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. investigate the response of surface ozone concentrations due to changes in sea surface temperature (SST) in three different ocean basins (North Pacific –NP, North Atlantic – NA, and North Indian Ocean, NIO). The authors use an Earth System Model (CESM) and perform a set of sensitivity studies in which they alter in turn the climatological SSTs of +/-1°C over the NP, NA and NIO. The imposed variation in SST leads to changes in surface ozone up to 5 ppbv. The authors focused on the summer season and they show that changes in transport associated to the SSTs anomalies are important in driving ozone anomaly at the surface. In general, increased SST reduces the intercontinental transport of O3. Overall the manuscript is relatively well written and logically organized. However, in the introduction several references to previous work are missing, several parts of the manuscript need to be clarified and the authors should try to be consistent when displaying the results (see comments below). Overall, the study presents interesting new aspects and fits well with the scope of the journal. However, before publication large improvements are needed.

We really appreciate the reviewer's thoughtful and valuable comments. Following the reviewer's suggestion, we have extended the introduction and discussion based on these important references in the revised manuscript. The IPR analysis used in our study has been described more thoroughly to make it easier to understand. Based on the reviewer's comments, we have further clarified the text and improved the quality of relevant figures: high-resolution plots are provided in the revised manuscript, which are more distinguishable and understandable. We believe it substantially helps to improve our manuscript by addressing these issues. Please see our response to each comment below.

Major comments:

1) I suggest including a sensitivity test in which the SSTs in all 3 basins is increased. It will be interesting to see the effect of a generalized warming. I would also recommend to include a discussion of the results for winter season (see specific comments below)

Good suggestion. Following the reviewer's suggestion, we conducted 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), denoted as "All-W" and "All-C", respectively. Their effects on surface O₃ distributions are

compared with the sum of results from three individual warming or cooling cases during boreal summer (see Figure S5 or S6 below) and winter (see Figure S7 or S8 below). It shows that the responses of surface O₃ to a generalized SST anomaly over all three ocean basins generally resemble the sum of results from individual oceans. Slight differences are observed over the extratropical North Pacific for the warming cases in boreal summer.

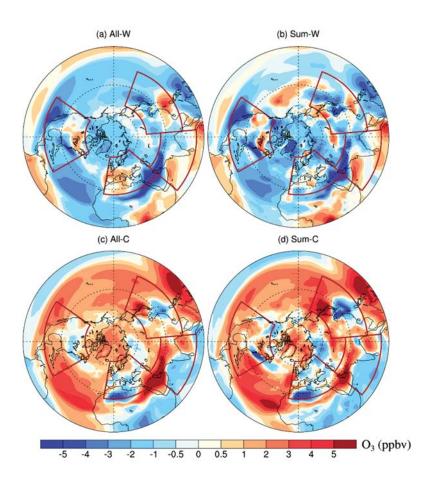


Figure S5. Left column: Changes in the summertime (June-August) surface O₃ concentrations (ppbv) in the Northern Hemisphere induced by 1°C warming (a) and 1°C cooling (b) in all three ocean basins (i.e., the North Pacific, North Atlantic and North Indian Ocean) relative to the CTRL. Right column: Sum of changes in the summertime (June-August) surface O₃ concentrations (ppbv) from the 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 the CTRL, denoted as (b) Sum-W and (d) Sum-C, respectively. The four major regions of interest (i.e., NA, EU, EA and SA) are marked with polygons.

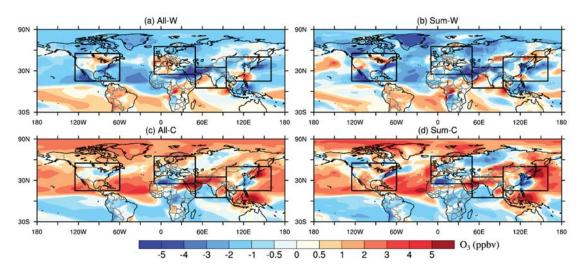


Figure S6. As in Figure S5 but using the Mercator projection.

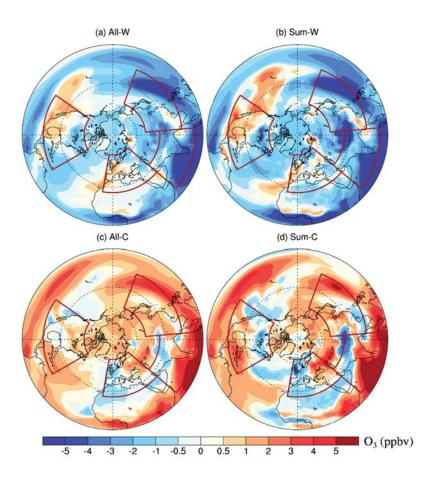


Figure S7. Same as Figure S5 but for the wintertime (December-February).

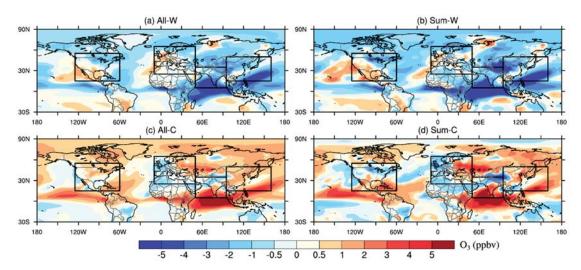


Figure S8. As in Figure S7 but using the Mercator projection.

Considering nonlinear relationships existed in the climate system and O₃ formation processes, the response of surface O₃ to a generalized SST anomaly over all three oceans can not perfectly match the sum of individual results. Regardless those slight difference, our results indicate that the effect of a generalized SST warming on surface O₃ can be decomposed into individual regional ocean forcings. This may help to interpret the responses of surface O₃ to a global-wide warming documented in previous studies (Doherty et al., 2013;Jacob and Winner, 2009;Wu et al., 2008). In the revised manuscript, we made the following revisions in Section 3 (*Page 10,Lines 283-291*):

"...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 different regional SST changes (see Figures S5 and S7 in the supplementary material)."

We also diagnosed the results for winter season, it shows that the responses of surface O₃ and other relevant variables are generally insignificant over land, especially over the four continental regions of interest (i.e., NA, EU, EA and SA). Therefore, we decided to focus mainly on boreal summers. More detailed discussion is provided in our reply to specific comments below.

2) I had major problems in understanding several figures: sometimes the figures are too "crowded" and it is not possible to distinguish the continents (e.g. figures. 4, 6, 9); the panels are often very small in particular in figures 1, 3, 4, 6, 9. The few figures that are clear are the one using the Mercator (or equirectangular) type of projection (e.g., Figures 5, 7, 8, S4, S5). I recommend the authors to be consistent and use the Mercator projection for all figures.

Furthermore, the authors should also be consistent with the use of colorbars: sometimes they use white for values that are not significant (e.g. figure 1), other times for small values (e.g. figure 3), other times they don't use it at all (e.g. figure 4). I recommend not using white color bins for small values, especially if they are significant as in figure 3. See specific comments. I struggle to understand the lack of consistency in making the plots (type of projections, type of colorbar, choice of significant levels, choice of how to display the significant values, etc) that makes it harder for the reader to follow.

Thanks for this really helpful suggestions. We agree that the quality of some figures are low. In our revised manuscript, high-resolution figures are provided that are more distinguishable. We also improved these figures (Figures 6 and 9) that look too "crowded" by removing unnecessary information. The continental outlines in all figures were thicker and darker than the old version. As for the type of projections, we compared the performance of different projections and decided to consistently use polar projection to exhibit our results in hemispheric scale (e.g., Figures 1, 3 and 4 as well as other figures in the supplementary material) while use Mercator projection for regional analysis (e.g., Figures 5-8). The polar projection looks better than Mercator projection when showing the continuous cross-regional relationship along the hemispheric-scale general circulation. Figures using the Mercator projection are also provided in the supplementary material. Please see Figures S2, S14 and S15 below for some examples. Following the reviewer's suggestions, we also enhanced the consistency of these figures. We have removed white color bins for small values (e.g., Figure 3) and consistently used the same symbols to mark results that are significant at the 0.05 level (e.g., Figure 1). We also substantially increased the quality of figures which contain smaller plots. Now these figures can be read more easily and clearly. Following figures show you some examples about these improvements. Please see the revised manuscript for all figures.

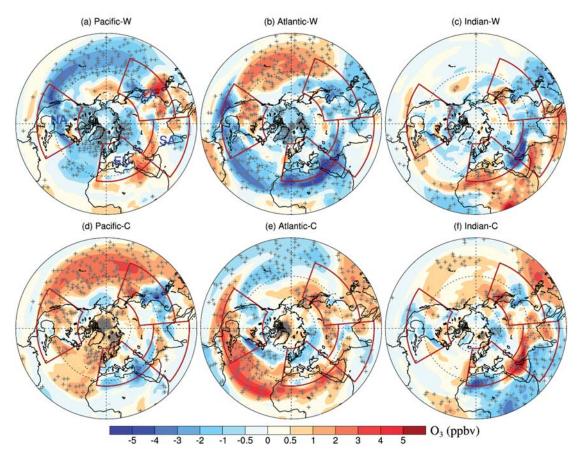


Figure 1. Changes in the summertime (June-August) surface O₃ concentrations (ppbv) in the Northern Hemisphere induced by 1°C warming (top) and 1°C cooling (bottom) in the North Pacific Ocean (left), North Atlantic Ocean (center), and North Indian Ocean (right) relative to the CTRL. The four major regions 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).

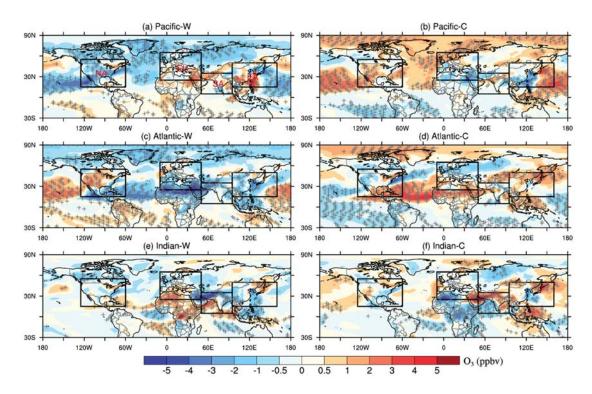


Figure S2. Changes in the summertime (June-August) surface O₃ concentrations (ppbv) in the Northern Hemisphere induced by 1°C warming (top) and 1°C cooling (bottom) in the North Pacific Ocean (left), North Atlantic Ocean (center), and North Indian Ocean (right) relative to the CTRL. The four major regions 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 polygons. 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.

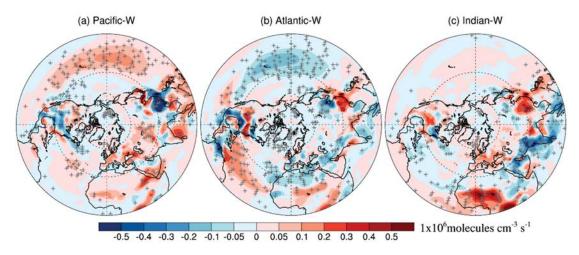


Figure 3. Perturbations of the surface net O₃ production rate (1x10⁶ molecules cm⁻³ s⁻¹

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).

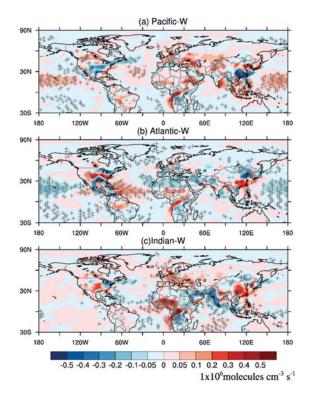


Figure S14. Perturbations of the surface net O_3 production rate (1x10⁶ molecules cm⁻³ s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

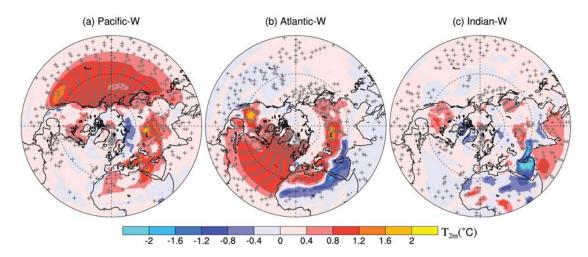


Figure 4. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL in the Northern Hemisphere in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data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

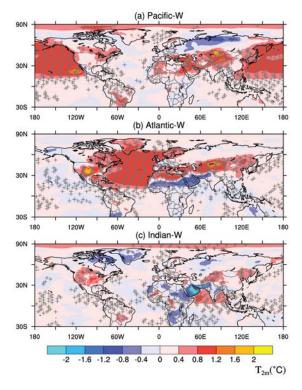


Figure S15. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

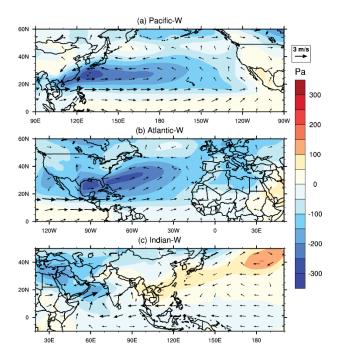


Figure 5. Changes in the surface pressure (color contours, Pa) and 850-hPa wind (arrows, m s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer.

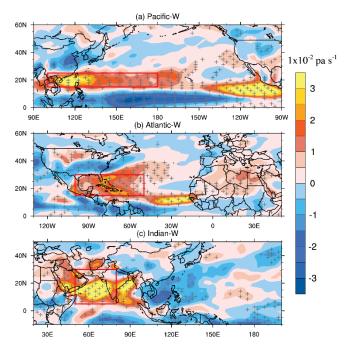


Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. Red polygons denote the

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.

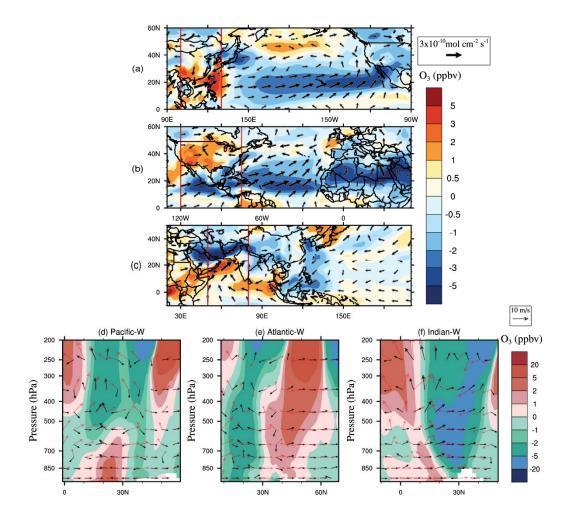


Figure 7. Top three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, 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.

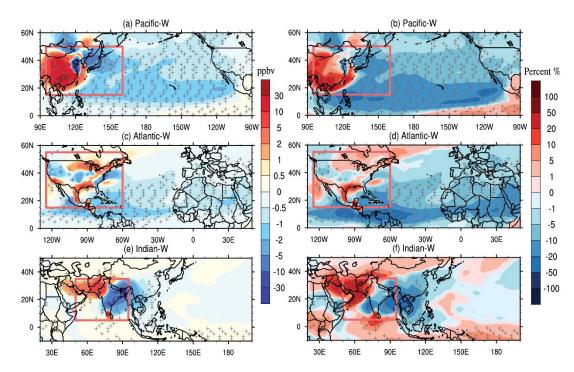


Figure 8. Left-hand panel: Difference in the surface concentration (ppbv) of a CO-like tracer emitted from (a) East Asia for Pacific-W, (c) North America for Atlantic-W and (e) the South Asia for Indian-W relative to the CTRL in the boreal summer. Right-hand panel: The percentage changes in the surface 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.

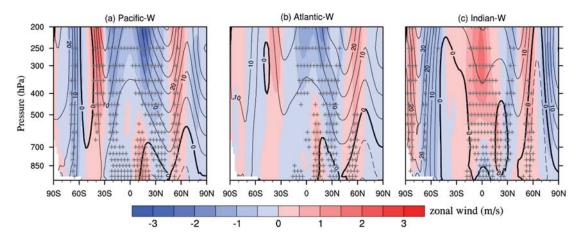


Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential

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.

3) In the introduction the authors do not cite several works done previously on the intercontinental transport of O3 and the meteorological factors affecting it, citing only 4 papers, which are not even the first to address these issues. Please make sure to include the references suggested in "specific comments" and possibly also extend the introduction/discussion.

Good suggestion! We have cited the references following the reviewer's suggestion and expanded our introduction and discussion accordingly. Please see the improved text below and refer to our response to specific comments for more details.

In the Introduction section (see *P3*, *L62-67* and *L72-83*):

"...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)."

. . .

"Atmospheric circulation considerably determines the timescale and pathway of O₃ transport (Bronnimann et al., 2000; Auvray and Bey, 2005; Hess and Mahowald, 2009). The efficiency of O₃ transport varies coherently with atmospheric circulations on different scales. Knowland et al. (2015) demonstrated the important role of mid-latitude storms in redistributing O₃ concentrations during springtime. The North Atlantic Oscillation (NAO) significantly affects surface and tropospheric O₃ 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 O₃ burdens over North America. The shift in the position of the jet stream associated with climate change was found to strongly affect summertime surface O₃ variability over eastern North America (Barnes and Fiore, 2013)."

General Minor comments:

- Why do the authors pick 11 years? Is 11 years enough to capture interannual O3 variability? The SSTs are fixed and in general 15-20 years should be enough to capture interannual atmospheric variability. I'm not sure about 11 though.

Good question. We originally preformed 11-year simulations referring to previous studies. For example, Doherty et al. (2013) conducted a 2000-year long unforced simulation to provide a comprehensive measure of model internal variability. They concluded that 5 years is long enough to capture the climate change signal with fixed SSTs. Given that the SST anomaly prescribed in our simulations (i.e., ±1°C) is comparable to the climate change effects of a specific ocean, we originally thought that 11 years are enough to capture the relevant signal in our study (see Figure 1 and Figure R1 for example). However, following the reviewer's suggestion, we extended our simulations to 21 years with the first year used for model spin-up. It shows that the 20-year averaged results are generally consistent with the 11-year averaged results except for a few minor differences (e.g., see Figure 1 and Figure R1 below). In our revised manuscript, we redo our calculation and regenerate all plots based on the 20-year averaged results.

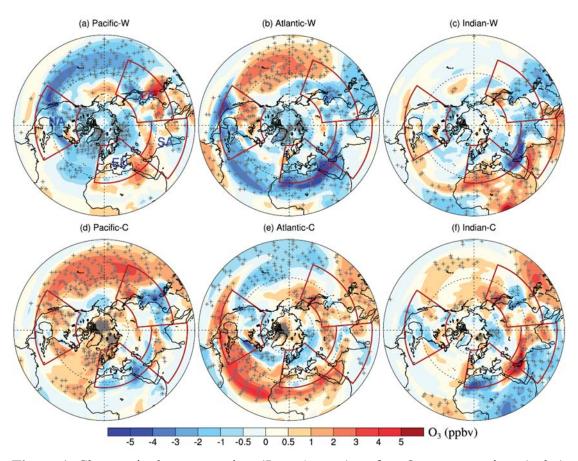


Figure 1. Changes in the summertime (June-August) surface O₃ concentrations (ppbv)

in the Northern Hemisphere induced by 1°C 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).

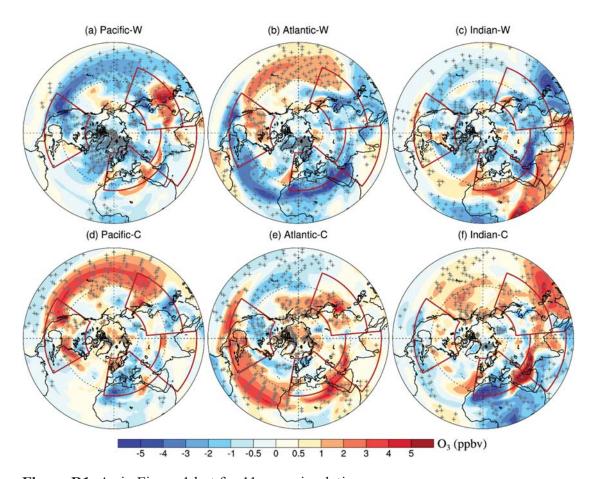


Figure R1. As in Figure 1 but for 11-year simulations

- The authors use different significance levels throughout the manuscript (0.01 or 0.05). Please, pick one and use that for all the analysis.

Good suggestion. We pick the significant level of 0.05 to be consistent in our revised manuscript.

- I suggest not overusing sentences in which part of the text is in parenthesis in order to avoid writing another sentence (e.g.: LL24-25 Increasing 25 (**decreasing**) SST by 1 °C in one of the regions of focus induces decreases (**increases**)). It makes it hard to follow. I think adding another sentence makes it much more easy to read (especially in the main text where there is no word limits).

We follow reviewer's suggestion and revised the relevant texts to avoid the overuse of parenthesis.

In the Abstract (*P1*, *L24-27*):

"... 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."

In Section 3 (P9-10, L264-280):

"...Surface O_3 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). 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 O₃ over the North Pacific, North America and the North Atlantic of approximately 1 ppbv (Table S1) but may enhance the surface O₃ by nearly 3 ppbv over South China. Similarly, in the "Atlantic-W" case, the surface O_3 levels decrease by $1\sim2$ ppbv over the North Atlantic and Europe but increase (~1 ppbv) 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 in the supplementary material."

- It is not necessary to include the figure captions in the text. Sentences like "Figure 2 shows ..." belong to figure captions not the main text, and make it hard to follow. Please, discuss directly the results and point to the figure that shows them in the running text, e.g. Larger anomalies (i.e., up to 5ppbv) are simulated in locations including the east

coast of China, the Indian subcontinent, and remote oceans (Figure 1 and Figure S2).

We have changed the text accordingly in our revised manuscript (e.g., P11, L305-308).:

"...In this study, the SST-induced, process-level O₃ 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)."

Specific comments:

LL59-61 Beside the missing reference pointed out in the short comment by Dr. Meiyun Lin there are several other key references missing that are related to the O3 long-range transport: Parrish et al., 1993; Fehsenfeld et al., 1996; Wild and Akimoto, 2001; Creilson et al., 2003; Simmonds et al., 2004.

Good suggestion. We have added these important references related to the O_3 long-range transport in our revised manuscript (P3, L62-67):

"... 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)."

LL66 Here as well, the authors do not cite several studies on the topic (e.g., Bronnimann et al. 2000; Hess and Mahowald, 2009; Pausata et al. 2012).

We have cited these valuable studies and expanded our introduction (see P3, L72-83):

"...Atmospheric circulation considerably determines the timescale and pathway of O₃ transport (Bronnimann et al., 2000; Auvray and Bey, 2005; Hess and Mahowald, 2009). The efficiency of O₃ transport varies coherently with atmospheric circulations on different scales. Knowland et al. (2015) demonstrated the important role of mid-latitude storms in redistributing O₃ concentrations during springtime. The North Atlantic Oscillation (NAO) significantly affects surface and tropospheric O₃ 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 O₃ burdens over North America. The shift in the position of the jet stream associated with climate change was found to strongly affect summertime surface O₃

variability over eastern North America (Barnes and Fiore, 2013)."

L104 remove spaces before and after comma.

We removed these spaces (P4, L119-121).

"...The mechanisms responsible for SST variability includes ocean circulation variability, wind stress, and ocean-atmosphere feedbacks (Frankignoul, 1985; Deser et al., 2010)."

L113 remove the hyphen after impacts.

We removed this mistake in our revised manuscript (*P5*, *L135-136*):

"... Except for the ENSO impacts, very few studies to date have directly addressed the linkage between SST and O_3 ."

L114 ENSO is an oscillation; hence "ENSO spring" does not mean anything. Please specify the ENSO phase the authors are referring to.

We changed "ENSO spring" to "strong La Niña spring" for clear clarification (*P5*, *L128-131*):

"...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."

LL114-115 indulge a bit more and provide the explanation of how ENSO affects stratospheric intrusions in western US. Otherwise the reader is forced to look it up.

Good suggestion. We revised this sentence in the introduction section to briefly explain the impacts of ENSO on stratospheric intrusions in western US (*P5*, *L128-131*):

"...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."

L194 mention also here at least some of the individual processes accounted for.

We revised this sentence in Section 2.3 (*P8*, *L222-226*) to give some examples of the individual processes:

"... 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)."

LL233-234 the sentence is unclear.

Here we means the responses of surface O_3 to SST changes in different cases behave differently in terms of spatial distribution. Different oceans warming can impact the surface O_3 at specific regions. In our revised manuscript, we changed this sentence in Section 3 (*P10*, *L282-283*) for clarification:

"Our simulations reveal that different oceans can exert distinct region-specific effects on the O_3 distribution."

LL253-254 It's not clear to me how the authors can conclude that the change in CHEM is "therefore" causing the increase in ozone at the surface over NA due to warmer Atlantic SSTs. See also comment on figure 2.

Good question. The integrated process rate (IPR) method can decompose the contribution of different physical and chemical processes to O₃ evolution. Typically a positive change in IPR is responsible for the increase of surface O₃ due to the change of SST, which is sometimes balanced by certain negative IPR changes. For instance, the increase in VDIF is always accompanied with a commensurate decrease in DRYD, resulting in an insignificant net change in TURB (here TURB=VDIF+DRYD). We redraw these plots and merge VDIF and DRYD into TURB and DEEP and SHAL into CONV (see Figure 2). Now it shows that the increase of CHEM tends to play the dominate role in enhancing the surface O₃ concentrations over North America.

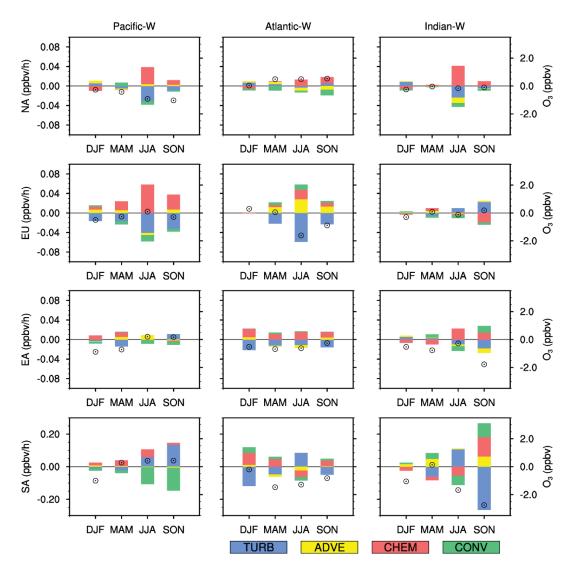


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second 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 S10 in the supplementary material.

To clarify our analysis, we added more descriptions on the IPR analysis in both Section 2.3 and Section 4.1 in the revised manuscript:

In Section 2.3 (*P8-9*, *L226-241*):

"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 O₃ 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 O₃ due to gasphase photochemistry. DRYD represents the dry deposition fluxes of O₃, which is an important sink for O₃. 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 O3 fluxes from these six processes matches the change in the O3 concentration. The IPR performance is verified by comparing the predicted hourly O₃ 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."

In Section 4.1 (*P10-11*, *L296-316*):

"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). 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). In most cases, VDIF and DRYD are the key processes controlling the O_3 variation. The downward transport of O_3 through diffusion 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). More detailed IPR results are shown in Figures S10 and S11 in the supplementary material."

L264 "inconsistent surface O3 response": do the authors mean "opposite surface O3 response"?

We replaced "inconsistent" with "distinct" (see P12, L340-342):

"These opposite changes in VDIF over upwind and downwind regions lead to distinct surface O_3 responses."

L270 I understand the authors' point on investigating only summer since it's the seasons with higher O3 concentration at the surface. However, during winter and spring the ozone at the surface is mainly affect by changes in long-range transport and stratosphere-troposphere exchange. Hence, it is important to understand how the warming in the SST in different basins can affect long range and stratosphere-troposphere exchange. I would suggest expanding the analysis to also winter.

Thanks for this helpful suggestion. In this study, we had investigated both the SST-O₃ relationship in both summer and winter seasons (see Figure 1 and Figure S3). It shows that in boreal winter, the warming of different oceans generally induces a widespread decrease of surface O₃. However, significant changes (up to 5 ppbv) mainly happen over remote oceanic regions. Over land, the O₃ response to SST changes is generally insignificant (see Figure S3). Besides surface O₃, the responses of meteorological fields to SST changes are also significant only over remote oceans (see Figure R3). Similar to the summer case, physical transport is the key process modulating surface O₃ during winters. As shown in Figure R2 and R3, these vertical and horizontal wind field changes are more robust over oceans than the polluted continents. Since the main focus of this study is to examine how O₃ air pollution in a populated continent is affected by regional SST changes, we therefore pay most attention to boreal summers than other seasons. In the revised manuscript, we follow the reviewer's suggestions and add a brief discussion in Section 3 (see *P10*, *L276-280*) for wintertime response:

"...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 in the supplementary material."

We further clarified why we only focus on summertime in Section 4.1 (P12, L344-347):

"In the following subsections, the mechanisms of the SST-O₃ relationship for the four polluted continents are further explored. Here we focus on boreal summers since the surface O₃ response to SST changes is more robust during this period than other seasons."

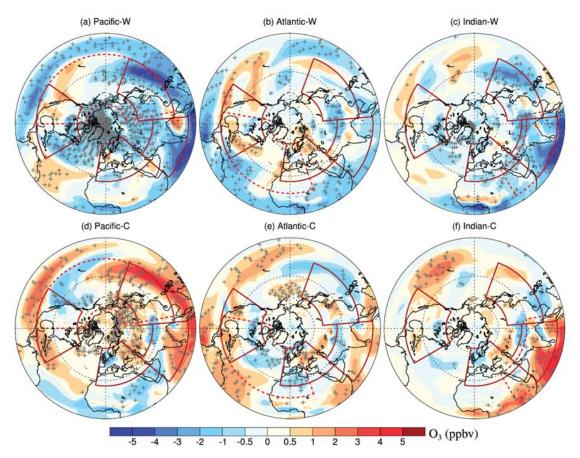


Figure S3. Changes in the wintertime (December-February) surface O₃ concentrations (ppbv) in the Northern Hemisphere for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W, (d) Pacific-C, (e) Atlantic-C and (f) Indian-C relative to the CTRL. The four major regions of interest (i.e., NA, EU, EA and SA) are marked with solid polygons. Red dashed lines mark the regions where the SST has been changed. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20

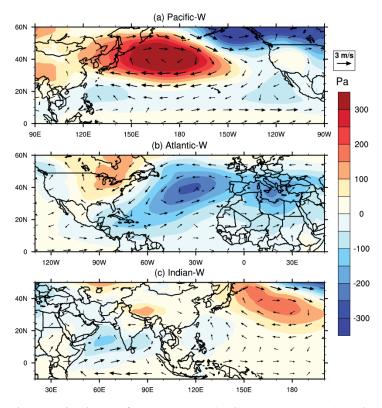


Figure R2. Changes in the surface pressure (color contours, Pa) and 850-hPa wind (arrows, m s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal winter.

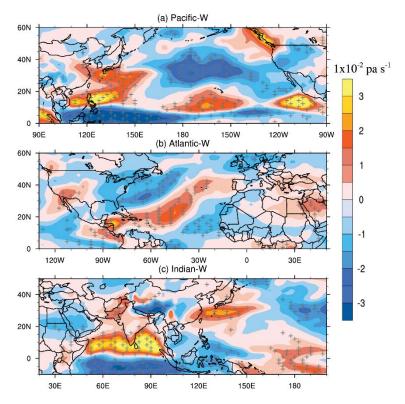


Figure R3. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal winter. Positive values indicate upward motion. Red polygons denote the regions where the surface pressure responses 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.

L291 "is believed": Beliefs do not belong to science. Please rephrase it and provide references to support the *belief*.

The effect of the North Indian Ocean warming on cloud formation has been well-documented in precious studies (Chaudhari et al., 2016;Roxy et al., 2015;Xi et al., 2015). We have rephrased this sentence in Section 4.2 (*P13*, *L367-372*) as below:

"...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)...."

LL356-357 The authors stated that the O3 changes at the surface over North America (Fig. 7 "b", which is actually c) are negligible. However, they look quite large (regionally) to me: over the Great Lakes, California and Baja California peninsula; also along the east coast of Unites States the changes are not that small. Furthermore, the changes aloft (that the authors define "large") are of the same order of magnitude that the changes seen at the surface.

Good question. We agree that the surface O_3 changes over NA (~1-2 ppbv) are large and significant for regional air quality management. In the revised manuscript, we have revised this description in Section 4.3 (*P16*, *L447-448*):

"... O_3 changes are observed to be larger in the upper troposphere than at the surface (Figure 7e)..."

We also reordered the plots in Figure 7:

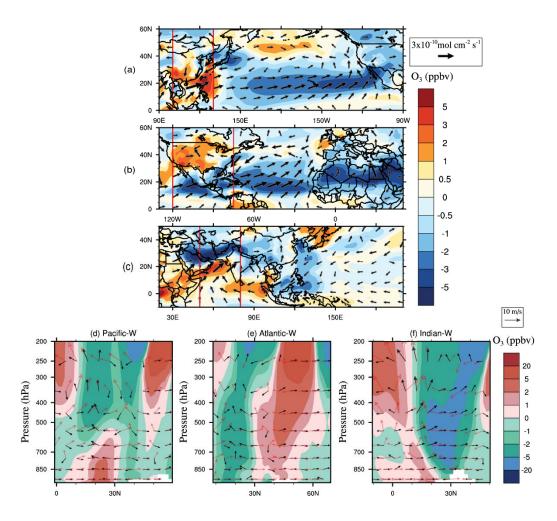


Figure 7. Top three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, 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.

L357-359 Given the above-mentioned comments, I am not sure how the authors could state that the changes seen in figure 7c are mostly due to enhanced photochemical production. This comment is also related to my previous comments on LL253-254.

Good question. Following our detailed response to the previous comments on L253-

254, the IPR analysis helps to identify the key processes associated to the SST induced O₃ evolution. For example, the warming of the North Atlantic Ocean leads to 1~2 ppbv increase in surface O₃ over North America. Ignoring VDIF and DRYD (they tend to offset each other in most cases, resulting in an insignificant net change in TURB, see Figure 2). Therefore, the change of CHEM is the dominant factor leading to the surface O₃ increase over North America. Please refer to our response to previous comments on L253-254 for more detail. Figure 7, on the other hand, further indicates that changes in the horizontal fluxes of O₃ over North America show no significant effect on the corresponding increase of surface O₃ levels.

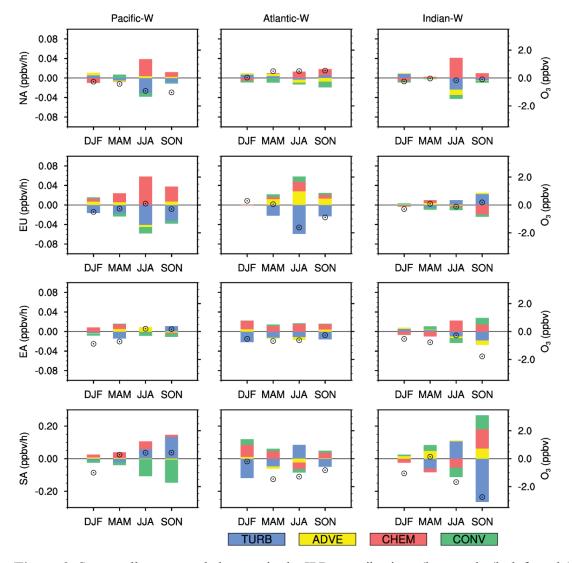


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second 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 S10 in the supplementary material.

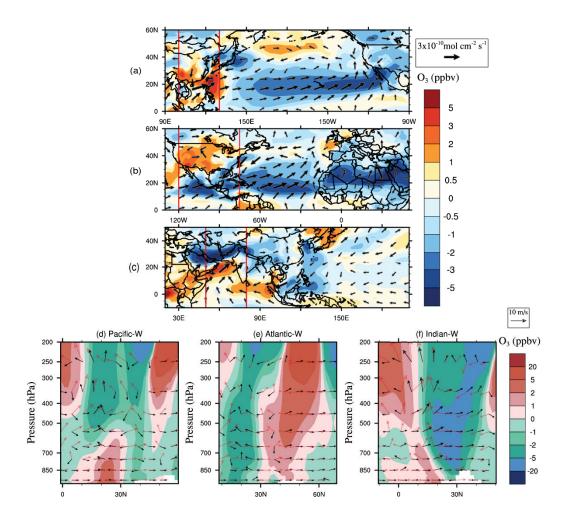


Figure 7. Top three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, 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.

In the revised manuscript, we rephrased this analysis to clarify our statement (P15-16, L444-450):

"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."

LL366-368 please refer to figure 2 as well.

Thanks, referred (see *P16*, *L456-460*).

"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). However, over the nearby Indian subcontinent, the suppressed convection tends to decrease surface O_3 in that region (Figure 2)."

L369 The IPR analysis show suppressed deep-convection. However, the warming of the Indian Ocean strengthens the Indian Summer Monsoon, as also stated by the authors (e.g. LL290-292), hence I wonder why the deep-convection is weakened. Please comment on that.

Good question. Changes in DEEP in the IPR analysis indicates that a warming of Indian Ocean tends to reduce surface O₃ over South Asia (Figure S10), but increase surface O₃ over North Indian Ocean (Figure S11). This is because that the warming of the North Indian Ocean enhances the deep convection above it while suppress the deep-convection over the Indian subcontinent. According to previous studies (e.g., Hartmann, 2015;Lau et al., 1997;Lau and Nath, 1994), the SST increase over the Indian Ocean strengthens deep-convection above it. A low-pressure anomaly is observed centered over the Arabian Sea (Figure 5). It consequently strengthens the southwesterly flow towards the Indian subcontinent, as a part of the Indian Summer Monsoon. On the other

hand, the enhanced upward movement of moist air above the Indian Ocean enhances cloud formation. This tends to block solar radiation reaching the earth surface and cools the surface air over the Indian subcontinent. A remarkable reduction of surface solar radiation and air temperature are shown in Figure S17 and Figure 4, respectively. This decrease in surface temperature over the Indian subcontinent may suppress the development of deep-convection there.

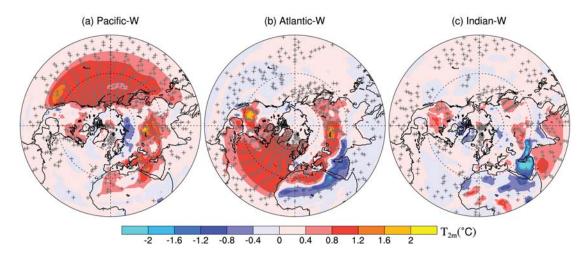


Figure 4. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL in the Northern Hemisphere in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data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

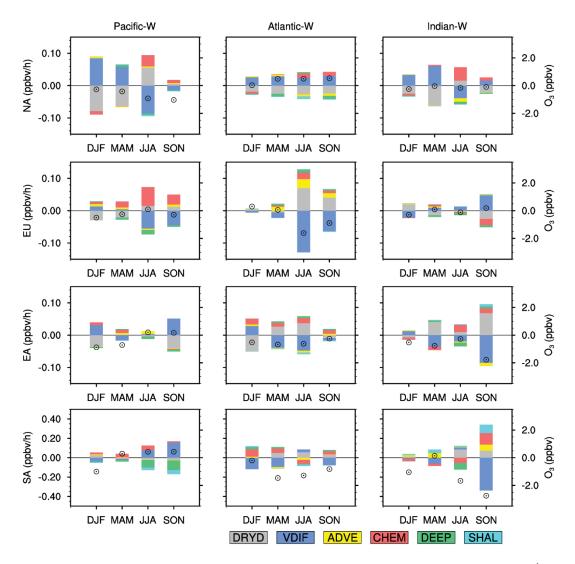


Figure S10. Seasonally averaged changes in the IPR contributions (bars, ppbv h⁻¹, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second row), EA (third row) and SA (last row). IPR contributions from the six processes (i.e., DRYD, VDIF, ADVE, CHEM, DEEP and SHAL) are represented by different colors.

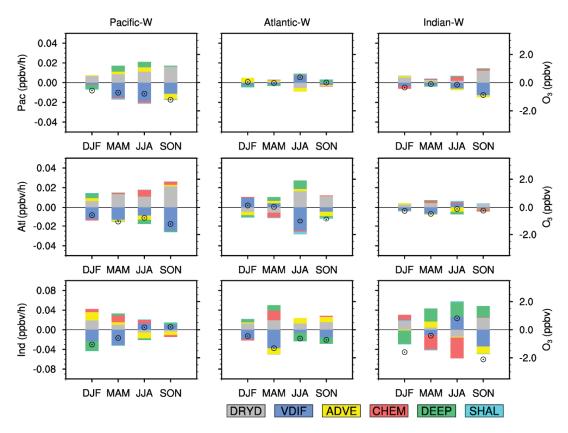


Figure S11. Same as Figure S6 but for three ocean basins defined in our study.

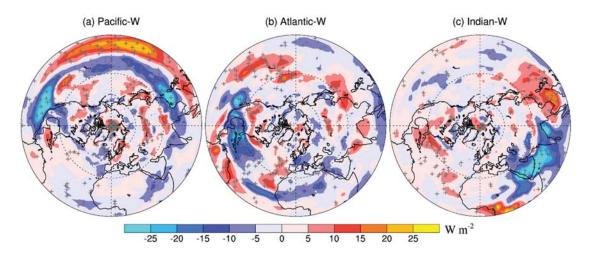


Figure S17. Perturbations of the surface solar radiations (W m⁻²) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test.

We have clarified the corresponding text in Section 4.2 (see *P13*, *L361-380*):

"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). Associated with this temperature decrease is a remarkable reduction in the solar radiation received at the continent below (more than 15 W/m², Figure S17). 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 blocks the solar radiation reaching the surface of the Indian subcontinent and reduce 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 S16), which may further suppress the development of deep convection over the Indian subcontinent."

LL396-400 Beside the fact that Figure 9 is difficult to read. The reduction in geopotential height over the Arabian Sea seems actually to increase the southwesterly flow towards the Indian subcontinent. Furthermore, the land sea contrast may play a very small role in enhancing or weakening the strength of the Indian Summer Monsoon (Molnar et al. 2010). Hence, an in depth analysis should be done before claiming that the change in land-sea contrast is what weakens the "thermal wind". Furthermore, the changes in temperature does not show a clear decrease in land-sea contrast, since there is a warming of SST, a cooling of the Tibert Plateau and northwestern Indian subcontinent, and a warming north of that cooling. Hence I really don't see the authors' point.

In any case, the sentence is not very clear and should be reformulated: "This nonuniform increase in air temperature (i.e., more significant at mid-latitudes) weakens the meridional temperature gradient, resulting in a reduction of thermal winds." What is more significant at mid-latitudes? The nonuniform increase in temperature? Or the fact that the temperature increases more there than the ocean? Or what?

Good question. The original Figure 9 contains many variables (i.e., air temperature, wind pattern and geopotential height at 500 hPa) that make it difficult to read. The changes of westerly wind are also hard to distinguish in Figure 9. In the revised

manuscript, we illustrated our result in a more clear way. The zonal averaged changes in zonal wind and geopotential height are now shown in Figure 9, which is more distinguishable.

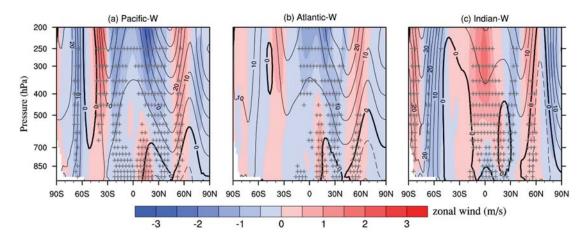


Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential height (contour, m) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. Black solid and dashed lines in the contours indicate positive and negative geopotential height anomalies, 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.

We agree that the increase in southwesterly flow towards the Indian subcontinent could not be simply explained by the changes in land-sea thermal contrast. According to our analysis, the warming of the North Indian Ocean creates a warm-core cyclonic anomaly centered over the Arabian Sea, which is responsible for the enhancement of the southwesterly flow towards the Indian subcontinent. A detail description is provided in our response to the comments on L369. The "thermal wind" theory here was used to explain the weakening of the westerly wind at mid-latitudes associated with the basin-scale SST warming. We find that a basin-scale SST warming, especially for the North Pacific and North Atlantic, tends to increase the air temperature (Figure S16) and geopotential height (Figure 9) more significantly at mid-latitudes than elsewhere. Consequently, the meridional temperature and geopotential height gradients are decreasing in the tropical-to-mid-latitude troposphere while increasing at higher latitudes. It tends to decrease the zonal westerly wind at lower-middle latitudes (25°N - 45 °N) in the Northern Hemisphere while increase it at higher latitudes (Figure 9).

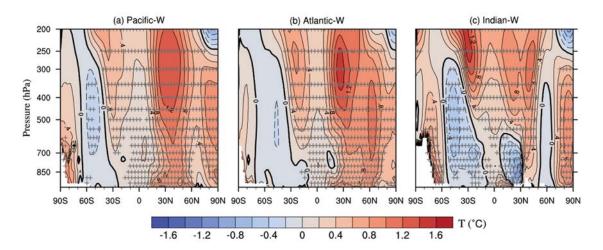


Figure S16. Vertical-meridional distributions of changes in the air temperature (contours, °C) for (a) Pacific_W (zonal averaged from 100°E-90°W), (b) Atlantic_W (zonal averaged from 100°W-180°W) and (c) Indian_W (zonal averaged from 30°E-100°E) relative to the CTRL in boreal summer. Black solid and dashed lines in the contours indicate positive and negative air temperature anomalies, respectively (contour interval: 0.2 °C). The + symbol denotes areas where the changes of air temperature are significant at the 0.05 level, evaluated by Student's t-test.

In the revised manuscript, we clarified our analysis in Section 5 (P17, L489-498):

"...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 O₃ transport."

LL383-385 Referring to figure 8b, the authors state: "Similarly, for the North American tracer, a warming of North Atlantic SSTs by 1°C slightly increases (~2%) concentrations in North America but decreases (3-4%) concentrations over downwind Europe". To me it looks like a slight decrease over Europe and quite an increase over large areas of North America. Please correct/clarify.

We previously showed absolute change. If switching to the percentage change (see the Figure 8 below), the pattern would be different. To avoid confusion, we decide to show both absolute and percentage changes in Figure 8 and remove the word "slightly" in this sentence in Section 5 (*P16*, *L471-474*):

"...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)."

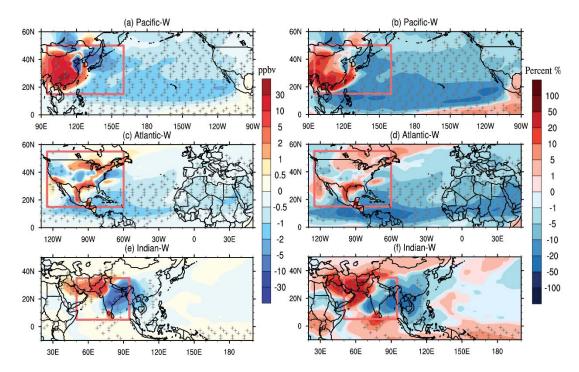


Figure 8. Left-hand panel: Difference in the surface concentration (ppbv) of a CO-like tracer emitted from (a) East Asia for Pacific-W, (c) North America for Atlantic-W and (e) the South Asia for Indian-W relative to the CTRL in the boreal summer. Right-hand panel: The percentage changes in the surface 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.

We have rephrased this sentence in Section 6 (*P19*, *L551-552*) following the reviewer's suggestion:

"... We further show that air temperature is an important factor controlling the surface O_3 responses to SST anomalies."

Figure 2: It is not clear to me how one can get the changes in O3 from the IPR analysis. It seems that the positive anomalies counterbalance the negative ones (if so this should be made clear, readers may not be familiar with the IPR analysis you are presenting). Therefore I wonder how can the total O3 anomalies be negative or positive (the circle)? It's not clear to me how to read the figure. Please clarify.

Why don't the authors plot in figure 2 only the CONV and the TURB and instead place the figure with the full analysis in the supplementary?

Good suggestion. The IPR analysis helps to identify the key processes that cause O₃ changes. Since some processes always offset the others (e.g., VERD and DRYD), we follow the reviewer's suggestion and merge those processes and show the CONV and the TURB instead (see the new Figure 2 below), and put the more detailed decomposition in the supporting information. In addition, we added more descriptions in the revised Section 2.3 and Section 4.1:

In Section 2.3 (*P8-9*, *L221-241*)

"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 gasphase 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 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 S_3 , the hourly surface O_3 changes are well represented by the sum of these fluxes in the model."

In Section 4.1 (*P10-11*, *L296-316*), we have:

"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). 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 O₃ concentration changes but rather to help examine the relative importance of different transport and chemical processes in driving the sensitivity of O₃ to SST forcing. In this study, the SST-induced, process-level O₃ 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). In most cases, VDIF and DRYD are the key processes controlling the O₃ variation. The downward transport of O_3 through diffusion 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 O₃ concentrations. In addition, we merge SHAL and DEEP as CONV to represent the total contribution of convective transport to surface O₃ (Figures 2 and S9)."

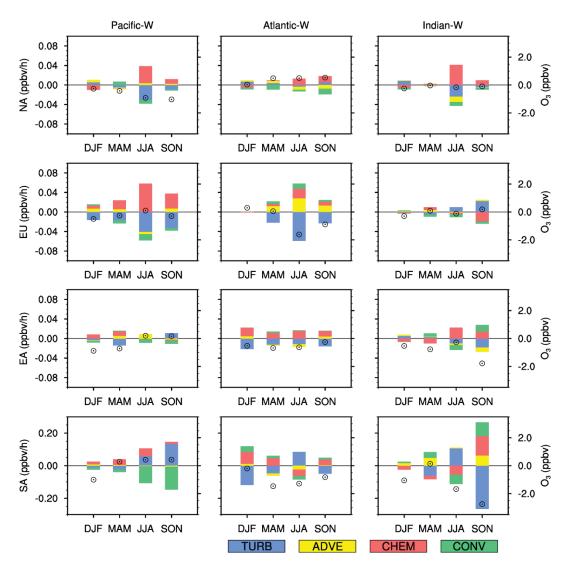


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second 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 S10 in the supplementary material.

Figure 3: the authors use 0.05 as significance level while in figure 1 was 0.01. Please pick one level. In figure 1 white colors were used for non-significant values, please be consistent. Furthermore, in figure 3 sometime white areas present significant changes.

Good suggestion. We used 0.05 as the significance level for all relevant figures. We have also removed white color bins used for small values and non-significant values. Please see the revised Figure 3 below for an example.

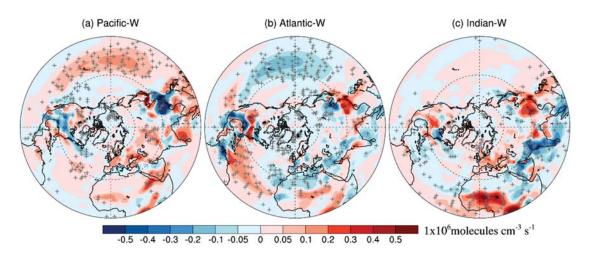


Figure 3. Perturbations of the surface net O₃ production rate (1x10⁶ molecules cm⁻³ s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data (plots using the Mercator projection are shown in Figure S14 in the supplementary material).

Figure 4: The panels are small and it's hard to see the continents. Please use Mercator projection.

Thanks for this suggestion. As we discussed previously (see our response to major comment 2), we compared the performance of different projections and decided to consistently use polar projection since it is easier to interpret the hemispheric flow patterns although it is true that it's hard to see continents. We realized that the difficulty of reading the original Figure 4 were mainly caused by the low figure quality and blurry continental outlines. In the revised version, high-resolution figure is provided. In addition, the continental outlines are thicker and darker than before. Please see the revised Figure 4 below:

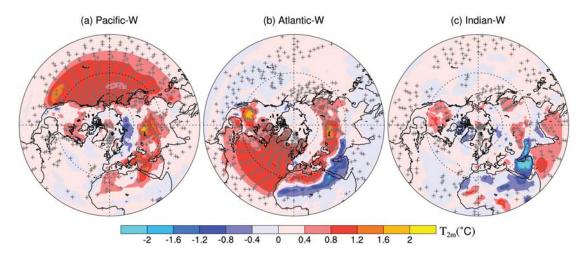


Figure 4. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL in the Northern Hemisphere in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data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

On the other hand, plots using the Mercator projection are also provided in the supplementary material. Please see Figure S15 below:

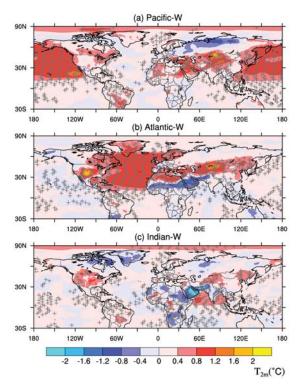


Figure S15. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-

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.

Figure 5: I think it would be better to show both the upwind and downwind area around the basin, i.e. in panel a) please show also the western coast of North and Central America; in panel b) please show the European coast. Finally, the authors plot the wind pattern but do not specify the level: is it at the surface or 850 hPa, ...? Please clarify it. Furthermore, I would suggest not to use the surface level but rather a low-middle atmosphere level (850 or 700 hPa).

Good suggestion. We have used the same map projection for Figure 5-8 that shows both the upwind and downwind area around the basin. Please refer to our response to major comment 2 for more details. The wind pattern at 850 hPa is depicted in Figure 5.

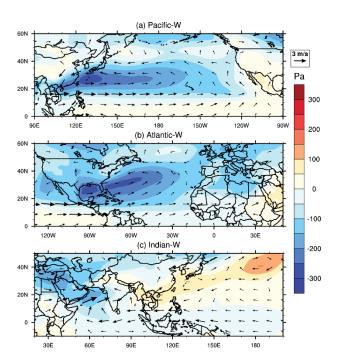


Figure 5. Changes in the surface pressure (color contours, Pa) and 850-hPa wind (arrows, m s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer.

Figure 6: Impossible to understand it without major efforts.

Thanks for bringing this problem up. Figure 6 shows the spatial pattern of vertical velocity changes at 500 hPa. We agree that the old version was hard to read because of

the low figure quality. We have optimized this figure with large improvement. We also changed the map projection of Figure 6 to make it comparable with Figures 5, 7 and 8. Please see the revised Figure 6 below:

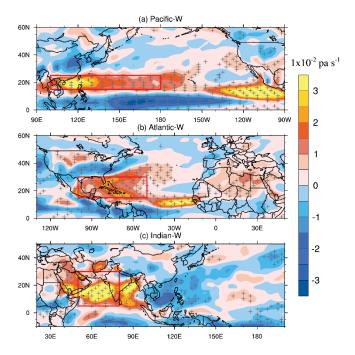


Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. Red polygons denote the regions where the surface pressure responses 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: as for figure 5 I don't understand the choice of the domain shown for each of the sensitivity experiment. Furthermore, panel b) should be switched with panel c). Furthermore, the authors should also here be consistent with the choice of the domain to show. I would advice to adopt the domain (or a similar one) used in figure 8.

Good suggestion! We have used the same map projection for Figures 5-8 that shows both the upwind and downwind areas around the ocean basin and revised mistakes in the original figure panel:

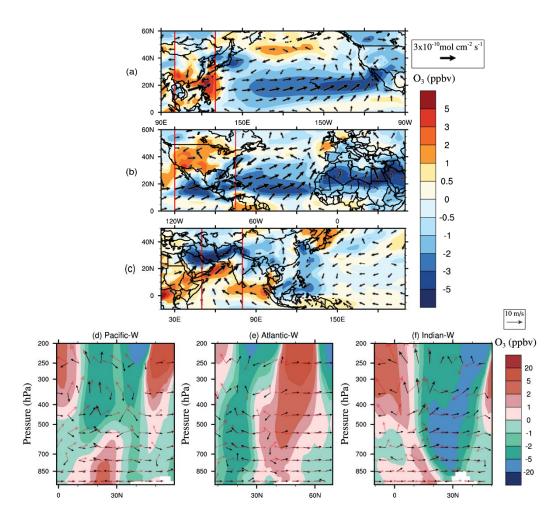


Figure 7. Top three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, 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 S3: which season?

This Figure is referring to the boreal summer. We have revised the caption of Figure S16 (i.e., the Figure S3 in old version) and clearly clarified the relevant season.

"Figure S16. Vertical-meridional distributions of changes in the air temperature (contours, °C) for (a) Pacific_W (zonal averaged from 100°E-90°W), (b) Atlantic_W (zonal averaged from 100°W-180°W) and (c) Indian_W (zonal averaged from 30°E-100°E) relative to the CTRL in boreal summer. Black solid and dashed lines in the contours indicate positive and negative air temperature anomalies, respectively (contour interval: 0.2 °C). The + symbol denotes areas where the changes of air temperature are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data."

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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 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), 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).

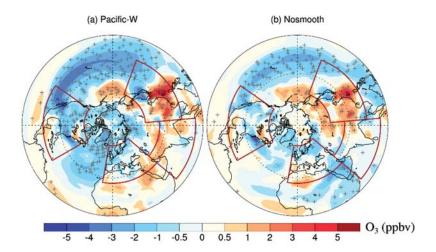


Figure R1. Changes in the summertime (June-August) surface ozone concentrations (ppbv) in the Northern Hemisphere for (a) Pacific-W and (b) Nosmooth relative to the CTRL. The four major regions of interest (i.e., NA (15°N–55 °N; 60°W–125°W), EU (25°N–65 °N;10°W-50 °E), EA (15 °N–50 °N; 95°E–160 °E) and SA (5 °N–35 °N; 50 °E–95°E)) are marked with red solid polygons. 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.

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).

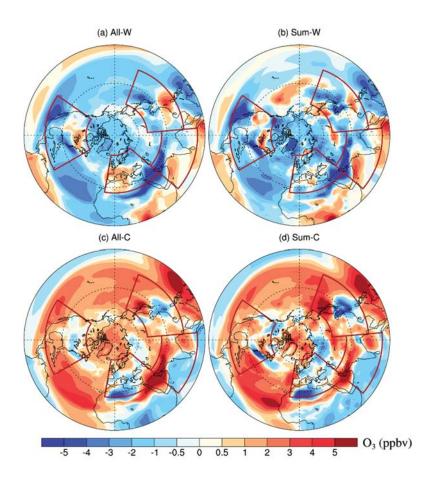


Figure S5. Left column: Changes in the summertime (June-August) surface O₃ concentrations (ppbv) in the Northern Hemisphere induced by 1°C warming (a) and 1°C cooling (b) in all three ocean basins (i.e., the North Pacific, North Atlantic and North Indian Ocean) relative to the CTRL. Right column: Sum of changes in the summertime (June-August) surface O₃ concentrations (ppbv) from the 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 the CTRL, denoted as (b) Sum-W and (d) Sum-C, respectively. The four major regions of interest (i.e., NA, EU, EA and SA) are marked with polygons.

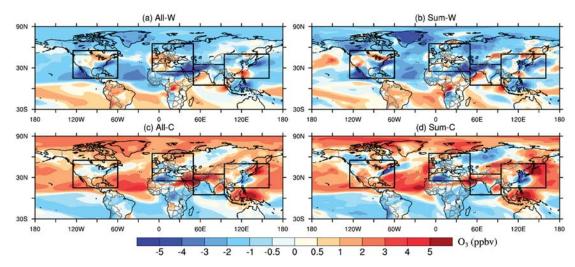


Figure S6. As in Figure S5 but using the Mercator projection.

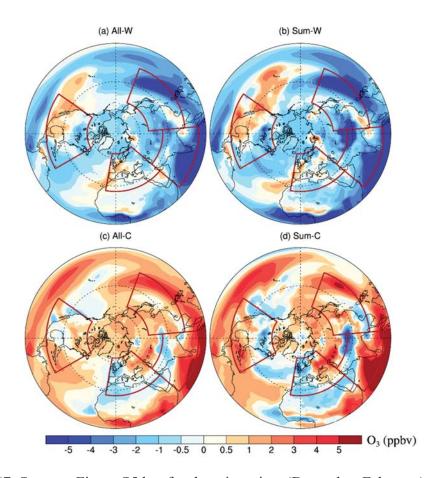


Figure S7. Same as Figure S5 but for the wintertime (December-February).

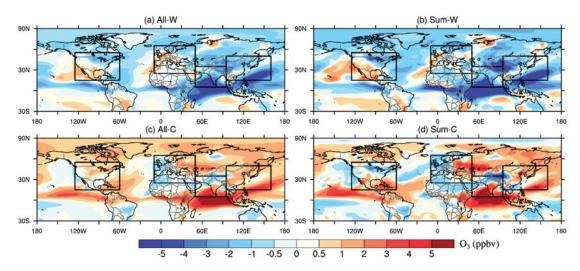


Figure S8. As in Figure S7 but using the Mercator projection.

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 (*P5*, *L135-138* and *L140-144*):

"...Except for the ENSO impacts, very few studies to date have directly addressed the linkage between SST and O₃. Therefore, a comprehensive understanding of the response of surface O₃ 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, $15^{\circ}N-55^{\circ}N$; $60^{\circ}W-125^{\circ}W$), Europe (EU, $25^{\circ}N-65^{\circ}N$; $10^{\circ}W-50^{\circ}E$), East Asia (EA, $15^{\circ}N-50^{\circ}N$; $95^{\circ}E-160^{\circ}E$) and South Asia (SA, $5^{\circ}N-35^{\circ}N$; $50^{\circ}E-95^{\circ}E$), defined in Fiore et al. (2009)) with respect to nearby basin-scale SST changes."

In Section 2.2 (*P7*, *L192-206*), we have:

"We first 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 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)."

In Section 3 (*P10*, *L282-291*):

"Our simulations reveal that different oceans can exert distinct region-specific effects on the O_3 distribution. 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 different regional SST changes (see Figures S5 and S7 in the supplementary material)."

In the Summary section (P20, L569-574), 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 this evolution. 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."

References:

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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 is an interesting modelling study that examines how surface ozone is influenced by warmer SSTs over the Pacific, Atlantic and Indian oceans. With a one degree warming across these basins, the changes in seasonal-mean ozone in the oceanic basin and its surrounding continents are rather large at 1-5 ppb. An increase in SST leads to lower surface ozone over the Pacific and Atlantic oceans but a more mixed response over the Indian ocean. The authors probe the contribution of chemistry and transport processes to these ozone changes. The paper is mostly well written but a number conclusions lack clarity and are not well-substantiated for reasons relating to poor and inconsistent figure quality and interpretation as outlined below. Hence the manuscript, needs much improvement before publication.

We greatly appreciate the reviewer for these detailed and valuable comments. In our revised manuscript, we have significantly improved the quality and consistency of the figures. The IPR analysis in this study has been described more clearly and all the relevant text has been clarified. We have also expanded our explanation and discussion by adding more sensitivity tests. By addressing the reviewer's comments, we believe our manuscript has considerably improved. Please see our response to each comment below:

Major comments:

1) (i) As noted by the other reviewer, the map projections used vary by figure in a non-logical fashion, hence it is extremely difficult to compare results across different figures and hence verify the conclusions in the text. For example, the vertical velocity changes in Figure 6 versus the surface pressure pattern changes in Figure 5 versus the changes in ozone concentrations in Figure 7 (See specific comments also). (ii) In addition, the continental outlines and hence oceanic basins are too difficult to distinguish if they are visible at all. (iii) Finally, most figure panels are too small to be legible- except for Figures S2-S5 which are hugely improved on the other figures (although the continental outlines are still hard to see in Figure S2).

Thanks for pointing out this. In our revised manuscript, we have fixed these problems and consistently used map projection. Specifically, we use Polar projection to show hemispheric scale results (e.g., Figures 1-4) and the Mercator projection to show basin-scale results (e.g., Figures 5-8). To make it more comparable, we also redraw Figures 1-4 using the Mercator projection and put them into the supplementary material (e.g., Figures S2, S14 and S15). Please see some examples of the improved figures below:

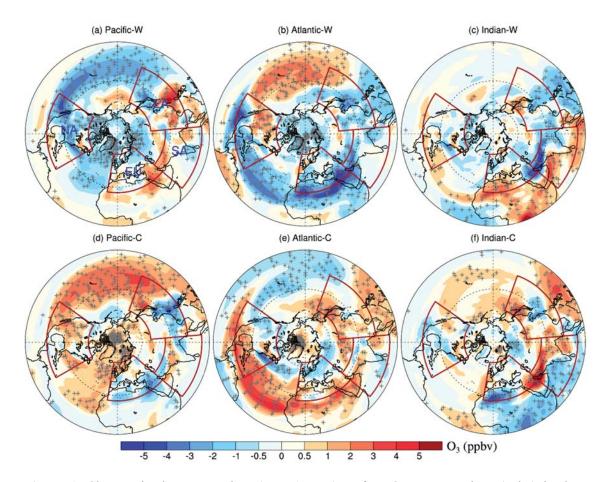


Figure 1. Changes in the summertime (June-August) surface O₃ concentrations (ppbv) in the Northern Hemisphere induced by 1°C warming (top) and 1°C cooling (bottom) in the North Pacific Ocean (left), North Atlantic Ocean (center), and North Indian Ocean (right) relative to the CTRL. The four major regions 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).

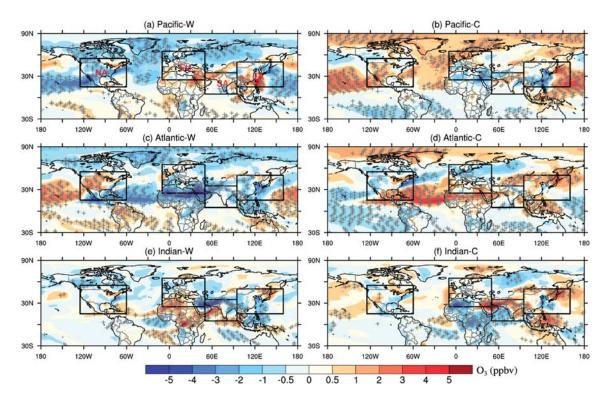


Figure S2. Changes in the summertime (June-August) surface O₃ concentrations (ppbv) in the Northern Hemisphere induced by 1°C warming (top) and 1°C cooling (bottom) in the North Pacific Ocean (left), North Atlantic Ocean (center), and North Indian Ocean (right) relative to the CTRL. The four major regions 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 polygons. 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.

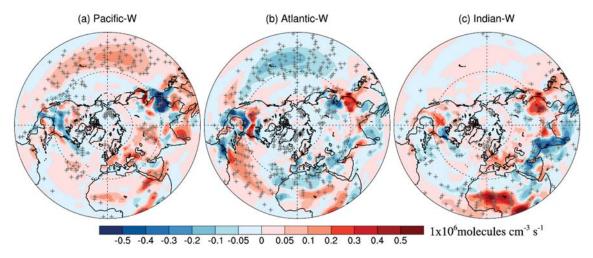


Figure 3. Perturbations of the surface net O_3 production rate $(1x10^6 \text{ molecules cm}^{-3} \text{ s}^{-1})$ for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The +

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).

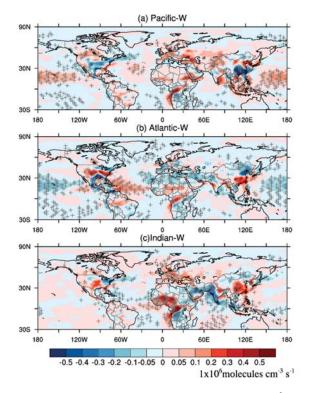


Figure S14. Perturbations of the surface net O₃ production rate (1x10⁶ molecules cm⁻³ s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

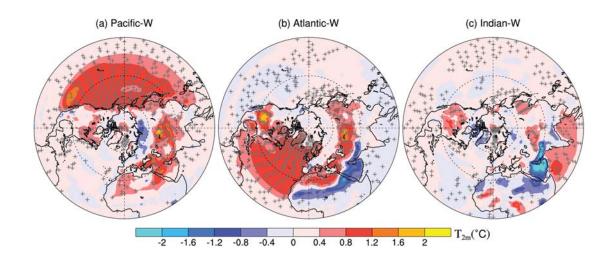


Figure 4. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL in the Northern Hemisphere in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data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

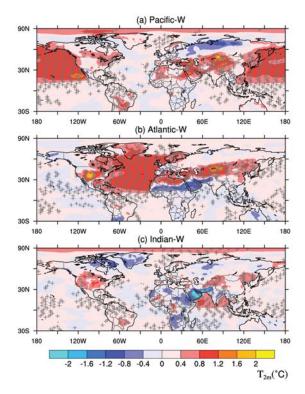


Figure S15. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

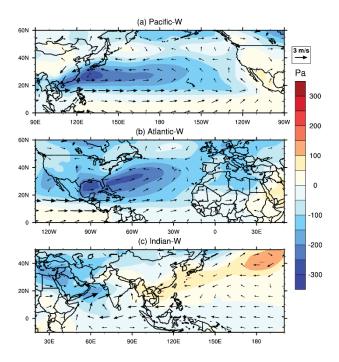


Figure 5. Changes in the surface pressure (color contours, Pa) and 850-hPa wind (arrows, m s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer.

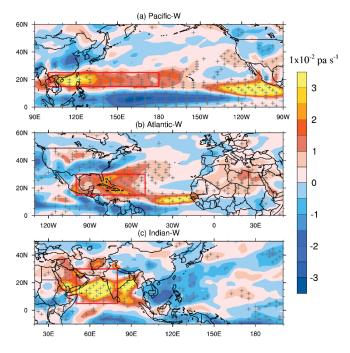


Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. Red polygons denote the regions where the surface pressure responses 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.

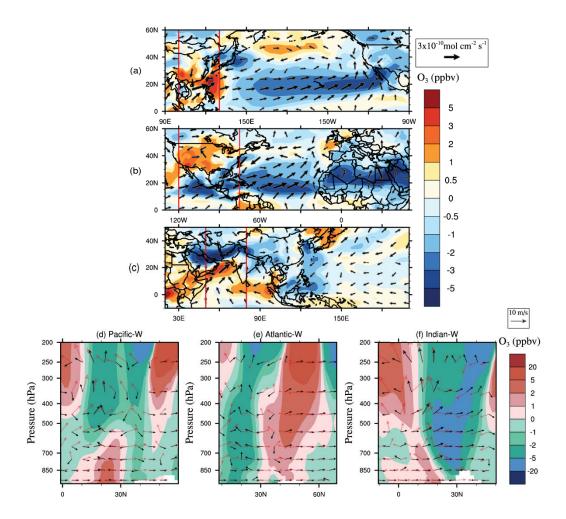


Figure 7. Top three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, 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.

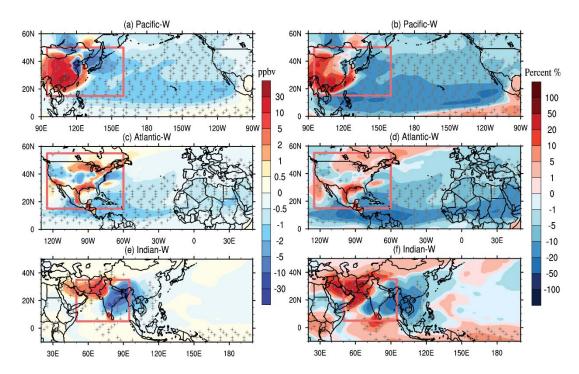


Figure 8. Left-hand panel: Difference in the surface concentration (ppbv) of a CO-like tracer emitted from (a) East Asia for Pacific-W, (c) North America for Atlantic-W and (e) the South Asia for Indian-W relative to the CTRL in the boreal summer. Right-hand panel: The percentage changes in the surface 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.

The continental outlines in all figures are now thicker and darker than previous ones. Please see Figure 4 above as an example and refer to the revised manuscript for more details.

Some figures have a set of small plots. In the previous version, we uploaded low-quality PDF plots. In this revised version, we have significantly improved the figure quality. Please see Figure 9 below for example:

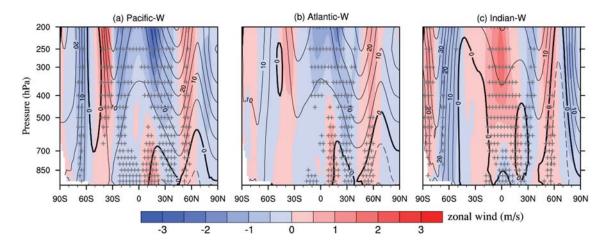


Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential height (contour, m) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. Black solid and dashed lines in the contours indicate positive and negative geopotential height anomalies, 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.

2) The seasonal mean surface ozone changes are quite large. This message could be brought out much more clearly. It would be beneficial to see some discussion of the magnitude of these surface ozone responses through comparison with previous papers even if these only relate to the effects of changes in air temperature or climate on surface ozone, as the further impacts of atmospheric circulation changes can be outlined.

Good suggestion. We add some discussion about the magnitude of these surface ozone responses in our revised manuscript, and compare our results with previous works (see *P9*, *L248-260*):

"The responses of surface O_3 concentrations to basin-scale SST changes (i.e., ± 1 °C) are mainly within 3 ppbv in the Northern Hemisphere (Table 1 and Table S1), though large anomalies (i.e., up to 5ppbv) are also observed over the east coast of China, the Indian subcontinent, and certain oceanic areas (Figure 1 and Figure S2). This SST-O3 sensitivity is comparable to previous findings. For instance, Bloomer et al. (2009) reported a positive O_3 -temperature relationship of $2.2\sim3.2$ 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 of surface O_3 by no more than 1 ppbv in response to 20% reductions in anthropogenic emissions within a continental

region. Our study indicates that basin-scale SST change alone may exert significant effects on the surface O_3 above specific ocean basin and its surrounding continents."

3) (i) The IPR analysis needs to be described more thoroughly and the processes selected would benefit with expanded definitions. In particular, gas-phase chemistry (CHEM) should be defined more clearly as later in the manuscript various other terms are used: net chemical production (Figure 3); photochemistry (line 265). Also vertical diffusion (VDIF) and dry deposition (DRYD) are combined into one term TURB- but these terms act in opposite directions in Figure 2. It would be useful to provide a brief outline as to why these terms are expected to act in opposite directions. (ii) All IPR related figures- Figures 2/S1 are very difficult to read. In addition the relationship between the fluxes and concentrations as plotted on figure 2 is unclear, and appears sensitive to the scaling's used on the right and left hand y-axes. See specific comments 3-8 below. (iii) The text discussing IPR results in section 4.1 is generally confusing and not well substantiated: often the season being referred to is not provided and general statements are sometimes given that only seem applicable to results in boreal summer. The text is section 4.3 also needs to be clarified and tightened in a good number of places- see specific comments. iv) For Figure 2/Table 1, it would be highly beneficial to also have results for the direct effect of a change in SSTs on regional surface ozone in that surface basin before any discussion of upwind or downwind continents. This would aid with interpretation as to the dilution of the ozone response with regional averaging.

(i)Good suggestion. The IPR analysis calculates the accumulated contributions of individual processes (e.g., net chemical production (production minus loss), advection, vertical diffusion, dry deposition, etc.) to the changes of O₃ concentrations. It is a widely used tool for air pollution diagnostics (Li et al., 2012; Zhang and Wu, 2013; Tao et al., 2015). In this study, we implemented the IPR scheme in the model to examine the O₃ flux in individual processes, including gas-phase chemistry, advection, vertical diffusion, dry deposition, shallow convection and deep convection. The wet deposition and aqueous-phase chemistry are ignored because of the low solubility and production rate of O₃ in water (Jacob, 1999). The sum of the IPR archived fluxes in CHEM. ADVE, VDIF, DRYD, SHAL and DEEP matches well the changes of O₃ concentration (Figure S1). Here the CHEM represents the net production (or production minus loss) flux of O₃ from gasphase chemistry, which is consistent with the net production rate shown in Figure 3. DRYD represents the removal rate of O₃ by dry deposition. VDIF represents the transport of O₃ to the surface due to vertical diffusion. Both DRYD and VDIF are closely dependent on turbulent mixing. The efficiencies of O₃ vertical diffusion and the corresponding dry deposition are all positively related to the strength of turbulence, but with opposite sign. We therefore define a new term TURB to represent the sum of DRYD and VDIF, which can represent the overall effect of

turbulence change on surface O_3 . In the revised manuscript, we added more descriptions about the IPR method in Section 2.3 and Section 4.1. Please also refer to our reply to the specific comments for more details.

In Section 2.3 (P8, L226-238):

"...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 O3 concentrations in every grid box. Wet deposition and aqueous-phase chemistry are ignored here due to the low solubility and negligible chemical production of O3 in water (Jacob, 1999). Therefore, CHEM represents the net production (production minus loss) rate of O3 due to gas-phase photochemistry. DRYD represents the dry deposition fluxes of O3, which is an important sink for O3. The other IPR terms (i.e., ADVE, VDIF, SHAL and DEEP) represent contributions from different transport processes. The IPR scheme tracks and archives the O3 flux in each grid from every process during each model time step. The sum of the O3 fluxes from these six processes matches the change in the O3 concentration. The IPR performance is verified by comparing the predicted hourly O3 changes with the sum of the individual fluxes from the six processes. As shown in Figure S1, the hourly surface O3 changes are well represented by the sum of these fluxes in the model."

In Section 4.1 (P10-11,L296-316), we also add more explanations about the IPR results:

"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). 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). In most cases, VDIF and DRYD are the key processes controlling the O_3 variation. The downward transport of O_3 through diffusion 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). More detailed IPR results are shown in Figures S10 and S11 in the supplementary material."

(ii) As we have mentioned above, each of the IPR processes (i.e., CHEM. ADVE, VDIF, DRYD, SHAL and DEEP) archived hourly and the sum of them matches well the time-varying O₃ concentration changes. Figure S1 in the supporting materials (see below) demonstrates the performance of the IPR scheme.

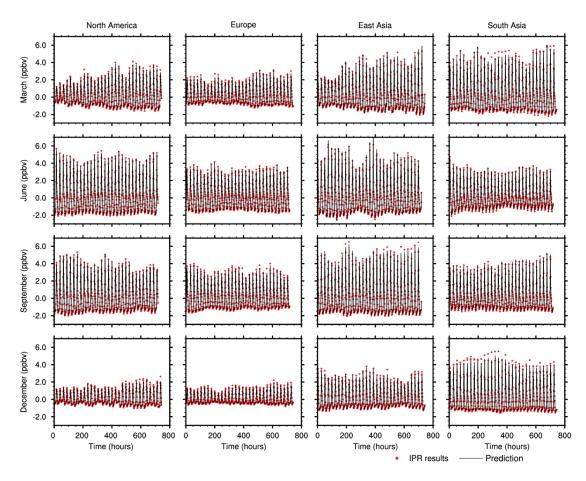


Figure S1. Hourly time-series of predicted O₃ changes (black lines) and the sum of IPR results (red dots) averaged over the four major regions of interest (i.e., America (15°N–55 °N; 60°W–125°W), Europe (25°N–65 °N;10°W-50 °E), East Asia (15 °N–50 °N; 95°E–160 °E) and South Asia (5 °N–35 °N; 50 °E–95°E)) during March (first row), June (second row), September (third row) and

December (last row) of random modeling years in the CTRL.

Figure 2, on the other hand, demonstrates the seasonally averaged results rather than the time-series shown in Figure S1. Positive changes of fluxes are generally counterbalanced by the negative ones because the climatological O_3 concentrations reside in an equilibrium when averaged over a long period of time. Therefore, the net flux changes could not be directly compared with surface O_3 changes between two climatological cases. Here we compared these IPR fluxes individually to identify the impact of basin-scale SST changes on each O_3 evolution process. Typically, the positive change of a particular IPR process is mainly responsible for the increase of surface O_3 , which may further induces O_3 loss process to counteract these factors. With this information, we can explore the relative importance of different processes closely linked to the SST changes, which helps to explain the variability of surface O_3 over different regions.

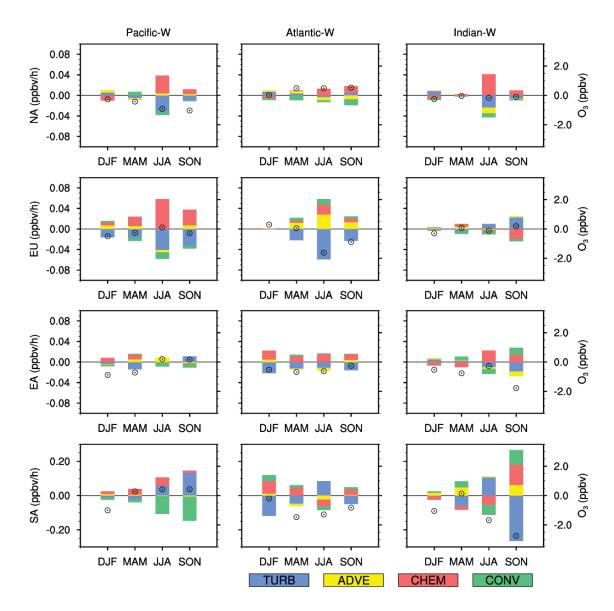


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second 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 S10 in the supplementary material.

(iii) We agree that some text needs more clarification. This study focuses mainly on summertime since both surface O₃ levels and their response to SST changes are highest during this period. We also find that an increase in SST in the North Pacific or North Atlantic tends to elevate the VDIF

of O₃ over upwind regions (i.e., East Asia and North America, respectively) but suppress it over downwind regions during JJA. In the revised manuscript, we have clarified the seasons we discussed and improved the consistency of the analysis. Please see our response to specific comments below.

iv) Good suggestion! We have added a similar table (Table S1) and figure (Figure S9) to examine the effect of SST changes on O₃ distribution over different ocean basins.

Table S1. Regionally and seasonally averaged (only ocean grid boxes are included) changes in surface O₃ concentrations (ppbv) over three ocean basins in the Northern Hemisphere (i.e., the North Pacific Ocean (15°N-65°N;100°E-90°W), the North Atlantic Ocean (15°N-65°N; 100°W-20°E) and the North Indian Ocean (5°N-30°N; 30°E-100°E)) 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 Pacific	-0.56*	-0.71*	-0.78*	-1.22*
		North Atlantic	-0.55*	-1.08*	-0.74*	-1.25*
		North India	-1.05*	-0.59	0.16	0.20
	-1°C	North Pacific	0.32	0.55*	1.04*	1.00*
		North Atlantic	0.43*	0.53*	0.75*	0.80^{*}
		North India	0.77^{*}	-0.06	-0.03	0.16
North Atlantic	+1°C	North Pacific	0.05	-0.02	0.38*	0.01
		North Atlantic	0.14	0.04	-1.00*	-0.86*
		North India	-0.45*	-1.31*	-0.63*	-0.72*
	-1°C	North Pacific	0.11	0.32	0.11	-0.30
		North Atlantic	-0.02	-0.14	0.76*	0.43*
		North India	0.39	0.59	0.38*	0.48
North India	+1°C	North Pacific	-0.34	-0.11	-0.14	-0.88*
		North Atlantic	-0.25	-0.46	-0.11	-0.23
		North India	-1.59*	-0.42	0.81*	-2.11*
	-1°C	North Pacific	0.32	0.32	0.50*	0.52*
		North Atlantic	-0.07	-0.42	0.11	-0.37*
		North India	1.32*	0.89*	-0.38*	1.84*

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

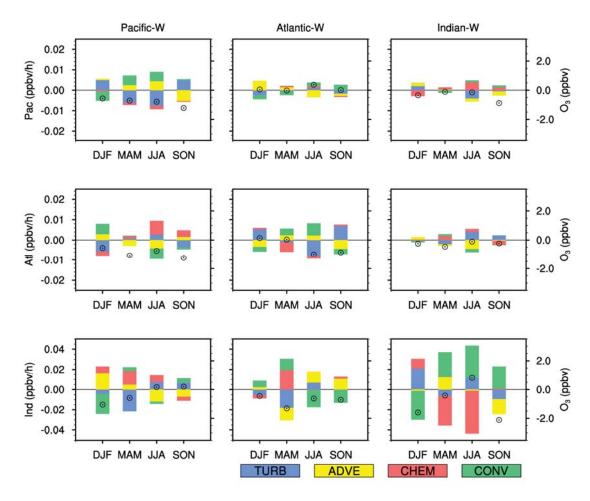


Figure S9. Seasonally averaged changes in the IPR contributions (bars, ppbv h⁻¹, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over North Pacific (15°N-65°N; 100°E-90°W, demoted as "Pac", first row), North Atlantic (15°N-65°N; 100°W-20°E, demoted as "Atl", second row) and North Indian Ocean (5°N-30°N; 30°E-100°E, demoted as "Ind", third row). IPR contributions from the four processes (i.e., TURB, ADVE, CHEM and CONV) are represented by different colors.

It shows that during boreal summers, the warming of North Pacific or North Atlantic leads to a widespread decrease of surface O_3 , while the warming of North Indian Ocean increases local surface O_3 . The IPR results indicate that the warming of the North Pacific or North Atlantic induce a reduction of TURB (mainly caused by the decrease of VDIF) and CHEM, which are responsible for the significant decrease of surface O_3 in JJA (Figure S19). The North Indian Ocean warming, on the other hand, enhances the CONV and TURB locally, leading to an increase of local surface O_3 in JJA. In the revised manuscript, more discussions on these effects are given:

In Section 3 (*P9-10*, *L266-276*):

"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 "Atlantic-W" case, the surface O_3 levels decrease by 1~2 ppbv over the North Atlantic and Europe but increase (~1 ppbv) 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)."

In Section 4.1 (*P12*, *L331-335* and *L337-340*):

"...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). 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)."

4) As noted above for the IPR results, but also in general, the text on the various contributions or roles of intercontinental transport versus that of chemistry is difficult to follow in a number of places and some conclusions appear over-stated. For example, the abstract discusses "suppression of O3 intercontinental transport due to increased stagnation at mid-latitudes induced by SST changes". Stagnation is a localised process largely determined by boundary layer processes and entrainment. Hence, the authors should be cautious in their interpretation based on large-scale changes in wind vectors and vertical velocity to infer changes in stagnation/ventilation. Perhaps clear definitions of what is meant by these terms would be useful. See specific comments below.

Thanks for bringing this issue up. We agree that stagnation/ventilation were improperly used here. Throughout the analysis, we find that the basin-scale SST increase not only strengthens upward motions over the low-latitudes oceans, but also lead to decreases of upward velocity over midlatitudes (Figure S23). Previous studies have revealed that strengthened deep convection will

trigger large-scale subsidence over the nearby regions through modulating large-scale circulations, which may suppress convective air movement there (Lau et al., 1997;Roxy et al., 2015;Ueda et al., 2015). Here we also demonstrate a weaker vertical temperature gradient associated with regional SST warming (Figure S16). Both factors (i.e., large-scale subsidence and weaker vertical temperature gradient) tend to stabilize the atmosphere that may inhibit vertical air transport. In our revised manuscript, we further examine the vertical transport of O₃ based on the IPR analysis (shown in Figure S12). It shows a widespread reduction of vertical diffusion transport of O₃ to the surface (i.e. VDIF) except for the upwind regions. We also find that SST increases of a specific ocean in the Northern Hemisphere, especially for the North Pacific and North Atlantic oceans, tend to increase the air temperature (Figure S16) and geopotential height (Figure 9) more significantly at mid-latitudes than other latitudes. Consequently, the meridional geopotential height gradient is decreasing in the tropical-to-mid-latitude troposphere while increasing at higher latitudes. It tends to decrease the westerly wind at lower-middle latitudes (25°N - 45 °N) in the Northern Hemisphere (Figure 9). Based on these facts, the warming of SST over a specific ocean may stabilize the troposphere at mid-latitudes that suppress the O₃ intercontinental transport. This effect is also supported by the CO-tracer analysis, which shows a significant reduction of intercontinental transport (Figure 8). We have discussed these processes in detail in the revised manuscript.

Here we revised the texts as below:

In Abstract (*P2*, *L35-37*):

"... 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."

In Section 4.3 (P15, L422-427), we have:

"...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). 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."

In the summary section (*P19-20, L563-567*):

"...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 ."

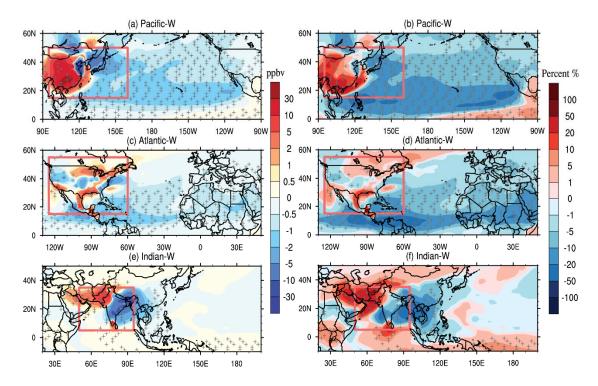


Figure 8. Left-hand panel: Difference in the surface concentration (ppbv) of a CO-like tracer emitted from (a) East Asia for Pacific-W, (c) North America for Atlantic-W and (e) the South Asia for Indian-W relative to the CTRL in the boreal summer. Right-hand panel: The percentage changes in the surface 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.

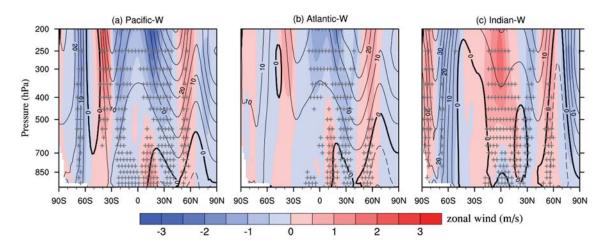


Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential height (contour, m) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. Black solid and dashed lines in the contours indicate positive and negative geopotential height anomalies, 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.

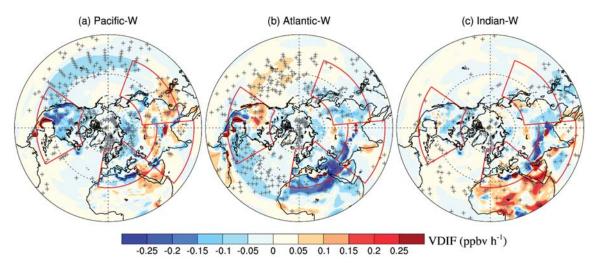


Figure S12. Changes in VDIF (ppbv h⁻¹) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. The four major regions of interest (i.e., NA, EU, EA and SA) are marked by red solid lines. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

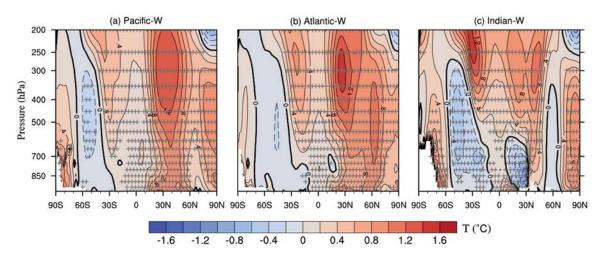


Figure S16. Vertical-meridional distributions of changes in the air temperature (contours, °C) for (a) Pacific_W (zonal averaged from 100°E-90°W), (b) Atlantic_W (zonal averaged from 100°W-180°W) and (c) Indian_W (zonal averaged from 30°E-100°E) relative to the CTRL in boreal summer. Black solid and dashed lines in the contours indicate positive and negative air temperature anomalies, respectively (contour interval: 0.2 °C). The + symbol denotes areas where the changes of air temperature are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

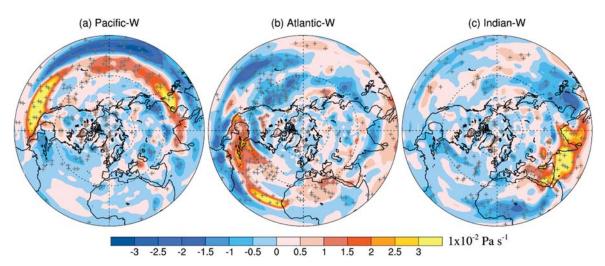


Figure S23. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. The + symbols indicate areas where results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

5) A number of references in the text are rather old, and some updated references would be highly beneficial. See specific comments below. Also with multi-references the logical order is unclear-chronological order is most commonly used.

Good suggestion! We have updated our references by citing more recent studies. Please see our response to specific comments below. We have also reordered references chronologically.

Specific comments:

1) As also noted by the other reviewer the frequent use of parenthesis to state a key result dilutes the message of the sentence and makes for a confusing read. Please rephrase when key points are being made in the abstract and main text (lines 223-230).

Good suggestion. We rephrased these key results following the reviewer's suggestions, see below or the revised abstract and text:

In Abstract (*P1*, *L24-27*):

"...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."

In Section 3 (*P9-10*, *L264-280*):

"...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). An increase in summertime SST over a specific ocean basin tends to increase the surface O₃ 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 O₃ over the North Pacific, North America and the North Atlantic of approximately 1 ppbv (Table S1) but may enhance the surface O₃ by nearly 3 ppbv over South China. Similarly, in the "Atlantic-W" case, the surface O₃ levels decrease by 1~2 ppbv over the North Atlantic and Europe but increase (~1 ppbv) over North America and the North Pacific. For the North Indian Ocean, positive SST anomalies tend to increase the surface O₃ over the Indian Ocean and Africa but decrease the surface O₃ over South and East Asia (Figure 1). During the boreal winter, a widespread decrease in surface O₃ 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 O₃ to basin-scale SST changes is typically insignificant. Details are shown in Figure S3 in the supplementary material."

2) Line 207/Table 1 – as noted above it would be beneficial to first show a similar table that examines the effect of SST changes within each basin and on other ocean basins.

Good suggestion. We add such a table in the supplementary material, see Table S1 below.

Table S1. Regionally and seasonally averaged (only ocean grid boxes are included) changes in surface O₃ concentrations (ppbv) over three ocean basins in the Northern Hemisphere (i.e., the North Pacific Ocean (15°N-65°N; 100°E-90°W), North Atlantic Ocean (15°N-65°N; 100°W-20°E) and North Indian Ocean (5°N-30°N; 30°E-100°E)) 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 Pacific	-0.56*	-0.71*	-0.78*	-1.22*
		North Atlantic	-0.55*	-1.08*	-0.74*	-1.25*
		North India	-1.05*	-0.59	0.16	0.20
	-1°C	North Pacific	0.32	0.55*	1.04*	1.00*
		North Atlantic	0.43*	0.53*	0.75*	0.80*
		North India	0.77*	-0.06	-0.03	0.16
North Atlantic	+1°C	North Pacific	0.05	-0.02	0.38*	0.01
		North Atlantic	0.14	0.04	-1.00*	-0.86*
		North India	-0.45*	-1.31*	-0.63*	-0.72*
	-1°C	North Pacific	0.11	0.32	0.11	-0.30
		North Atlantic	-0.02	-0.14	0.76*	0.43*
		North India	0.39	0.59	0.38*	0.48
North India	+1°C	North Pacific	-0.34	-0.11	-0.14	-0.88*
		North Atlantic	-0.25	-0.46	-0.11	-0.23
		North India	-1.59*	-0.42	0.81*	-2.11*
	-1°C	North Pacific	0.32	0.32	0.50*	0.52*
		North Atlantic	-0.07	-0.42	0.11	-0.37*
		North India	1.32*	0.89*	-0.38*	1.84*

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

We also revised the text accordingly, see Section 3 (P9-10, L264-276) or below:

"...Surface O_3 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 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 "Atlantic-W" case, the surface O_3 levels decrease by 1~2 ppbv over the North Atlantic and Europe but increase (~1 ppbv) 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)."

3) Line 242- what is meant by atmospheric turbulence intensity and explain to the reader how this relates to VDIF and DRYD.

Good question. Both VDIF and DRYD processes are dynamically determined by the strength of turbulence. Stronger turbulence enhances the downward transport of O₃ to the ground level, which also induces more O₃ dry deposition. Therefore, DRYD tends to behave concurrently with VDIF, but with an opposite sign. We have clarified this in our revised manuscript (see *P11*, *L308-313*):

"...In most cases, VDIF and DRYD are the key processes controlling the O_3 variation. The downward transport of O_3 through diffusion 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."

4) Line 248 "reducing it over North America. For the Pacific W panels in Figure 2 a reduction in VDIF is only seen in in summer in North America; VDIF increases just as strongly in North America in winter and spring.

This sentence only refers to summers. We have clarified this in Section 4.1 of the revised manuscript (*P11*, *L323-325*).

"...In the "Atlantic-W" case, increases in VDIF are also observed over the upwind regions (i.e., North America) in JJA."

5) Line 248- "similar increases in VDIF are simulated over North America. Similar to?

We state the increases in VDIF over North America in JJA is a "similar increase" because it also happens over upwind regions associated with the North Atlantic warming that is similar to the East Asia in the "Pacific-W" case. We realize that the word "similar" may induce confusion and we delete it in the revised manuscript (*P11*, *L323-325*).

"...In the "Atlantic-W" case, increases in VDIF are also observed over the upwind regions (i.e., North America) in JJA."

6) Line 253- "the increase of CHEM tends to dominate the surface O3 increase over North America." This is not obvious from Figure 2 (and it is unclear which season/s are being discussed), and is unintuitive without a clearer definition of CHEM, and how fluxes relate to concentrations in Figure 2.

Here we show the key processes enhancing the surface O₃ over North America in JJA in the Atlantic_W case. CHEM tracks the surface O₃ flux due to net chemical production (i.e., production minus loss). In the revised Figure 2, we replaced VDIF and DRYD by TURB, and DEEP and SHAL by CONV. Now it is easier to see that a warmer SST over the Atlantic enhances O₃ chemical production, which positively contributes to the surface O₃ increase over North America. Please see the revised Figure 2 below:

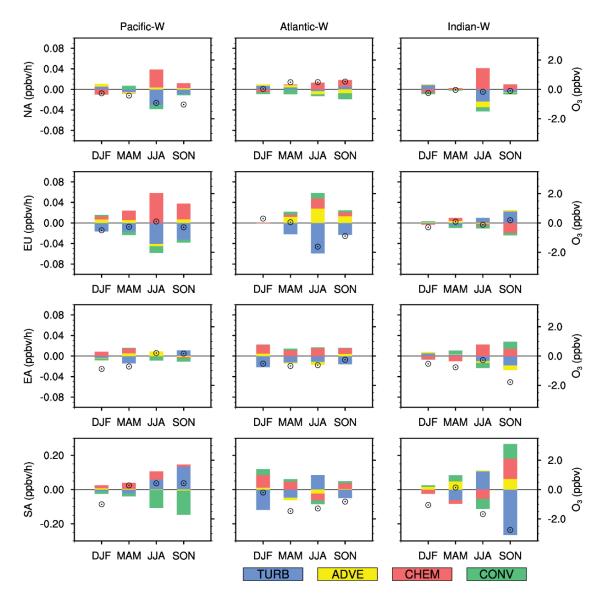


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second 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 S10 in the supplementary material.

In the revised manuscript, we have rephrased this sentence (see P11, L323-328):

"...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 O₃ increase over North America in JJA."

7) Line 254- "TURB is more important ... leasing to reduced surface O3 concentrations." Again the positive and negative fluxes in JJA and SON look to balance so why are there reduced ozone concentrations. Line 257- as above the fluxes look as though they balance (especially in JJA) but ozone concentrations are reduced.

As mentioned in our reply to comment (3), the IPR scheme in our study tracks all processes that are related to the O₃ formation. It 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 relevant analysis (usually measuring the difference between two equilibrium states), the net change of all IPRs approaches zero. The multi-year seasonally averaged positive and negative fluxes are balanced with each other after model spin-up. Typically, the positive change of a particular IPR process is mainly responsible for the increase of surface O₃, which may further induce O₃ removal to counteract this forcing. Therefore, here the IPR analysis is not used to budget SST induced O₃ concentration changes. Instead, it helps to screen out the key processes driving the sensitivity of O₃ to a SST forcing. In the revised manuscript, we have clarified this issue and focused mainly on the individual process-level responses to SST changes. Please see our revised text in Section 4.1 (see *P10-11*, *L296-316* and *L318-335*) or below:

"IPR analysis is used to evaluate the contribution of different physicochemical processes to O₃ 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). 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 O₃, which may further induce O₃ removal to balance this forcing in a new equilibrium. Therefore, here, the IPR analysis is used not to budget the SST-induced O₃ concentration changes but rather to help examine the relative importance of different transport and chemical processes in driving the sensitivity of O₃ to SST forcing. In this study, the SST-induced, process-level O₃ 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). In most cases, VDIF and DRYD are the key processes controlling the O₃ variation. The downward transport of O₃ through diffusion is an important source of surface O₃, 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 O₃ 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). More detailed IPR results are shown in Figures S10 and S11 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), which is insignificant if averaged over the whole East Asia region. Meanwhile, this Pacific warming considerably reduces VDIF over North America (Figure S10). The corresponding decrease in TURB over North America mainly determines the surface O₃ 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 O₃ 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 O₃ 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 O₃ above these regions in JJA (Figure S11). The North Indian Ocean warming, on the other hand, enhances DEEP and VDIF, leading to a local increase in surface O_3 in JJA."

8) Lines 260-263- It would be helpful to define remote versus downwind. Remote is used in this sentence and downwind in the following sentence. If North America is the remote continent in the Pacific W simulation then VDIF is only suppressed in summer, but not in winter and spring.

Good suggestion. To avoid confusion, we have replaced "remote" with "downwind" for consistency (*P12*, *L337-342*):

"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). These opposite changes in VDIF over upwind and downwind regions lead to distinct surface O_3 responses."

9) Line 266- "change in photochemistry. . . advection . . . dominates the feedbacks of Indian Ocean warming- CHEM appears as a substantial component in the lowermost right hand panel of Figure 2.

Good question! As shown in Figure 2, the North Indian warming leads to substantial decreases of CHEM and CONV, which are responsible for the reduction of surface O₃ over South Asia. We have clarified this sentence in Section 3 of the revised manuscript (*P12*, *L329-331*):

"...In the "Indian-W" case, both CHEM and CONV are reduced over South Asia in JJA, leading to overall reductions in surface O₃ over the Indian subcontinent (Figure 2)."

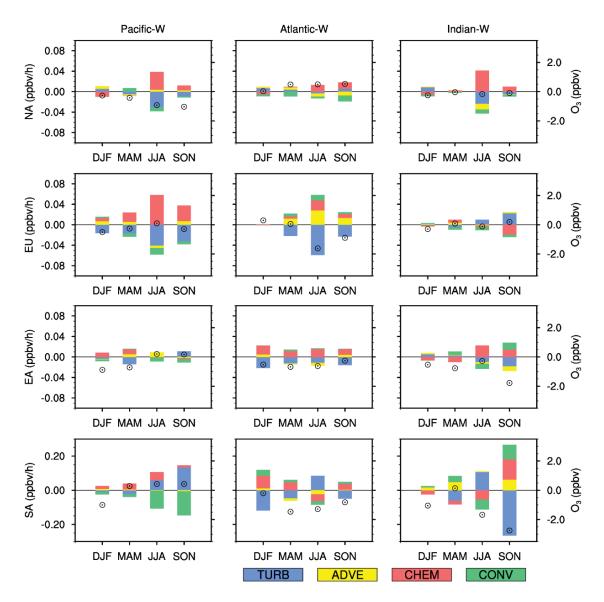


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second 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 S10 in the supplementary material.

10) Line 275- "Peak changes are confined to the polluted region because of their high precursor emissions". This is not obviously related. Please explain this statement more clearly. The examples that follow to the end of the paragraph referring to Figure 3 (the regions discussed are hard to see) do not clearly substantiate this.

As shown in Figure 3, changes of O₃ net production rate are generally highest over North America, East and South Asia where O₃ precursors' emissions are high. In addition, significant change also happens in tropical Africa, when North Indian SST is warmer. Therefore, we agree with the reviewer that our original explanation is not clear. Please see our revised text in Section 4.2 (*P12*, *L350-357*) or below:

"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."

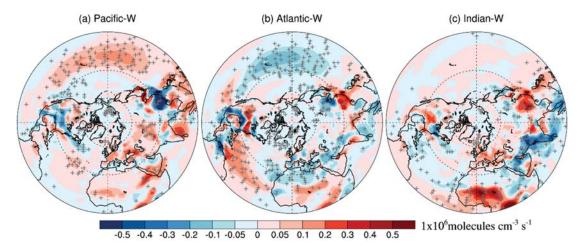


Figure 3. Perturbations of the surface net O_3 production rate $(1x10^6 \text{ molecules cm}^{-3} \text{ s}^{-1})$ for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The +

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).

11) Lines 290 to end of paragraph- "Increase in SST facilitates moist convection... Lau. . .1997)"- Figure 2 for summer for the Indian W influence on SA suggests a decrease in deep convection. Please clarify? Here the references used are rather old. This is an interesting find, an increase in SST would be normally accompanied by an increase in surface air temperature directly above the ocean, but yet figure S3 shows cooling but with warming above. Hence it would be beneficial to provide an expanded interpretation of this finding, compare the results with those from more recent papers on how elevated SSTs in the Indian Ocean region or tropics affect surface air temperature and convection.

Good suggestion. Figure S11 shows that a warmer North Indian Ocean increases the DEEP (i.e., the contribution of deep convection to surface O₃) while Figure S10 suggests a decrease in the DEEP over South Asia. Actually, this SST increase facilitates deep convection over the North Indian Ocean while suppresses deep convection over the Indian subcontinent. Generally, an increase in SST for tropical oceans are more likely to enhance evaporation and vertical movement of warm moist air above its surface (Lau and Nath, 1994; Lau et al., 1997; Hartmann, 2015). However, its effects on convection over nearby and remote regions are rather complicated. In this case, the SST increase over the Indian Ocean strengthens deep-convection above it according to our analysis as well as previous studies (Roxy, 2014;Xi et al., 2015;Chaudhari et al., 2016). The enhanced upward movement of warm moist air above the Indian Ocean promotes cloud formation in the upper troposphere. As demonstrated in our study, there is a remarkable reduction of solar radiation received at the surface (Figure S17) and a significant decrease of surface air temperature (Figure 4) over Indian subcontinent. This decrease of surface temperature over the Indian subcontinent suppresses the development of deep-convection above. Additionally, the latent heat release from convective activity warms the air temperature in upper troposphere significantly (Sabeerali et al., 2012;Xi et al., 2015), leading to opposite changes of air temperature between upper and lower troposphere over South Asia. We have discussed these processes with some most recent references in Section 4.2 of the revised manuscript (P13, L363-380).

"...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). Associated with this temperature decrease is a remarkable reduction in the solar radiation received at the continent below (more than 15 W/m², Figure S17). 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 blocks the solar radiation reaching the surface of the Indian subcontinent and reduce 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 S16), which may further suppress the development of deep convection over the Indian subcontinent."

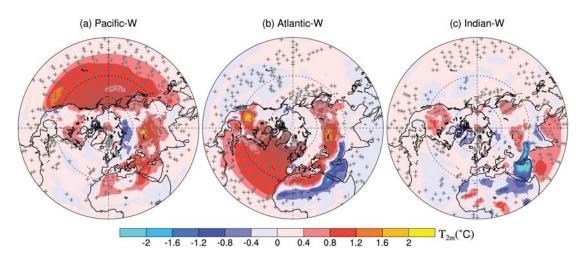


Figure 4. Changes in the surface air temperature (°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL in the Northern Hemisphere in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data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

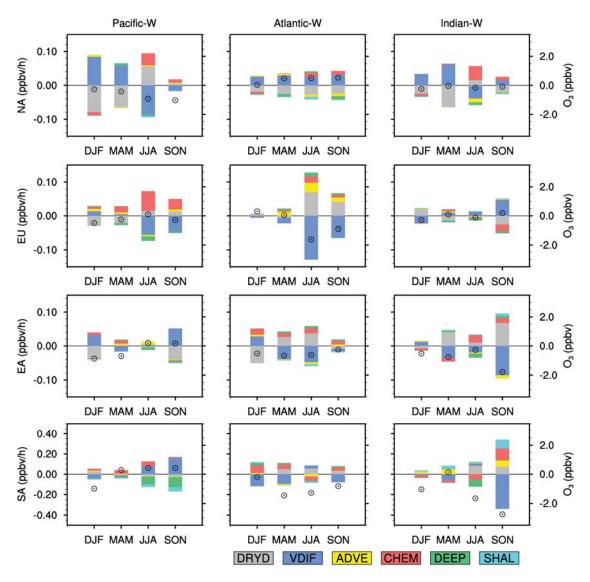


Figure S10. Seasonally averaged changes in the IPR contributions (bars, ppbv h⁻¹, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second row), EA (third row) and SA (last row). IPR contributions from the six processes (i.e., DRYD, VDIF, ADVE, CHEM, DEEP and SHAL) are represented by different colors.

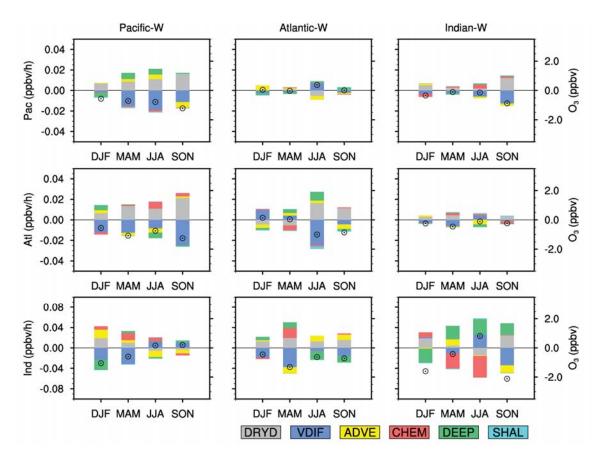


Figure S11. Same as Figure S10 but for the three ocean basins defined in our study.

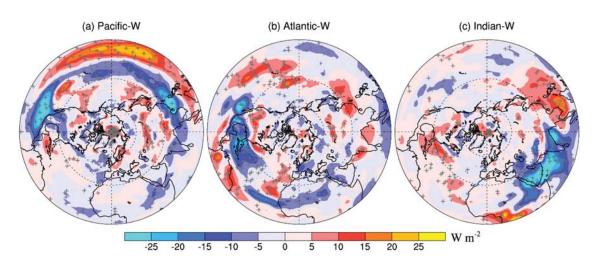


Figure S17. Perturbations of the surface solar radiations (W m⁻²) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test.

12) Line 297- The text relating ozone production -temperature relationships to net surface ozone production relationships with temperature should be clarified: it is the ozone not the ozone

production that is related to temperature in the references cited, and as the authors note both ozone production and destruction rates will increase with temperature (directly and indirectly through higher humidity).

Good suggestion. Figure S19 compares the O₃ chemical production rate and destruction rate. The former usually dominates the net chemical production over continental regions, whereas the latter is relatively important over oceans. This indicates that although both ozone production and destruction rates increase with temperature, the response of O₃ is more relevant to the changes of O₃ production rate over continental regions. In contrast, the effects of humidity on O₃ destruction and concentrations are more important over coastal and oceanic areas. Please see the revised text in Section 4.2 (*P13-14*, *L382-389*) or below:

"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 S19, 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 S21), which enhances O_3 destruction over most coastal and oceanic areas."

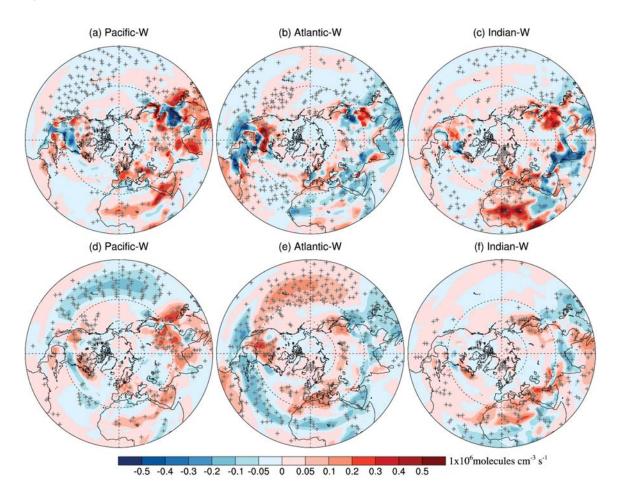


Figure S19. Top row: Perturbations of the surface chemical O_3 production rate $(1x10^6 \text{ molecules cm}^{-3} \text{ s}^{-1})$ for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Bottom row: Perturbations of surface chemical O_3 loss rate $(1x10^6 \text{ molecules cm}^{-3} \text{ s}^{-1})$ for (d) Pacific-W, (e) Atlantic-W, and (f) 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.

13) Line 318- As shown in Figure 6. . . surface pressure reduction is closely associated with enhanced upward motion. Please use the same map in Figures 5 and 6 in order to see this association.

Good suggestion. Now we use the same map for Figures 5 and 6 (see below), and put the original Figure 6 (using polar projection) into the supporting information (i.e., Figure S23):

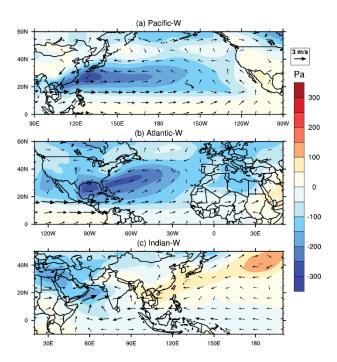


Figure 5. Changes in the surface pressure (color contours, Pa) and 850-hPa wind (arrows, m s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer.

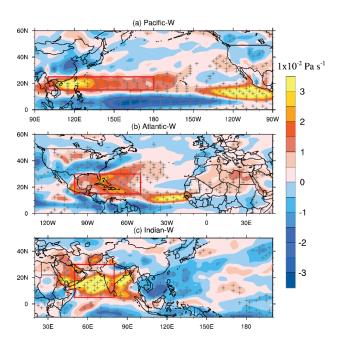


Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, $1x10^{-2}$ Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. Red polygons denote the regions where the surface pressure responses 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.

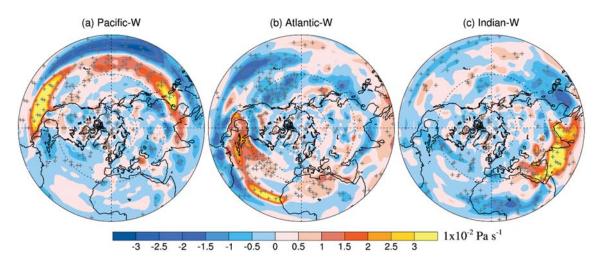


Figure S23. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. The + symbols indicate areas where results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

14) Line 329- "This effect is confirmed by widespread decreases of upward vertical velocity". Again it is hard to see if vertical velocity reductions are occurring only over the adjacent regions to the regions where the authors suggest enhanced convection may occur.

As shown in Figure 6, a basin-scale SST increase strengthens the upward motions locally. Adjacent to these anomalous upward motion, significant decreases of upward velocity are observed. For example, decreases of upward velocity are shown over East Asia associated with the North Pacific warming, and similarly the changes over North America associated with the North Atlantic warming. Additionally, decreases of upward vertical velocity are also demonstrated over higher latitudes and remote regions (shown in Figure S23). For example, the warming of the North Atlantic may induces a decrease of upward vertical velocity over the North Pacific. These effects have been reported in previous studies (Lau et al., 1997;Roxy et al., 2015;Ueda et al., 2015). We have optimized the relevant figures to verify our conclusion, please refer to our response to the former comment for the relevant figures (Figure 6 and Figure S23). We also modified the text below in Section 4.3 (*P14-15*, *L416-422*) for clarification:

"...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)."

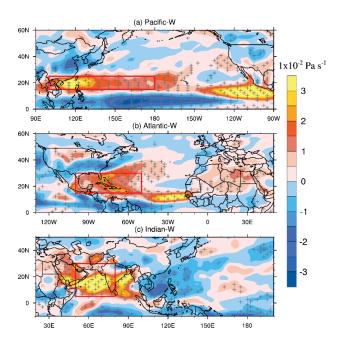


Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive

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.

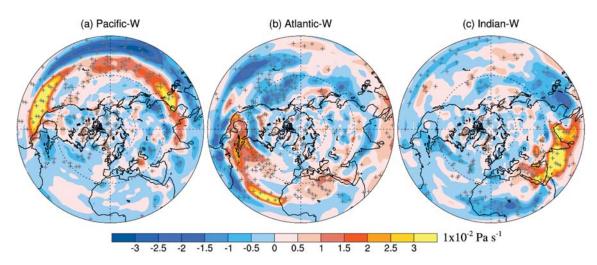


Figure S23. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. The + symbols indicate areas where results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

15) Line 333-end of paragraph. This first sentence of the paragraph discusses atmospheric stability based on zonal mean large-scale temperatures changes between the upper and lower troposphere (a weaker vertical temperature gradient; Figure S3) and stagnation/ventilation which are local processes often related to surface winds. Hence these processes may not be as simply related as suggested. In addition, a differential ozone response over clean and polluted regions seem unlikely to be associated with change in atmospheric stability associated with large-scale increases in upper tropospheric temperature. The final sentence of the paragraph needs substantiated especially given the link proposed in the previous section between clean regions with reduced net ozone production due to greater destruction.

Referring to our response to major comment 4, we agree that stagnation/ventilation were improperly used here and have rephrased our explanation in the revised manuscript. The basin-scale SST increase not only strengthens upward motions over the low-latitudes of the specific ocean, but also leads to the decrease of upward velocity over mid-latitudes (Figure S23). Previous studies have revealed that strengthened deep convection will trigger large-scale subsidence over other regions through modulating large-scale circulations, which may suppress convective air movement there (Lau et al., 1997;Roxy et al., 2015;Ueda et al., 2015). Here we also demonstrate a

weaker vertical temperature gradient associated with regional SST warming (Figure S16). Both factors (i.e., large-scale subsidence and weaker vertical temperature gradient) tend to slow down vertical air movement. In the revised manuscript, we further examined the vertical transport of O₃ based on the IPR analysis (shown in Figure S12). It shows a wide-spread reduction of transport of O₃ by vertical diffusion to the surface (i.e. VDIF). Considering that the responses of O₃ destructions to SST anomalies are more important over oceans than land (referring to our reply to previous comment 12), it is reasonable for us to infer that this reduced vertical transport may also exert a negative effect on surface O₃ over clean continents. Please see the relevant figures below. A detail explanation is provided in Section 4.3 of our revised manuscript (*P14-15*, *L416-430*):

"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). 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). 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 S12."

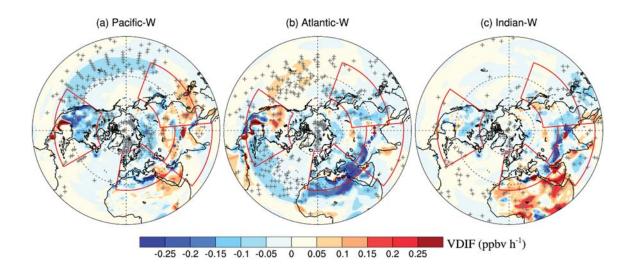


Figure S12. Changes in VDIF (ppbv h⁻¹) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. The four major regions of interest (i.e., NA, EU, EA and SA) are marked by red solid lines. The + symbols denote areas where the results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

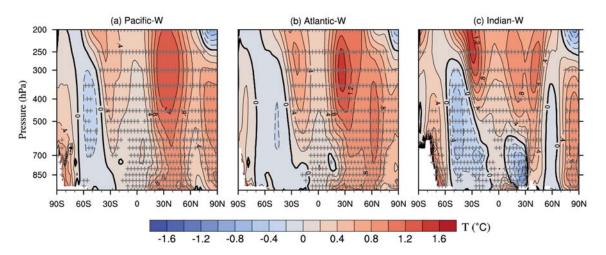


Figure S16. Vertical-meridional distributions of changes in the air temperature (contours, °C) for (a) Pacific_W (zonal averaged from 100°E-90°W), (b) Atlantic_W (zonal averaged from 100°W-180°W) and (c) Indian_W (zonal averaged from 30°E-100°E) relative to the CTRL in boreal summer. Black solid and dashed lines in the contours indicate positive and negative air temperature anomalies, respectively (contour interval: 0.2 °C). The + symbol denotes areas where the changes of air temperature are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

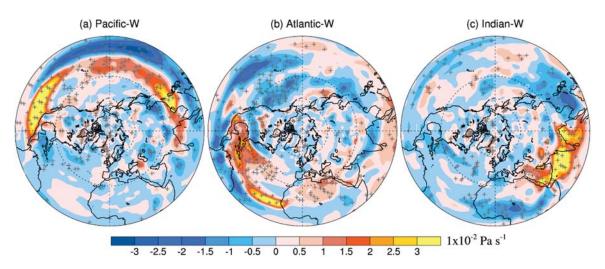


Figure S23. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer.

Positive values indicate upward motion. The + symbols indicate areas where results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

16) Line 341- please explain how a reduction in low pressure weakens the East Asian monsoon?

Good question. As shown in Figure 5, this low pressure induces a cyclonic anomaly in the lower troposphere over the subtropical northwestern Pacific. This weakens southwesterly winds to East China and thus the East Asian summer monsoon. However, East Asian summer monsoon is a rather complicated phenomenon when considering its onset, withdrawal, and relationship with precipitation. These factors are beyond the focus of this study. We therefore removed this discussion in Section 4.3. Now we have (see *P15*, *L433-437*):

"...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)."

17) Line 349/Line 357- if the IPR analysis refers to Figure 2 there seem to be a few inconsistencies – the influence of Pacific W on EA then VDIF appears to have the strongest role, yet advective transport is discussed here? The influence of the Atlantic W on NA then CHEM seems only to have a small contribution in Figure 2 and not be the main contribution discussed here. Furthermore, the logic of the argument that physical transport is not important because of large changes in the upper troposphere but small changes at the surface is unclear.

Thanks for pointing out this. The IPR analysis here does not refer to Figure 2. In Figure 2, we averaged IPR changes over East Asia. In this sentence, the IPR analysis only focus on southern China. The warming of the North Pacific increases the surface O₃ over upwind region in JJA, but this effect is insignificant when averaging over the whole East Asia (shown in Table 1). Correspondingly, the increase of VDIF over EA is also insignificant in JJA based on the IPR analysis. As shown in Figure 1, the increases of surface O₃ are highest over southern China while accompanied with a slight decrease over the North China. We find that the wind pattern anomaly induced by the warming of the North Pacific may be responsible for this dipole changes through modulating surface O₃ transport. The IPR analysis indicates that the increase in advective transport is mainly responsible for the surface O₃ increase in southern China. These results are not shown in the manuscript.

As for the cases of "Atlantic-W", we find that the VDIF and DRYD are two processes that changes significantly over NA (Figure S10). However, they tend to offset each other in most places, resulting in an insignificant overall change in TURB (please see our revised Figure 2). Therefore, the change of CHEM are higher than TURB and dominates the surface O_3 increase over NA in "Atlantic-W" (as discussed in Section 4.1). Please refer to our response to specific comment 6 for more details. Here we further demonstrated the changes in horizontal fluxes of O_3 over NA in "Atlantic-W", which shows no significant effect on the increase of surface O_3 (Figure 7b). Therefore, we conclude that the response of ground-level O_3 over North America to the North Atlantic warming is mainly caused by the enhanced chemical production, rather than physical transport. The discussion of the difference in O_3 changes between upper troposphere and surface is not a supporting argument. It only indicates that O_3 transport maybe more important over upper troposphere.

We have clarified our descriptions in Section 4.3 (P15, L432-442 and L444-450) as follows:

"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."

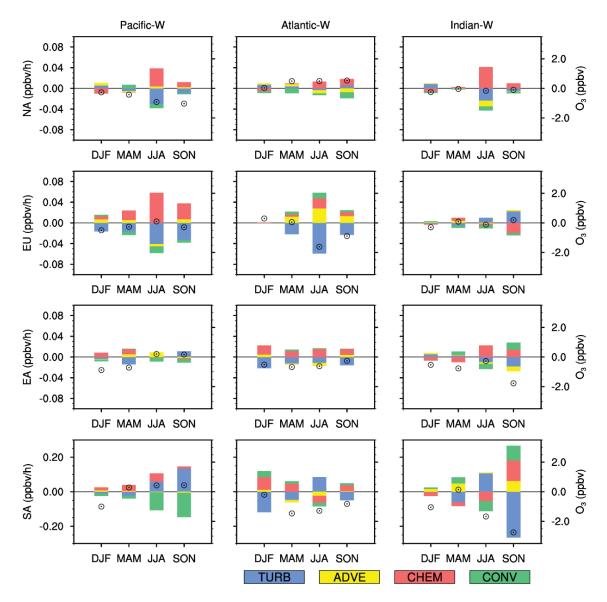


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second 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 S10 in the supplementary material.

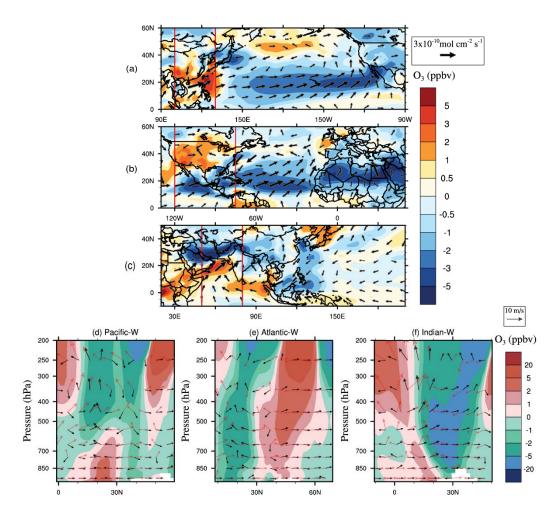


Figure 7. Top three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, 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.

18) Line 380 to end of paragraph- The results in figures 8 (CO tracer) and 7 (ozone concentrations) look similar and reinforce each other except over the Indian Ocean. Please comment on this.

Good suggestion! Given that the CO-like tracers added in our simulation are idealized with fixed decay lifetime, their concentration changes associated with SST anomalies can be completely

attributed to the corresponding changes of air mass. As shown in Figure 8a, changes in surface concentrations of CO-like tracer emitted from EA resembles that of surface O₃ depicted in Figure 7a. This indicates that changes in transport associated with North Pacific warming play an important role in redistributing the surface O₃ over East Asia. As for the CO-like tracer emitted from NA in the "Atlantic-W" case, it shows a slight increase at the surface over the NA source. However, the spatial pattern of its concentration change (Figure 8c) is not consistent with that of O₃ (Figure 7b). This is because the increase of surface O₃ over NA is mainly caused by the enhanced chemical production, which deviates it substantially from the CO-like transport pattern. The response of the South Asia CO tracer to North Indian Ocean warming also shows an inconsistent spatial pattern with that of surface O₃ concentrations, suggesting the importance of chemistry on surface O₃. Therefore, the diagnosis of CO-like tracers not only infers the response of O₃ long-range transport to SST anomalies, but can also help to verify our previous arguments. We have improved this text and added more discussions in the revised manuscript (*P17*, *L477-487*).

"...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."

19) Line 395 to end of paragraph- Is significance plotted in figure 9? The text cannot be followed well here with the current figure quality. The conclusion on vertical diffusion is hard to follow, given text in previous sections discussing areas of both enhanced convection and subsidence in the ocean basin and downwind.

Thanks for pointing this problem out. We have improved the quality of Figure 9. The changes of zonal wind depicted in Figure 9 have been verified to be significant at 0.05 level with the Student t-test using 20 years of data.

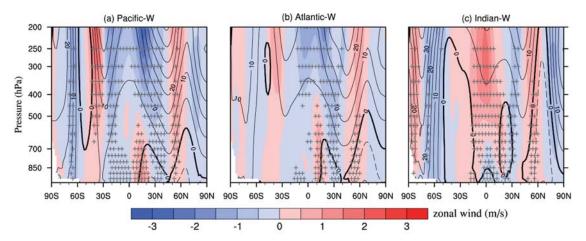


Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential height (contour, m) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. Black solid and dashed lines in the contours indicate positive and negative geopotential height anomalies, 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.

This discussion here mainly focuses on the circulation pattern changes induced by basin-scale SST changes. Our result shows that the response of air temperature to a warmer North Pacific or North Atlantic mainly happens in the mid-latitudes (Figure 9). Thus, the associated meridional temperature gradient is decreasing at lower latitudes while increasing at higher latitudes. Consequently, it weakens the westerly wind at lower mid-latitudes (25°N - 45 °N) while intensifies it at higher latitudes. As for vertical transport (mainly discussed in Section 4.3), a warmer basin-scale SST facilitates the deep convection over tropical regions while suppresses the upward transport at higher latitudes. The decreases of upward velocity at mid-latitudes are also demonstrated in Figure 6 and Figure S6. A detail description is provided in Section 4.3 (see our response to specific comments 13 and 15). Therefore, here we attribute the decreases in CO tracer transport to remote regions (Figure 8a and 8c) to the suppressed vertical transport and weakened westerlies at mid-latitudes. We have clarified this text in our revised manuscript (see *P17*, *L489-502*).

"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 O₃ 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."

20) Line 435- "90% of surface O3"- first mention of this in the text.

Thanks for pointing out this. We have rephrased this in the revised manuscript (P19, L542-545).

"...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."

Minor comments:

1) Line 49- it would be useful to state why ground-level ozone affects food security. Also it would be useful to provide a more up to date reference than 2006 for WHO.

Good suggestion! We have specified the ground-level ozone effects on human health and food security. We have also provided more recent references (see *P2*, *L50-52*):

"High ground-level ozone (O₃) concentrations adversely impact human health by inducing respiratory diseases and threaten food security by lowering crop yields (Brown and Bowman, 2013; Organization, 2013; Chuwah et al., 2015)."

2) Line 54- again can the authors use a more recent reference than Vingarzan et al. 2004.

We have added several more recent references here (P2, L57-58)

"These precursors originate from both natural and anthropogenic sources (Vingarzan, 2004; Simon et al., 2014; Jiang et al., 2016)."

3) Line 66- an enhanced description of what is meant by atmospheric circulations would be useful e.g. Barnes and Fiore (2013) specifically discuss the effect of the Jetstream in the northern midlatitudes at 500 hPa. Other processes to mention are mid-latitude cyclones and the North

Atlantic Oscillation for the N. Atlantic. Some further useful references include: Creilson et al. 2003; Christiadios et al. 2012; Knowland et al. 2014. Line et al. (2012/14) and references therein are useful for circulations relating to atmospheric circulations in the N. Pacific.

Creilson, J. K., Fishman, J., and Wozniak, A. E.: Intercontinental transport of tropo-spheric ozone: a study of its seasonal variability across the North Atlantic utilizing tro-pospheric ozone residuals and its relationship to the North Atlantic Oscillation, Atmos. Chem. Phys., 3, 2053–2066, doi:10.5194/acp-3-2053-2003, 2003

Christoudias, T., Pozzer, A., and Lelieveld, J.: Influence of the North Atlantic Oscillation on air pollution transport, Atmos. Chem. Phys., 12, 869–877, doi:10.5194/acp-12-869-2012, 2012 Knowland, K. E., Doherty, R. M., and Hodges, K. I.: The effects of spring- time mid-latitude storms on trace gas composition determined from the MACC reanal- ysis, Atmos. Chem. Phys., 15, 3605-3628, doi:10.5194/acp-15-3605-2015, 2015.

Good suggestion. These references are valuable and closely related to our work. We have cited them and expanded our introduction with more detailed descriptions (see *P3*, *L72-83*).

"...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)."

4) Line 79- what is meant by "SST is an indicator for both marine and terrestrial mete- orology"?

Here we want to emphasize the important role of SST played in the climate system. The SST anomalies have been widely used to indicate the climate variability and have great implications for climate predictions. We have rephrased this in the revised manuscript (*P4*, *L95-96*):

"Sea surface temperature (SST) is an important indicator that characterizes the state of the climate system."

5) Line 83- perhaps the reference to the text book is unnecessary.

We agree that the reference to the text book is unnecessary here and have removed it.

6) Line 87- some recent references from the IPCC AR5 report will be relevant here.

We have updated our references with more recent studies (P4, L101-104).

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

7) Line 92- it would be more useful to the reader to refer to the specific chapter in IPCC AR5- the science of climate change that discusses SST changes rather than broadly reference the IPCC synthesis report.

Good suggestion! We have changed our reference to the Chapter 2 in IPCC AR5 (P4, L107-108):

"SSTs are generally increasing due to the impacts of anthropogenic forcings on global climate change (IPCC, 2013, Chapter 2)."

8) Line 102- is "according to observations" needed?

We agree that it is an unnecessary description here and has removed it in our revised manuscript (*P4*, *L117-119*).

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

9) Line 105 "Emissions of aerosols.. complicate regional SST variability because of their climate effects"- this sentence is unclear.

Here we state that regional SST can be significantly influenced by the aerosol and GHGs emitted from anthropogenic and natural sources through modulating the solar radiation at an oceanic surface. These processes may contribute to the SST variability, especially in the regional scale. For example, the responses of SST to volcanic eruptions have been identified to vary between regions. We have clarified these sentences in the revised manuscript (*P4-5*, *L121-124*).

"...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)."

10) Line 113- besides Lin et al. 2014, Liu et al. (2005) is also a valuable reference here in relation to ENSO and pollution transport from East Asia. Liu, J., D. L. Mauzerall, and L. W. Horowitz (2005), Analysis of seasonal and interannual variability in transpacific transport, J. Geophys. Res., 110, D04302, doi:10.1029/2004JD005207.

Good suggestion. Liu et al. (2005) evaluated the meteorological component of the seasonal and interannual variability of transpacific transport. It is a valuable reference that relates the transpacific pollution transport to ENSO. Their analysis is mainly based on idealized tracer with constant emissions and chemical lifetimes. This finding has valuable implication for the ENSO effects on O₃ long-rang transport. We have discussed it in our revised manuscript (*P5*, *L133-135*).

"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 O₃."

11) Line 119- it would be useful to first discuss the surface ozone response for the specific ocean basin relative to the experiment and then discuss effects on surrounding continents. The four continental regions used in Fiore et al. (2009) and elsewhere should be defined here, as they are used throughout the text.

Good suggestion. We agree that it is beneficial to provide some discussion about the surface O₃ changes above ocean basins associated with regional SST anomalies. We have added some descriptions and explanations about those effects. The relevant table and figure are placed in the supplementary material. Please refer to our reply to major comment 3 for details. The major focus of this study is on the responses of surface O₃ over polluted continents to regional SST changes. The surface O₃ levels over these regions are much higher than remote oceans, which may negatively impacts human health and threatens food security. This SST-O₃ relationship over the populated continents may help air quality management. We have also clearly defined the four continental regions in the revised manuscript (*P5*, *L140-144*).

"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."

12) Line 157 typo- AEROCOM

Thanks for catching this typo. We have corrected it (*P6*, *L178-180*).

"...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)."

13) Line 161 – "scientifically" is unnecessary.

We have removed this unnecessary description (P7, L184-185).

"... The performance of CESM in simulating tropospheric O_3 has been validated by comparing with ozonesondes and satellite observations (Tilmes et al., 2014)."

14) Line 251- "similar increases in VDIF" compared to?

We have clarified this in the revised manuscript (P11, L323-325).

"...In the "Atlantic-W" case, increases in VDIF are also observed over the upwind regions (i.e., North America) in JJA."

15) Line 273- explain how net production rate in this section related to CHEM in the previous section.

Good question. The CHEM refers to the net cumulated contributions of the chemical production and loss of O₃ during a specific period. The net production rate is calculated by chemical production rate minus loss rate of O₃. Therefore, the CHEM and net-production rate are consistent with each other but indicate the O₃ change at different timescale. We have clearly defined these two variables in our revised manuscript.

In Section 2.3 (P8, L230-233):

"...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."

In Section 4.2 (*P12*, *L350-351*):

"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."

16) Line 305 – rephrase "jointly destructs O3 production".

We have rephrased this (P14, L389-391):

"...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 O₃ production in that region."

17) Lines 320-323, "Given that . . .)." This sentence contains a number of grammar errors. The following sentence starting line 323 seems to state that the pressure difference induced by warmer SSTs would be greater at lower latitude but notes this is not shown here in Figure 5. Please comment further on this or remove.

Thanks for pointing out this problem. We have revised this sentence to correct grammars errors. We also linked the warmer SST directly to the enhanced upward motion instead of the surface pressure changes (as showed in Figure 6). Please see the revised text in Section 4.3 (*P14*, *L408-414*) or below:

"...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."

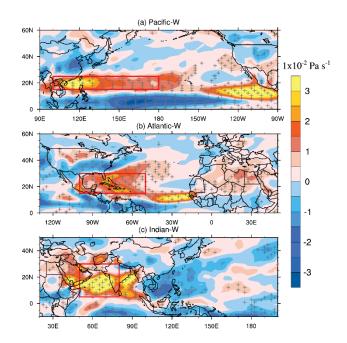


Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. Red polygons denote the regions where the surface pressure responses 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.

18) Line 363- Mediterranean?

As shown in Figure 5, the warming of the North Indian SST leads to a low-pressure anomaly that spreads to the Saudi Arabia and eastern Mediterranean. However, its effects on the Europe has proved to be insignificant (as shown in Table 1). To avoid confusion, we revised this sentence in Section 4.3 (*P16*, *L452-453*) and confined our analysis to the Indian Ocean and the Indian Subcontinent.

"The North Indian SST warming leads to a low-pressure anomalies centered over the Arabian Sea (Figure 5c)."

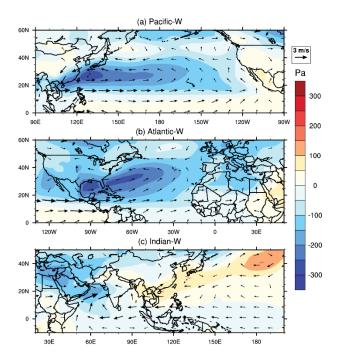


Figure 5. Changes in the surface pressure (color contours, Pa) and 850-hPa wind (arrows, m s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer.

19) Line 367-"Downward diffusion from the upper troposphere"- please clarify what is meant here as this is not a region of STE.

According to the IPR analysis, we find that the surface O_3 increase over the Indian Ocean associated with North Indian warning is mainly attributed to the enhanced vertical transport of O_3 to the surface through deep convection and vertical diffusion processes (Figure S11). It is not

related to the STE. We have clarified this in Section 4.3 of our revised manuscript (*P16*, *L456-459*).

"...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)."

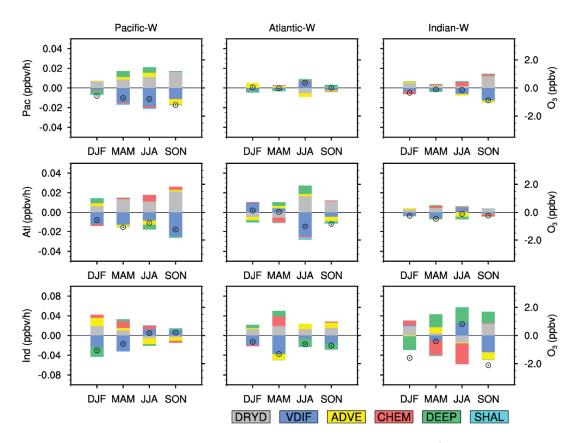


Figure S11. Seasonally averaged changes in IPR contributions (bars, ppbv h⁻¹, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over North Pacific (demoted as Pac, first row), North Atlantic (demoted as Atl, second row) and North Indian Ocean (demoted as Ind, third row) defined in our study, respectively. IPR contributions from six processes (i.e., CHEM, ADVE, VDIF, DRYD, SHAL and DEEP) are represented by different colors.

20) Line 374- why only at mid-latitudes? Figure S3 shows large temperature increases in temperature above all 3 basins.

Good question. As we have discussed in Section 4.3, the SST warming over a specific ocean basin tends to enhance the deep convection over tropical oceans. Strengthened deep convection further trigger large-scale subsidence over other areas through modulating large-scale circulations, which may suppress air convective movement there (Lau et al., 1997;Roxy et al., 2015;Ueda et al., 2015). As depicted in Figure S23, decreases of upward vertical velocity are significant over midlatitudes and other oceans. As shown in Figure S16 (i.e., the Figure S3 in old version), the warming of air temperature are more significant over free troposphere at mid-latitudes, which leads to a remarkable decrease in the vertical air temperature gradient. This weakens vertical movement of air pollution.

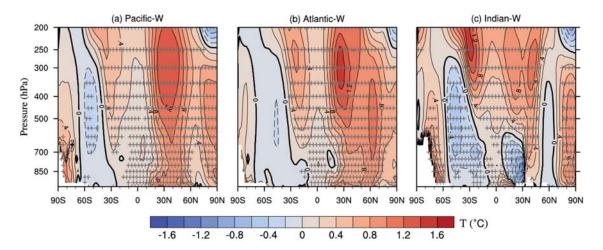


Figure S16. Vertical-meridional distributions of changes in the air temperature (contours, °C) for (a) Pacific_W (zonal averaged from 100°E-90°W), (b) Atlantic_W (zonal averaged from 100°W-180°W) and (c) Indian_W (zonal averaged from 30°E-100°E) relative to the CTRL in boreal summer. Black solid and dashed lines in the contours indicate positive and negative air temperature anomalies, respectively (contour interval: 0.2 °C). The + symbol denotes areas where the changes of air temperature are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

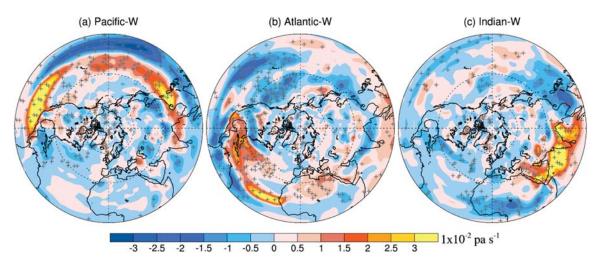


Figure S23. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal summer. Positive values indicate upward motion. The + symbols indicate areas where results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.

21) Line 439/line 440 – rephrase "increasing influence on surface O3 concentrations" as this is confusing e.g. regional surface ozone over SA decreases under the Indian W simulation.

We have rephrased this sentence in Section 6 (*P19*, *L545-547*).

- "...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."
- 22) Line 465 natural variability is not discussed in this paper (although used for significance testing so it is odd to mention here).

The natural variability mentioned here is referred to the variability existed in the SST. As we have discussed in the Introduction section, regional SST exhibits natural periodic or irregular oscillations with timescales ranging from months to decades. In this study, we used idealized SST anomalies to generally compare the role of SST over different oceans in modulating the surface O₃ distributions. Our results highlight the sensitivity of the surface O₃ distribution to basin-wide SST changes. To provide a more realistically understanding of this SST-O₃ relationship, further studies are necessary and realistic SST variability should be taken into account. We have rephrased the relevant sentences in our summary section for clarification (*P20*, *L569-574*).

"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 this

evolution. 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."

23) Figure 6 refers to Figure 7 re surface pressure- should the reference be to Figure 5?

Thanks for pointing out this mistake. We have corrected the caption of Figure 6.

"Figure 6. Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² 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."

24) Figure 7 – swap panels b) and c) to be consistent with text.

We have corrected this in the Figure 7.

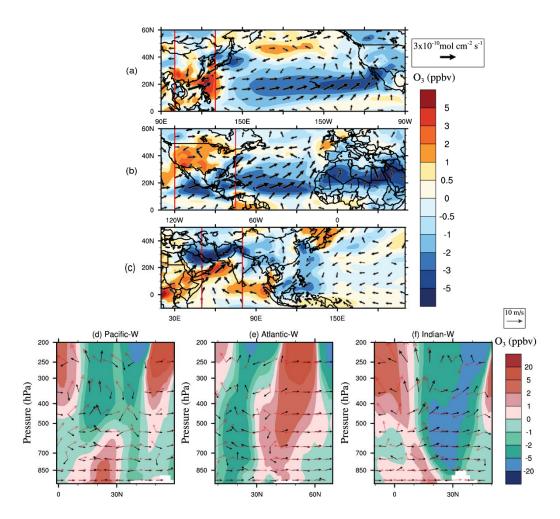


Figure 7. Top three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, 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.

25) Figures 8, 9, S3: the season is omitted from the figure caption.

Thanks for pointing out these errors. These figures are all referring to boreal summers. We have revised the captions of these figures and clarified the relevant season:

"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."

"Figure S16. Vertical-meridional distributions of changes in the air temperature (contours, °C) for (a) Pacific_W (zonal averaged from 100°E-90°W), (b) Atlantic_W (zonal averaged from 100°W-180°W) and (c) Indian_W (zonal averaged from 30°E-100°E) relative to the CTRL in boreal summer. Black solid and dashed lines in the contours indicate positive and negative air temperature anomalies, respectively (contour interval: 0.2 °C). The + symbol denotes areas where the changes of air temperature are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data."

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Response to Dr. Meiyun Lin

(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.)

1. The manuscript is missing an importance reference on the connection among ENSO, intercontinental pollution transport, and ozone variability.

Meiyun Lin, L.W. Horowitz, S. J. Oltmans, A. M. Fiore, Songmiao Fan (2014): Tropospheric ozone trends at Manna Loa Observatory tied to decadal climate variability, Nature Geoscience, 7, 136-143, doi:10.1038/NGEO2066.

This paper used observations at Mauna Loa Observatory in Hawaii, multi-decadal model hindcasts (including those driven by observed SSTs) and idealized CO-like tracers to show that the eastward extension and equatorward shift of the subtropical jet stream during El Nino enhances long-range transport of Asian pollution towards the eastern North Pacific, raising free tropospheric ozone over the subtropical North Pacific region. La Nina manifests in an opposite way. They also found that long-range transport of Asian pollution has weakened in the 2000s due to more frequent La Nina-like conditions (the negative phase of the Pacific Decadal Oscillation).

It seems like that the ozone response to Pacific cooling shown in your Figure 1 resembles the response to El Nino discussed in the above paper. Their findings should be summarized in the Introduction and discussed in Section 5.

Thanks Meiyun for your thoughtful and valuable comments. Your paper presented an excellent work regarding the interactions among ENSO, SST anomaly, pollution transport and ozone variability, and fits well the scope of this study. We have summarized your key conclusions in the Introduction (see *P5*, *L131-133* in our revised manuscript or below). In our study, we also find that SST increase in North Pacific tends to weaken the zonal westerly wind at lower-middle latitudes (25°N - 45 °N) in the Northern Hemisphere while strengthen it at higher latitudes (Figure 9, see the revised manuscript or below), which further influence the O₃ long-range transport. The corresponding cooling in North Pacific, on the other hand, may exert an opposite effect. Since our analysis mainly focus on the surface O₃ changes in boreal summers while your findings are more relevant to O₃ changes in spring and autumn, we will do more analysis about these seasons and compare directly with your work in the follow-up studies.

"...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)."

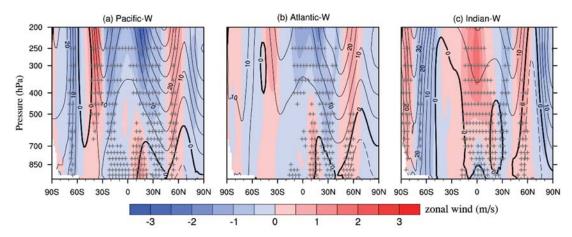


Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential height (contour, m) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the CTRL in the boreal summer. Black solid and dashed lines in the contours indicate positive and negative geopotential height anomalies, 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.

2. Lines 113-115 (Page 4), the description for the findings of Lin et al. (2015, Nature Communications) is not quite accurate. You stated "Lin et al. (2015) had found that more frequent deep stratospheric intrusions appear during ENSO springs". By "ENSO springs", it is not clear which phase of the ENSO. Please change that to "during strong La Nina springs".

Thanks for mentioning this. We have clarified the relevant text (see *P5*, *L128-131*) based on your suggestions:

"...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."

References:

Lin, M., Horowitz, L. W., Oltmans, S. J., Fiore, A. M., and Fan, S.: Tropospheric ozone trends at Mauna Loa Observatory tied to decadal climate variability, Nat Geosci, 7, 136-143, 2014.

Lin, M., Fiore, A. M., Horowitz, L. W., Langford, A. O., Oltmans, S. J., Tarasick, D., and Rieder, H. E.: Climate variability modulates western US ozone air quality in spring via deep stratospheric intrusions, Nat Commun, 6, 2015.

Response of the Global Surface Ozone Distribution to 1 Northern Hemisphereic Sea Surface Temperature Changes: 2 Implications for Long-Range Transport 3 4 Kan Yi¹, Junfeng Liu¹, George Ban-Weiss², Jiachen Zhang², 5 Wei Tao1, Shu Tao1 6 7 [1] Laboratory for Earth Surface Processes, College of Urban and Environmental 8 Sciences, Peking University, Beijing, China 9 10 [2] Sonny Astani Department of Civil and Environmental Engineering, University of 11 Southern California, U.S.A. 12 13 Correspondence to: Junfeng Liu (E-mail: <u>ifliu@pku.edu.cn</u>) 14 15 16 17 Abstract 18 19 The response of surface ozone (O₃) concentrations to basin-scale warming and cooling 20 of Northern HemispherieHemisphere oceans is investigated using the Community 21 Earth System Model (CESM). Idealized, spatially uniform sea surface temperature (SST) anomalies of +/- 1°_ °C are individually superimposed onto the North Pacific, 22 North Atlantic, and North Indian oceans, individually. Oceans. Our simulations suggest 23 <u>large</u> seasonal and regional variability of surface O₃ in response to SST anomalies, 24 25 especially in the boreal summer. The responses of surface O₃ associated with basinscale SST warming and cooling have similar magnitude but are opposite in sign. 26 Increasing (decreasing)the SST by 1 °C°C in one of the regions of focus induces oceans 27 generally decreases (increases) inthe surface O₃ concentrations, ranging from 1 to 5 28 29 ppbv. With fixed emissions, SST increases of a specific ocean basin in the Northern Hemisphere tend to increase the summertime surface O₃ concentrations over upwind 30 continents regions, accompanied with by a widespread reduction over downwind 31

regionscontinents. We implement the integrated process rate (IPR) analysis (IPR) in

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CESM and find that meteorological O₃ transport in response to SST changes is the key process causing surface O₃ perturbations in most cases. During the boreal summer, basin-scale SST warming facilitates the vertical transport of O₃ to the surface over upwind regions while significantly reducing the vertical transport over downwind continents that are downwind. This process, as confirmed by tagged CO-like tracers, implicates indicates a considerable suppression of O₃-intercontinental O₃ transport due to increased stagnationtropospheric stability at lower mid-latitudes induced by SST increases. Changes in O₃-changes. On the other hand, the responses of chemical O₃ production associated with to regional SST increases, on the other hand, warming can increase exert positive effects on surface O₃ levels over highly polluted continents, except for South Asia. In 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 O₃ production of O₃. Our findings indicate a robust linkage between basin-scale SST variability and continental surface O₃ pollution, which should be taken into account for considered in regional air quality management.

Keywords: SST anomaly, Surface O₃, Process analysis, Transport, CESM

1. Introduction

Ground level ozone (O₃) adversely impacts human health (WHO., 2006) and threatens food security (Chuwah et al., 2015). Considering its eco-toxicity, it is of great importance to understand the physical and chemical mechanisms that control atmospheric ozone concentrations. High ground-level ozone (O₃) concentrations adversely impact human health by inducing respiratory diseases and threaten food security by lowering crop yields (Brown and Bowman, 2013; Organization, 2013; Chuwah et al., 2015). Considering the eco-toxicity of O₃, understanding the physical and chemical mechanisms that control atmospheric O₃ concentrations is of great importance. Surface O₃ is produced in the atmosphere via photochemical

processing of multiple precursors including volatile organic compounds (VOCs), carbon monoxide (CO) and nitrogenoxides (NO, NO₂). These precursors originate from both natural and anthropogenic sources (Vingarzan, 2004).(Vingarzan, 2004;Simon et al., 2014;Jiang et al., 2016). In addition to local production, transport of O₃ and its precursors from upwind regions and the upper atmosphere can also influence surface O₃ abundance. Stratospheric intrusion events, which lead to vertical down-mixing of ozone-rich air, can significantly elevate surface O₃ during spring and summer (Lin et al., 2012;Zhang et al., 2014;Grewe, 2006)(Grewe, 2006;Lin et al., 2012b;Zhang et al., 2014). LongThe long-range transport of O₃ and its precursors havehas been extensively studied, and their inter-continental impacts have been evaluated withusing measurements and model simulations (Fiore et al., 2009;Brown-Steiner and Hess, 2011)(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 O₃ levels. Important meteorological factors that can impact both photochemistry and transport include atmospheric circulations, solar radiation, and relative humidity. Atmospheric circulations determine the timescale and pathway of O₃ transport (Auvray and Bey, 2005;Barnes and Fiore, 2013). air temperature, and relative humidity. Atmospheric circulation considerably determines the timescale and pathway of O₃ transport (Bronnimann et al., 2000;Auvray and Bey, 2005;Hess and Mahowald, 2009). The efficiency of O₃ transport varies coherently with atmospheric circulations on different scales. Knowland et al. (2015) demonstrated the important role of mid-latitude storms in redistributing O₃ concentrations during springtime. The North Atlantic Oscillation (NAO) significantly affects surface and tropospheric O₃ 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 O₃ burdens over North America. The shift in the position of the jet stream associated with

eastern North America (Barnes and Fiore, 2013). Increases in solar radiation and air temperature can increase the rate of the chemical production of O₃ and modulate the biogenic emissions of O₃ 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 O₃ 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 O₃ distribution. Understanding the mechanisms and feedbacks of the interactions between O₃ and climate has received increasing attention and will be essential for future surface O₃ mitigation (Jacob and Winner, 2009;Doherty et al., 2013).

Sea surface temperature (SST) is an indicator for both marine and terrestrial meteorology. 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). Numerous studies have shown that SST changes over different oceans and at different latitudes lead to significantly different meteorological sensitivities 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). SSTs are generally increasing due to the impacts of anthropogenic global climate change (Pachauri et al., 2014). 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 various modes of low-frequency SST variability according to observations (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). Emissions of aerosols and greenhouse gases (GHGs) from anthropogenic and natural sources further complicate regional SST variability because of their climate effects (Wu and Kinter, 2011; Hsieh et al., 2013; Rotstayn and Lohmann, 2002). 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).

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Considering the distinct roles of regional SST variability in modulating regional climate system, there is a need to explore the impact of regional SST change on surface O₃ distribution. To date very few studies have been conducted to address the linkage between SST-O₃ interactions except for the ENSO impacts. For example, Lin et al. (2015) had found that more frequent deep stratospheric intrusions appear during ENSO springs, which increase western US surface O₃ levels remarkably. Considering the distinct roles of regional SST variability in modulating regional climate systems, the impact of regional SST changes on the surface O₃ 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 O₃ 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 O₃-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 O₃. Except for the ENSO impacts, very few studies to date have directly addressed the linkage between SST and O₃. Therefore, a comprehensive understanding of the response of surface O₃ to SST changes in individual ocean basins is lacking and necessary.

To fill this gap, this study focuses on examining the sensitivity of O₃ 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)), and its response with respect to nearby basin-scale SST changes. We describe the design of numerical experiments and model configuration in Section 2. Surface O₃ responses to regional SST changes are given in Section 3. Relevant mechanisms governing the SST-O₃ relationships are discussed in Section 4. The impact of basin-scale SST changes on inter-continental transport of O₃ 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).

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The chemistry coupled in the CAM5 (i.e., CAM5-chem) is primarily based on the Model for O₃ 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 O₃ has been scientifically 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 O₃ over the Eastern US and Western Europe in the summer (Tilmes et al., 2014).

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2.2 Numerical Experiments

We first conduct a control simulation, hereafter referred to as CTRL, with prescribed 215 climatological SSTs averaged from 1981 to 2010 (see Hurrell et al. (2008)). We then 216 217 conduct six perturbation simulations with monthly SSTs that are uniformly increased 218 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 Ocean (15°N-65°N; 100°W-219 20°E) and the North Indian Oceans (5°N-30°N; 30°E-100°E); here 5°N is used to 220 attain a relatively larger domain size). The simulations are denoted as "Pacific-W", 221 "Atlantic-W", and "Indian-W" for the three warming cases and "Pacific-C", "Atlantic-222 C", and "Indian-C" for the three cooling cases. We defined the latitudinal and 223 longitudinal ranges of these ocean basins mainly based on their geographical features. 224 The boundaries of the prescribed SST anomalies generally align with the edge of the 225 226 ocean basins, except along the southern side. In each perturbation simulation, southern boundaries of these oceanic regions arewe further linearly smoothed to prevent large 227 SST smooth the southern boundaries of these SST anomalies towards the equator to 228 remove the sharp SST anomaly gradients- at the edge, following a previous approach 229 230 (e.g., Taschetto et al., 2016; Seager and Henderson, 2016). Air pollution emissions, including biogenic emissions of VOCs, are held-fixed to distinguish the impacts of SST 231 variation on O₃ transport and photochemistry. All simulations are run for 1221 years 232 with the first year used for model spin-up. 233

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To explore the impacts of SST changes on inter-continental transport, an explicit emission tagging technique is used in our simulations following previous studies (Doherty et al., 2013;Shindell et al., 2008).(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 O₃ (Doherty et al., 2013)₅ and used to single out changes in O₃ transport induced by SST anomalies.

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2.3 Integrated process rate (IPR) analysis

To provide a process-level explanation onfor the response of surface O₃ to regional SST changes, an integrated process rate (the IPR) method is applied. This technique method calculates the accumulated contributions of individual processes to model (e.g., chemical production and loss, advection, vertical diffusion, dry deposition, etc.) to O₃ predictions during runtime, which the model simulation and has been widely used for air pollution diagnostics (Tao et al., 2015; Li et al., 2012; Zhang and Wu, 2013). (Li et al., 2012; Zhang and Wu, 2013; Tao et al., 2015). In this study, we add added the IPR scheme to the CESM modeling framework to track hourly contributions from 6the contribution of six physicochemical processes, including (i.e., gas-phase chemistry (CHEM), advection (ADVE), vertical diffusion (VDIF), dry deposition (DRYD), shallow convection (SHAL) and deep convection (DEEP). The wet)) to O₃ concentrations in every grid box. Wet deposition and aqueous-phase chemistry are not considered ignored here due to the negligible low solubility and negligible chemical production rate of O₃ in water (Jacob, 1999). The performance of Therefore, CHEM represents the net production (production minus loss) rate of O₃ due to gas-phase photochemistry. DRYD represents the dry deposition fluxes of O₃, which is an important sink for O₃. The other IPR is verified throughterms (i.e., ADVE, VDIF, SHAL and DEEP) represent contributions from different transport processes. The IPR scheme tracks and archives the O₃ flux in each grid from every process during each model time step. The sum of the O₃ fluxes from these six processes matches the change in the O₃ concentration. The <u>IPR performance is verified by</u> comparing the predicted hourly O₃ changes with the sum of the individual burdensfluxes from the 6six processes during runtime. As shown in Figure S1, the hourly surface O₃ abundance ischanges are well represented by the sum of these processes fluxes in the model.

271 3. The response Response of surface O₃ concentrations to SST changes The responses of surface O₃-concentrations to basin-scale SST changes (i.e., ±1°C) 272 are investigated over four highly populated continental regions (Table 1). Seasonally 273 274 and regionally averaged surface O₃ changes (i.e., DJF (December, January, February), 275 MAM (March, April, May), JJA (June, July, August) and SON (September, October, November)) for each SST perturbation simulation are mostly within 3 ppbv relative to 276 CTRL. Larger anomalies (i.e., up to 5ppbv) are simulated in locations including the east 277 coast of China, the Indian subcontinent, and remote oceans (Figure 1 and Figure S2). 278 279 This magnitude of O₃ change is comparable to intercontinental changes in ozone in response to 20% reductions in anthropogenic emissions within a continental region 280 (Fiore et al., 2009). 281 282 283 Seasonally (i.e., DJF (December, January, February), MAM (March, April, May), JJA (June, July, August) and SON (September, October, November)) and regionally 284 averaged surface O₃ changes in each SST perturbation simulation for the four highly 285 populated continental regions and three ocean basins defined in our study are given in 286 Tables 1 and S1, respectively. The responses of the surface O₃ concentrations to basin-287 scale SST changes (i.e., ± 1 °C) are mainly below 3 ppbv in the Northern Hemisphere 288 (Tables 1 and S1), though larger anomalies (i.e., up to 5 ppbv) are also observed over 289 the eastern coast of China, the Indian subcontinent, and certain oceanic areas (Figures 290 1 and S3). This SST-O3 sensitivity is comparable to previous findings. For instance, 291 Bloomer et al. (2009) reported a positive O₃-temperature relationship of 2.2~3.2 292 ppbv/°C across the rural eastern United States. Wu et al. (2008) found that summertime 293 surface O₃ may increase by 2-5 ppbv over the northeastern United States in the 2050s. 294 Additionally, Fiore et al. (2009) demonstrated an intercontinental decrease in surface 295 296 O₃ 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 297 exert significant effects on the surface O₃ above specific ocean basin and its 298 surrounding continents. 299

As shown in Figure 1, up to 5 ppbv seasonal changes of up to 5 ppbv in the mean surface O₃ concentration changes are found observed during boreal summers, mainly in coastal regions and remote oceans. Over Southern China, increases in Northern Pacific SSTs lead to increases in surface O₃ of nearly 3 ppbv, accompanied by decreases in North America (~1 ppbv, shown in Table 1). In the "Atlantic-W" case, ground-level O₃ increases by nearly 1 ppbv over North America, but decreases by 1~2 ppbv over Europe. Positive (negative)Surface O₃ changes in response to positive and negative SST anomalies generally pronounce a consistent spatial pattern but are opposite in the Northern Indian Ocean lead to increases (decreases) insign, suggesting robust relationships between surface O₃ over the Indian Oceanlevels and Africa, but decreases (increases) over South and East Asia (TableSST anomalies (Figure 1). Generally, we find that anAn increase (decrease) in summertime SST over a specific ocean basin tends to increase (decrease)the surface O₃ concentrations inconcentration over the upwind regions but reduce (rise) that this concentration over downwind or remote continents. During boreal winters, For instance, a 1 °C warming over the North Pacific leads to a widespread decrease (increase) of surface O₃ is observed in surface O₃ over the North Pacific, North America and the North Atlantic of approximately 1 ppbv (Table S1) but may enhance the surface O₃ by nearly 3 ppbv over South China. Similarly, in the "Atlantic-W" case, the surface O₃ levels decrease by 1~2 ppbv over the North Atlantic and Europe but increase (~1 ppbv) over North America and the North Pacific. For the North Indian Ocean, positive SST anomalies tend to increase the surface O₃ over the Indian Ocean and Africa but decrease the surface O₃ over South and East Asia (Figure 1). During the boreal winter, a widespread decrease in surface O₃ associated with the warming (cooling) of different oceans 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 O₃ to basin-scale SST changes is typically shown in Figure S2S3 insignificant. **Details** are in the supporting information supplementary material.

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Our simulations reveal that ehanges in SSTsdifferent oceans can impacteixert distinct region-specific complex changes in effects on the O₃ distribution. 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 O₃ 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 O₃ to a hemispheric SST anomaly generally resemble the sum of responses to different regional SST changes (see Figures S5 and S7 in the supplementary material). We now focus on analyze the processes that impact the dependency dependence of SST on ozone distributions the O₃ distribution using the simulations that increase the SST.

4. Mechanism forof SST-induced surface O₃ changes.

4.1 Process-level response to SST changes

Figure 2 shows IPR analysis is used to evaluate the contribution of different physicochemical processes to O₃ 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). 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 O₃, which may further induce O₃ removal to balance this forcing in a new equilibrium. Therefore, here, the IPR analysis is used not to budget the SST—induced O₃ concentration changes but rather to help examine the relative importance of different transport and chemical processes in driving the sensitivity of O₃ to SST forcing. In this study, the SST-induced, process-level O₃ changes are spatially averaged over the four populated continental regions of interest (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). In most cases, vertical diffusion (VDIF) and dry deposition (DRYD) are the

key processes controlling the O₃ variation. Since both processes are closely dependent on the atmospheric The downward transport of O₃ through diffusion is an important source of surface O₃, while DRYD acts as a sink. Both processes are simultaneously <u>determined by the strength of turbulence intensity. Here,</u> we define here a new term TURB as the sum of **DRYFDRYD** and VDIF, which can represent capture the overall intensity changes effect of turbulence on surface O_3 variation. Meanwhile concentrations. In addition, we also combine the shallow convection (merge SHAL) and deep convection (DEEP) as CONV to represent the total contribution of convective eontribution. transport to surface O3 (Figures 2 and S9). More detailed IPR results are shown in Figures S10 and S11 in the supplementary material.

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In the "Pacific-W" case, a SST anomaly of +1 °C C SST warming over the North Pacific increases VDIF over eastern China in JJA (Figure S12), which is insignificant if averaged over the whole East Asia while significantly reducing itregion. Meanwhile, this Pacific warming considerably reduces VDIF over North America- (Figure S10). The corresponding decrease of TURB inover North America accounts for nearly 80 % of mainly determines the surface O₃ reduction during in JJA and SON, while reductions of the reduction in CONV are responsible for the remainder exerts an additional negative impact (Figure 2). In the "Atlantic-W" run, similarcase, increases in VDIF are simulatedalso observed over the upwind regions (i.e., North America) in JJA. However, it is these increases are accompanied by commensurate decreases in DRYD, resulting in an insignificant overall change in TURB. The (Figure 2). Therefore, the increase of in CHEM therefore tends to dominate is mainly responsible for the surface O₃ increase over North America. Relatively, in JJA. TURB is more relatively important over Europe (only in JJA and SON-only), leading to reduced surface O₃ abundances. For abundance. <u>In the</u> "Indian-W" <u>case</u>, both CHEM and <u>DEEPCONV</u> are reduced over South Asia in JJA and SON. This reduction, though partially balanced by increases in ADVE and SHAL, leads, leading to overall reductions in surface O₃ 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 O₃ above these regions in JJA (Figure S11). The North Indian Ocean warming, on the other hand, enhances DEEP and VDIF, leading to a local increase in surface O₃ in JJA.

The IPR analysis indicates that, in general, increases in SSTsan SST increase in the North Pacific or North Atlantic areis more likely to elevateenhance the vertical diffusion of O₃ over upwind regions (i.e., East Asia andor North America, respectively) but suppress it over remote this diffusion over the ocean basin as well as downwind continents, especially in boreal summer JJA (Figure S12). These opposite changes in VDIF over upwind and downwind regions lead to inconsistent distinct surface O₃ responses. Changes in photochemistry usually CHEM enhance surface O₃ formation except for in most cases. An exception is in South Asia, where advection CHEM and convection DEEP dominate the feedbacks of reduction in surface O₃ over the region in JJA associated with the North Indian Ocean warming. In the following subsections, the mechanisms responsible of the SST-O₃ relationship for the effects of SST changes in different oceans on modulating relevant chemical and physical processes four polluted continents are further explored. We will Here we focus on boreal summers since both the surface O₃ levels and their response to SST changes are highest is more robust during this period than other seasons.

4.2 Response of O₃-photochemical O₃ production to SST

increaseincreases

Figure 3 shows changes in the net-production rate (i.e., chemical production rate minus loss rate) of O₃ at the surface in JJA associated with basin-scale SST increases. Peak are shown in Figure 3. The peak changes are mainly confined to the polluted regions owing to their high precursor emissions where O₃ precursors are abundant (e.g., South and East Asia and North America). For example, an increase in a warmer North Pacific SSTsSST exerts a positive (negative) impact on net O₃ production in the northern (southern) regions of East Asia. Similarly, the warming of the North

Atlantic promotes a dipole impact on the surface O₃ production over North America, while the warming of <u>the</u> North Indian Ocean significantly decreases the <u>net</u> O₃ net-production rate over South Asia.

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As emissions are fixed in all simulations, the change in net O₃ production is driven by SST induced meteorological changes (e.g., air temperature, air humidity, and solar radiation). Figure 4 illustrates that anAn 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 2m2 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 $1-2^{\circ}$ °C. This temperature decrease is not only limited to the surface, but also spreads to 600hPa600 hPa (Figure S3S16). Associated with this temperature decrease, there is a remarkable reduction of the solar radiation received at the continent beneathbelow (more than 15 W/m², Figure S4).S17). 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 is believed tends to facilitates trengthen the moist convection and that eventually facilitates cloud formation in the upper troposphere, blocking (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 blocks the solar radiation reaching the earth-surface of the Indian subcontinent and reduce 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 S16), which may further suppress the development of deep convection over the Indian subcontinent. This is consistent with previous findings that moist convection is more sensitive to the SST changes in the tropical oceans rather than mid- or highlatitude oceans (Lau et al., 1997; Lau and Nath, 1994).

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A positive relationship_ between air temperature and O₃ chemical production has been well-documented previously (Jacob and Winner, 2009; Rasmussen et al., 2012a), and thus the SST induced warming or cooling of air temperature is thought to be largely responsible for the change in net O₃ production rate (Figure 3). Previous studies have indicated that air temperature positively affects both O₃ production and destruction rates (Zeng et al., 2008; Pusede et al., 2015). As shown in Figure S19, changes in the net O₃ production rate are mainly dominated by O₃ production over continents but by O₃ destruction over oceans. An increase in SST leads to a widespread enhancement of the air temperature, resulting in a positive change in the net O₃ production over most continental regions (Figure 3). On the other hand, the SST increases also enhance evaporation of ocean water and increase humidity above the respective ocean and its eoastal areas (Figure S5), However, a warmer SST also increases the air humidity (Figure S21), which enhances O₃ 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 O₃ production in that region.

4.3 Response of O₃-physical O₃ transport to SST increase increases

In Section 4.1, our IPR analysis highlights multiple important physical processes (i.e., vertical diffusion, convection and advection) on that are important in modulating surface O_3 concentrations. However, the role and relative importance of each process exhibits exhibit large spatial heterogeneity. In this section, we explore the key factors controlling O_3 -physical O_3 transport in response to basin-scale SST changes.

Figure 5 shows the surface pressure and wind pattern changes induced by a basin-wide SST increase. 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. Given that an SST threshold (about 26°-28°C) is required for deep convection to generate, tropical oceans where meet this threshold are pronounced large-scale deep convections and more sensitivity to SST anomalies (Johnson and Xie, 2010; Graham and Barnett, 1987). Previous studies have identified an SST threshold (approximately 26°-28°C) to generating deep convection (Graham and Barnett, 1987; Johnson and Xie, 2010). Therefore, this low-pressure anomaly mainly occurs at low-latitudes whereas the SST increase imposed at higher latitudes would have relatively less effect on surface pressure changes (not shown here). 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 adjacent regionsnearby regions through the modulation of large-scale circulation patterns, which may suppress convective air movement over nearby continents (Lau et al., 1997)-transport (Lau et al., 1997;Roxy et al., 2015;Ueda et al., 2015). This effect is confirmed verified by the widespread decreases of in upward vertical 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). Meanwhile, we also find that the air temperature increase associated with in response to regional SST warming is more significant inabove the upper versus lower troposphere, which leads to a decrease in the vertical air—temperature gradient (shown in—Figure S3S16). These factors tend to enhance atmospheric stability and lead to a more stagnant climate at mid-latitudes that

restrains restrain the vertical exchange of air pollutants. The corresponding decrease in air ventilation contributes to the at mid-latitudes, which facilitates surface O₃ accumulation over polluted continental regions in JJA₇ but may weaken the intrusion of O₃ from the upper troposphere to the surface in most clean regions. We believe this effect unpolluted areas. This process helps to be responsible for explain the wide—spread widespread decrease of in surface O₃ over unpolluted regions associated in clean regions with aan SST increase, as described in Section 3.—, and can be further verified by the wide-spread reduction in VDIF shown in Figure S12.

The surface pressure anomalies induced by SST changes can play a dominant role in modulating surface O₃ transport at specific locations. For example, the low-pressure anomaly centered over the east coast of Asiasubtropical northwestern Pacific in the "Pacific-W" case tends to weakencauses the East Asian summer monsoonconvergence of wind in the lower troposphere (Figure 5a). Consequently, surface O₃ pollution is enhanced in Southernsouthern China due to an increase ofin O₃ transport from the-more polluted Northernnorthern China (Figure 7a). Figure 7a also shows the The vertical distribution of the corresponding O₃ changes, zonally averaged over 100°E-130°E. It also shows that the increase ofin O₃ over Southernsouthern China is limited tooccurs below 700hPa, accompanied by a noticeable decrease in the North and decreases above-700hPa as well as over nearby northern China (Figure 7d). The IPR analysis also indicates that the increase in the South China, with the depressed and downward turbulent/convective transport being are mainly responsible for the remainder. surface O₃ increase in southern China.

In the "Atlantic-W" runcase, the SST warming—induced surface pressure anomalies lead to substantial O₃ redistribution, especially over the North Atlantic Ocean (Figure 7b). Ozone overFor North American is simulated to have large America, the changes in horizontal O₃ fluxes have no significant effect on the O₃ concentration increase. In addition, O₃ changes are observed to be larger in the upper troposphere and negligible

changes than at the surface. Therefore, as (Figure 7e). As demonstrated in Section 4.1, the response of ground-levellower-altitude O₃ over North America to the North Atlantic warming is mainly caused by enhanced photochemicalchemical production, rather than physical transport.—

As shown in Figure 5c, the The North Indian SST warming leads to two surface a low-pressure anomalies, with one anomaly centered over the Arabian Sea and the other centered over the Mediterranean (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. Effects The effects of this process on O₃ concentrations are observed to be more significant in the upper troposphere (Figure 7e7f). According to the IPR analysis, the surface O₃ increase over the Indian Ocean is mainly caused by the downwardenhanced vertical transport of O₃ to the surface through deep convection and vertical diffusion of O₃ from upper troposphere processes (Figure S11). However, over the nearby Indian subcontinent, the suppressed deep convection accounts for nearly 20% oftends to decrease surface O₃ reduction there in that region (Figure 2).

5. Implication for O₃ long-range transport

The above findings indicate that, in general, a basin-scale SST increase (decrease)-in the Northern Hemisphere is more likely to enhance (reduced) atmospheric stability at mid-latitudes, which may suppress (promote) air pollutants from lofting to the free troposphere. This process potentially has large effects on O₃ intercontinental transport. We followFollowing previous work (e.g., (Doherty et al., 2013) and Doherty et al., 2013; (Fang et al., 2011)) and Fang et al., 2011), we use passive CO-like tracers to demonstrate the potential effect of regional SST changes on long-range O₃ transport. The surface changes of CO tracers originating from East Asia in the "Pacific W" run, North America in the "Atlantic W" run, and South Asia in the "Indian W" run are displayed in Figure 8. A warming of North Pacific SSTs by 1°C tends to increase the East Asian

CO tracer concentrations by nearly 86% at the surface. This (Figure 8b), which is accompanied withby a significant reduction (> 5%) of $(\sim 4\%)$ in eastward transport to North America (Figure 8a). Similarly, for the North American tracer, a warming of North Atlantic SSTs by 1°C slightly increases ($\sim \frac{2\%}{1}$) the concentrations in North America but decreases (3-4 %) the concentrations over downwind Europe. These results suggest that the warming (cooling) of SSTs tend to block (promote) trans-Pacific or trans-Atlantic transport. (Figure 8d). The response of the Indian 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 8ee). 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 O₃ over East Asia (Figure 7a), indicating the dominant effect of physical transport on the O₃ 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 O₃ (see Figures 7 and 8). This further indicates the distinct roles that different basin-scale SSTs play in nearby air quality.

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The changes in upper tropospheric circulation generally shows that warming of SSTs in different oceans tend to increase the 500-hPa geopotential height at mid-latitudes and decrease it over polar areas (Figure 9). Additionally, the increase in North Indian SSTs lead to geopotential height reductions above the Arabian Sea. The spatial pattern of geopotential height anomalies at 500hPa is roughly consistent with that of air temperature changes (Figure 9). This nonuniform increase in air temperature (i.e., more significant at mid-latitudes) weakens the meridional temperature gradient, resulting in a reduction of thermal winds. Therefore, the decreases in CO tracer transport to remote

regions can be well explained by both suppressed vertical ventilation and weakened westerlies at mid-latitudes. Further investigations of zonal wind suggest that an increase in SST over different oceans consistently decreases the westerly winds at lower midlatitudes (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 basinscale 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 O₃ 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 responsible for the dipole of the South Asian CO tracer changes over the source region depicted in Figure 8e.

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In addition, we also find a hemispheric-scale decrease of peroxyacetylnitratein peroxyacetyl nitrate (PAN), a reservoir of O₃ precursors (NO_x and HO_x) that facilitates the long-range transport of O₃, during the warming of different oceans (Figure S6S25). This decrease is likely to be caused by the increase of in the thermal decomposition of PAN responding in response to the air temperature rise increase (Doherty et al., 2013;Jacob and Winner, 2009;Doherty et al., 2013).

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Thus, it is reasonable to infer <u>that</u>, in general <u>that</u>, the increased thermal decomposition of PAN, <u>the</u> weakened mid-latitude westerlies, and <u>the</u> reduced vertical <u>ventilation air</u> <u>transport</u> may exert a joint reducing effect on <u>the</u> intercontinental transport of O₃

forduring basin-scale SST increases.

6. Summary

In this paper, we investigate the responses of surface O₃ to basin-scale SST anomalies in the Northern Hemisphere. The latest version of CESM (version 1.2.2) is used in our North Pacific, North Atlantic and North Indian Ocean Ceans, respectively. The responses of surface O₃ 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 O₃ changes. The regionally and seasonally averaged surface O₃ changes over four continental regions (i.e., NA, EU, EA and SA) pronounce wide seasonal and regional variability (varying from 1-to 3 ppbv). The warming of the North Pacific leads to <u>>nearly</u> 3 ppbv increases in <u>the</u> surface O₃ over <u>Southernsouthern</u> China in summer, with corresponding decreases over North America (~1 ppbv). Similarly, the North Atlantic SST warming elevates the surface O₃ pollution over North America while reducing the surface O₃ (nearly 1-2 ppbv) over Europe. Changes to in the North Indian SSTsSST exert significant impacts (1-3ppbv3 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 ozoneO₃ balance. The increase ofin SST in different ocean basins tendtends to increase the contributions of vertical diffusion to surface O₃ over upwind regions while greatly restraining that over other remotedownwind continents. These processes generally lead to a-widespread decrease ofdecreases in surface O₃, which are partially offset by increases in air temperature-dependent photochemical production rates. Specifically, the photochemicalchemical production changes account are mainly responsible for -90% of the surface O₃ increase 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. Increases Decreases in advective and the convective transport of O₃ to the ground-level are significant over South Asia surface associated with North Indian warming, which exerts an increasing influence are significant over South Asia and exert a negative impact on surface O₃ concentrations. Advective transport also exerts an increasing influence has a positive effect on surface O₃ in Southern China in the "Pacific-W" case.—

We further revealshow that air temperature is an important factor controlling the surface O₃ responses to SST anomalies. Reductions in the surface O₃ photochemicalchemical production in SouthernSouth 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 ofin air temperature associated with basin-scale SST warming is more likely to promote O₃ production over other highly polluted regions.

On the other hand, SST increases overat low latitudes of over different oceans enhance deep convection in the summer, which promotes convergence at the surface, as well as upward ventilation in themotions at low-latitudes. Corresponding The corresponding surface pressure anomalies centered over the east coast of Eastern East Asia associated with the North Pacific warming and over the Arabian Sea associated with the North Indian warming tend to increase the surface O₃ above through exchanging exchanges with the surrounding highly polluted air. The basin-scale SST increases in the North Northern Hemisphere promote a more stagnant climate reduce the tropospheric temperature gradient at mid-latitudes that restrains vertical transport of O₃ over continental regions as well as weakened mid-latitude continents and weakens the westerlies: at lower mid-latitudes. The response of the CO-tracer analysisalso suggests that these factors may have joint jointly exert a negative effects on long-range the intercontinental transport of surface O₃.

Overall, our study highlights the sensitivity of the surface O₃ distribution O₃ evolution to basin-wide SST changes over different oceans—in the Northern Hemisphere as well as and identifies the key chemical andor dynamical factors that control it. We recommend that regional air quality—this evolution. However, to provide a more comprehensive understanding of the SST-O₃ relationship, further studies using realistic SST variability are necessary. This study may aid in the management of O₃ pollution should consider by considering the influence of natural specific SST variability—and future increases in SSTs on ozone concentrations.

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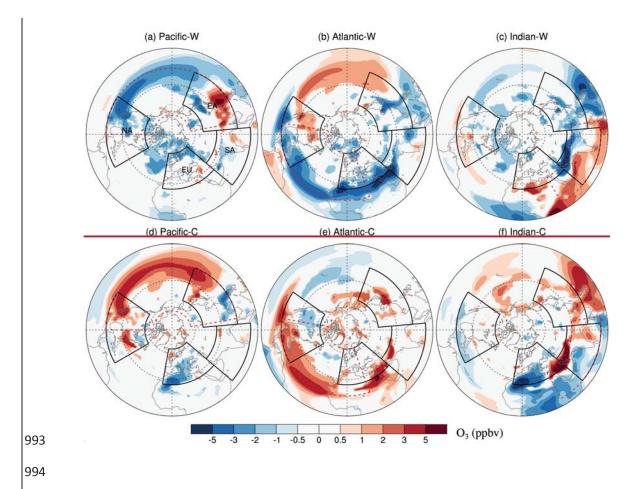
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Ozone (ppbv)			DJF	MAM	JJA	SON
North Pacific	+1° C	North America	-0. 50 ** <u>27</u> *	-0. 78 ** <u>42</u> *	- 1.01 ** <u>0.92</u> *	0.91** 1.03*
		Europe	-0.50 ** *	-0. 71 ** <u>26</u>	-0. 23 <u>10</u>	-0. 40 <u>29</u>
		East Asia	-0. 96 ** <u>88</u> *	-0. 92 ** <u>71</u> *	0. 45 <u>20</u>	0. 00 <u>17</u>
		South Asia	-1. 37 ** <u>00</u> *	0. 22 <u>30</u>	-0. 63 <u>43</u>	0. 58 <u>43</u> *
	-1° C	North America	0. 58** 00	0. 38 <u>57</u> *	0. 58 ** <u>55</u> *	0.49** <u>82</u> *
		Europe	0. 40 * <u>19</u>	0. 35 <u>15</u>	-0. 86 ** <u>47</u> *	0. 41 <u>47</u> *
		East Asia	0. 50 * <u>30</u>	<u>-</u> 0. 18 <u>17</u>	-0. 12 22	<u>-</u> 0. 00 <u>67</u> *
		South Asia	0. 02 <u>04</u>	- 1.22 * <u>0.24</u>	-0. 05 <u>03</u>	-0. 56<u>40</u>
North Atlantic	+1° C	North America	-0. 07 <u>03</u>	0. 20 49	0. 62 ** <u>50</u> *	0.4 6 ** <u>53</u> *
		Europe	0. 37 <u>30</u> *	0. 02 <u>06</u>	-1. 77 ** <u>61</u> *	-0. 83 ** <u>89</u> *
		East Asia	-0. 50 ** <u>52</u> *	-0. 61 ** <u>68</u> *	-0. 26 62*	-0. 06 <u>25</u>
		South Asia	-0.41 <u>20</u>	-1. 62 ** <u>46</u> *	- 0.96 ** <u>1.28</u> *	-0. 60 <u>82</u> *
	-1° C	North America	<u>-</u> 0. 27 <u>07</u>	<u>-</u> 0. 32 <u>10</u>	-0. 35 <u>10</u>	-0. 35 * <u>17</u>
		Europe	-0. 33 <u>00</u>	0. 24 <u>00</u>	0. 52 <u>07</u>	- 0.06
		East Asia	0. 11 <u>16</u>	<u>-</u> 0. 59 ** <u>08</u>	0. 64 <u>80</u> *	<u>-0.4160*</u>
		South Asia	-0. 08 <u>20</u>	-0. 55 <u>40</u>	0. 95 * <u>30</u>	<u>-</u> 0. 21 <u>10</u>
North India	+1° C	North America	-0. 40 * <u>25</u>	-0. 31 <u>04</u>	-0. 11 <u>16</u>	-0. 08 <u>10</u>
		Europe	-0.30	0. 29 <u>08</u>	-0. 07 <u>12</u>	0. 42 <u>19</u>
		East Asia	-0. 67** 53*	-0. 63 ** <u>77</u> *	-0. 57** 28	-1. 56 ** <u>78</u> *
		South Asia	-1. 30 ** <u>00</u> *	0. 12 <u>14</u>	-2.06** <u>1.67</u> *	-2. 33 ** <u>75</u> *
	-1° C	North America	-0. 07 <u>04</u>	-0. 10 <u>17</u>	0. 31<u>04</u>	-0. 12 <u>25</u>
		Europe	-0. 28 <u>05</u>	-0. 09 <u>07</u>	-0. 52 <u>13</u>	-0. 27 <u>24</u>
		East Asia	<u>-</u> 0. 22 <u>06</u>	0. 69 ** <u>15</u>	0. 22 <u>55</u> *	1.60** <u>0.33</u>
		South Asia	-0. 01 <u>03</u>	0. 21 <u>57</u>	1. 94** 70*	1. 70 ** <u>31</u> *

^{*}significant at the 0.1 level from Student t-test using 11 years model result

^{**}significantSignificant at the 0.05 level from Student's t-test using 1120 years of model resultresults.



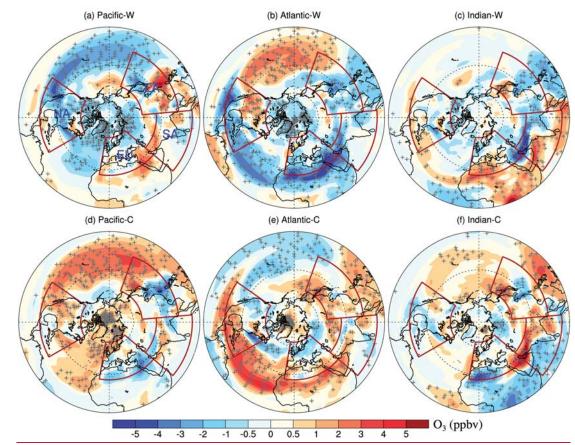


Figure 1. Changes in the summertime (June-August) surface ozoneO₃ concentrations (ppbv) in the Northern Hemisphere induced by 1°C1°C warming (top) and 1°C°C cooling (bottom) in the North Pacific Ocean (left), North Atlantic Ocean (center), and North Indian Ocean (right) relative to the CTRL. Four The 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 blackred polygons. Only The + symbols denote areas where results are significant at the 0.405 level, evaluated with aby Student's t-test using 1120 years of data are depicted (plots using the Mercator projection are shown in Figure S2 in the supplementary material).

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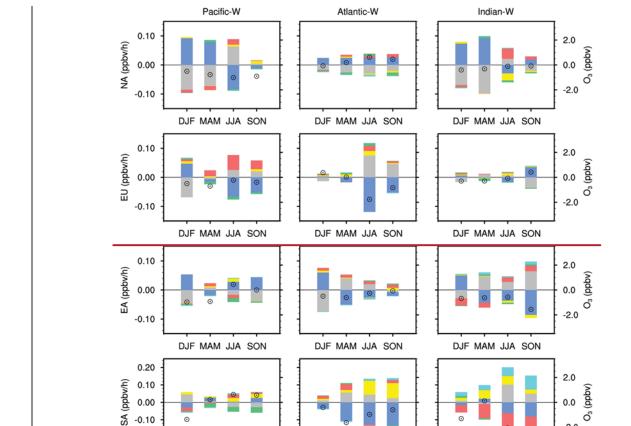
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-0.20

DJF MAM JJA SON



DJF MAM JJA SON

DRYD VDIF ADVE

-2.0

DJF MAM JJA SON

DEEP

CHEM

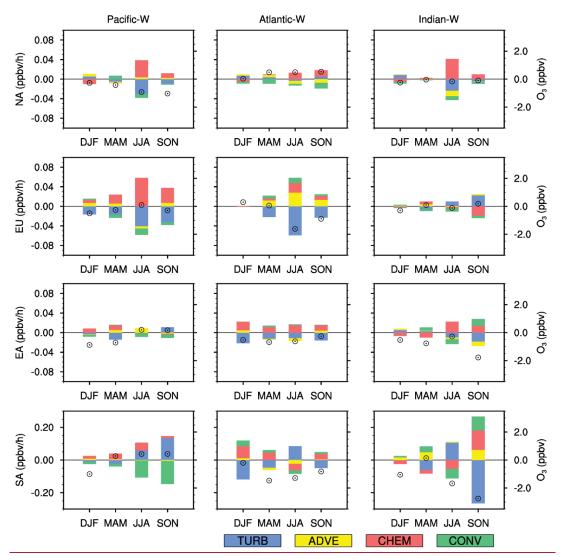


Figure 2. Seasonally averaged changes in the IPR contributions (bars, ppbv/h, left scale) and surface O₃ concentrations (hollow circles, ppbv, right scale) for Pacific-W (left), Atlantic-W (middle) and Indian-W (right) relative to the CTRL. Values are regionally averaged over NA (first row), EU (second row), EA (third row) and SA (last row), respectively.). TURB is defined as the sum of VDIF and DRYD. CONV is the sum of DEEP and SHAL. IPR contributions from sixthe four processes (i.e., gas-phase chemistry (TURB, ADVE, CHEM), advection (ADVE), vertical diffusion (VDIF), dry deposition (DRYD), shallow convection (SHAL) and deep convection (DEEP)) and CONV) are represented by different colors. A more detailed IPR result is shown in Figure S10 in the supplementary material.

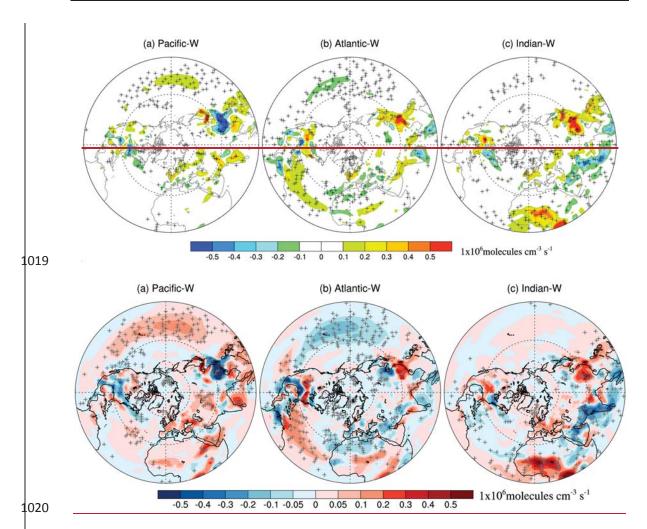


Figure 3. Perturbations of <u>the surface net O3 net-production rate</u> (1x10⁶ molecules cm³ s⁻¹) relative to CTRL for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W <u>relative to the CTRL</u> in <u>the boreal summer.</u> The + symbols denote areas where <u>the results are significant at the 0.05 level as, evaluated with aby Student's t-test using 1120 years of data-(plots using the Mercator projection are shown in Figure S14 in the supplementary material).</u>

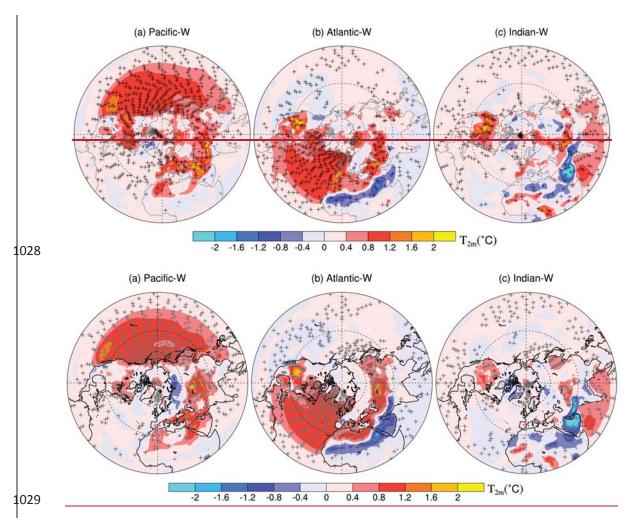
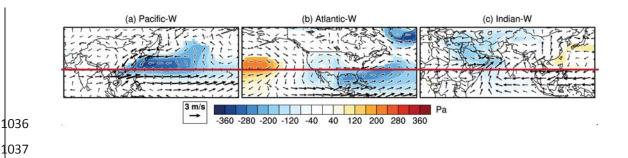


Figure 4. —The differenceChanges in the surface air temperature (°C°C) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL in the Northern Hemisphere in the boreal summer. The + symbols denote areas where the results are significant at the 0.05 level-as, evaluated with aby Student's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



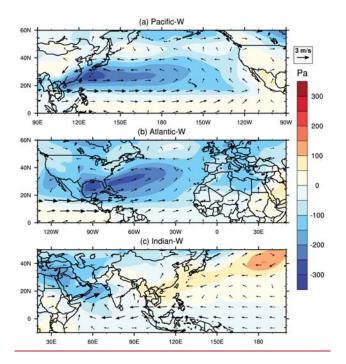
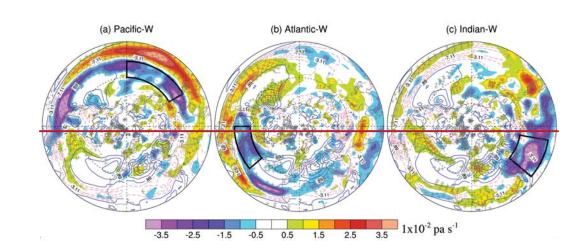


Figure 5. –Changes in <u>the</u> surface pressure (color contours, Pa) and <u>850-hPa</u> wind <u>pattern</u> (arrows, m/_s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to <u>the</u> CTRL in <u>the</u> boreal summer. As for surface pressure changes, only results <u>significant at the 0.05 level evaluated with a Student t-test are depicted.</u>



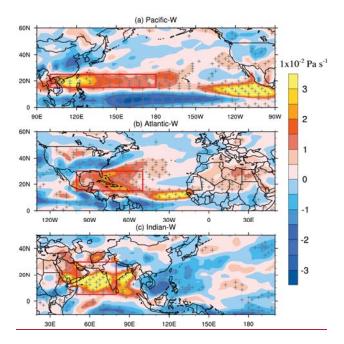
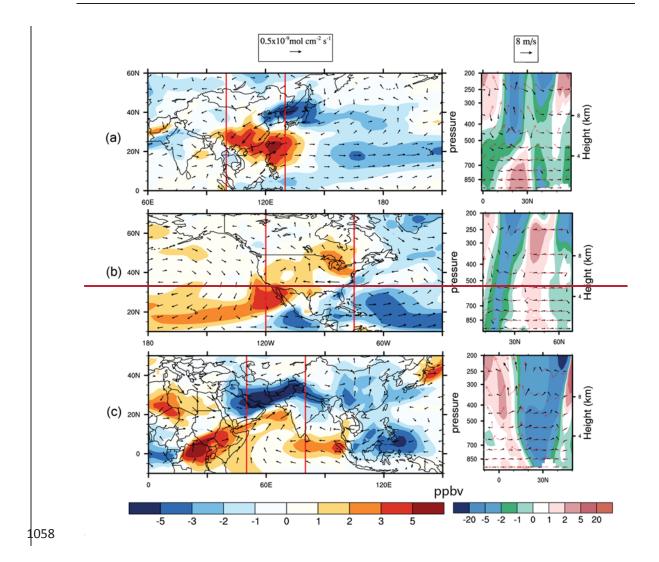


Figure 6. The spatial Spatial pattern of vertical velocity changes at 500 hPa (color contours, 1x10⁻² Pa s⁻¹) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to CTRL. Contours with blue solid lines and red dashes indicate positive and negative downward vertical velocity in the control case, respectively (Contour interval: 2x10⁻² Pa s⁻¹). Blackthe 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 75 a-c). The + symbols indicate areas where the results are significant at the 0.05 level-as, evaluated with aby Student's t-test using 1120 years of data.



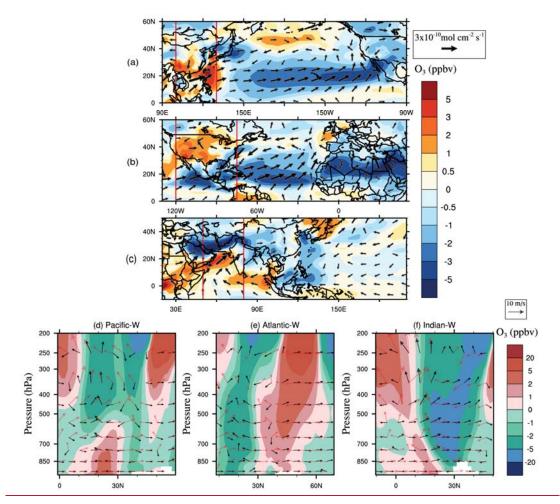


Figure 7. Left columnTop three rows: Changes in O₃ concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm⁻² s⁻¹) at the surface level in boreal summer for (a) Pacific-W-(top)₅₂ (b) Atlantic-W-(middle)₅₂ (c) Indian-W (bottom) relative to the CTRL. Right column: Longitude averaged vertical and latitudinal distributions of in the boreal summer. Last row: zonal average of the tropospheric O₃ changes (color contours, ppbv) and), wind velocityfluxes in CTRL (red arrows, m/s⁻¹) and itsthe wind flux perturbation (black arrows, m/s) corresponding to the left. in (d) Pacific-W, (e) Atlantic-W, (f) Indian-W relative to the CTRL in the boreal summer. The red rectangles in the left column (a), (b) and (c) denote the longitudinal range used for average the zonal averages in (d), (e) and (f), respectively. The vertical wind velocity is amplified 1000 times to bemake it comparable withto the horizontal wind velocity and distinct in the panels.

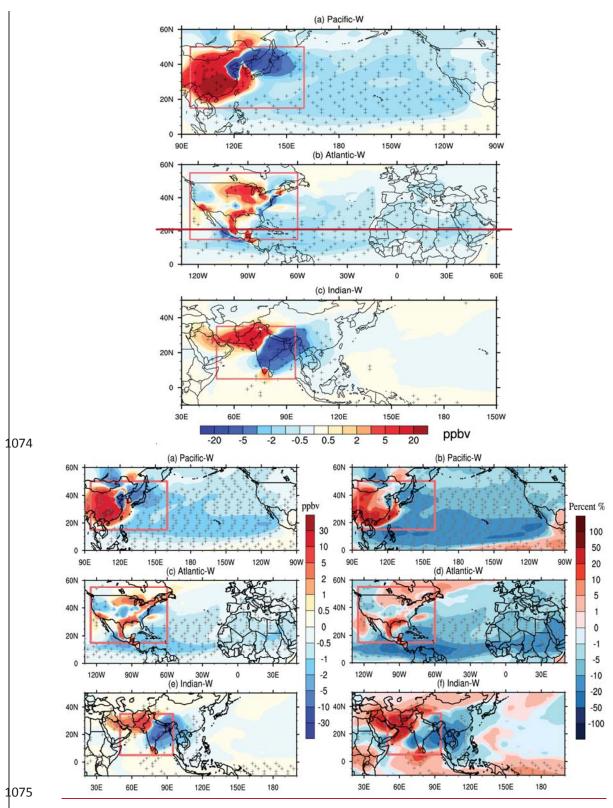


Figure 8. The difference in Left-hand panel: Difference in the surface concentration (ppbv) of a CO-like tracer emitted infrom (a) the East Asia for Pacific-W, (b) thec) North America for Atlantic-W and (ee) the Indian South Asia for Indian-W relative to CTRL: the CTRL in the boreal summer. Right-hand 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 with aby Student's t-test using 20 years of data.



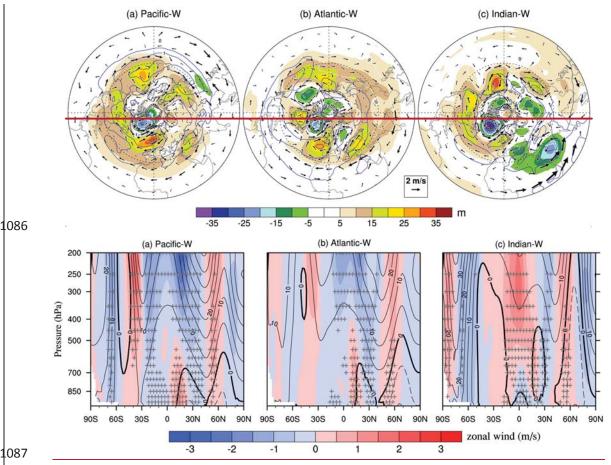


Figure 9. Changes Zonally averaged changes in geopotential height zonal wind (color contour, m), air temperature (/s) and geopotential height (contour, °C) and wind pattern anomalies (arrows, m s +) at 500hPa) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to CTRL. Bluethe CTRL in the boreal summer. Black solid lines and red dashed lines in the contours indicate positive and negative air temperature geopotential height anomalies, respectively (Contour contour interval: 5 m). The + symbol denotes areas where the zonal wind changes are significant at the 0.5 ° C). 05 level, evaluated by Student's t-test using 20 years of data.