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

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Abstract

The response of surface ozone (O_3) concentrations to basin-scale warming and cooling of Northern Hemisphere oceans is investigated using the Community Earth System Model (CESM). Idealized, spatially uniform sea surface temperature (SST) anomalies of +/- 1 °C are individually superimposed onto the North Pacific, North Atlantic, and North Indian Oceans. Our simulations suggest large seasonal and regional variability of surface O_3 in response to SST anomalies, especially in the boreal summer. The responses of surface O_3 associated with basin-scale SST warming and cooling have similar magnitude but are opposite in sign. Increasing the SST by 1 °C in one of the oceans generally decreases the surface O_3 concentrations from 1 to 5 ppbv. With fixed emissions, SST increases in a specific ocean basin in the Northern Hemisphere tend to increase the summertime surface O_3 concentrations over upwind regions, accompanied by a widespread reduction over downwind continents. We implement the integrated process rate (IPR) analysis in CESM and find that meteorological O_3 transport in response to SST changes is the key process causing surface O_3 perturbations in most

cases. During the boreal summer, basin-scale SST warming facilitates the vertical transport of O_3 to the surface over upwind regions while significantly reducing the vertical transport over downwind continents. This process, as confirmed by tagged CO-like tracers, indicates a considerable suppression of intercontinental O_3 transport due to increased tropospheric stability at lower mid-latitudes induced by SST changes. On the other hand, the responses of chemical O_3 production to regional SST warming can exert positive effects on surface O_3 levels over highly polluted continents, except South Asia, where intensified cloud loading in response to North Indian SST warming depresses both the surface air temperature and solar radiation, and thus photochemical O_3 production. Our findings indicate a robust linkage between basin-scale SST variability and continental surface O_3 pollution, which should be considered in regional air quality management.

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

1. Introduction

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

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

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

92 climate has received increasing attention and will be essential for future surface O_3
93 mitigation (Jacob and Winner, 2009; Doherty et al., 2013).
94
95 Sea surface temperature (SST) is an important indicator that characterizes the state of
96 the climate system. Its variations strongly perturb the mass and energy exchange
97 between the ocean and atmosphere (Small et al., 2008; Gulev et al., 2013), which
98 influence atmospheric circulation, atmospheric temperature and specific humidity
99 (Sutton and Hodson, 2005; Frankignoul and Sennéchael, 2007; Li et al., 2008) from
100 regional to global scales (Glantz et al., 1991; Wang et al., 2000; Goswami et al., 2006).
101 It also affects cloud formation and consequently influences incoming solar radiation
102 (Deser et al., 1993; Fallmann et al., 2017). Numerous studies have shown that SST
103 changes over different oceans and at different latitudes lead to significantly different
104 meteorological and climate responses (Webster, 1981; Lau and Nath, 1994; Lau, 1997;
105 Sutton and Hodson, 2007; Sabeerali et al., 2012; Ueda et al., 2015). Details on the SST-
106 climate relationships over individual oceanic regions are summarized in Kushnir et al.
107 (2002).
108
109 The Intergovernmental Panel on Climate Change Fifth Assessment Report (IPCC, 2013)
110 provides strong evidences in Chapter 2 that global SSTs are generally increasing due to
111 the impacts of anthropogenic forcings on global climate change. In addition, regional
112 SST exhibits natural periodic or irregular oscillations with timescales ranging from
113 months to decades. The El Niño/Southern Oscillation (ENSO) is the most influential
114 natural SST variability that originates in the tropical Pacific and has worldwide climate
115 impacts (Philander, 1983; Wang et al., 2012). The Pacific decadal oscillation (PDO),
116 defined by ocean temperature anomalies in the northeast and tropical Pacific Ocean, is
117 another long-lived, El Niño-like pattern that persists for several decades (Mantua and
118 Hare, 2002). Over the Indian Ocean, SST anomalies feature a seesaw structure between
119 the western and eastern equatorial regions, known as the Indian Ocean Dipole (IOD)
120 mode (Saji et al., 1999). The North Atlantic Ocean exhibits various modes of low-
121 frequency SST variability (Kushnir, 1994; Wu and Liu, 2005; Fan and Schneider, 2012;

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

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

To fill this gap, this study focuses on examining the sensitivity of O_3 evolution over four polluted continental regions in the Northern Hemisphere (i.e., North America (NA, 15°N–55 °N; 60°W–125°W), Europe (EU, 25°N–65 °N; 10°W-50 °E), East Asia (EA, 15 °N–50 °N; 95°E–160 °E) and South Asia (SA, 5 °N–35 °N; 50 °E–95°E), defined in Fiore et al. (2009)) with respect to nearby basin-scale SST changes. We describe the design of numerical experiments and model configuration in Section 2. Surface O_3 responses to regional SST changes are given in Section 3. Relevant mechanisms governing the SST-O_3 relationships are discussed in Section 4. The impact of basin-

scale SST changes on inter-continental transport of O_3 is described in Section 5. Conclusions are drawn in Section 6.

2. Methodology

2.1 Model description and configuration

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

The chemistry coupled in the CAM5 (i.e., CAM5-chem) is primarily based on the Model for O_3 and Related chemical Tracers, version 4 (MOZART-4), which resolves 85 gas-phase species, and 196 gas-phase reactions (Emmons et al., 2010; Lamarque et al., 2012). A three-mode (i.e., Aitkin, accumulation and course) aerosol scheme for black carbon (BC), primary organic matter (POM), second organic aerosol (SOA), sea salt, dust and sulfate was used in our simulations following Liu and Ghan (2010). The lightning parameterization is modified according to Price et al. (1997) and tropospheric photolysis rates are calculated interactively following Tie et al. (2005). Gaseous dry deposition is calculated using the resistance-based parameterization of Wesely (1989), Walmsley and Wesely (1996), and Wesely and Hicks (2000). The parameterizations of in-cloud scavenging and below-cloud washout for soluble species are described in

detail by Giorgi and Chameides (1985) and Brasseur et al. (1998), respectively. Anthropogenic emissions of chemical species are from the IPCC AR5 emission datasets (Lamarque et al., 2010), whose injection heights and particle size distributions follow the AEROCOM protocols (Dentener et al., 2006). The emissions of natural aerosols and precursor gases are prescribed from the MOZART-2 (Horowitz et al., 2003) and MOZART-4 (Emmons et al., 2010) datasets. All emission datasets are available from the CESM data inventory (https://svn-ccsm-inputdata.cgd.ucar.edu/trunk/inputdata/). The performance of CESM in simulating tropospheric O_3 has been validated by comparing with ozonesondes and satellite observations (Tilmes et al., 2014). The deviations between model and observations are within the range of about 25%. In general, the model can capture the surface ozone distribution and variability well, but may overestimate O_3 over the Eastern US and Western Europe in the summer (Tilmes et al., 2014).

2.2 Numerical experiments

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

1 °C SST warming and 1 °C SST cooling superimposed onto all three ocean basins (i.e., the North Pacific, North Atlantic and North Indian Ocean) in the Northern Hemisphere, denoted as "All-W" and "All-C", respectively. Air pollution emissions, including biogenic emissions of VOCs, are fixed to distinguish the impacts of SST variation on O_3 transport and photochemistry. All simulations are run for 21 years with the first year used for model spin-up.

To explore the impacts of SST changes on inter-continental transport, an explicit emission tagging technique is used in our simulations following previous studies (Shindell et al., 2008; Doherty et al., 2013). Artificial CO-like tracers emitted from four continental regions, i.e., North America (NA, 15°N–55 °N; 60°W–125°W), Europe (EU, 25°N–65 °N; 10°W-50 °E), East Asia (EA, 15 °N–50 °N; 95°E–160 °E) and South Asia (SA, 5 °N–35 °N; 50 °E–95°E), are tracked individually. These tracers are idealized with a first-order decay lifetime of 50 days, which is similar to O_3 (Doherty et al., 2013) and used to single out changes in O_3 transport induced by SST anomalies.

2.3 Integrated process rate (IPR) analysis

To provide a process-level explanation for the response of surface O_3 to regional SST changes, the IPR method is applied. This method calculates the accumulated contributions of individual processes (e.g., chemical production and loss, advection, vertical diffusion, dry deposition, etc.) to O_3 predictions during the model simulation and has been widely used for air pollution diagnostics (Li et al., 2012; Zhang and Wu, 2013; Tao et al., 2015). In this study, we added the IPR scheme to the CESM framework to track the contribution of six physicochemical processes (i.e., gas-phase chemistry (CHEM), advection (ADVE), vertical diffusion (VDIF), dry deposition (DRYD), shallow convection (SHAL) and deep convection (DEEP)) to O_3 concentrations in every grid box. Wet deposition and aqueous-phase chemistry are ignored here due to the low solubility and negligible chemical production of O_3 in water (Jacob, 1999). Therefore, CHEM represents the net production (production minus loss) rate of O_3 due to gas-phase photochemistry. DRYD represents the dry deposition fluxes of O_3, which

is an important sink for O_3. The other IPR terms (i.e., ADVE, VDIF, SHAL and DEEP) represent contributions from different transport processes. The IPR scheme tracks and archives the O_3 flux in each grid from every process during each model time step. The sum of the O_3 fluxes from these six processes matches the change in the O_3 concentration. The IPR method has been widely used in air quality studies to examine the cause of pollution episodes (Wang et al., 2010; Li et al., 2012). When applied in climate sensitivity analysis (usually measuring the difference between two equilibriums), the net change of all IPRs approaches zero. Typically, the positive changes in IPRs are mainly responsible for the increase in surface O_3, which may further induce O_3 removal to balance this forcing in a new equilibrium. Therefore, here, the IPR analysis is used not to budget the SST-induced O_3 concentration changes but rather to help examine the relative importance of different transport and chemical processes in driving the sensitivity of O_3 to SST forcing. Its performance is verified by comparing the predicted hourly O_3 changes with the sum of the individual fluxes from the six processes. As shown in Figure S1, the hourly surface O_3 changes are well represented by the sum of these fluxes in the model.

3. Response of surface O_3 concentrations to SST changes

Seasonally and regionally averaged surface O_3 changes in each SST perturbation simulation for the four highly populated continental regions and three ocean basins defined in our study are given in Tables 1 and S1, respectively. The responses of the surface O_3 concentrations to basin-scale SST changes (i.e., ±1 °C) are mainly below 3 ppbv in the Northern Hemisphere (Tables 1 and S1), though larger anomalies (i.e., up to 5 ppbv) are also observed over the eastern coast of China, the Indian subcontinent, and certain oceanic areas (Figures 1 and S2). This SST-O3 sensitivity is comparable to previous findings. For instance, Bloomer et al. (2009) reported a positive O_3-temperature relationship of 2.2~3.2 ppbv/°C across the rural eastern United States. Wu et al. (2008) found that summertime surface O_3 may increase by 2-5 ppbv over the northeastern United States in the 2050s. Additionally, Fiore et al. (2009) demonstrated

an intercontinental decrease in surface O_3 of no more than 1 ppbv in response to 20 % reductions in anthropogenic emissions within a continental region. Our study indicates that basin-scale SST changes alone may exert significant effects on the surface O_3 above specific ocean basin and its surrounding continents.

As shown in Figure 1, seasonal changes of up to 5 ppbv in the mean surface O_3 concentration are observed during boreal summers, mainly in coastal regions and remote oceans. Surface O_3 changes in response to positive and negative SST anomalies generally exhibit a consistent spatial pattern but are opposite in sign, suggesting robust relationships between surface O_3 levels and SST anomalies (Figure 1). An increase in summertime SST over a specific ocean basin tends to increase the surface O_3 concentration over the upwind regions but reduce this concentration over downwind continents. For instance, a 1 °C warming over the North Pacific leads to a widespread decrease in surface O_3 over the North Pacific, North America and the North Atlantic of approximately 1 ppbv (Table S1) but may enhance the surface O_3 by nearly 3 ppbv over South China. Similarly, the SST warming over the North Atlantic decrease the surface O_3 levels by 1~2 ppbv over the North Atlantic and Europe but increase (~1 ppbv) that over North America and the North Pacific. 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 S2 in the supplementary material.

Our simulations reveal that different oceans can exert distinct region-specific effects on the O_3 distribution. The effects of three individual warming/cooling cases (i.e., Pacific-W, Atlantic-W and Indian-W/Pacific-C, Atlantic-C and Indian-C) on surface O_3 distributions are further summed up to compare with the combined warming/cooling cases(i.e., ALL-W/ALL-C). The responses of surface O_3 to a hemispheric SST anomaly

generally resemble the sum of responses to individual regional SST changes (see Figures S3 and S4 in the supplementary material). This indicates that the effect of a generalized SST warming on surface O_3 can be decomposed into individual regional SST forcings. We now analyze the processes that impact the dependence of SST on the O_3 distribution using simulations that increase the SST.

4. Mechanism of SST-induced surface O_3 changes

4.1 Process-level response to SST changes

In this study, IPR analysis is used to evaluate the contribution of different physicochemical processes to O_3 evolution. 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 S5). In most cases, VDIF and DRYD are the key processes controlling the O_3 variation. The downward transport of O_3 through diffusion (VDIF) is an important source of surface O_3, while DRYD acts as a sink. Both processes are simultaneously determined by the strength of turbulence. Here, we define a new term TURB as the sum of DRYD and VDIF, which can capture the overall effect of turbulence changes on surface O_3 concentrations. In addition, we merge SHAL and DEEP as CONV to represent the total contribution of convective transport to surface O_3 (Figures 2 and S5). More detailed IPR results are shown in Figures S6 and S7 in the supplementary material.

In the "Pacific-W" case, a 1 °C SST warming over the North Pacific increases VDIF over eastern China in JJA (Figure S8), which is insignificant if averaged over the whole East Asia region. Meanwhile, this Pacific warming considerably reduces VDIF over North America (Figure S6). The corresponding decrease in TURB over North America mainly determines the surface O_3 reduction in JJA and SON, while the reduction in CONV exerts an additional negative impact (Figure 2). In the "Atlantic-W" case, increases in VDIF are also observed over the upwind regions (i.e., North America) in JJA. However, these increases are accompanied by commensurate decreases in DRYD,

329 resulting in an insignificant overall change in TURB (Figure 2). Therefore, the increase
330 in CHEM is mainly responsible for the surface O_3 increase over North America in JJA.
331 TURB is more relatively important over Europe (only in JJA and SON), leading to
332 reduced surface O_3 abundance. In the "Indian-W" case, both CHEM and CONV are
333 reduced over South Asia in JJA, leading to overall reductions in surface O_3 over the
334 Indian subcontinent (Figure 2). The IPR analysis over the ocean basins shows that the
335 warming of the North Pacific or North Atlantic induces reductions in VDIF and CHEM,
336 which are responsible for the significant decrease in surface O_3 above these regions in
337 JJA (Figure S7). The North Indian Ocean warming, on the other hand, enhances DEEP
338 and VDIF, leading to a local increase in surface O_3 in JJA.
339
340 The IPR analysis indicates that, in general, an SST increase in the North Pacific or
341 North Atlantic is more likely to enhance the vertical diffusion of O_3 over upwind
342 regions (i.e., East Asia or North America, respectively) but suppress this diffusion over
343 the ocean basin as well as downwind continents in JJA (Figure S8). These opposite
344 changes in VDIF over upwind and downwind regions lead to distinct surface O_3
345 responses. Changes in CHEM enhance surface O_3 formation in most cases. An
346 exception is in South Asia, where CHEM and DEEP dominate the reduction in surface
347 O_3 over the region in JJA associated with the North Indian Ocean warming. In the
348 following subsections, the mechanisms of the SST-O_3 relationship for the four polluted
349 continents are further explored. Here we focus on boreal summers since the surface O_3
350 response to SST changes is more robust during this period than other seasons.
351

352 **4.2 Response of photochemical O_3 production to SST increases**

353 Changes in the net production rate (i.e., chemical production rate minus loss rate) of O_3
354 at the surface in JJA associated with basin-scale SST increases are shown in Figure 3.
355 The peak changes are mainly confined to regions where O_3 precursors are abundant
356 (e.g., South and East Asia and North America). For example, a warmer North Pacific
357 SST exerts a positive (negative) impact on net O_3 production in the northern (southern)

regions of East Asia. Similarly, the warming of the North Atlantic promotes a dipole impact on the surface O_3 production over North America, while the warming of the North Indian Ocean significantly decreases the net O_3 production rate over South Asia.

As emissions are fixed in all simulations, the change in net O_3 production is driven by SST induced meteorological changes (e.g., air temperature, air humidity, and solar radiation). An increase in SST of 1 °C in any ocean basin leads to a widespread enhancement of the surface air temperature (i.e., the air temperature at 2 m) over most continental areas (Figure 4). An exception is the North Indian Ocean, where an increase in SST tends to cool the Indian subcontinent by 1-2 °C. This temperature decrease is not only limited to the surface but also spreads to 600 hPa (Figure S9). Associated with this temperature decrease is a remarkable reduction in the solar radiation received at the surface (more than 15 W/m^2, Figure S10). 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 reduces the solar radiation reaching the surface of the Indian subcontinent and thus 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 S9), which may further suppress the development of deep convection over the Indian subcontinent.

Previous studies have indicated that air temperature positively affects both O_3 production and destruction rates (Zeng et al., 2008; Pusede et al., 2015). As shown in Figure S11, 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

388 to a widespread enhancement of the air temperature, resulting in a positive change in
389 the net O_3 production over most continental regions (Figure 3). However, a warmer SST
390 also increases the air humidity (Figure S12), which enhances O_3 destruction over most
391 coastal and oceanic areas. In addition, over South Asia, a warming of the North Indian
392 Ocean decreases solar radiation and air temperature, and simultaneously increases air
393 humidity, which jointly exert negative effects on O_3 production in that region.
394

395 **4.3 Response of physical O_3 transport to SST increases**

396 In Section 4.1, our IPR analysis highlights multiple physical processes (i.e., vertical
397 diffusion, convection and advection) that are important in modulating surface O_3
398 concentrations. However, the role and relative importance of each process exhibit large
399 spatial heterogeneity. In this section, we explore the key factors controlling physical O_3
400 transport in response to basin-scale SST changes.
401

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

Strengthened deep convection will trigger large-scale subsidence over nearby regions through the modulation of large-scale circulation patterns, which may suppress convective transport (Lau et al., 1997; Roxy et al., 2015; Ueda et al., 2015). This effect is verified by the decreases in upward velocity at 500 hPa. As depicted in Figure 6, significant decreases in upward velocity occur over regions adjacent to the strengthened deep convection. Similar effects are also observed over higher latitudes or remote oceans (Figure S13). Meanwhile, the air temperature increase in response to regional SST warming is more significant above the lower troposphere, which leads to a decrease in the vertical temperature gradient (Figure S9). These factors tend to restrain the vertical exchange of air pollutants at mid-latitudes, which facilitates surface O_3 accumulation over polluted continental regions in JJA but may weaken the intrusion of O_3 from the upper troposphere to the surface in most unpolluted areas. This process helps to explain the widespread decrease in surface O_3 over unpolluted regions associated with an SST increase, as described in Section 3, and can be further verified by the wide-spread reduction in VDIF shown in Figure S8.

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

In the "Atlantic-W" case, the SST warming-induced surface pressure anomalies lead to

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

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

5. Implications for O_3 long-range transport

The above findings indicate that, in general, a basin-scale SST increase in the Northern Hemisphere is more likely to enhance atmospheric stability at mid-latitudes, which may suppress air pollutants from lofting to the free troposphere. This process potentially has large effects on O_3 intercontinental transport. Following previous work (e.g., Doherty et al., 2013; Fang et al., 2011), we use passive CO-like tracers to demonstrate the potential effect of regional SST changes on long-range O_3 transport. A warming of North Pacific SSTs by 1°C tends to increase the East Asian CO tracer concentrations by nearly 6% at the surface (Figure 8b), which is accompanied by a significant reduction (~4%) in eastward transport to North America. Similarly, for the North American tracer, a warming of North Atlantic SSTs by 1°C increases (~1%) the concentrations in North America but decreases (3-4 %) the concentrations over

downwind Europe (Figure 8d). The response of the South Asian CO tracer to North Indian Ocean warming also shows a decreasing tendency over downwind regions, but the patterns are more complicated over the source region in this case (Figure 8e). Because the CO-like tracers added in the simulation have a fixed decay lifetime, their concentration changes are completely caused by the SST-induced transport anomalies. The decrease in CO tracer concentrations over downwind regions suggests that the warming of basin-scale SST tends to suppress the long-range transport of air pollutants. Additionally, in the "Pacific-W" case, changes in the East Asian CO tracer (Figure 8a) generally resemble the changes in surface O_3 over East Asia (Figure 7a), indicating the dominant effect of physical transport on the O_3 distribution over East Asia. Regarding the North American CO tracer in response to the North Atlantic warming or the South Asian CO tracer in response to the North Indian Ocean warming, their concentration changes are spatially inconsistent with those of O_3 (see Figures 7 and 8). This further indicates the distinct roles that different basin-scale SSTs play in nearby air quality.

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

discussed in Section 4.3). The corresponding anomalous cyclonic circulation may be responsible for the dipole of the South Asian CO tracer changes over the source region depicted in Figure 8e.

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

Thus, it is reasonable to infer that, in general, the increased thermal decomposition of PAN, the weakened mid-latitude westerlies, and the reduced vertical air transport may exert a joint reducing effect on the intercontinental transport of O_3 during basin-scale SST increases.

6. Summary

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

surface O_3 (nearly 1-2 ppbv) over Europe. Changes in the North Indian SST exert significant impacts (1-3 ppbv) over South and East Asia during the entire year.

Process analysis indicates that dry deposition and vertical diffusion are two major processes governing the surface O_3 balance. The increase in SST in different ocean basins tends to increase the contributions of vertical diffusion to surface O_3 over upwind regions while greatly restraining that over downwind continents. These processes generally lead to widespread decreases in surface O_3, which are partially offset by increases in air temperature-dependent chemical production rates. Specifically, the chemical production changes are mainly responsible for the surface O_3 increases over North America in response to the North Atlantic SST warming but exert a negative effect on South Asia in response to the North Indian SST warming. Decreases in the convective transport of O_3 to the surface associated with North Indian warming are significant over South Asia and exert a negative impact on surface O_3 concentrations. Advective transport has a positive effect on surface O_3 in southern China in the "Pacific-W" case.

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

On the other hand, SST increases at low latitudes over different oceans enhance deep convection in summer, which promotes convergence at the surface, as well as upward motions at low latitudes. The corresponding surface pressure anomalies centered over the east coast of East Asia associated with the North Pacific warming and over the Arabian Sea associated with the North Indian warming tend to increase the surface O_3 above through exchanges with the surrounding highly polluted air. The basin-scale SST

565 increases in the Northern Hemisphere reduce the tropospheric temperature gradient at
566 mid-latitudes that restrains vertical transport of O_3 over continents and weakens the
567 westerlies at lower mid-latitudes. The response of the CO-tracer also suggests that these
568 factors may jointly exert a negative effect on the intercontinental transport of O_3.
569
570 This study highlights the sensitivity of O_3 evolution to basin-wide SST changes in the
571 Northern Hemisphere and identifies the key chemical or dynamical factors that control
572 this evolution. Idealized and spatially uniform SST anomalies are used to explore the
573 general mechanisms governing SST-O_3 relationships. We find that the SST changes
574 over tropical regions exert considerable impacts on surface O_3 levels. The increase in
575 tropical SST over different ocean basins enhances deep convection, which further
576 trigger large-scale subsidence over nearby and remote regions. These enhanced
577 convective activities also tend to release more latent heat over the upper troposphere
578 and significantly increases the air temperature there. These processes influence large-
579 scale circulation patterns and lead to opposite surface O_3 responses over upwind and
580 downwind regions related to a specific ocean basin. These finding provides valuable
581 implications for the potential surface O_3 change in response to future warming or
582 cooling of individual oceans.
583
584 Additionally, the sensitivity tests with 1ºC SST warming and cooling superimposed
585 onto all three ocean basins further show in general that the SST forcing on O_3
586 distribution is geographically additive. A number of studies have used the decomposed
587 SST anomalies for different regions to identify their relevant roles in a particular
588 climate response (e.g., Sutton and Hodson, 2005; Camargo et al., 2013; Ueda et al.,
589 2015). A linear assumption that the influence of large-scale SST anomaly pattern on the
590 atmosphere can be generally constructed by the linear combination of the influences of
591 individual SST patches have been verified by previous studies, especially for the
592 tropical regions where the signal-to-noise is higher (e.g., Fan et al., 2016; Seager and
593 Henderson, 2016). Therefore, our study also helps to understand the roles of different
594 ocean basins in the Northern Hemisphere played in modulating surface O_3 distributions

595 in a global-wide SST warming condition associated with climate change.

596

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

606

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

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

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

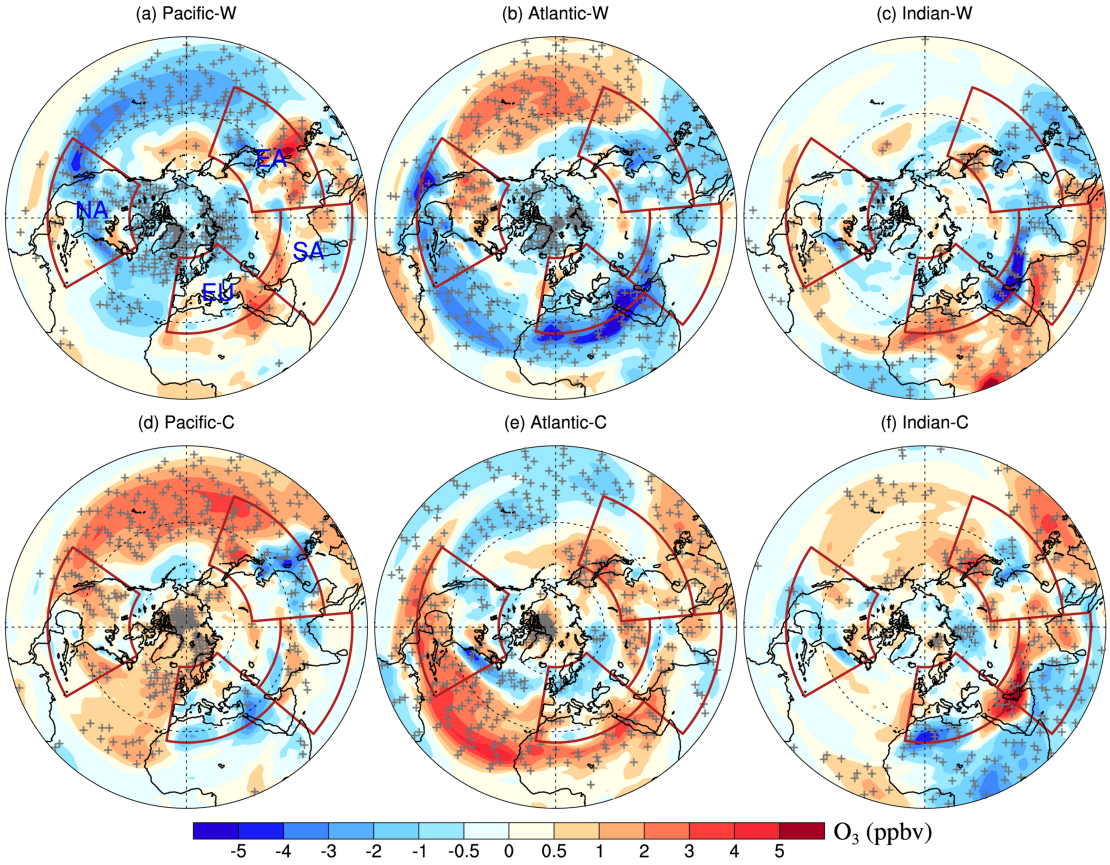
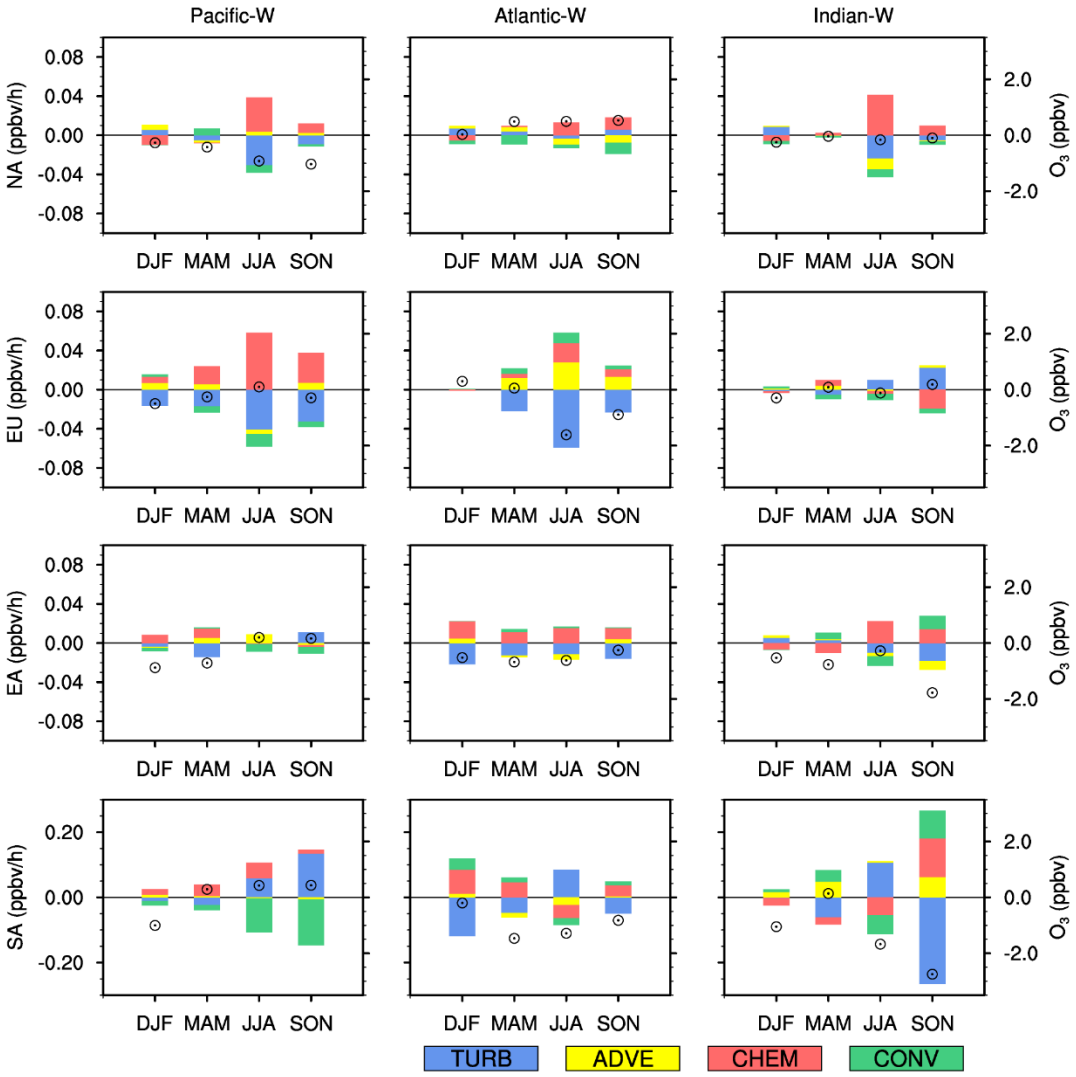


Figure 1. Changes in the summertime (June-August) surface O_3 concentrations (ppbv) in the Northern Hemisphere induced by 1ºC warming (top) and 1°C cooling (bottom) in the North Pacific Ocean (left), North Atlantic Ocean (center), and North Indian Ocean (right) relative to the CTRL. The four major regions of interest (i.e., NA (15°N–55°N; 60°W–125°W), EU (25°N–65°N; 10°W-50 °E), EA (15°N–50°N; 95°E–160 °E) and SA (5°N–35°N; 50°E–95°E)) are marked with red polygons. The + symbols denote areas where results are significant at the 0.05 level, evaluated by Student's t-test using 20 years of data.



951

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

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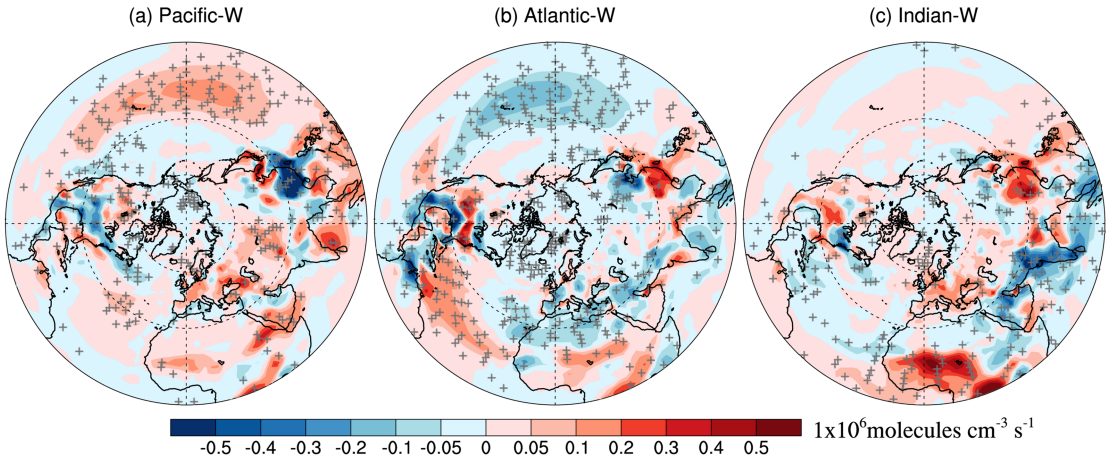

961

962 **Figure 3**. Perturbations of the surface net O_3 production rate ($1x10^6$ molecules cm^{-3} s^{-1}) for (a) Pacific-W, (b) Atlantic-W, and (c) Indian-W relative to the CTRL in the boreal
963
964 summer. The + symbols denote areas where the results are significant at the 0.05 level,
965 evaluated by Student's t-test using 20 years of data.

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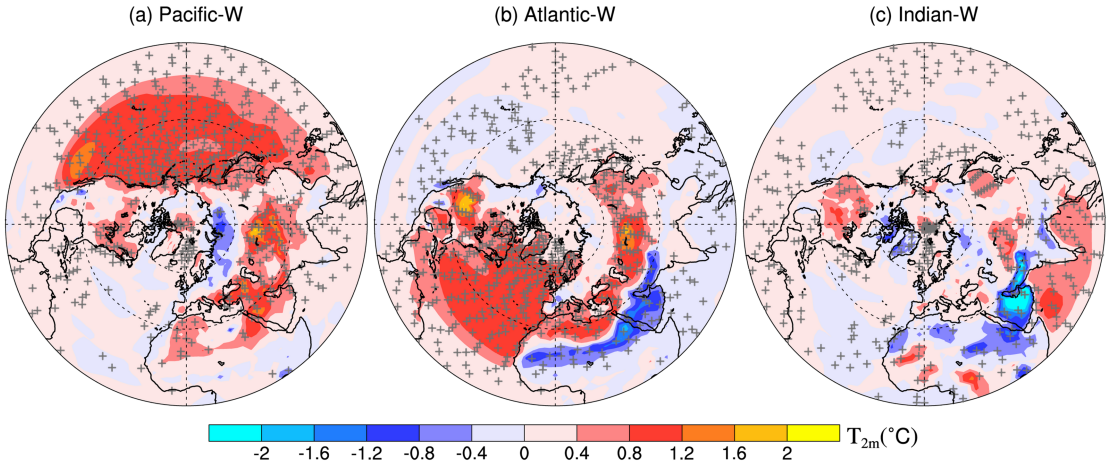

967

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

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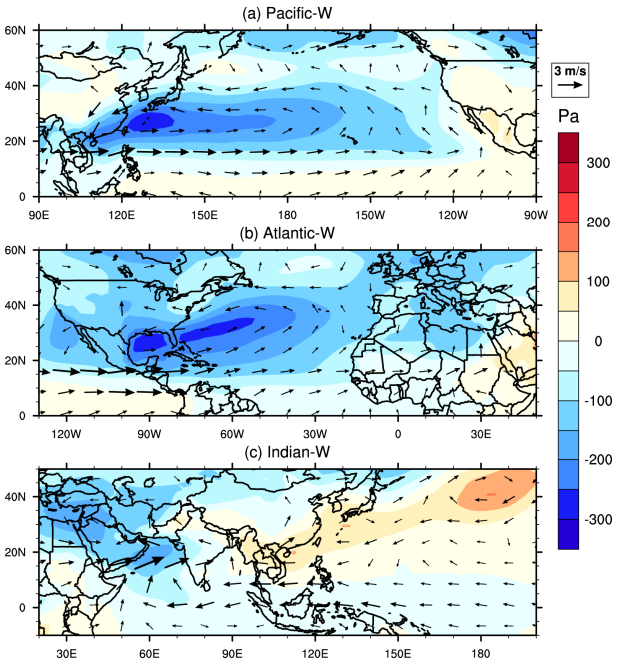

974

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

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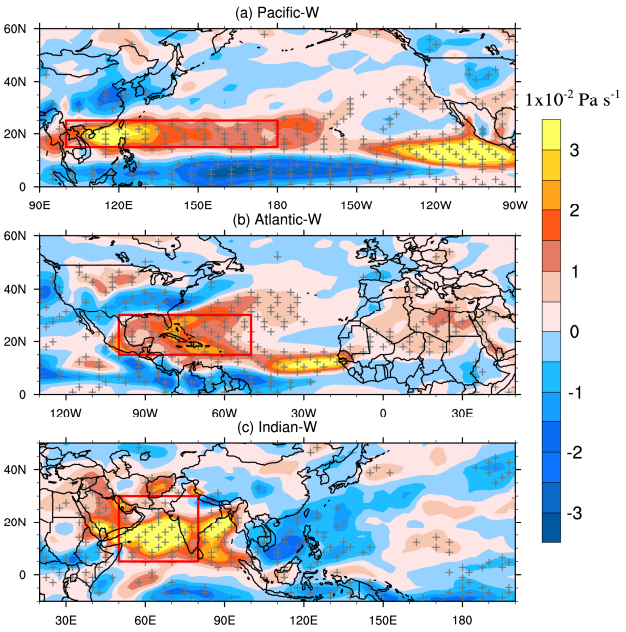

979

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

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

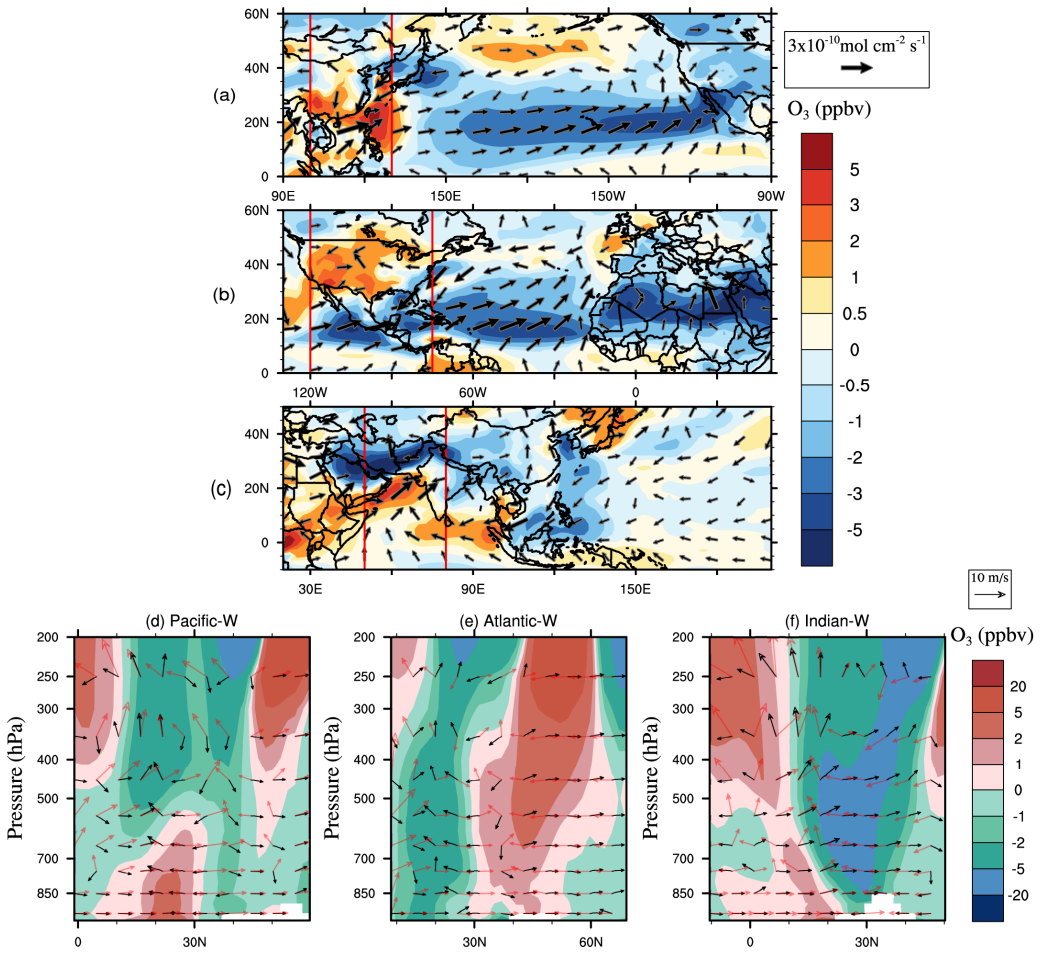

Figure 7. Top three rows: Changes in O_3 concentrations (color contours, ppbv) and horizontal fluxes (arrows, mol cm^{-2} s^{-1}) at the surface level for (a) Pacific-W, (b) Atlantic-W, (c) Indian-W relative to the CTRL in the boreal summer. Last row: zonal average of the tropospheric O_3 changes (color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

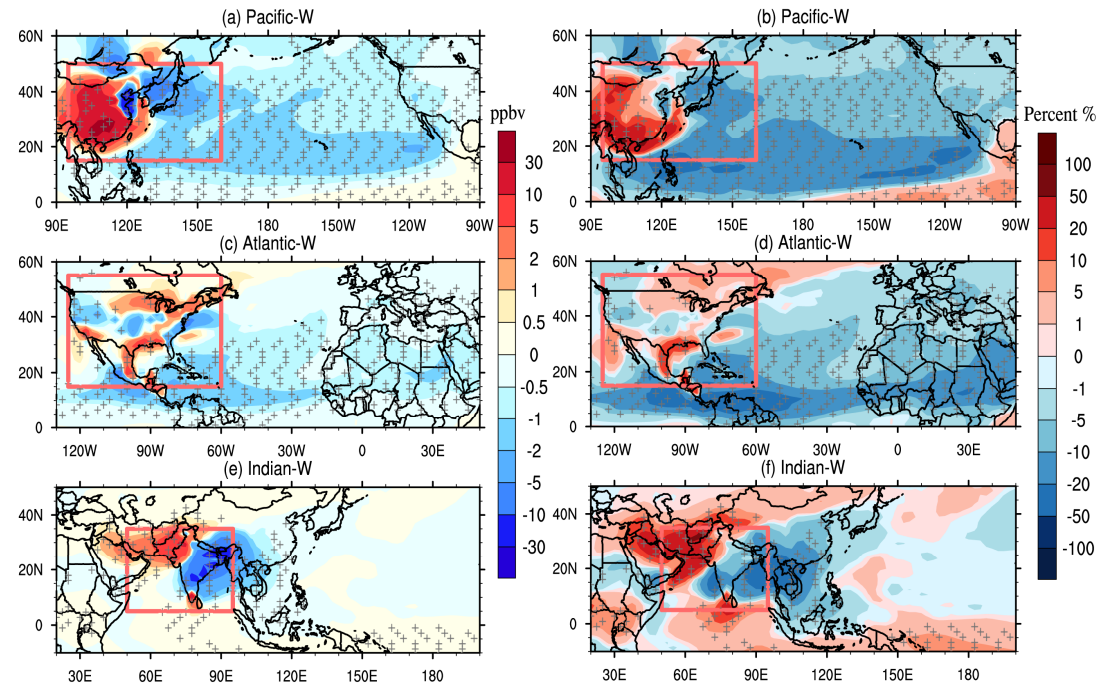

Figure 8. Left-hand panel: Difference in the surface concentration (ppbv) of a CO-like tracer emitted from (a) East Asia for Pacific-W, (c) North America for Atlantic-W and (e) the South Asia for Indian-W relative to the CTRL in the boreal summer. Right-hand panel: The percentage changes in the surface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

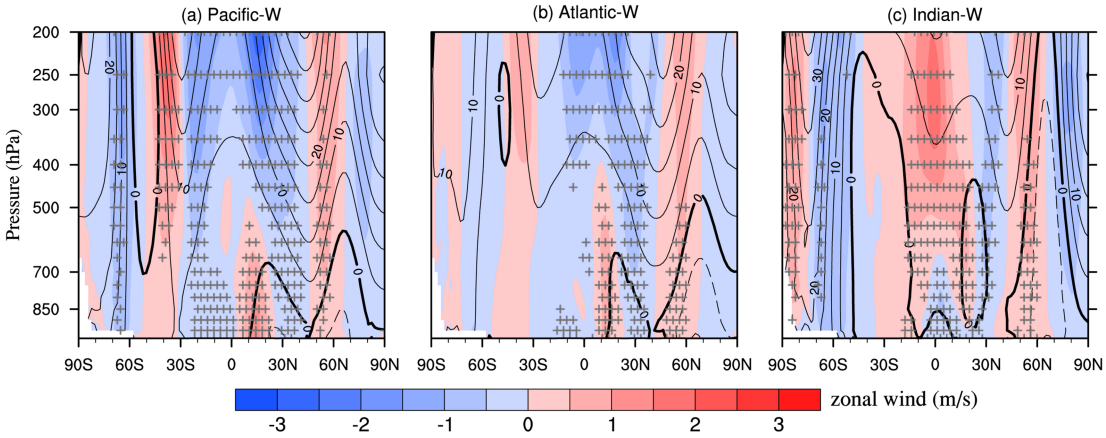

Figure 9. Zonally averaged changes in zonal wind (color contour, m/s) and geopotential

1012 height (contour, m) for (a) Pacific-W, (b) Atlantic-W and (c) Indian-W relative to the
1013 CTRL in the boreal summer. Black solid and dashed lines in the contours indicate
1014 positive and negative geopotential height anomalies, respectively (contour interval: 5
1015 m). The + symbol denotes areas where the zonal wind changes are significant at the
1016 0.05 level, evaluated by Student's t-test using 20 years of data.