Response to Anonymous Referee #1

(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.)

Yi et al. have addressed most of the concerns raised by the reviewers. I have only one suggestion that may help in better highlighting the value of those idealized experiments: The authors have performed the additional experiment in which the SST changes are applied over the entire tropical regions. However, the authors barely present it in the manuscript. I think the authors should discuss it more, in particular, in my opinion, it would make more sense to have this simulation as the main experiment. Then the authors can use the other sensitivity studies to understand the changes seen in the main experiment. Otherwise, it is difficult to understand the rationale behind the warming of only in one basin rather than another.

We really appreciate the reviewer's thoughtful and valuable suggestions. We agree that the additional experiments with SST changes superimposed onto all three ocean basins provide valuable complements for our study. The results from these additional experiments indicate that the effect of a generalized SST warming on surface O_3 can be decomposed into individual regional SST forcings. It also helps to understand the responses of surface O_3 to a global-wide SST change. In our revised manuscript, we have expanded our discussion about these additional experiments (See Page 10-11, L295-304; P20-21, L584-595).

"Our simulations reveal that different oceans can exert distinct region-specific effects on the O_3 distribution. The effects of three individual warming/cooling cases (i.e., Pacific-W, Atlantic-W and Indian-W/ Pacific-C, Atlantic-C and Indian-C) on surface O_3 distributions are further summed up to compare with the combined warming/cooling (i.e., ALL-W/ALL-C). The responses of surface O_3 to a hemispheric SST anomaly generally resemble the sum of responses to individual regional SST changes (see Figures S3 and S4 in the supplementary material). This indicates that the effect of a generalized SST warming on surface O_3 can be decomposed into individual regional SST forcings."

…

"Additionally, the sensitivity tests with 1ºC SST warming and cooling superimposed onto all three ocean basins further show in general that the SST forcing on O_3 distribution is geographically additive. A number of studies have used the decomposed SST anomalies for different regions to identify their relevant roles in a particular climate response (e.g., Sutton and Hodson, 2005; Camargo et al., 2013; Ueda et al., 2015). A linear assumption that the influence of large-scale 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 (e.g., Fan et al., 2016; Seager and Henderson, 2016). Therefore, our study also helps to understand the roles of different

ocean basins in the Northern Hemisphere played in modulating surface O_3 distributions in a global-wide SST warming condition associated with climate change."

Few minor suggestions:

L98 SST changes affect solar radiation indirectly through changes in atmospheric circulation and cloud cover. How it is written it seems that SST directly affect solar radiation. (same in LL360-361)

Thanks for pointing it out. We have rephrased this for clarification (P4, L95-102).

"Sea surface temperature (SST) is an important indicator that characterizes the state of the climate system. Its variations strongly perturb the mass and energy exchange between the ocean and atmosphere (Small et al., 2008; Gulev et al., 2013), which influence atmospheric circulation, atmospheric temperature and specific humidity (Sutton and Hodson, 2005; Frankignoul and Sennéchael, 2007; Li et al., 2008) from regional to global scales (Glantz et al., 1991; Wang et al., 2000; Goswami et al., 2006). It also affects cloud formation and consequently influences incoming solar radiation (Deser et al., 1993; Fallmann et al., 2017). "

As for L362-364 (original L360-361), we stated "As emissions are fixed in all simulations, the change in net O_3 production is driven by SST induced meteorological changes (e.g., air temperature, air humidity, and solar radiation)." Here we just listed some meteorological variables that may affect by SST anomalies. Therefore, we think that it is unnecessary to distinguish weather it is directly or indirectly.

LL101-102 "Numerous studies have shown that SST changes over different oceans and at different latitudes lead to significantly different meteorological sensitivities …" what do the authors mean by "meteorological sensitivities"?

Here we mean that the sensitivities of meteorological conditions to SST changes are quite different over different oceans. In our revised manuscript, we rephrased this sentence for clarification (see P4, L102-105):

"Numerous studies have shown that SST changes over different oceans and at different latitudes lead to significantly different meteorological and climate responses (Webster, 1981;Lau and Nath, 1994;Lau, 1997;Sutton and Hodson, 2007;Sabeerali et al., 2012;Ueda et al., 2015)."

LL117-118 "The North Atlantic Ocean pronounces various modes of low-frequency SST variability" what do the authors mean by "pronounces"?

We have replaced "pronounces" with "exhibits" (P4, L120-121):

"…The North Atlantic Ocean exhibits various modes of low-frequency SST variability (Kushnir, 1994;Wu and Liu, 2005;Fan and Schneider, 2012;Taboada and Anadon, 2012)."

LL126-136 The authors stated that ozone stratospheric intrusions associated with ENSO for winter and spring. However, the authors mentioned "Stratospheric intrusion events, which lead to vertical down mixing of ozone-rich air, can significantly elevate surface O3 during spring and summer" (LL60-61). Is there any study relating tropical SST and O3 stratospheric intrusion in summer?
Actually to my knowledge in summer stratospheric intrusions are few and not very important. Taken from Lin et al. 2012 cited by the authors in L62:
"The impacts of deep STT events on surface O3 are strongest at Northern Hemisphere extratropical latitudes in late winter and spring, likely reflecting a combination of peak O3 abundances at the tropopause [e.g., Prather et al., 2011], more frequent storms [e.g., Holton et al., 1995] and a longer O3 lifetime than in summer, and stronger surface heating that enhances the turbulent mixing between the free troposphere (FT) and the planetary boundary layer (PBL) compared to early winter."

Good comment. There are several studies that reveals O_3 stratospheric intrusion events during summertime (e.g., Ding and Wang, 2006; Zanis et al., 2014). However, these phenomena are mainly happened over Central Asia. We agree that compared with spring, stratospheric intrusions are few and less important in summer over most regions. We have revised this sentence in L61 to avoid confusion:

"Stratospheric intrusion events, which lead to vertical down-mixing of ozone-rich air, can significantly elevate surface O_3 during spring (Grewe, 2006;Lin et al., 2012;Zhang et al., 2014)."

LL192-193 Specify climatological monthly SSTs.

Thanks! We revised this sentence in P7, L196-197:

"We first conduct a control simulation, hereafter referred to as CTRL, with prescribed climatological monthly SSTs averaged from 1981 to 2010 (see Hurrell et al. (2008))."

Section 2.2
The author should present the all-basin experiments here rather than in LL282-291.

Good suggestion. We have moved the description of the all-basin experiments to Section 2.2 (see P7-8, L210-213):

"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."

LL264-266 "Surface O3 changes in response to positive and negative SST anomalies generally pronounce a consistent spatial pattern but are opposite in sign, suggesting robust relationships between surface O3 levels and SST anomalies (Figure 1)." What do the authors mean by "pronounce"? produce?

We have replaced "pronounce" with "exhibit" (P10, L277-279):

"Surface O_3 changes in response to positive and negative SST anomalies generally exhibit a consistent spatial pattern but are opposite in sign, suggesting robust relationships between surface O_3 levels and SST anomalies (Figure 1)."

LL297-316 This part should go on the methods section 2.3

Good suggestion. We have moved this part below to the Section 2.3 (see P9, L242-253).

"The IPR method has been widely used in air quality studies to examine the cause of pollution episodes (Wang et al., 2010;Li et al., 2012). When applied in climate sensitivity analysis (usually measuring the difference between two equilibriums), the net change of all IPRs approaches zero. Typically, the positive changes in IPRs are mainly responsible for the increase in surface O_3, which may further induce O_3 removal to balance this forcing in a new equilibrium. Therefore, here, the IPR analysis is used not to budget the SST-induced O_3 concentration changes but rather to help examine the relative importance of different transport and chemical processes in driving the sensitivity of O_3 to SST forcing."

L367 "at the continent below" change to "surface"

We revised this sentence in P13, L368-370:

"Associated with this temperature decrease is a remarkable reduction in the solar radiation received at the surface (more than 15 W/m^2, Figure S10)."

L375 Clouds do not block but reduce solar radiation at the surface.

Thanks for pointing out this. We have rephrased this in the revised manuscript (P13, L377-379).

"Meanwhile, the corresponding increase in cloud cover reduces the solar radiation reaching the surface of the Indian subcontinent and thus the air temperature of lower troposphere in that region."

L504 "The corresponding anomalous cyclone should be responsible" change to read as

something like:
"The corresponding anomalous cyclonic circulation may be responsible"

We have rephrased this sentence in P18, L506-508:

"The corresponding anomalous cyclonic circulation may be responsible for the dipole of the South Asian CO tracer changes over the source region depicted in Figure 8e."

L528 again "pronounce" that I believe should be "produce"

We have replaced "pronounce" with "produce" for clarification (P18, L529-531)

"The regionally and seasonally averaged surface O_3 *changes over four continental regions (i.e., NA, EU, EA and SA) produce wide seasonal and regional variability (varying from 1 to 3 ppbv)."*

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.)

Second review of: "Title: Response of the Global Surface Ozone Distribution to Northern Hemisphere Sea Surface Temperature Changes: Implications for Long-Range Transport" by Yi et al.

This paper reports on the response of continental ozone distributions to basin-wide increases or decreases in SST. For the most part it is well written and clear and represents a thorough analysis. Nevertheless at this point I cannot recommend publication for essentially the same reasons articulated in my first review. It is too bad the authors did not design their simulations to be more relevant to the atmosphere-ocean system. In their reply the authors essentially re-articulated again what initially stated in the paper. They did include a few additional sensitivity simulations, but overall my main objections remain the same.

My main objection has to do with the size and justification for the SST modifications the authors use in their study. They state in their reply that "in previous studies, idealized, uniform SST anomalies have been widely used to explore 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)". I have briefly examined the studies listed and in none of them did the authors use basin wide SST perturbations. Taschetto et al., added warming patches to understand the atmospheric teleconnections associated with El Nino. In addition to: "further facilitate this comparison, two additional "semi-realistic experiments were carried out …". Fan et al. (2016) changed the SST in small patches to understand the impact on the Asian monsoon. Hu and Veres (2016) examined the patches of SST in relation to the AMO oscillation. The patches were relevant to those seen during the AMO. Kushnir et al. (2002) used SST perturbations from idealized GCM experiments to "capture only the salient features of observed anomaly patterns." "The prescribed SST anomalies are stripped of details to capture the "essential features" of the observed patterns". In each of these cases: (i) the SST was prescribed to capture in a basic way observed SST patterns and the resulting atmospheric dynamics and (ii) the authors could relate their simulations to observations. In the present paper the SST perturbations are not related to any observed pattern. Furthermore the key results showing differences in upwind and downwind responses are not related specifically to observed changes.

I had originally thought that the results might possibly relate to the SST changes expected in a future climate. In climate change experiments large-scale SST forcing is sometimes used (e.g. Shaw and Voigt, 2015: Nature Geoscience 8, 560–566 (2015) doi:10.1038/ngeo2449). However, in these simulations the climate response is a "tug of war" between that due to changes in the SST and that due to direct radiative forcing.

In the present simulations the direct radiative forcing due to climate change is not simulated. Moreover, even in the case of climate induced SST warming I don't see why the SST perturbations would only include the N.H.

Furthermore, it is apparent the results are sensitive to the SST perturbation used. For example, the authors claim that when the SST perturbation raises the ocean temperature above 29oC the increased convection changes the circulation. What if the ocean temperatures had only been modified north of 25o: would the same results be expected? The authors state in the response that their SST forcing is straightforward since their objective is to examine mid-latitude air quality. However, in fact mid-latitude circulation patterns are sensitive to tropical SSTs through well known teleconnection patterns.

To conclude: (i) I don't see a physical justification to the SST perturbations applied in this study (in contrast to the above referenced studies). (ii) I don't see how the present study sheds light on any observed phenomena, neither interannual variability or climate change. Or the authors did not clearly point out the connections. (iii) As the SST perturbations are rather arbitrary (why not put the southern boundary at 25o N, or at the equator?) the results may differ in unknown ways when other SST perturbations are used or when more realistic SST perturbation patterns are used.

We appreciate the reviewer's thoughtful comments. Those references (i.e., Hu and Veres, 2016; Kushnir et al., 2002; Fan et al., 2016; Taschetto et al., 2016) listed in our previous reply are a few examples showing the approach used in this study, i.e., applying idealized, uniform SST anomalies to explore ocean-atmosphere relationships, has been used previously. Besides these examples, a lot more similar references exist which conducted idealized model experiments. For example, Li et al., (2008) superimposed an idealized SST warming spreading the whole Indian Ocean basin (from 30 °N–30 °S) to explore the influence of this warming on the East Asian summer monsoon. Chen et al., (2013) raised the SST uniformly in an idealized aquaplanet model to analysis the mechanisms of the atmospheric response to climate forcing. Hu et al., (2014) conducted two experiments—one with uniformly increased global SSTs, the other with linearly increased global SSTs—to explore the relevant atmospheric responses using a coupled chemistry-climate model. There references show that model experiments with prescribed idealized SST anomalies are widely approved for scientific study and not all of them were based on observed SST patterns. As we stated in our previous reply, the realistic SST is undergoing a variety of changes in different spatial and temporal scales. Instead of focusing on the responses of surface O_3 to a specific real SST signal, our study is designed to show the broad picture of how regional surface O_3 concentrations response to SST anomalies in individual ocean basins and understand the mechanisms that govern this SST-O_3 relationship. In our model experiments, we uniformly increased/decreased SST over different ocean basins in the Northern Hemisphere. It quite difficult to reach our goal if applying the observed SST anomalies since the perturbation patterns are changing from one decade to another and there is no representative SST patterns we can use to derive some implications for future environmental policy. As a theoretical analysis, the method we used here is consistent with our research objective. Therefore, we don't think that our study is lack of foundations compared with other studies.

On the other hand, basin-scale warming or cooling anomalies are actually existed in different ocean basins. For example, the Atlantic Multidecadal Oscillation (AMO) exhibits periodical warming or cooling SST anomaly over the whole North Atlantic (Sutton and Hodson, 2005, 2007). El Niño has been revealed to induce a basin-wide SST increase in the Indian Ocean, especially over the North region (Du et al., 2009; Roxy et al., 2014). Our study could provide valuable implications (though not perfect) for the responses of surface O_3 to these observed SST anomalies. We agree that the teleconnection effects of tropical SSTs on mid-latitude climate has been well-documented in previous studies (e.g., Roxy et al., 2015; Lau, 1997; Lau et al., 1997). However, how surface O_3 responses to tropical SST anomalies is still questionable owning to complex chemical and physical processes existed in the O_3 formation. The connection between O_3 distribution and tropical SST anomalies can't be established directly without further investigations. In this study, we successfully explored the roles of different chemical and physical processes played in governing the SST-O_3 relationships in different oceans. We further showed that the tropical SST warming may suppress intercontinental O_3 transport and lead to opposite surface O_3 responses over upwind and downwind regions. Based on the solid theoretical foundations and a state-or-art climate-chemistry model, our study revealed a general mechanism for the SSTs over different oceans on modulating the surface O_3 distributions.

What's more, as we stated in our previous reply, sensitivity tests with 1ºC SST warming and cooling superimposed onto all three ocean basins showed in general that the SST forcing on O_3 distribution is geographically additive. Therefore, this study also aids in understanding the roles of different ocean basin in the Northern Hemisphere on surface O_3 distribution in a global-wide SST warming condition associated with climate changes. We only focused on ocean basins over the Northern Hemisphere because that these are close to the most populated and polluted regions in the world (i.e., NA, EU, EA, SA). We will study the more remote effects from the Southern Hemispheric oceans in the follow-up works.

In our model experiments, we imposed idealized SST anomalies onto three different ocean basins in the North Hemisphere, individually. The boundaries of the prescribed SST anomalies generally align with the edge of the ocean basins that constrained by the continental line except for the southern side. We defined a southern boundary of 15°N for the North Pacific and North Atlantic oceans and 5°N for the North Indian Ocean. 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. This method helps us to constrain our prescribed SST anomalies in the Northern Hemisphere, which is consistent with the goal of this study. We believe that it is a suitable approach to conduct our experiments. Previous studies have shown that the atmospheric response to extratropical SST anomalies is small compared to internal (unforced) atmospheric variability (Kushnir et al., 2002). In our study, we also found that the SST changes over the tropical regions play a more dominate role on modulating the surface O_3 distributions than extratropical regions. If the SST perturbation was only prescribed over extratropical regions (e.g., above 25 °N), it is more likely to exert insignificant effects on the surface O_3. On the other hand, as we showed in our previous reply, the responses of surface O_3 to different SST perturbations are broadly consistent in spatial patterns. Previous studies (e.g., Zhou et al., 2017; Li et al., 2015; Lau et al., 1997 and references therein) showed that the atmospheric response to SST changes over the tropical oceans are mainly locally driven and thermally direct owning to deep convection. The atmospheric responses are generally linearly related to tropical SST anomalies (Fan et al., 2016;Seager and Henderson, 2016). Therefore, if we expand the southern boundary to the equatorial line, the atmospheric response would be more significant. Directly expanding the southern boundary to the equatorial line without any smoothness will product sharp SST anomaly gradient at the edge, which may not be a proper way to prescribe the SST anomaly, referring to previous studies (e.g., Sutton and Hodson, 2007; Hu and Veres, 2016; Seager and Henderson, 2016; Taschetto et al., 2016). As a model experiment, the magnitude of the signal may always changes along with the forcing we imposed to the model. However, the mechanisms driving the O_3-SST relationships are solid, which are independent to ways how the SST are prescribed.

In summary, this study mainly focuses on exploring the sensitivity of surface O_3 distributions to SST anomalies in different ocean basins and investigating the general mechanisms that govern this SST-O_3 relationship rather than a specific realistic phenomenon. We believe that the SST perturbations applied in this study is a suitable way to attain our purpose. Similar approaches have been widely applied in previous studies to conduct scientific research. Basin-scale SST warming or cooling anomalies were also observed in the real world. As an idealized model study, our analysis is based on solid theoretical foundations and a state-of-art model. Therefore, the mechanism that governs the SST-O_3 relationship, as revealed in this study, is also applicable in the real world. It could provide valuable implications for the responses of surface O_3 to potential regional or hemispheric SST changes. However, cautions should be taken in interpreting our results in the real world since that observed surface O_3 and SST variabilities are much more complicate. In our revised manuscript, we add more discussions to explain our approach and caveat the uncertainties of our findings (Please see P20-21, L570-605).

"This 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 this evolution. Idealized and spatially uniform SST anomalies are used to explore the general mechanisms governing the regional SST-O_3 relationships. We find that the SST

changes over tropical regions exert considerable impacts on surface O_3 levels. The increase in tropical SST over different ocean basins enhances deep convection, which further trigger large-scale subsidence over nearby and remote regions. These enhanced convective activities also tend to release more latent heat over the upper troposphere and significantly increase the air temperature there. These processes influence large-scale circulation patterns and lead to opposite surface O_3 responses over upwind and downwind regions related to a specific ocean basin. These findings provide valuable implications for the potential surface O_3 change in response to future warming or cooling of individual oceans.

Additionally, the sensitivity tests with 1ºC SST warming and cooling superimposed onto all three ocean basins further show in general that the SST forcing on O_3 distribution is geographically additive. A number of studies have used the decomposed SST anomalies for different regions to identify their relevant roles in a particular climate response (Sutton and Hodson, 2005; Camargo et al., 2013; Ueda et al., 2015). A linear assumption that the influence of large-scale 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 (e.g., Fan et al., 2016; Seager and Henderson, 2016). Therefore, our study also helps to understand the roles of different ocean basins in the Northern Hemisphere played in modulating surface O_3 distributions in a global-wide SST warming condition associated with climate change.

Overall, this study may guide the management of regional O_3 pollution by considering the influence of specific SST variability. However, cautions should be taken in interpreting our results in the real world since observed surface O_3 variabilities are induced by various factors including O_3 precursor emissions and atmospheric conditions. Realistic SST anomalies over different oceans are more complicate (usually not uniformly distributed) and often inter-correlated with each other (Fan et al., 2016). They may exert jointly effects on modulating surface O_3 distributions. To provide more *precise understanding about the SST-O_3 relationship over a specific region, additional sensitivity tests regarding smaller patches of SST variability are also necessary. "*

Response to Anonymous Referee #3

(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 has undergone much improvement and the authors have invested a good deal of time to carefully consider and respond to all the reviewer comments, and write an authoritative paper. My one recommendation is not send a response to reviewers file that is 2000 MB in size as I was unable to download the file. You need only show revised figures once and refer back to them.

We greatly appreciate the reviewer for these detailed and valuable comments. We will pay attention to this problem and avoid providing large files afterward.

Minor comments (pages/line numbers refer to the revised document with track changes):

Generally I'm not sure so many (26) supplementary figures are needed, if their sole purpose is to show a spatial pattern on a different projection.

Good suggestion! Considering that figures using polar projection are distinguishable enough in our manuscript, we decided to remove the same plots using the Mercator projection in the supplementary material. The number of supplementary figures has been reduced from 26 to 14.

Spaces after ; throughout.

Thanks! we have carefully revised these typos existed in the manuscript.

Page 2, line 58- reference Organisation 2013? Perhaps WHO 2013?

We have revised this citation (P2, L50-52):

"High ground-level ozone (O_3) concentrations adversely impact human health by inducing respiratory diseases and threaten food security by lowering crop yields (Brown and Bowman, 2013; WHO, 2013; Chuwah et al., 2015)."

Page 4, line 120- IPCC chapter 2 is not the correct way to cite this chapter. The citation will be given at the start of the IPCC chapter.

Thanks! We have revised it (P4, L109-111):

"The Intergovernmental Panel on Climate Change Fifth Assessment Report (IPCC, 2013) provides strong evidence in Chapter 2 that global SSTs are generally increasing due to the impacts of anthropogenic forcings on global climate change."

Page 10, line 283- perhaps omit the season definitions here and leave only in the figures caption, as otherwise this detracts from finding the main point of the first sentence in the results.

Good suggestion. We moved the season definitions from this sentence (P9, L259-261) to the Caption of Table 1.

"Seasonally and regionally averaged surface O_3 changes in each SST perturbation simulation for the four highly populated continental regions and three ocean basins defined in our study are given in Tables 1 and S1, respectively."

"Table 1. *Seasonally (i.e., DJF (December, January, February), MAM (March, April, May), JJA (June, July, August) and SON (September, October, November)) and regionally averaged (only land grid boxes are included) changes in surface O_3 concentrations (ppbv) for basin-scale SST perturbation cases relative to the control simulation."*

Page 11, line 319- for consistency, rephrase "Similarly in the Atlantic W case" as the previous sentence does not use that same type of terminology.

We have rephrased this sentence in P10, L285-287:

"Similarly, the SST warming over the North Atlantic decrease the surface O_3 levels by 1~2 ppbv over the North Atlantic and Europe but increase (~1 ppbv) that over North America and the North Pacific."

Page 13, line 360- Add "VDIF" after "through diffusion"

We have revised it in P11, L312-313.

"The downward transport of O_3 through diffusion (VDIF) is an important source of surface O_3, while DRYD acts as a sink."

Page 21, line 613, rephrase "should".

We have rephrased this sentence in P18, L506-508:

"The corresponding anomalous cyclonic circulation may be responsible for the dipole of the South Asian CO tracer changes over the source region depicted in Figure 8e."

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Response of the Global Surface Ozone Distribution to Northern Hemisphere Sea Surface Temperature Changes: Implications for Long-Range Transport

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Abstract

The response of surface ozone (O_3) concentrations to basin-scale warming and cooling of Northern Hemisphere oceans is investigated using the Community Earth System Model (CESM). Idealized, spatially uniform sea surface temperature (SST) anomalies of +/- 1 °C are individually superimposed onto the North Pacific, North Atlantic, and North Indian Oceans. Our simulations suggest large seasonal and regional variability of surface O_3 in response to SST anomalies, especially in the boreal summer. The responses of surface O_3 associated with basin-scale SST warming and cooling have similar magnitude but are opposite in sign. Increasing the SST by 1 °C in one of the oceans generally decreases the surface O_3 concentrations from 1 to 5 ppbv. With fixed emissions, SST increases in a specific ocean basin in the Northern Hemisphere tend to increase the summertime surface O_3 concentrations over upwind regions, accompanied by a widespread reduction over downwind continents. We implement the integrated process rate (IPR) analysis in CESM and find that meteorological O_3 transport in response to SST changes is the key process causing surface O_3 perturbations in most cases. During the boreal summer, basin-scale SST warming facilitates the vertical transport of O_3 to the surface over upwind regions while significantly reducing the vertical transport over downwind continents. This process, as confirmed by tagged CO-like tracers, indicates a considerable suppression of intercontinental O_3 transport due to increased tropospheric stability at lower mid-latitudes induced by SST changes. On the other hand, the responses of chemical O_3 production to regional SST warming can exert positive effects on surface O_3 levels over highly polluted continents, except South Asia, where intensified cloud loading in response to North Indian SST warming depresses both the surface air temperature and solar radiation, and thus photochemical O_3 production. Our findings indicate a robust linkage between basin-scale SST variability and continental surface O_3 pollution, which should be considered in regional air quality management.

Keywords: SST anomaly, Surface O_3, Process analysis, Transport, CESM

1. Introduction

High ground-level ozone (O_3) concentrations adversely impact human health by inducing respiratory diseases and threaten food security by lowering crop yields (Brown and Bowman, 2013; WHO~~Organization~~, 2013; Chuwah et al., 2015). Considering the eco-toxicity of O_3, understanding the physical and chemical mechanisms that control atmospheric O_3 concentrations is of great importance. Surface O_3 is produced in the atmosphere via photochemical processing of multiple precursors including volatile organic compounds (VOCs), carbon monoxide (CO) and nitrogen oxides (NO, NO_2). These precursors originate from both natural and anthropogenic sources (Vingarzan, 2004; Simon et al., 2014; Jiang et al., 2016). In addition to local production, transport of O_3 and its precursors from upwind regions and the upper atmosphere can also influence surface O_3 abundance. Stratospheric intrusion events, which lead to vertical down-mixing of ozone-rich air, can significantly elevate surface

O_3 during spring and summer (Grewe, 2006; Lin et al., 2012b; Zhang et al., 2014). The long-range transport of O_3 and its precursors has been extensively studied, and their inter-continental impacts have been evaluated using measurements and model simulations (Parrish et al., 1993; Fehsenfeld et al., 1996; Wild and Akimoto, 2001; Creilson et al., 2003; Simmonds et al., 2004; Fiore et al., 2009; Brown-Steiner and Hess, 2011; Lin et al., 2012a; Lin et al., 2014).

Both photochemistry and dynamic transport collectively affect surface O_3 levels. Important meteorological factors that can impact both photochemistry and transport include atmospheric circulations, solar radiation, air temperature, and relative humidity. Atmospheric circulation considerably determines the timescale and pathway of O_3 transport (Bronnimann et al., 2000; Auvray and Bey, 2005; Hess and Mahowald, 2009). The efficiency of O_3 transport varies coherently with atmospheric circulations on different scales. Knowland et al. (2015) demonstrated the important role of mid-latitude storms in redistributing O_3 concentrations during springtime. The North Atlantic Oscillation (NAO) significantly affects surface and tropospheric O_3 concentrations over most of Europe by influencing the intercontinental transport of air masses (Creilson et al., 2003; Christoudias et al., 2012; Pausata et al., 2012). Lamarque and Hess (2004) indicated that the Arctic Oscillation (AO) can modulate springtime tropospheric O_3 burdens over North America. The shift in the position of the jet stream associated with climate change was found to strongly affect summertime surface O_3 variability over eastern North America (Barnes and Fiore, 2013). Increases in solar radiation and air temperature can increase the rate of the chemical production of O_3 and modulate the biogenic emissions of O_3 precursors (Guenther, 1993; Sillman and Samson, 1995; Peñuelas and Llusià, 2001), especially over highly polluted regions (Ordónez et al., 2005; Rasmussen et al., 2012; Pusede et al., 2015). Increases in humidity can enhance the chemical destruction of O_3 and shorten its atmospheric lifetime (Johnson et al., 1999; Camalier et al., 2007). Therefore, changes in meteorological conditions on various spatial and temporal scales play key roles in determining the surface O_3 distribution. Understanding the mechanisms and feedbacks of the interactions between O_3 and climate has received increasing attention and will be essential for future surface O_3 mitigation (Jacob and Winner, 2009; Doherty et al., 2013).

Sea surface temperature (SST) is an important indicator that characterizes the state of the climate system. Its variations strongly perturb the mass and energy exchange between the ocean and atmosphere (Small et al., 2008; Gulev et al., 2013), which ~~further~~ influence atmospheric circulation, ~~solar radiation,~~ atmospheric temperature and specific humidity (Sutton and Hodson, 2005; Frankignoul and Sennéchael, 2007; Li et al., 2008) from regional to global scales (Glantz et al., 1991; Wang et al., 2000; Goswami et al., 2006). It also affects cloud formation and consequently influences incoming solar radiation (Deser et al., 1993; Fallmann et al., 2017). Numerous studies have shown that SST changes over different oceans and at different latitudes lead to significantly different meteorological and climate responses (Webster, 1981; Lau and Nath, 1994; Lau, 1997; Sutton and Hodson, 2007; Sabeerali et al., 2012; Ueda et al., 2015). Details on the SST-climate relationships over individual oceanic regions are summarized in Kushnir et al. (2002).

The Intergovernmental Panel on Climate Change Fifth Assessment Report (IPCC, 2013) provides strong evidences in Chapter 2 that global SSTs are generally increasing due to the impacts of anthropogenic forcings on global climate change ~~(IPCC, 2013, Chapter 2).~~. In addition, regional SST exhibits natural periodic or irregular oscillations with timescales ranging from months to decades. The El Niño/Southern Oscillation (ENSO) is the most influential natural SST variability that originates in the tropical Pacific and has worldwide climate impacts (Philander, 1983; Wang et al., 2012). The Pacific decadal oscillation (PDO), defined by ocean temperature anomalies in the northeast and tropical Pacific Ocean, is another long-lived, El Niño-like pattern that persists for several decades (Mantua and Hare, 2002). Over the Indian Ocean, SST anomalies feature a seesaw structure between the western and eastern equatorial regions, known as the Indian Ocean Dipole (IOD) mode (Saji et al., 1999). The North Atlantic Ocean ~~pronounces~~exhibits various modes of low-frequency SST variability (Kushnir, 1994;

Wu and Liu, 2005; Fan and Schneider, 2012; Taboada and Anadon, 2012). The mechanisms responsible for SST variability includes ocean circulation variability, wind stress, and ocean-atmosphere feedbacks (Frankignoul, 1985; Deser et al., 2010). Aerosols and greenhouse gases (GHGs) emitted from anthropogenic and natural sources also contribute to regional SST variability through modulation of the solar radiation received by the ocean surface (Rotstayn and Lohmann, 2002; Wu and Kinter, 2011; Hsieh et al., 2013; Ding et al., 2014; Meehl et al., 2015).

Considering the distinct roles of regional SST variability in modulating regional climate systems, the impact of regional SST changes on the surface O_3 distribution needs to be explored. Lin et al. (2015) found that more frequent deep stratospheric intrusions appear over the western US during strong La Niña springs because of the meandering of the polar jet towards this region. This process can remarkably increase surface O_3 levels in the western US. The La Niña-like decadal cooling of the eastern equatorial Pacific Ocean in the 2000s weakened the long range transport of O_3-rich air from Eurasia towards Hawaii during spring (Lin et al., 2014). Liu et al. (2005) revealed that El Niño winters are associated with stronger transpacific pollutant transport, which also has implications for the long-range transport of O_3. Except for the ENSO impacts, very few studies to date have directly addressed the linkage between SST and O_3. Therefore, a comprehensive understanding of the response of surface O_3 to SST changes in individual ocean basins is lacking and necessary.

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), defined in Fiore et al. (2009)) with respect to nearby basin-scale SST changes. We describe the design of numerical experiments and model configuration in Section 2. Surface O_3 responses to regional SST changes are given in Section 3. Relevant mechanisms governing the SST-O_3 relationships are discussed in Section 4. The impact of basin-scale SST changes on inter-continental transport of O_3 is described in Section 5. Conclusions are drawn in Section 6.

2. Methodology

2.1 Model description and configuration

The Community Earth System Model (CESM, v1.2.2) developed by the National Center for Atmospheric Research (NCAR) is used in this study, configured with the Community Atmosphere Model version 5.0 (CAM5) and the Community Land Model version 4.0 (CLM4). The ocean and sea ice components are prescribed with climatological SST and sea ice distributions. Moist turbulence is parameterized following the Bretherton and Park (2009) scheme. Shallow convection is parameterized using the Park and Bretherton (2009) scheme. The parameterization of deep convection is based on Zhang and McFarlane (1995) with modifications following Richter and Rasch (2008), Raymond and Blyth (1986), and Raymond and Blyth (1992). The cloud microphysical parameterization is following a two-moment scheme described in Morrison and Gettelman (2008) and Gettelman et al. (2008). The microphysical effect of aerosols on clouds are simulated following Ghan et al. (2012). The parameterization of cloud macrophysics follows Conley et al. (2012).

The chemistry coupled in the CAM5 (i.e., CAM5-chem) is primarily based on the Model for O_3 and Related chemical Tracers, version 4 (MOZART-4), which resolves 85 gas-phase species, and 196 gas-phase reactions (Emmons et al., 2010; Lamarque et al., 2012). A three-mode (i.e., Aitkin, accumulation and course) aerosol scheme for black carbon (BC), primary organic matter (POM), second organic aerosol (SOA), sea salt, dust and sulfate was used in our simulations following Liu and Ghan (2010). The lightning parameterization is modified according to Price et al. (1997) and tropospheric photolysis rates are calculated interactively following Tie et al. (2005). Gaseous dry deposition is calculated using the resistance-based parameterization of Wesely (1989), Walmsley and Wesely (1996), and Wesely and Hicks (2000). The parameterizations of in-cloud scavenging and below-cloud washout for soluble species are described in detail by Giorgi and Chameides (1985) and Brasseur et al. (1998), respectively. Anthropogenic emissions of chemical species are from the IPCC AR5 emission datasets (Lamarque et al., 2010), whose injection heights and particle size distributions follow the AEROCOM protocols (Dentener et al., 2006). The emissions of natural aerosols and precursor gases are prescribed from the MOZART-2 (Horowitz et al., 2003) and MOZART-4 (Emmons et al., 2010) datasets. All emission datasets are available from the CESM data inventory (https://svn-ccsm-inputdata.cgd.ucar.edu/trunk/inputdata/). The performance of CESM in simulating tropospheric O_3 has been validated by comparing with ozonesondes and satellite observations (Tilmes et al., 2014). The deviations between model and observations are within the range of about 25%. In general, the model can capture the surface ozone distribution and variability well, but may overestimate O_3 over the Eastern US and Western Europe in the summer (Tilmes et al., 2014).

2.2 Numerical experiments

We first conduct a control simulation, hereafter referred to as CTRL, with prescribed climatological monthly SSTs averaged from 1981 to 2010 (see Hurrell et al. (2008)). We then conduct six perturbation simulations with monthly SSTs that are 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), North Atlantic (15°N-65°N; 100°W-20°E) and North Indian Oceans (5°N-30°N, 30°E-100°E; here 5°N is used to attain a relatively larger domain size). The simulations are denoted as “Pacific-W”, “Atlantic-W” and “Indian-W” for the three warming cases and “Pacific-C”, “Atlantic-C” and “Indian-C” for the three cooling cases. We defined the latitudinal and longitudinal ranges of these ocean basins mainly based on their geographical features. The boundaries of the prescribed SST anomalies generally align with the edge of the ocean basins, except along the southern side. In each perturbation simulation, we ~~further~~ linearly smooth the southern boundaries of these SST anomalies towards the equator to remove the sharp SST anomaly gradients at the edge~~,~~, following a previous approach (e.g., Taschetto et al., 2016; Seager and Henderson, 2016). 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. Air pollution emissions, including biogenic emissions of VOCs, are fixed to distinguish the impacts of SST variation on O_3 transport and photochemistry. All simulations are run for 21 years with the first year used for model spin-up.

To explore the impacts of SST changes on inter-continental transport, an explicit emission tagging technique is used in our simulations following previous studies (Shindell et al., 2008; Doherty et al., 2013). Artificial CO-like tracers emitted from four continental regions, 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), are tracked individually. These tracers are idealized with a first-order decay lifetime of 50 days, which is similar to O_3 (Doherty et al., 2013) and used to single out changes in O_3 transport induced by SST anomalies.

2.3 Integrated process rate (IPR) analysis

To provide a process-level explanation for the response of surface O_3 to regional SST changes, the IPR method is applied. This method calculates the accumulated contributions of individual processes (e.g., chemical production and loss, advection, vertical diffusion, dry deposition, etc.) to O_3 predictions during the model simulation and has been widely used for air pollution diagnostics (Li et al., 2012; Zhang and Wu, 2013; Tao et al., 2015). In this study, we added the IPR scheme to the CESM framework to track the contribution of six physicochemical processes (i.e., gas-phase chemistry (CHEM), advection (ADVE), vertical diffusion (VDIF), dry deposition (DRYD), shallow convection (SHAL) and deep convection (DEEP)) to O_3 concentrations in every grid box. Wet deposition and aqueous-phase chemistry are ignored here due to the low solubility and negligible chemical production of O_3 in water (Jacob, 1999). Therefore, CHEM represents the net production (production minus loss) rate of O_3 due to gas-phase photochemistry. DRYD represents the dry deposition fluxes of O_3, which is an important sink for O_3. The other IPR terms (i.e., ADVE, VDIF, SHAL and DEEP) represent contributions from different transport processes. The IPR scheme tracks and archives the O_3 flux in each grid from every process during each model time step. The sum of the O_3 fluxes from these six processes matches the change in the O_3 concentration. The IPR method has been widely used in air quality studies to examine the cause of pollution episodes (Wang et al., 2010; Li et al., 2012). When applied in climate sensitivity analysis (usually measuring the difference between two equilibriums), the net change of all IPRs approaches zero. Typically, the positive changes in IPRs are mainly responsible for the increase in surface O_3, which may further induce O_3 removal to balance this forcing in a new equilibrium. Therefore, here, the IPR analysis is used not to budget the SST-induced O_3 concentration changes but rather to help examine the relative importance of different transport and chemical processes in driving the sensitivity of O_3 to SST forcing. ~~The IPR~~Its performance is verified by comparing the predicted hourly O_3 changes with the sum of the individual fluxes from the six processes. As shown in Figure S1, the hourly surface O_3 changes are well represented by the sum of these fluxes in the model.

3. Response of surface O_3 concentrations to SST changes

Seasonally ~~(i.e., DJF (December, January, February), MAM (March, April, May), JJA (June, July, August) and SON (September, October, November))~~ and regionally averaged surface O_3 changes in each SST perturbation simulation for the four highly populated continental regions and three ocean basins defined in our study are given in Tables 1 and S1, respectively. The responses of the surface O_3 concentrations to basin-scale SST changes (i.e., ± 1 °C) are mainly below 3 ppbv in the Northern Hemisphere (Tables 1 and S1), though larger anomalies (i.e., up to 5 ppbv) are also observed over the eastern coast of China, the Indian subcontinent, and certain oceanic areas (Figures 1 and ~~S3~~S2). This SST-O3 sensitivity is comparable to previous findings. For instance, Bloomer et al. (2009) reported a positive O_3-temperature relationship of 2.2~3.2 ppbv/°C across the rural eastern United States. Wu et al. (2008) found that summertime surface O_3 may increase by 2-5 ppbv over the northeastern United States in the 2050s. Additionally, Fiore et al. (2009) demonstrated an intercontinental decrease in surface O_3 of no more than 1 ppbv in response to 20 % reductions in anthropogenic emissions within a continental region. Our study indicates that basin-scale SST changes alone may exert significant effects on the surface O_3 above specific ocean basin and its surrounding continents.

As shown in Figure 1, seasonal changes of up to 5 ppbv in the mean surface O_3 concentration are observed during boreal summers, mainly in coastal regions and remote oceans. Surface O_3 changes in response to positive and negative SST anomalies generally ~~pronounce~~exhibit a consistent spatial pattern but are opposite in sign, suggesting robust relationships between surface O_3 levels and SST anomalies (Figure 1). An increase in summertime SST over a specific ocean basin tends to increase the surface O_3 concentration over the upwind regions but reduce this concentration over downwind continents. For instance, a 1 °C warming over the North Pacific leads to a widespread decrease in surface O_3 over the North Pacific, North America and the North Atlantic of approximately 1 ppbv (Table S1) but may enhance the surface O_3 by nearly 3 ppbv over South China. Similarly, ~~in~~ the “SST warming over the North Atlantic~~ W~~” ~~case,~~ decrease the surface O_3 levels ~~decrease~~ by 1~2 ppbv over the North Atlantic and Europe but increase (~1 ppbv) that over North America and the North Pacific. For the North Indian Ocean, positive SST anomalies tend to increase the surface O_3 over the Indian Ocean and Africa but decrease the surface O_3 over South and East Asia (Figure 1). During the boreal winter, a widespread decrease in surface O_3 associated with the warming of different oceans is observed. Significant changes (e.g., up to 5 ppbv) mainly occur over remote ocean areas. Over populated continents, the response of the surface O_3 to basin-scale SST changes is typically insignificant. Details are shown in Figure ~~S3~~S2 in the supplementary material.

Our simulations reveal that different oceans can exert distinct region-specific effects on the O_3 distribution. The effects of three individual warming/cooling cases (i.e., Pacific-

W, Atlantic-W and Indian-W/Pacific-C, Atlantic-C and Indian-C) on surface O_3 distributions are further summed up to compare with the combined warming/cooling cases(i.e., ALL-W/ALL-C).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. The effects of these combined warming and cooling cases on surface O_3 distributions are respectively compared with the sum of the three individual warming cases (i.e., Pacific-W, Atlantic-W and Indian-W) and three individual cooling cases (i.e., Pacific-C, Atlantic-C and Indian-C). The responses of surface O_3 to a hemispheric SST anomaly generally resemble the sum of responses to differentindividual regional SST changes (see Figures S5S3 and S7S4 in the supplementary material). This indicates that the effect of a generalized SST warming on surface O_3 can be decomposed into individual regional SST forcings. We now analyze the processes that impact the dependence of SST on the O_3 distribution using simulations that increase the SST.

4. Mechanism of SST-induced surface O_3 changes

4.1 Process-level response to SST changes

In this study, IPR analysis is used to evaluate the contribution of different physicochemical processes to O_3 evolution. This type of analysis has been widely used in air quality studies to examine the cause of pollution episodes (Wang et al., 2010;Li et al., 2012)The. When applied in climate sensitivity analysis (usually measuring the difference between two equilibriums), the net change of all IPRs approaches zero. Typically, the positive changes in IPRs are mainly responsible for the increase in surface O_3, which may further induce O_3 removal to balance this forcing in a new equilibrium. Therefore, here, the IPR analysis is used not to budget the SST-induced O_3 concentration changes but rather to help examine the relative importance of different transport and chemical processes in driving the sensitivity of O_3 to SST forcing. In this study, the SST-induced, process-level O_3 changes are spatially averaged over four populated continental regions (i.e., NA, EU, EA and SA, Figure 2) and three ocean basins (i.e., the North Pacific, North Atlantic and North Indian Oceans, Figure ~~S9~~S5). In most cases, VDIF and DRYD are the key processes controlling the O_3 variation. The downward transport of O_3 through diffusion (VDIF) is an important source of surface O_3, while DRYD acts as a sink. Both processes are simultaneously determined by the strength of turbulence. Here, we define a new term TURB as the sum of DRYD and VDIF, which can capture the overall effect of turbulence changes on surface O_3 concentrations. In addition, we merge SHAL and DEEP as CONV to represent the total contribution of convective transport to surface O_3 (Figures 2 and ~~S9~~S5). More detailed IPR results are shown in Figures ~~S10~~S6 and ~~S11~~S7 in the supplementary material.

In the "Pacific-W" case, a 1 °C SST warming over the North Pacific increases VDIF over eastern China in JJA (Figure ~~S12~~S8), which is insignificant if averaged over the whole East Asia region. Meanwhile, this Pacific warming considerably reduces VDIF over North America (Figure ~~S10~~S6). The corresponding decrease in TURB over North America mainly determines the surface O_3 reduction in JJA and SON, while the reduction in CONV exerts an additional negative impact (Figure 2). In the "Atlantic-W" case, increases in VDIF are also observed over the upwind regions (i.e., North America) in JJA. However, these increases are accompanied by commensurate decreases in DRYD, resulting in an insignificant overall change in TURB (Figure 2). Therefore, the increase in CHEM is mainly responsible for the surface O_3 increase over North America in JJA. TURB is more relatively important over Europe (only in JJA and SON), leading to reduced surface O_3 abundance. In the "Indian-W" case, both CHEM and CONV are reduced over South Asia in JJA, leading to overall reductions in surface O_3 over the Indian subcontinent (Figure 2). The IPR analysis over the ocean basins shows that the warming of the North Pacific or North Atlantic induces reductions in VDIF and CHEM, which are responsible for the significant decrease in surface O_3 above these regions in JJA (Figure ~~S11~~S7). The North Indian Ocean warming, on the other hand, enhances DEEP and VDIF, leading to a local increase in surface O_3 in JJA.

The IPR analysis indicates that, in general, an SST increase in the North Pacific or

North Atlantic is more likely to enhance the vertical diffusion of O_3 over upwind regions (i.e., East Asia or North America, respectively) but suppress this diffusion over the ocean basin as well as downwind continents in JJA (Figure ~~S12~~S8). These opposite changes in VDIF over upwind and downwind regions lead to distinct surface O_3 responses. Changes in CHEM enhance surface O_3 formation in most cases. An exception is in South Asia, where CHEM and DEEP dominate the reduction in surface O_3 over the region in JJA associated with the North Indian Ocean warming. In the following subsections, the mechanisms of the SST-O_3 relationship for the four polluted continents are further explored. Here we focus on boreal summers since the surface O_3 response to SST changes is more robust during this period than other seasons.

4.2 Response of photochemical O_3 production to SST increases

Changes in the net production rate (i.e., chemical production rate minus loss rate) of O_3 at the surface in JJA associated with basin-scale SST increases are shown in Figure 3. The peak changes are mainly confined to regions where O_3 precursors are abundant (e.g., South and East Asia and North America). For example, a warmer North Pacific SST exerts a positive (negative) impact on net O_3 production in the northern (southern) regions of East Asia. Similarly, the warming of the North Atlantic promotes a dipole impact on the surface O_3 production over North America, while the warming of the North Indian Ocean significantly decreases the net O_3 production rate over South Asia.

As emissions are fixed in all simulations, the change in net O_3 production is driven by SST induced meteorological changes (e.g., air temperature, air humidity, and solar radiation). An increase in SST of 1 °C in any ocean basin leads to a widespread enhancement of the surface air temperature (i.e., the air temperature at 2 m) over most continental areas (Figure 4). An exception is the North Indian Ocean, where an increase in SST tends to cool the Indian subcontinent by 1-2 °C. This temperature decrease is not only limited to the surface but also spreads to 600 hPa (Figure ~~S16~~S9). Associated with this temperature decrease is a remarkable reduction in the solar radiation received at the continent belowsurface (more than 15 W/m^2, Figure S17S10). Previous studies have indicated that moist convection is more sensitive to the SST changes in the tropical oceans than in mid- or high- latitude oceans (Lau and Nath, 1994; Lau et al., 1997; Hartmann, 2015). The SST increase over the North Indian Ocean tends to strengthen the moist convection that eventually facilitates cloud formation in the upper troposphere (Roxy et al., 2015; Xi et al., 2015; Chaudhari et al., 2016). The latent heat released from convective activities significantly warms the air temperature over the upper troposphere (Sabeerali et al., 2012; Xi et al., 2015). Meanwhile, the corresponding increase in cloud cover blocksreduces the solar radiation reaching the surface of the Indian subcontinent and reducethus the air temperature of lower troposphere in that region. These processes lead to opposite air temperature changes between upper and lower troposphere over South Asia in response to the North Indian warming (as shown in Figure S16S9), which may further suppress the development of deep convection over the Indian subcontinent.

Previous studies have indicated that air temperature positively affects both O_3 production and destruction rates (Zeng et al., 2008; Pusede et al., 2015). As shown in Figure S19S11, changes in the net O_3 production rate are mainly dominated by O_3 production over continents but by O_3 destruction over oceans. An increase in SST leads to a widespread enhancement of the air temperature, resulting in a positive change in the net O_3 production over most continental regions (Figure 3). However, a warmer SST also increases the air humidity (Figure S21S12), which enhances O_3 destruction over most coastal and oceanic areas. In addition, over South Asia, a warming of the North Indian Ocean decreases solar radiation and air temperature, and simultaneously increases air humidity, which jointly exert negative effects on O_3 production in that region.

4.3 Response of physical O_3 transport to SST increases

In Section 4.1, our IPR analysis highlights multiple physical processes (i.e., vertical diffusion, convection and advection) that are important in modulating surface O_3

concentrations. However, the role and relative importance of each process exhibit large spatial heterogeneity. In this section, we explore the key factors controlling physical O_3 transport in response to basin-scale SST changes.

The changes in the surface pressure and wind pattern induced by a basin-wide SST increase are shown in Figure 5. Generally, a warming of any ocean basin will lead to a low-pressure anomaly centered to its west at low-latitudes, which is caused by SST-induced convective activity. Additionally, the warming of the Indian Ocean induces an anticyclonic anomaly over the subtropical western Pacific, which has been documented in previous studies (Yang et al., 2007; Li et al., 2008). As shown in Figure 6, the surface pressure reduction induced by SST warming in any ocean basin is closely associated with enhanced upward motions, suggesting a substantial enhancement in deep convection over tropical oceans. Previous studies have identified an SST threshold (approximately 26°−28°C) to generating deep convection (Graham and Barnett, 1987; Johnson and Xie, 2010). Therefore, the sensitivity of deep convection to an SST anomaly is strongly dependent on the distribution of base SST. The enhanced upward motion in response to a uniform increase in basin-scale SST mainly occurs over regions with high climatological SST (Figure 6). Regions with a low climatological SST have little effects on the vertical movement of air masses.

Strengthened deep convection will trigger large-scale subsidence over nearby regions through the modulation of large-scale circulation patterns, which may suppress convective transport (Lau et al., 1997; Roxy et al., 2015; Ueda et al., 2015). This effect is verified by the decreases in upward velocity at 500 hPa. As depicted in Figure 6, significant decreases in upward velocity occur over regions adjacent to the strengthened deep convection. Similar effects are also observed over higher latitudes or remote oceans (Figure ~~S23~~S13). Meanwhile, the air temperature increase in response to regional SST warming is more significant above the lower troposphere, which leads to a decrease in the vertical temperature gradient (Figure ~~S16~~S9). These factors tend to restrain the vertical exchange of air pollutants at mid-latitudes, which facilitates surface

O_3 accumulation over polluted continental regions in JJA but may weaken the intrusion of O_3 from the upper troposphere to the surface in most unpolluted areas. This process helps to explain the widespread decrease in surface O_3 over unpolluted regions associated with an SST increase, as described in Section 3, and can be further verified by the wide-spread reduction in VDIF shown in Figure S12S8.

The surface pressure anomalies induced by SST changes can play a dominant role in modulating surface O_3 transport at specific locations. For example, the low-pressure anomaly centered over the subtropical northwestern Pacific in the "Pacific-W" case causes the convergence of wind in the lower troposphere (Figure 5a). Consequently, surface O_3 pollution is enhanced in southern China due to an increase in O_3 transport from more polluted northern China (Figure 7a). The vertical distribution of the corresponding O_3 changes also shows that the increase in O_3 over southern China occurs below 700hPa, accompanied by noticeable decreases above 700hPa as well as over nearby northern China (Figure 7d). The IPR analysis also indicates that the increases in advective transport and downward turbulent transport are mainly responsible for the surface O_3 increase in southern China.

In the "Atlantic-W" case, the SST warming-induced surface pressure anomalies lead to substantial O_3 redistribution, especially over the North Atlantic Ocean (Figure 7b). For North America, the changes in horizontal O_3 fluxes have no significant effect on the O_3 concentration increase. In addition, O_3 changes are observed to be larger in the upper troposphere than at the surface (Figure 7e). As demonstrated in Section 4.1, the response of lower-altitude O_3 over North America to the North Atlantic warming is mainly caused by enhanced chemical production, rather than physical transport.

The North Indian SST warming leads to a low-pressure anomaly centered over the Arabian Sea (Figure 5c). The warming of the North Indian Ocean strengthens the upward motion of air at low-latitudes and further induces a convergence of highly polluted air over the Indian Ocean. The effects of this process on O_3 concentrations are observed to be more significant in the upper troposphere (Figure 7f). According to the IPR analysis, the surface O_3 increase over the Indian Ocean is mainly caused by the enhanced vertical transport of O_3 to the surface through deep convection and vertical diffusion processes (Figure ~~S11~~S7). However, over the nearby Indian subcontinent, the suppressed convection tends to decrease surface O_3 in that region (Figure 2).

5. Implications for O_3 long-range transport

The above findings indicate that, in general, a basin-scale SST increase in the Northern Hemisphere is more likely to enhance atmospheric stability at mid-latitudes, which may suppress air pollutants from lofting to the free troposphere. This process potentially has large effects on O_3 intercontinental transport. Following previous work (e.g., Doherty et al., 2013; Fang et al., 2011), we use passive CO-like tracers to demonstrate the potential effect of regional SST changes on long-range O_3 transport. A warming of North Pacific SSTs by 1°C tends to increase the East Asian CO tracer concentrations by nearly 6% at the surface (Figure 8b), which is accompanied by a significant reduction (~4%) in eastward transport to North America. Similarly, for the North American tracer, a warming of North Atlantic SSTs by 1°C increases (~1%) the concentrations in North America but decreases (3-4 %) the concentrations over downwind Europe (Figure 8d). The response of the South Asian CO tracer to North Indian Ocean warming also shows a decreasing tendency over downwind regions, but the patterns are more complicated over the source region in this case (Figure 8e). Because the CO-like tracers added in the simulation have a fixed decay lifetime, their concentration changes are completely caused by the SST-induced transport anomalies. The decrease in CO tracer concentrations over downwind regions suggests that the warming of basin-scale SST tends to suppress the long-range transport of air pollutants. Additionally, in the "Pacific-W" case, changes in the East Asian CO tracer (Figure 8a) generally resemble the changes in surface O_3 over East Asia (Figure 7a), indicating the dominant effect of physical transport on the O_3 distribution over East Asia. Regarding the North American CO tracer in response to the North Atlantic warming or the South

Asian CO tracer in response to the North Indian Ocean warming, their concentration changes are spatially inconsistent with those of O_3 (see Figures 7 and 8). This further indicates the distinct roles that different basin-scale SSTs play in nearby air quality.

Further investigations of zonal wind suggest that an increase in SST over different oceans consistently decreases the westerly winds at lower mid-latitudes (25°N - 45 °N) in the Northern Hemisphere but increases these winds at higher latitudes (Figure 9). In general, increases in the geopotential height induced by basin-scale SST warming are more significant at mid-latitudes than at other latitudes, which is consistent with the air temperature changes. Consequently, the meridional geopotential height gradient is decreasing at lower latitudes but increasing at higher latitudes, leading to corresponding changes in the westerly winds. The latitude band at 25°N - 45 °N covers many polluted regions (i.e., North America and East Asia). A weakened westerly wind may reduce long-rang O_3 transport. As demonstrated in Section 4.3, the basin-scale SST increases also exert negative effects on the upward transport of air masses at mid-latitudes. Therefore, the decreases in CO tracer concentrations over downwind regions (Figure 8a and 8c) can be explained by both suppressed vertical transport and weakened westerly winds. In the “Indian-W” case, the SST increase over North India leads to a low-pressure anomaly above the Arabian Sea due to the enhanced deep convection (as discussed in Section 4.3). The corresponding anomalous ~~cyclone should be~~cyclonic circulation may be responsible for the dipole of the South Asian CO tracer changes over the source region depicted in Figure 8e.

In addition, we also find a hemispheric-scale decrease in peroxyacetyl nitrate (PAN), a reservoir of O_3 precursors (NO_X and HO_X) that facilitates the long-range transport of O_3, during the warming of different oceans (Figure ~~S25~~S14). This decrease is likely caused by the increase in the thermal decomposition of PAN in response to the air temperature increase (Jacob and Winner, 2009; Doherty et al., 2013).

Thus, it is reasonable to infer that, in general, the increased thermal decomposition of

PAN, the weakened mid-latitude westerlies, and the reduced vertical air transport may exert a joint reducing effect on the intercontinental transport of O_3 during basin-scale SST increases.

6. Summary

In this paper, we investigate the responses of surface O_3 to basin-scale SST anomalies in the Northern Hemisphere. The latest version of CESM (version 1.2.2) is used in our simulation, forced with climatological and stationary SST anomalies (± 1 °C) in the North Pacific, North Atlantic and North Indian Oceans, respectively. The responses of surface O_3 associated with these SST changes are evaluated. Results of similar magnitude but opposite sign are observed for the SST warming versus cooling simulations for each ocean basin, suggesting robust connections between the SST anomalies and surface O_3 changes. The regionally and seasonally averaged surface O_3 changes over four continental regions (i.e., NA, EU, EA and SA) ~~pronounce~~produce wide seasonal and regional variability (varying from 1 to 3 ppbv). The warming of the North Pacific leads to nearly 3 ppbv increases in the surface O_3 over southern China in summer, with corresponding decreases over North America (~1 ppbv). Similarly, the North Atlantic SST warming elevates the surface O_3 pollution over North America while reducing the surface O_3 (nearly 1-2 ppbv) over Europe. Changes in the North Indian SST exert significant impacts (1-3 ppbv) over South and East Asia during the entire year.

Process analysis indicates that dry deposition and vertical diffusion are two major processes governing the surface O_3 balance. The increase in SST in different ocean basins tends to increase the contributions of vertical diffusion to surface O_3 over upwind regions while greatly restraining that over downwind continents. These processes generally lead to widespread decreases in surface O_3, which are partially offset by increases in air temperature-dependent chemical production rates. Specifically, the chemical production changes are mainly responsible for the surface O_3 increases over

North America in response to the North Atlantic SST warming but exert a negative effect on South Asia in response to the North Indian SST warming. Decreases in the convective transport of O_3 to the surface associated with North Indian warming are significant over South Asia and exert a negative impact on surface O_3 concentrations. Advective transport has a positive effect on surface O_3 in southern China in the "Pacific-W" case.

We further show that air temperature is an important factor controlling the surface O_3 responses to SST anomalies. Reductions in the surface O_3 chemical production in South Asia associated with North Indian SST warming can be explained by the corresponding SST-induced decreases in ground-level air temperature and solar radiation. Meanwhile, the widespread increase in air temperature associated with basin-scale SST warming is more likely to promote O_3 production over other highly polluted regions.

On the other hand, SST increases at low latitudes over different oceans enhance deep convection in summer, which promotes convergence at the surface, as well as upward motions at low latitudes. The corresponding surface pressure anomalies centered over the east coast of East Asia associated with the North Pacific warming and over the Arabian Sea associated with the North Indian warming tend to increase the surface O_3 above through exchanges with the surrounding highly polluted air. The basin-scale SST increases in the Northern Hemisphere reduce the tropospheric temperature gradient at mid-latitudes that restrains vertical transport of O_3 over continents and weakens the westerlies at lower mid-latitudes. The response of the CO-tracer also suggests that these factors may jointly exert a negative effect on the intercontinental transport of O_3.

~~Overall, our~~This 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 this evolution. Idealized and spatially uniform SST anomalies are used to explore the general mechanisms governing SST-O_3 relationships. We find that the SST changes over tropical regions exert considerable impacts on surface O_3 levels.

The increase in tropical SST over different ocean basins enhances deep convection, which further trigger large-scale subsidence over nearby and remote regions. These enhanced convective activities also tend to release more latent heat over the upper troposphere and significantly increases the air temperature there. These processes influence large-scale circulation patterns and lead to opposite surface O_3 responses over upwind and downwind regions related to a specific ocean basin. These finding provides valuable implications for the potential surface O_3 change in response to future warming or cooling of individual oceans.

Additionally, the sensitivity tests with 1ºC SST warming and cooling superimposed onto all three ocean basins further show in general that the SST forcing on O_3 distribution is geographically additive. A number of studies have used the decomposed SST anomalies for different regions to identify their relevant roles in a particular climate response (e.g., Sutton and Hodson, 2005; Camargo et al., 2013; Ueda et al., 2015). A linear assumption that the influence of large-scale 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 (e.g., Fan et al., 2016; Seager and Henderson, 2016). Therefore, our study also helps to understand the roles of different ocean basins in the Northern Hemisphere played in modulating surface O_3 distributions in a global-wide SST warming condition associated with climate change.

Overall, this study may guide the management of regional O_3 pollution by considering the influence of specific SST variability. However, to provide a more comprehensive understanding of the SST-O_3 relationship, further studies using realistic SST variability are necessary. This study may aid in the management of O_3 pollution by considering the influence of specific SST variability.cautions should be taken in interpreting our results in the real world since observed surface O_3 variabilities are induced by various factors including O_3 precursor emissions and atmospheric conditions. Realistic SST anomalies over different oceans are more complicate (usually not uniformly distributed) and often inter-correlated with each other (Fan et al., 2016). They may exert jointly effects on modulating surface O_3 distributions. To provide more precise understanding about the SST-O_3 relationship over a specific region, additional sensitivity tests regarding smaller patches of SST variability are necessary.

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Table 1. ~~Regionally and s~~Seasonally (i.e., DJF (December, January, February), MAM (March, April, May), JJA (June, July, August) and SON (September, October, November)) and regionally averaged (only land grid boxes are included) changes in surface O_3 concentrations (ppbv) for basin-scale SST perturbation cases relative to the control simulation. Positive (negative) changes that are significant at the 0.05 level evaluated by Student's t-test are marked in red (blue).

Ozone (ppbv)			DJF	MAM	JJA	SON
North Pacific	+1° C	North America	-0.27*	-0.42*	-0.92*	-1.03*
		Europe	-0.50*	-0.26	0.10	-0.29
		East Asia	-0.88*	-0.71*	0.20	0.17
		South Asia	-1.00*	0.30	0.43	0.43*
	-1° C	North America	0.00	0.57*	0.55*	0.82*
		Europe	0.19	0.15	-0.47*	0.47*
		East Asia	0.30	-0.17	-0.22	-0.67*
		South Asia	0.04	-0.24	0.03	-0.40
North Atlantic	+1° C	North America	0.03	0.49	0.50*	0.53*
		Europe	0.30*	0.06	-1.61*	-0.89*
		East Asia	-0.52*	-0.68*	-0.62*	-0.25
		South Asia	-0.20	-1.46*	-1.28*	-0.82*
	-1° C	North America	-0.07	-0.10	0.10	-0.17
		Europe	0.00	0.00	0.07	0.06
		East Asia	0.16	-0.08	0.80*	-0.60*
		South Asia	-0.20	-0.40	0.30	-0.10
North India	+1° C	North America	-0.25	-0.04	-0.16	-0.10
		Europe	-0.30	0.08	-0.12	0.19
		East Asia	-0.53*	-0.77*	-0.28	-1.78*
		South Asia	-1.00*	0.14	-1.67*	-2.75*
	-1° C	North America	0.04	0.17	0.04	0.25
		Europe	0.05	-0.07	-0.13	-0.24
		East Asia	-0.06	0.15	0.55*	0.33
		South Asia	-0.03	0.57	1.70*	1.31*

*Significant at the 0.05 level from Student's t-test using 20 years of model results.

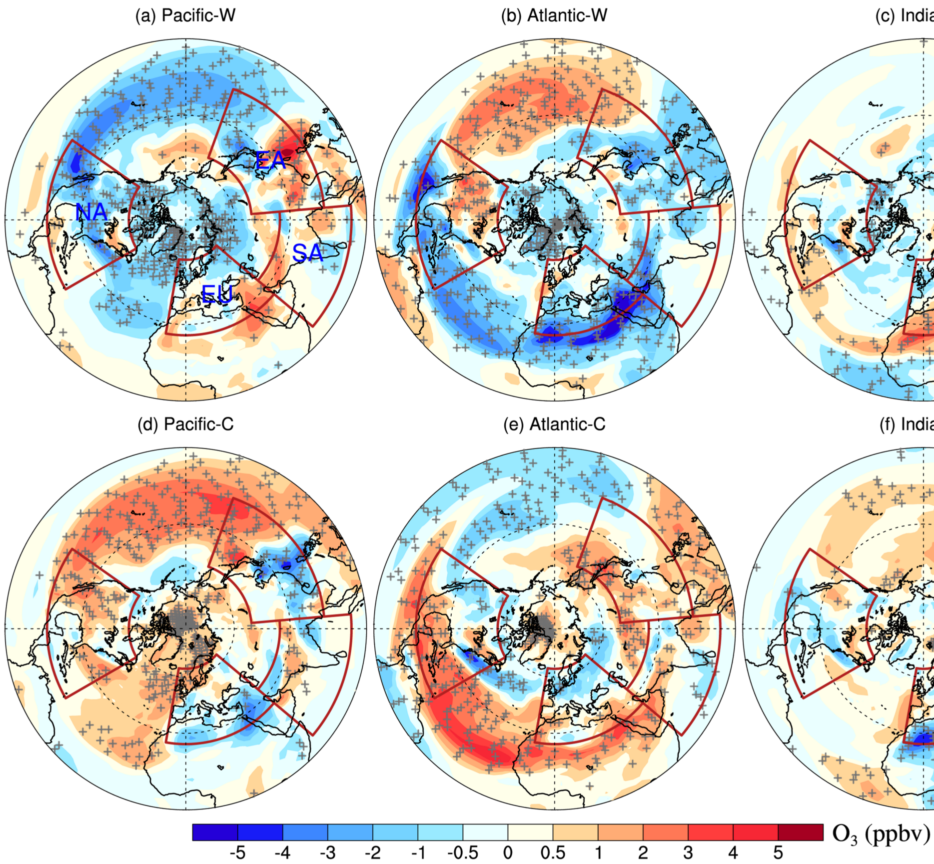

Figure 1. Changes in the summertime (June-August) surface O_3 concentrations (ppbv) in the Northern Hemisphere induced by 1ºC warming (top) and 1°C cooling (bottom) in the North Pacific Ocean (left), North Atlantic Ocean (center), and North Indian Ocean (right) relative to the CTRL. The four major regions 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. The + symbols denote areas where results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data~~ (plots using the Mercator projection are shown in Figure S2 in the supplementary material).~~.

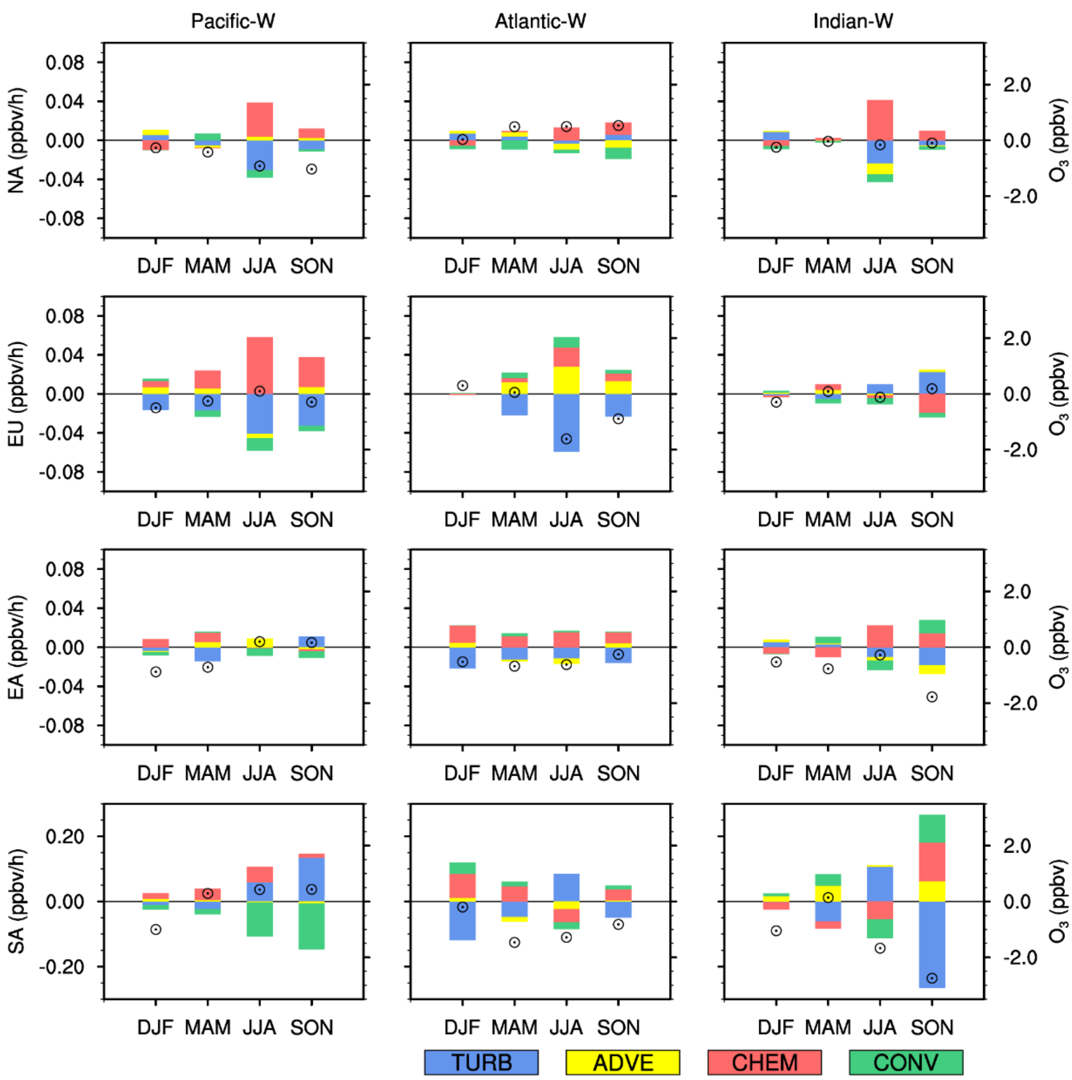

Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O_3 concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second row), EA (third row) and SA (last row). TURB is defined as the sum of VDIF and DRYD. CONV is the sum of DEEP and SHAL. IPR contributions from the four processes (i.e., TURB, ADVE, CHEM and CONV) are represented by different colors. A more detailed IPR result is shown in Figure S6~~10~~ in the supplementary material.

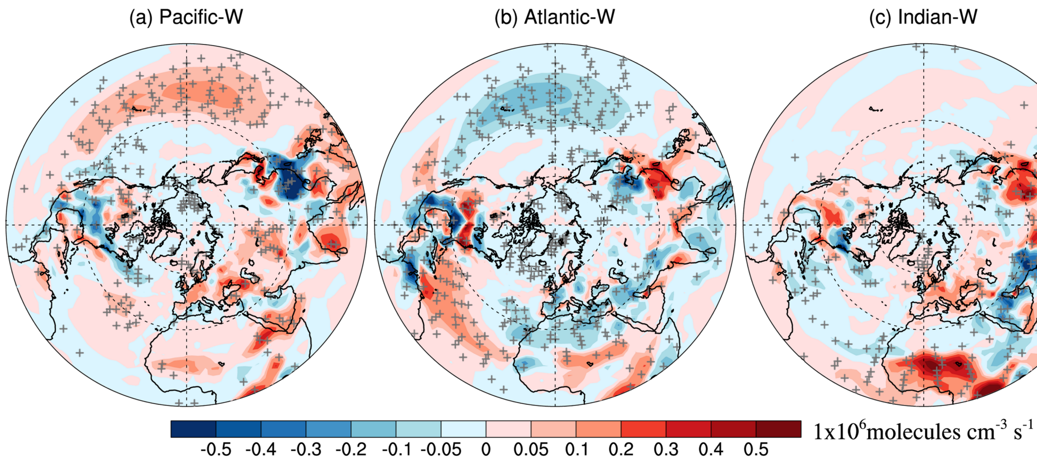

Figure 3. Perturbations of the surface net O_3 production rate ($1x10^6$ molecules cm^{-3} s^{-1}) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data ~~(plots using the Mercator projection are shown in Figure S14 in the supplementary material).~~.

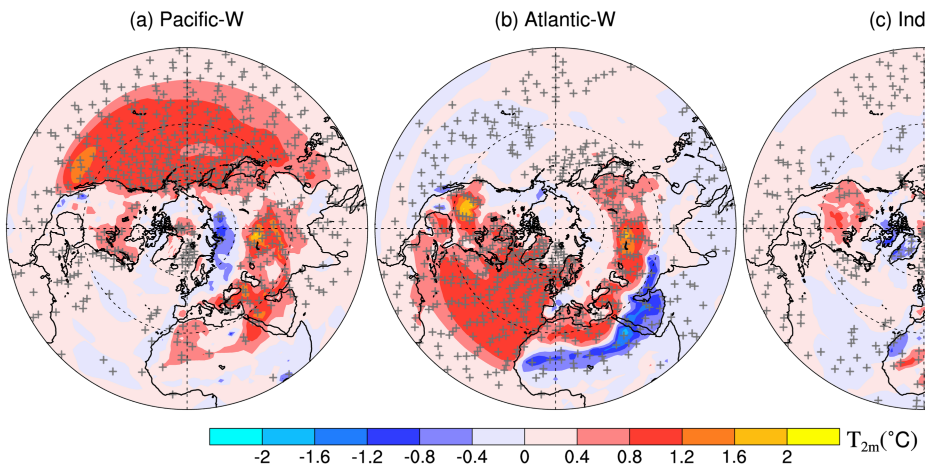

Figure 4. Changes in the surface air temperature (ºC) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL in the Northern Hemisphere in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data ~~(plots using the Mercator projection are shown in Figure S15 in the supplementary material).~~.

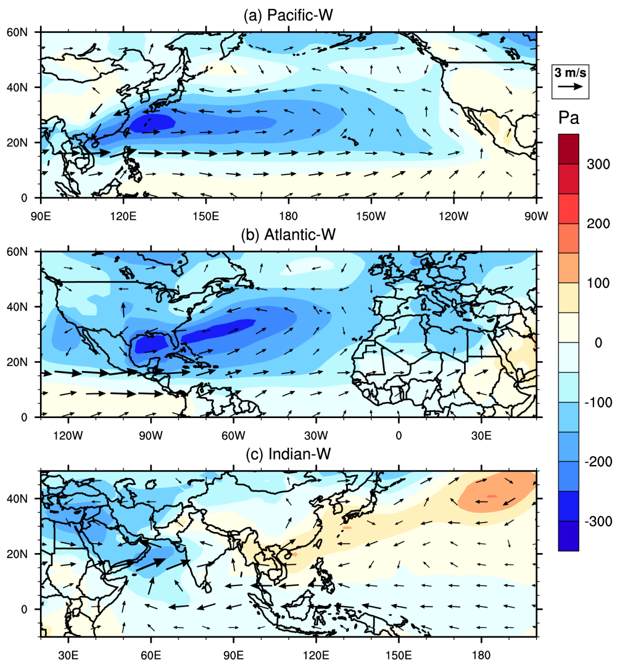

Figure 5. Changes in the surface pressure (color contours, Pa) and 850-hPa wind (arrows, m s^{-1}) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer.

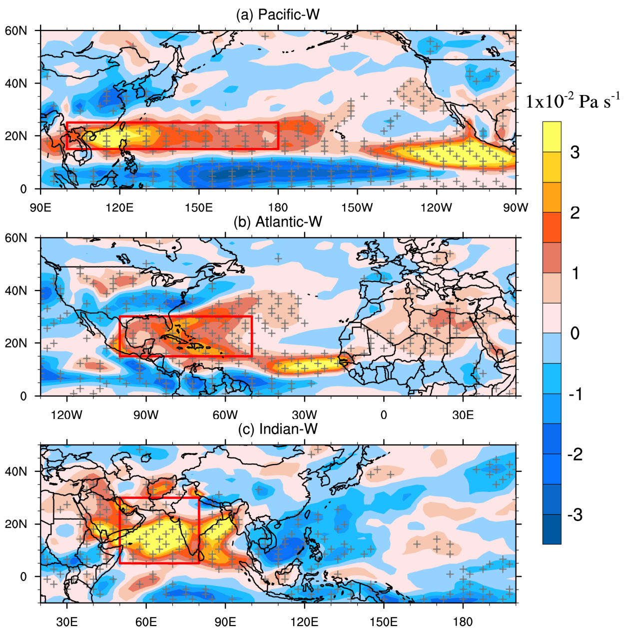

Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1×10^{-2} Pa s^{-1}) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. Red polygons denote the regions where the surface pressure responses to SST anomalies are significant (see

Figure 5 a-c). The + symbols indicate areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

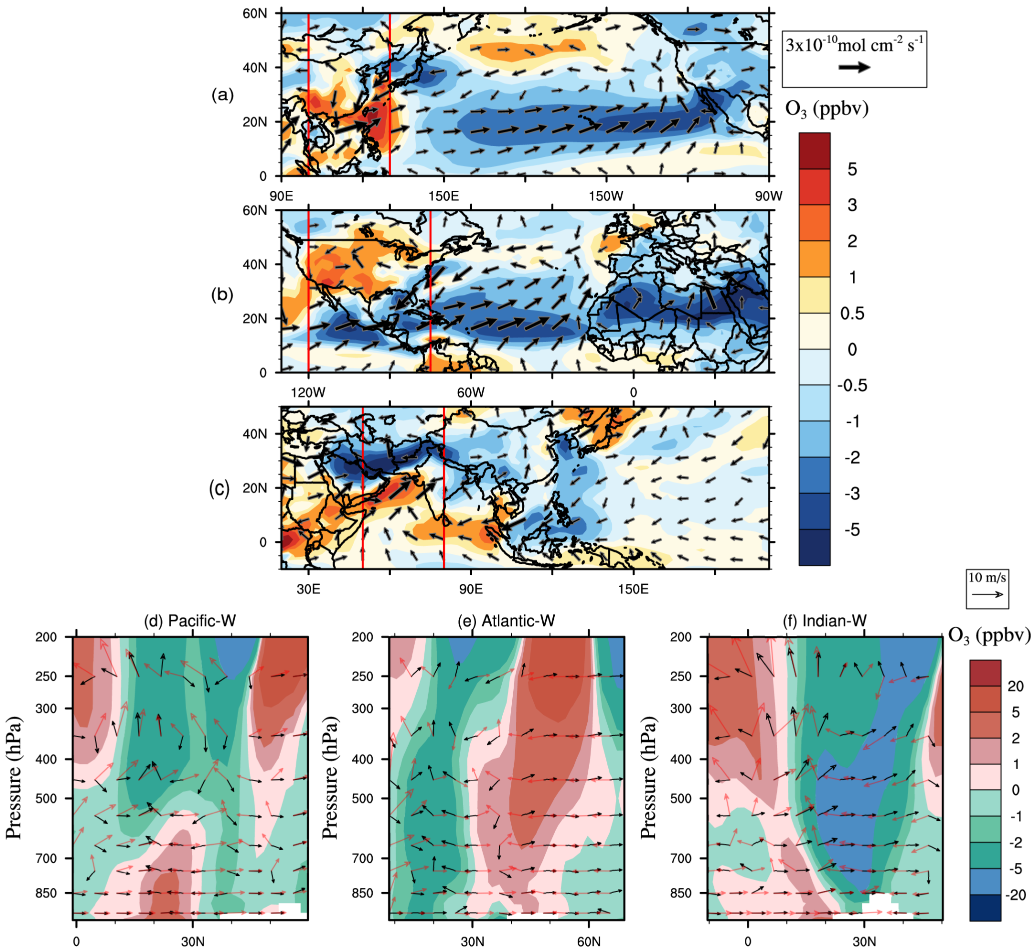

Figure 7. Top three rows: Changes in O_3 concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm^{-2} s^{-1}) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O_3 changes (color contours, ppbv), wind fluxes in CTRL (red arrows, m s^{-1}) and the wind flux perturbation (black arrows, m s^{-1}) in (d) Pacific-W, (e) Atlantic-W, (f) Indian-W relative to the CTRL in the boreal summer. The red rectangles in (a), (b) and (c) denote the longitudinal range used for the zonal averages in (d), (e) and (f), respectively. The vertical wind velocity is amplified 1000 times to make it comparable to the horizontal wind velocity.

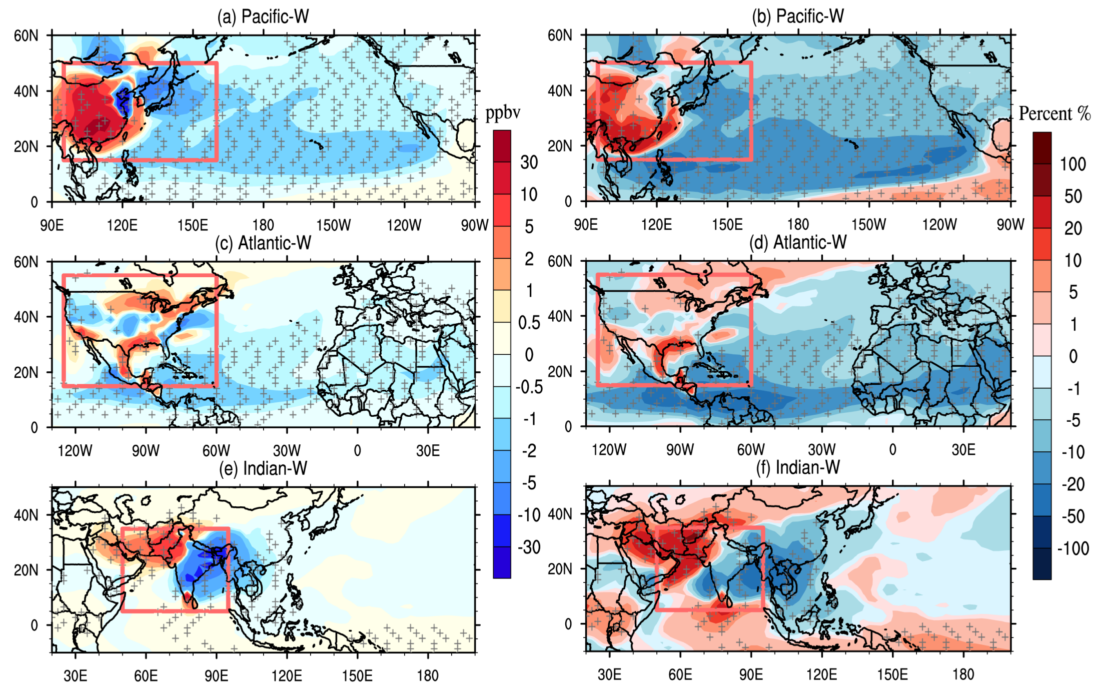

Figure 8. Left-hand panel: Difference in the surface concentration (ppbv) of a CO-like tracer emitted from (a) East Asia for Pacific-W, (c) North America for Atlantic-W and (e) the South Asia for Indian-W relative to the CTRL in the boreal summer. Right-hand panel: The percentage changes in the surface concentration of a CO-like tracer emitted from (b) East Asia for Pacific-W, (d) North America for Atlantic-W and (f) South Asia for Indian-W relative to the CTRL in the boreal summer. Red polygons denote the region where the CO-like tracer is emitted from. The + symbol denotes areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

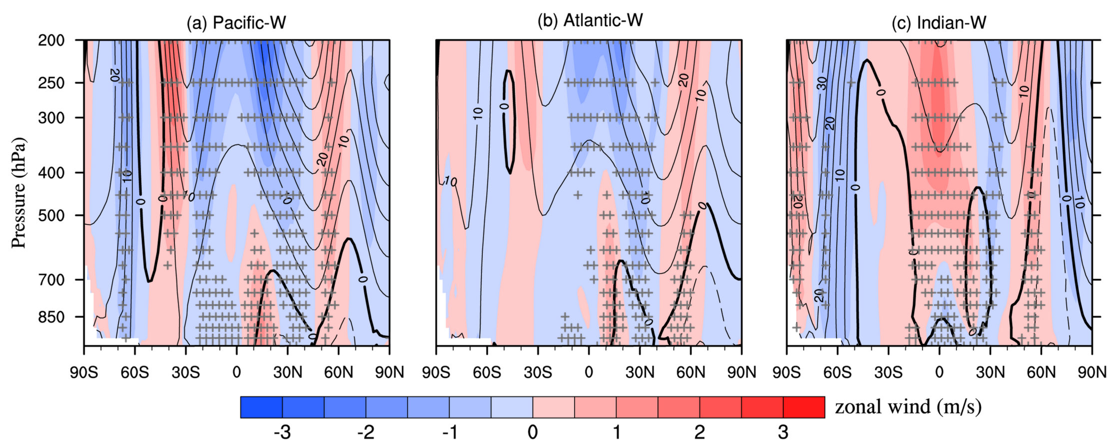

Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential

height (contour, m) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. Black solid and dashed lines in the contours indicate positive and negative geopotential height anomalies, respectively (contour interval: 5 m). The + symbol denotes areas where the zonal wind changes are significant at the 0.05 level, evaluated by Student’s t-test using 20 years of data.