”

Response: Removed.

9. Line 125: Please explain the impact

Response: This part has been revised as “The DT site was set up in a national nature reserve near the coast of Shanghai, where the observed pollutant levels have been reported to well reflect the impacts of megacities in the Yangtze River Delta (YRD) region on the remote atmosphere during the MIRAGE-Shanghai (Megacities Impact on Regional and Global Environment at Shanghai) field campaign”.

10. Line 158: Add “its” before “surrounding”

Response: Added.

11. Line 175: Change “require” to “requires”

Response: Changed.

12. Line 177: Add “to” before “be”

Response: Added.

13. Line 216: Change “cleaner” to “less polluted”

Response: Changed.

14. Line 228: Change “distinctions” to “variations”

Response: Changed.

15. Line 234: The observed O₃ at DT site need to be provided in the supplementary

materials.

Response: The mean diurnal variations of O₃ at DT during the period 2012–2017 have been added to the supplementary materials as Fig. S1. Please see below.

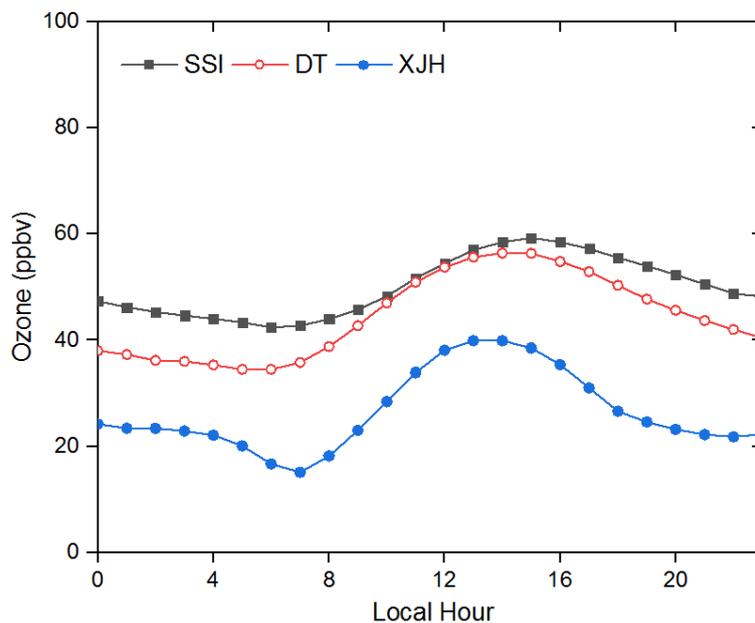


Fig. S1. Mean diurnal variations of O₃ at Sheshan Island (SSI, remote and oceanic), Dongtan (DT, rural), and Xujiahui (XJH, urban) station during the period 2012–2017.

16. Line 252: Actually, the study of Scheel et al. (1997) was conducted in Europe. Is 1.4 also a typical value of O_{3-max}/O_{3-min} in Chinese background sites? Please make sure of that.

Response: Thanks for pointing out that. The following description has been added to the second paragraph of Sect. 3.2:” For regional background sites in China, the typical values of O_{3-max}/O_{3-min} were usually in the range of 2–3 (Xu et al., 2008; Meng et al., 2009; Gu et al., 2020). In Lin’an, a continental background site in YRD region, the ratio was reported to increase as a result of NO_x emission changes during past decades, which could reach above 6 during summertime (Xu et al., 2008).”.

17. Line 292: How about the trend of O₃ observed at DT and Lin’an?

Response: Compared to those at SSI, observed extreme values of O₃ concentrations at DT and Lin’an were reported to exhibit more statistically significant ($\alpha < 0.05$) changes

response to the changes of anthropogenic emissions (e.g. NO_x) in past decades (Xu et al., 2008; Gao et al., 2017; Gu et al., 2020). We have revised this part in the manuscript.

18. Line 298: Change “nearly uncontaminated” to “least contaminated”

Response: Changed.

19. Line 330: Change “variations of” to “changes in”

Response: Changed.

20. Line 396-397: Please specify the source of this conclusion. Reference or methods need to be added.

Response: We have added Fig. S2 to the supplementary materials and revised this part as :” According to the cluster analysis results (Fig. S2), easterly winds from the ocean greatly affected the Shanghai region, accounting for 64–78% of the total flows in non-winter months during the period 2012–2017.”

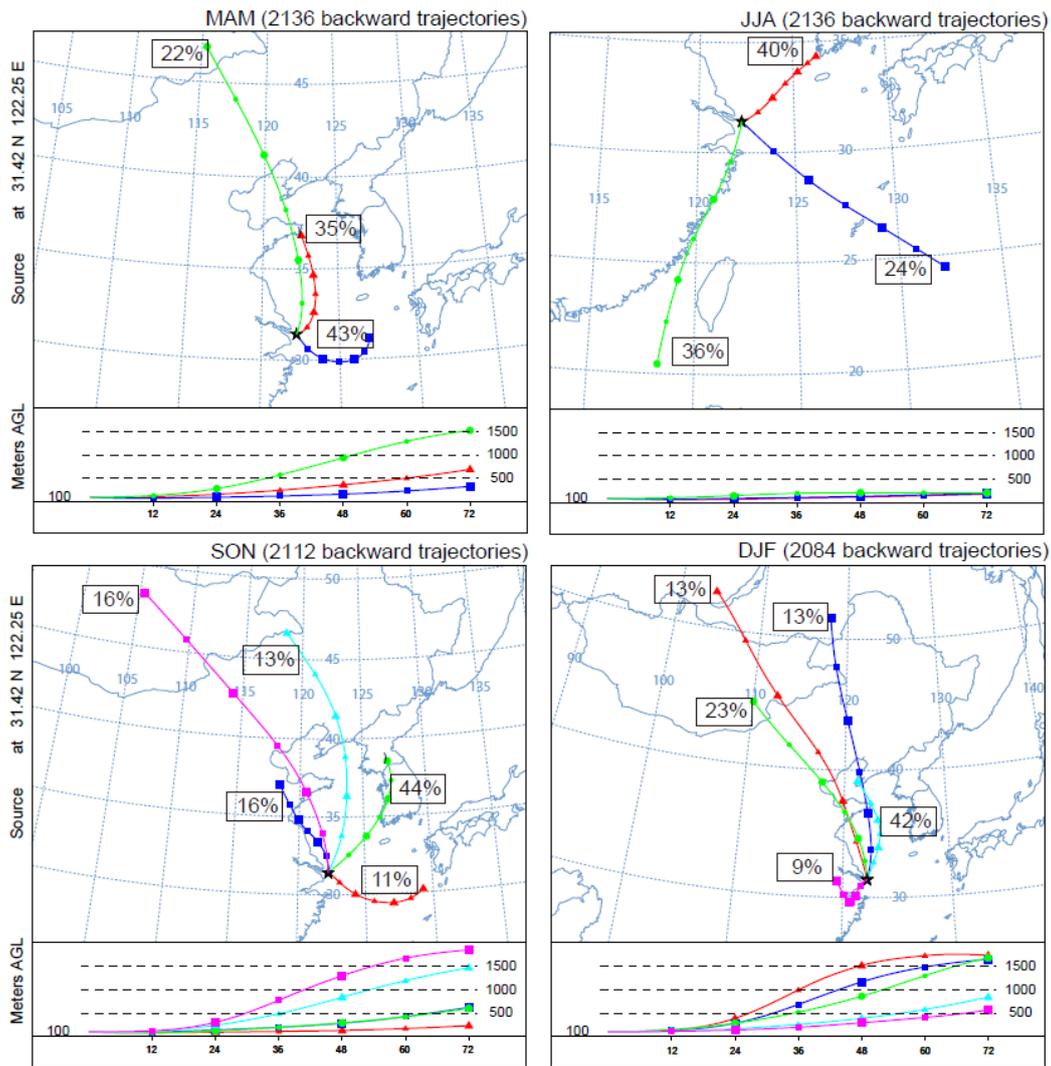


Fig. S2. Seasonal variations of the 72-h air mass backward trajectories arriving at the Sheshan Island (SSI) site using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model (Version 4, Draxler and Hess, 1998) driven by NCEP/NCAR Global Reanalysis Data ($2.5^{\circ} \times 2.5^{\circ}$). Trajectory clusters for MAM (March–May, left up), JJA (June–August, right up), SON (September–November, left bottom), and DJF (December–February, right bottom) were calculated based on the trajectories of 2012–2017 with steps of 12 h. The corresponding percentage occurrence values for different groups are presented as numbers in black squares.

21. Line 456: Do the 6-8 ppbv increases in O_3 occur in downtown Shanghai? Please specify it.

Response: This sentence has been revised as “Even so, simulated mean O_3

concentrations still exhibit 6–8 ppbv increases in downtown Shanghai in the BC_60 scenario, accounting for approximately 30% of the simulated O₃ concentrations in the BC_40 case.”.

22. Line 473: Change “mean concentrations” to “a mean value”

Response: Changed.

Reference

- Draxler, R. R. and Hess, G. D.: An overview of the HYSPLIT 4 modelling system for trajectories, dispersion, and deposition, *Austral. Meteorol. Mag.*, 47, 295–308, 1998.
- Gao, W., Tie, X., Xu, J., Huang, R., Mao, X., Zhou, G., and Chang, L.: Long-term trend of O₃ in a mega city (Shanghai), China: characteristics, causes, and interactions with precursors, *Sci. Total Environ.*, 603–604, 425–433, 2017.
- Gu, Y., Li, K., Xu, J., Liao, H., Zhou, G.: Observed dependence of surface ozone on increasing temperature in Shanghai, China. *Atmos. Environ.*, 221, 117108, 2020.
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- Xu, X., Lin, W., Wang, T., Yan, P., Tang, J., Meng, Z., and Wang, Y.: Long-term trend of surface ozone at a regional background station in eastern China 1991–2006: enhanced variability, *Atmos. Chem. Phys.*, 8, 2595–2607, 2008.