Response to Anonymous Referee #2

(Note: Reviewer comments are listed in grey, and responses to reviewer comments are in black. Pasted text from the new version of the paper is in italics.)

This paper presents a very detailed description of changes in ozone due to basin wide changes in SST. Changes can be up to 5 ppbv. The authors have provided great detail for the mechanisms behind the changes. The previous reviewers have commented on many aspects of this paper. Here I will restrict my comments to the overall methodology.

In short, for reasons explained in more detail below I am having difficulty interpreting the paper's results. The authors need to justify their methodology in detail, expand on some of the sensitivities of the solution to the methodology chosen and possibly run some addition simulations to put the results in context.

The authors set rectangular patches of ocean temperature warmer (or colder) by 1 degree within very large ocean-basin domains. The reasons for this setup are not well explained. How did they choose the size of the patch? How did they choose the southern and northern boundaries? The various patches are not similar in latitude or longitude nor do they apparently align with the edge of the ocean basins. We know the resulting circulation changes are sensitive to where the SST is perturbed. In particular there are large differences between the impact of SST perturbations in midlatitudes and those in the tropics. Teleconnections stemming from tropical ocean SST perturbations can have long-range impacts depending very much on the location of the perturbation. Changes in ocean temperature gradients are also likely to be important for the transport. We note that the simulations in this paper rather dramatically modify the ocean temperature gradients along all boundaries of the perturbation. While the authors smooth out the gradients I am not sure of the resulting impact. I would like to understand the impact of the details of their methodology on the solutions. As it stands I don't really see a strong justification to how they perturbed the ocean temperatures.

I am also having a difficult time interpreting the results. If the authors are interested in understanding the importance of SST perturbations on present day transport it would make sense to perturb the SST using realistic SST variability – perhaps an EOF analysis would be helpful here. This is because the result is sensitive to how the SST is perturbed. If the authors are interested in the importance of climate change it is also difficult to interpret the results. Perturbing the SST in one ocean basin is likely to alter the land-sea pressure gradients and transport in a different ways than changes under

CO2 influenced climate change. It may be possible to parse the impact of transport changes from climate change in terms of the perturbation simulations carried out by the authors but they have not done this.

So, I'm not sure what I ultimately have learned from the paper. The authors do a great job in providing details of transport changes due to SST modification: changes in stability, in clouds, in overall transport and other processes are important. It is not surprising that whole-scale changes in SST modify the transport and ozone chemistry. However, it is unclear to what extent these details are an artifact of their simulation setup and how they apply to the real world (either to interannual variations in SST or to climate induced variations). Specific details of the solution are likely to depend on how the SST has been changed. Thus I'm left with a very detailed analysis but I'm not sure what I have really learned.

We appreciate the reviewer's thoughtful comments. In this study, we mainly focus on exploring the sensitivity of surface O₃ distributions to SST anomalies in different ocean basins and investigating the mechanisms that govern this SST-O₃ relationship. Therefore, we used the Community Earth System Model (CESM) to investigate the response of surface O₃ concentrations at different continents to the basin-scale warming and cooling of individual Northern Hemispheric oceans. In our model experiments, idealized uniform sea surface temperature (SST) anomalies of +/- 1°C are superimposed onto North Pacific (15°N-65°N;100°E-90°W), North Atlantic (15°N-65°N; 100°W-20°E) and North Indian Oceans (5°N-30°N; 30°E-100°E), individually. The rectangular patches of SST changes in different cases can not be consistent in latitudinal and longitudinal ranges due to geographical differences in various ocean basins. We defined the latitudinal and longitudinal ranges of these oceans basin mainly based on their geographical features. The boundaries of prescribed SST anomalies generally align with the edge of the ocean basins that constrained by the continental line except for the southern side. The southern boundary of each ocean is relatively difficult to define since there is no apparent geographical boundary in the south. To constrain our prescribed SST anomalies in the Northern Hemisphere, we defined a southern boundary of 15°N for the North Pacific and North Atlantic oceans to be consistent. As for the North Indian Ocean, which is mainly located in the tropical regions, a lower southern boundary (i.e., 5°N) is chosen. If using 15°N as the southern boundary, the size of North Indian Ocean is too small. In each perturbation simulation, we further linearly smoothed the southern boundaries of these SST anomalies toward the equator to remove the sharp SST anomaly gradients at the edge.

To examine the sensitivity of the solution to the methodology we chosen to perturb the SST, we conducted an additional simulation based on the North Pacific warming case, in which no smoothness toward the equator is used, denoted as "Nosmooth". The corresponding changes of summertime surface O₃ in both perturbation simulations (i.e., with and without smoothing) are depicted in Figure R1. It shows that the responses of surface O₃ to these SST perturbations are broadly consistent in spatial patterns, though small differences do exist at specific places. According to previous studies (e.g., Lau et al., 1997;Li et al., 2015; Zhou et al., 2017 and references therein), the atmospheric response to SST changes over the tropical oceans are mainly locally driven and thermally direct owning to deep convection. There is no doubt that if we extend the southern boundary southward to the equatorial line, the atmospheric responses would be much larger, especially over the tropics. Since this study focuses more on qualitative understanding how the mid-latitude air quality is affected by the fluctuation of basin-scale SST changes in individual Northern Hemisphere oceans, the idealized SST anomaly employed here is a relatively straightforward way. In addition, smoothing out the gradient of prescribed SST anomalies is widely used in previous studies (e.g., Hu and Veres, 2016;Seager and Henderson, 2016;Sutton and Hodson, 2007;Taschetto et al., 2016).



Figure R1. Changes in the summertime (June-August) surface ozone concentrations (ppbv) in the Northern Hemisphere for (a) Pacific-W and (b) Nosmooth relative to CTRL. Four major regions of interest (i.e., NA ($15^{\circ}N-55^{\circ}N$; $60^{\circ}W-125^{\circ}W$), EU ($25^{\circ}N-65^{\circ}N$; $10^{\circ}W-50^{\circ}E$), EA ($15^{\circ}N-50^{\circ}N$; $95^{\circ}E-160^{\circ}E$) and SA ($5^{\circ}N-35^{\circ}N$; $50^{\circ}E-95^{\circ}E$)) are marked with red solid polygons. Red dashed lines mark the regions where the SST has been changed. The + symbols denote areas where results are significant at the 0.05 level as evaluated with a Student t-test using 20 years of data.

As we have mentioned in our Introduction section, the realistic SST is undergoing a variety of changes in different spatial and temporal scales. The relevant anomalies picked up from observation dataset are closely depended on the timescale we choose. SST anomalies over different oceans are often inter-correlated and difficult to separate from each other. The pattern of SST anomaly decomposed statistically (e.g., EOF analysis) is also largely depended on the duration of observation used. What's more, the prediction of future SST variability also remains great uncertainty. Hence, it is very difficult to define a representative SST anomaly pattern for each ocean basin. Since we are mainly interested in investigating the sensitivity of surface O₃ distributions to the SST changes in different ocean basins and the mechanisms governing the SST-O₃ relationships, this idealized SST anomaly setup are sufficient to address our questions. In previous studies, idealized, uniform SST anomalies have been widely used to explore the ocean-atmosphere relationships in the climate systems (e.g., Taschetto et al., 2016, Fan et al., 2016, Hu and Veres, 2016, Kushnir et al., 2002 and so on), which could simplify the interpretation and are easier for theoretical analysis.

In our revised manuscript, we further conducted two sensitivity tests with 1°C SST warming and cooling superimposed onto all three ocean basins (i.e., the North Pacific, North Atlantic and North Indian Ocean), denoted as All-W and All-C, respectively. Their effects on surface O₃ distributions are found to be comparable to the sum of the effects from individual oceans during boreal summers (see Figure S5 or S6 below) and winters (see Figure S7 or S8 below). This indicates that the SST forcing on O3 distribution is geographically additive. Local environmental policymakers may pay more attention to the SST variability over specific oceans. A lot of studies have used decomposed SST anomalies for different regions to identify their relevant roles in a particular climate response (Camargo et al., 2013;Sutton and Hodson, 2005;Ueda et al., 2015). A linear assumption that the influence of large SST anomaly pattern on the atmosphere can be generally constructed by the linear combination of the influences of individual SST patches have been verified by previous studies, especially for the tropical regions where the signal-to-noise is higher (Fan et al., 2016;Seager and Henderson, 2016).



Figure S5. Left column: Changes in the summertime (June-August) surface ozone concentrations (ppbv) in the Northern Hemisphere induced by 1 °C warming (a) and 1 °C cooling (b) in all three ocean basins (i.e., the North Pacific, North Atlantic and North Indian Ocean) relative to CTRL. Right column: sum of changes in the summertime (June-August) surface ozone concentrations (ppbv) from three warming cases (i.e., Pacific-W, Atlantic-W and Indian-W) and three cooling cases (i.e., Pacific-C, Atlantic-C and Indian-C) relative to CTRL, denoted as (b) Sum-W and (d) Sum-C, respectively. Four major regions of interest (i.e., NA (15°N–55 °N; 60°W–125°W), EU (25°N–65 °N;10°W-50 °E), EA (15 °N–50 °N; 95°E–160 °E) and SA (5 °N–35 °N; 50 °E–95°E)) are marked with red polygons.



Figure S6. Same as Figure S5 but using the Mercator projection.



Figure S7. Same as Figure S5 but for the wintertime (December-February).



Figure S8. Same as Figure S7 but using the Mercator projection.

Even through the response of atmosphere to SST changes in different scales have been extensively studies in the last several decades, their corresponding effects on air quality are rarely investigated directly. There are many important questions need to be addressed to provide a comprehensive understanding of the SST-O₃ relationship. In this study, we explored the responses of surface O₃ to idealized SST changes imposed in different ocean basins. The ultimate objective of this study is to differentiate roles of each Northern Hemispheric ocean played on the surface O₃ evolution over these highly populated regions (e.g., NA, EU, EA and IN). We provided a detailed analysis about the mechanisms modulating the SST-O₃ relationships at the process level. An opposite response pattern is observed between the upwind and downwind regions of an ocean. This though can not exactly match the real world conditions, but can provides useful implications for environmental management. Since the basin-scale warming or cooling has been frequently observed in different oceans, conducting idealized model experiments can capture the large-scale features of the observed anomaly patterns while ignores the local noisy variability. On the other hand, given the near-linear relationships existed in the ocean-atmosphere interactions, our analysis also illustrates the spatial extent of different oceans that modulate the surface O₃ in a changing climate system. In the revised manuscript, we have clarified our objectives as well as the ways how we perturb the SST in different oceans.

In the Introduction section:

"To fill this gap, this study focuses on examining the sensitivity of O_3 evolution over four polluted continental regions in the Northern Hemisphere (i.e., North America (NA, 15°N–55 °N; 60°W–125°W), Europe (EU, 25°N–65 °N; 10°W-50 °E), East Asia (EA, 15 °N–50 °N; 95°E–160 °E) and South Asia (SA, 5 °N–35 °N; 50 °E–95°E),

In the first paragraph of Section 2.2, we have:

"We firstly conduct a control simulation, hereafter referred to as CTRL, with prescribed climatological SSTs averaged from 1981 to 2010 (see Hurrell et al. (2008)). We then conduct six perturbation simulations with monthly SSTs uniformly increased or decreased by 1 °C in three ocean basins in the Northern Hemisphere: the North Pacific (15°N-65°N;100°E-90°W), the North Atlantic (15°N-65°N; 100°W-20°E) and the North Indian Ocean (5°N-30°N; 30°E-100°E, here 5°N is used in order to have a relatively larger domain size). The simulations are denoted as "Pacific-W", "Atlantic-W", "Indian-W" for three warming cases and "Pacific-C", "Atlantic-C", "Indian-C" for three cooling cases. We defined the latitudinal and longitudinal ranges of these ocean basins mainly based on their geographical features. The boundaries of prescribed SST anomalies generally align with the edge of the ocean basins except for the southern side. In each perturbation simulation, we further linearly smooth the southern boundaries of these SST anomalies toward the equator to remove the sharp SST anomaly gradients at the edge, following the previous approach (Taschetto et al., 2016; Seager and Henderson, 2016)."

In the last paragraph of Section 3:

"Our simulations reveal that different oceans can exert distinct region-specific effects on O₃ distributions. We further conduct two sensitivity tests with 1 °C SST warming and 1 °C SST cooling superimposed onto all three ocean basins (i.e., the North Pacific, North Atlantic and North Indian Ocean) in the Northern Hemisphere, denoted as All-W and All-C, respectively. Their effects on surface O₃ distributions are respectively compared with the sum of three individual warming cases (i.e., Pacific-W, Atlantic-W and Indian-W) and three cooling cases (i.e., Pacific-C, Atlantic-C and Indian-C). The responses of surface O₃ to a hemispheric SST anomaly generally resemble the sum of responses to different regional SST changes (see Figure S5 and S7 in the supplementary material)."

In the Summary section, we have:

"Overall, our study highlights the sensitivity of O_3 evolution to basin-wide SST changes in the Northern Hemisphere and identifies the key chemical or dynamical factors that control it. However, to provide a more comprehensive understanding of the SST-O₃ relationship, further studies using realistic SST variability are necessary. This may help the management of O_3 pollution by considering the influence of specific SST variability."

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