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3 **The Relationship between Lower Stratospheric Ozone in the**

4 **Southern High Latitude and Sea Surface Temperature in the**

5 **East Asia Marginal Seas**

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1 **Abstract**

2 Using satellite observations, reanalysis data, and model simulations, this study
3 investigates the effect of sea surface temperatures (SST) on interannual variations of
4 lower stratospheric ozone in the southern high latitude. It is found that the SST
5 variations across the East Asian marginal seas (5°S–35°N, 100°E–140°E) rather than
6 the tropical eastern Pacific Ocean, where ENSO occurs, have the most significant
7 correlation with the southern high latitude lower stratospheric ozone changes. Further
8 analysis reveals that planetary waves originating over the marginal seas can be
9 propagated to southern middle to high latitudes via two teleconnection pathways in
10 summer and one pathway in autumn. The anomalous propagation and dissipation of
11 ultra-long Rossby waves in the stratosphere strengthen/cool (weaken/warm) the
12 southern polar vortex which produces more (less) active chlorine and enhances
13 (suppresses) ozone depletion in the southern high latitude stratosphere on one hand,
14 and impedes (favors) the transport of ozone from the southern middle latitude
15 stratosphere to high latitude on the other. The model simulations also reveal that
16 approximately 17% of the decreasing trend in the southern high latitude lower
17 stratospheric ozone observed over the past five decades can be attributed to the
18 increasing trend in SST over the East Asian marginal seas.



1 1. Introduction

2 Ozone variations over recent decades exhibit not only a strong decreasing trend,
3 forced by changes in ozone-depleting substances superimposed on a changing climate,
4 but also interannual variability influenced by various external and internal climate
5 forcings (e.g. Manney et al. 1994; Müller et al., 1994, 2005; Weiss et al., 2001;
6 Hadjinicolaou et al., 2002; Tian and Chipperfield, 2005; Austin et al., 2006, 2010;
7 Eyring et al., 2010; Liu et al., 2011, 2013; Douglass et al., 2014). Ozone variations
8 can change the amount of harmful solar ultraviolet rays reaching the Earth's surface
9 (Kerr and McElroy, 1993) and even influence climate (Forster and Shine, 1997;
10 Thompson et al., 2011; Li et al., 2016; Xie et al., 2016). Therefore, clarifying the
11 processes that are responsible for ozone variability is crucial for understanding how
12 global climate interacts with ozone variations (Austin et al., 2006; Hess and Lamarque,
13 2007; Frossard et al., 2013; Rieder et al., 2013). Many previous studies have analyzed
14 the ozone variability caused by external processes such as volcanic aerosols (e.g.
15 Hofmann and Oltmans, 1993; Rozanov et al., 2002; Dhomse et al., 2015) and the
16 solar cycle (e.g. Chandra and McPeters, 1994; Rozanov et al., 2005; Dhomse et al.,
17 2016) and these studies showed that volcanic aerosols and solar variations can result
18 in considerable short- and long-term variations in ozone levels. Ozone variations can
19 also be caused by changes in the surface climate (Zhang et al., 2014). Other studies
20 have reported the effects of internal climate variability on ozone, including El Niño–
21 Southern Oscillation (ENSO; Ziemke and Chandra, 1999; Cagnazzo et al., 2009;
22 Randel et al., 2009; Xie et al., 2014a, 2014b; Zhang et al., 2015a, 2015b), Madden–
23 Julian Oscillation (MJO; Fujiwara et al., 1998; Tian et al., 2007; Liu et al., 2009;
24 Weare, 2010; Li et al., 2012), Arctic Oscillation (AO) or North Atlantic Oscillation
25 (NAO; Schnadt and Dameris, 2003; Lamarque and Hess, 2004; Creilson et al., 2005;



1 Steinbrecht et al., 2011), and Quasi-Biennial Oscillation (QBO; Bowman, 1989; Tung
2 and Yang, 1994; Dhomse, 2006; Li and Tung, 2014). These studies indicate that ozone
3 over different regions shows different variability due to the location-specific nature of
4 the processes that influence this variability.

5 The stratospheric ozone hole (Farman et al., 1985) over the Antarctic has been
6 shown to have an important impact on the Southern Hemisphere climate (Shindell and
7 Schmidt, 2004; Son et al., 2008, 2009, 2010, Perlwitz et al., 2008; Feldstein, 2011,
8 Kang et al., 2011, Polvani et al., 2011; Thompson et al., 2011; Cagnazzo et al., 2013;
9 Keeble et al., 2014; Previdi and Polvani, 2014). Although the principal mechanisms
10 responsible for the formation of the ozone hole are well understood (e.g., Solomon,
11 1990, 1999; Ravishankara et al., 1994, 2009), the factors or processes that generate
12 interannual variations in ozone levels in the southern high latitude stratosphere remain
13 under debate. Among various factors, the QBO has been reported to have a significant
14 impact on the interannual variations of the Antarctic ozone (Garcia and Solomon,
15 1987; Lait et al., 1989; Mancini et al., 1991; Gray and Ruth, 1993; Bodeker and
16 Scourfield, 1995; Shindell et al., 1997a). The September to March levels of ozone
17 over the Antarctic is also marginally correlated with the wintertime mean eddy heat
18 flux (Weber et al., 2003). Heat transport induced by upward propagating planetary
19 waves warms the polar vortex (Schoeberl and Hartmann, 1991), which reduces the
20 occurrence of polar stratospheric clouds (PSCs), a key prerequisite for the
21 heterogeneous chemistry that depletes Antarctic ozone. Subsequent efforts to
22 understand Antarctic ozone variations during individual years have considered
23 planetary wave activity, which account for much of the interannual variations of
24 ozone levels over the Northern Hemisphere (Hadjinicolaou et al., 1997; Fusco and
25 Salby, 1999; Salby and Callaghan, 2004, 2007a, 2007b; Hadjinicolaou and Pyle,



1 2004). Studies based on measurements (Bodeker and Scourfield, 1995), modeling
2 (Shindell et al., 1997a, 1997b), and reanalysis data (Huck et al., 2005) have shown
3 that interannual differences in the severity of Antarctic ozone depletion are
4 anti-correlated with Southern Hemisphere planetary wave activity. However, the
5 source of the planetary wave activity that modulates interannual variability in
6 southern high latitude stratospheric ozone is still not well understood.

7 Variations in tropical sea surface temperatures (SST) associated with El
8 Niño-Southern Oscillation (ENSO), are an important factor in the modulation of the
9 planetary wave activity in the Northern Hemisphere that affects the interannual
10 variability of temperature and ozone levels in the northern polar stratosphere (Sassi et
11 al., 2004; Manzini et al., 2006; Calvo et al., 2004, 2009; Cagnazzo et al., 2009; Hu
12 and Pan, 2009; Hurwitz et al., 2011a, b; Ren et al., 2010; Zubiaurre and Calvo, 2012;
13 Xie et al., 2012; Yao et al., 2015). The long-term trend in tropical SST also has
14 corresponding to the trend of temperature in the southern polar stratosphere (Grassi et
15 al., 2005, 2006; Hu and Fu, 2009; Li et al., 2010; Clem et al., 2016). Although ENSO
16 is reported to cause circulation and temperature anomalies in the southern high
17 latitude stratosphere, the interannual variability of the southern polar vortex and ozone
18 levels over the past three decades cannot be explained by ENSO variations alone
19 (Angell, 1988, 1990; Hurwitz et al., 2011a, 2011b; Lin et al., 2012; Wilson et al., 2014;
20 Evtushevsky et al., 2015; Yu et al., 2015; Yang et al., 2015; Welhouse et al., 2016).

21 Over recent decades, SST in the East Asian marginal seas has followed an
22 increasing trend with strong interannual variations (Zheng et al., 2014). Zhao et al.
23 (2015, 2016) pointed out that Rossby waves generated by variations in the SST of the
24 South China Sea can cross the equator and be propagated towards to southern middle
25 to high latitudes. It is likely that the Rossby waves generated by SST changes in the



1 vicinity of the East Asian marginal seas can cross the equator to the Southern
2 Hemisphere and regulate ozone levels in the southern high latitude stratosphere via
3 their influence on the southern stratospheric circulation. Therefore, it is worthwhile to
4 examine the potential connections between SST variations over the East Asian
5 marginal seas and southern high latitude lower stratospheric ozone variations. The
6 remainder of the paper is organized as follows. The data, method and model used are
7 introduced and briefly described in section 2. Section 3 analyzes the connection
8 between the East Asian marginal seas and southern high latitude lower stratospheric
9 ozone. Section 4 presents and discusses the simulations of the connection. Finally, the
10 results are summarized and conclusions drawn in section 5.

11

12 **2. Data, Model, and Methods**

13 The ozone data used in this study is obtained from the NASA Modern Era
14 Retrospective Analysis for Research and Applications (MERRA) dataset version 2
15 (Rienecker et al., 2011) and TOMCAT/SLIMCAT 3-D model simulations
16 (Chipperfield, 2006). The MERRA2 data uses 42 pressure levels from the surface up
17 to 0.1 hPa. The vertical resolution of MERRA2 is ~1–2 km in the UTLS and 2–4 km
18 in the middle and upper stratosphere. MERRA2 is assimilated by the Goddard Earth
19 Observing System Model, Version 5 (GEOS-5) with ozone from the Solar
20 Backscattered Ultra Violet (SBUV) radiometers from October 1978 to October 2004,
21 and thereafter from the Ozone Monitoring Instrument (OMI) and AURA Microwave
22 Limb Sounder (MLS) (Bosilovich et al., 2015). The MERRA2 reanalysis ozone data
23 compares well with satellite ozone observations (Rieder et al., 2014; Zhang et al.,
24 2015b) and shows a better representation of the QBO and stratospheric ozone
25 compared to MERRA1 (Coy et al., 2016). In the present study, the ozone data from a



1 3D offline chemical transport model, SLIMCAT (Feng et al., 2007, 2011), is also used.
2 The simulation performed in this study is driven by horizontal winds and temperatures
3 from meteorological analyses of the ERA-Interim data provided by European Centre
4 for Medium-Range Weather Forecasts (ECMWF) (Dee et al., 2011). The vertical
5 advection in the model is calculated from the divergence of the horizontal mass flux
6 (Chipperfield, 2006), and chemical tracers are advected by the conservation of
7 second-order moments (Prather, 1986). Figure 1 shows the ozone variations over the
8 region 200–50 hPa and 60–90 °S, where the variability and depletion of ozone
9 concentration is most pronounced in the Southern Hemisphere in the past five decades
10 (Austin and Wilson, 2006; Solomon 1990, 1999; Ravishankara et al., 1994, 2009),
11 from the two datasets. The ozone variations from MERRA2 are in good agreement
12 with those from SLIMCAT (Fig. 1a), and the difference between the two kinds of
13 ozone data is small (Fig. 1b).

14 SST data is obtained from HadISST dataset compiled by the UK Met Office
15 Hadley Centre for Climate Prediction and Research (Rayner et al., 2003).
16 Geopotential height, zonal wind, and temperature data are obtained from the ECMWF
17 ERA-Interim dataset.

18 We also use version 4 of the Whole Atmosphere Community Climate Model
19 (WACCM4) in this study since WACCM has been shown to simulate well the
20 stratospheric circulation, temperature and ozone variations (Garcia et al. 2007).
21 WACCM4 is part of the Community Earth System Model (CESM) framework
22 developed by the National Center for Atmospheric Research (NCAR). WACCM4 uses
23 a finite-volume dynamical core, with 66 vertical levels extending from the ground to
24 4.5×10^{-6} hPa (145 km geometric altitude), and a vertical resolution of 1.1–1.4 km in
25 the tropical tropopause layer and the lower stratosphere (below a height of 30 km).



1 The simulations presented in this paper are performed at a horizontal resolution of
 2 $1.9^\circ \times 2.5^\circ$ and with interactive chemistry (Garcia et al., 2007). More details
 3 regarding WACCM4 are provided in Marsh et al. (2013).

4 We calculate the statistical significance of the correlation between two
 5 auto-correlated time series using the two-tailed Student's t -test and the effective
 6 number (N^{eff}) of degrees of freedom (DOF; Bretherton et al. 1999). For this study, N^{eff}
 7 is determined using the following approximation (Li et al. 2012):

$$8 \quad \frac{1}{N^{\text{eff}}} \approx \frac{1}{N} + \frac{2}{N} \sum_{j=1}^N \frac{N-j}{N} \rho_{XX}(j) \rho_{YY}(j)$$

9 where N is the sample size, and ρ_{XX} and ρ_{YY} are the autocorrelations of two sampled
 10 time series, X and Y , respectively, at time lag j .

11 We use the formulae given by Andrews et al. (1987) to calculate the
 12 quasi-geostrophic 2D Eliassen–Palm (E–P) flux. The meridional (F_y) and vertical (F_z)
 13 components of the E–P flux, and the E–P flux divergence D_F , are expressed as:

$$14 \quad F_y = -\rho_0 a \cos \varphi \overline{\varphi u' v'}$$

$$15 \quad F_z = -\rho_0 a \cos \varphi \frac{Rf}{HN^2} \overline{v' T'}$$

$$16 \quad D_F = \frac{\nabla \cdot F}{\rho_0 a \cos \varphi} = \frac{\partial(F_y \cos \varphi) / a \cos \varphi \partial \varphi + \partial F_z / \partial z}{\rho_0 a \cos \varphi},$$

17 where ρ_0 is the air density, a is the radius of the Earth, R is the gas constant, f
 18 is the Coriolis parameter, H is the atmospheric scale height (7 km), u and v are
 19 the zonal and meridional wind components, respectively, and T is the temperature;
 20 the overbar denotes the zonal mean, and the prime symbol denotes departures from
 21 the zonal mean.

22



1 **3. The connection between the East Asian marginal seas and southern high**
2 **latitude lower stratospheric ozone**

3 Figure 2a shows the correlation coefficients between SST and southern high latitude
4 lower stratospheric ozone variations between 1979 and 2015 using ozone data from
5 the MERRA2 dataset and SST from HadISST dataset. Ozone data from SLIMCAT
6 simulations was further used to confirm the correlation coefficients (Fig. 2b). The
7 regions of significant correlation are generally different for the two ozone datasets
8 except for the East Asian marginal seas; i.e., 5°S–35°N, 100°E–140°E, where the
9 most significant correlations between Antarctic stratospheric ozone variations and
10 SST are seen in both datasets. Figure 2 implies an interannual connection between
11 SST in the East Asian marginal seas and southern high latitude lower stratospheric
12 ozone variations. Figure 2 also reveals that SST variations associated with ENSO are
13 not the main factor controlling the interannual variability of southern high latitude
14 lower stratospheric ozone.

15 Through the interannual connection in Fig. 2 possibly caused by the influence of
16 lower latitude SST on the south high latitude stratosphere, south high latitude
17 stratospheric ozone has also been shown to affect tropical climate (Son et al., 2008;
18 Kang et al., 2011; Thompson et al., 2011). Thus, it is first necessary to confirm the
19 causality of this connection. To investigate the SST variations across the marginal
20 seas of East Asia, we first define an SST index over the region with the most
21 significant correlations in Fig. 2, i.e., the ST_MSEA index. This index is a time series
22 that represents SST variations across the marginal seas of East Asia (Figure 3a). It is
23 calculated by averaging the SST variations in the region from 5°S–35°N at 100°E–
24 140°E, and then removing the seasonal cycle and linear trend. Fig. 3b and c show the
25 composite warm and cold SST anomalies for the events that occurred in the marginal



1 seas of East Asia between 1979 and 2015 (see Table 1).

2 The ST_MSEA index and southern high latitude lower stratospheric ozone
3 variations show a significant simultaneous correlation (Fig. 4). This implies that SST
4 variations in the marginal seas of East Asia have an impact on southern high latitude
5 lower stratospheric ozone, since there is a lag of several months associated with the
6 effect of southern high latitude lower stratospheric ozone on the tropical climate (Son
7 et al., 2008; Kang et al., 2011; Thompson et al., 2011).

8 It is well known that the SST changes in the eastern Pacific, the Indo-Pacific
9 warm pool, and the Atlantic can significantly influence the northern polar stratosphere
10 (Calvo et al., 2004, 2009; Hoerling et al., 2001, 2004; Cagnazzo et al., 2009; Hu and
11 Fu, 2009; Hu and Pan, 2009; Li et al., 2010; Hurwitz et al., 2011a, b; Lin et al., 2012;
12 Zubiaurre and Calvo, 2012; Xie et al., 2012; Li and Chen, 2014). SST variations in
13 some regions can excite Rossby wave trains and those waves can propagate into
14 northern middle and high latitude stratosphere (Gettelman et al., 2001; Sassi et al.,
15 2004; Manzini et al., 2006; Garc ía-Herrera et al., 2006; Taguchi and Hartmann, 2006;
16 Garfinkel and Hartmann, 2007, 2008; Free and Seidel, 2009). The mechanism that
17 allows SST variations in the East Asian marginal seas to affect the southern high
18 latitude stratosphere is also possibly related to tropospheric wave propagation from
19 northern lower latitude to southern middle and high latitudes.

20 Figure 5 shows the ray paths of waves generated by the SST anomalies over the
21 region 5°S–35°N, 100°E–140°E, at 300 hPa in four seasons. The wavenumbers along
22 these rays are between 1 and 5. The wave ray paths represent the climate
23 teleconnections; i.e., the propagation of stationary waves in realistic flows. The
24 calculation of the wave ray paths and application of the barotropic model is described
25 in detail by Li et al. (2015) and Zhao et al. (2015). We found that the Rossby waves



1 generated by SST anomalies in the marginal seas of East Asia could indeed propagate
2 to the middle to high latitudes of the Southern Hemisphere in summer and autumn
3 (Fig. 5b and c), but not in spring and winter (Fig. 5a and d) because the Rossby waves
4 motivated by the low-latitude SST anomalies move mostly northwards in spring and
5 winter. Meanwhile, we must note that the propagating paths of those waves in
6 summer and autumn are different (Fig. 5b and c). In summer, the first path of rays
7 originates over the marginal seas of East Asia, crosses the Indian Ocean to arrive over
8 tropical Africa or even South America, and then reflects equatorward to the middle to
9 high latitudes of the Southern Hemisphere. The second path of rays originates over
10 the marginal seas of East Asia reflects directly into the southern Indian Ocean and
11 reaches the Southern Hemisphere. In autumn, the first path disappears, and only the
12 rays that follow the second path reach the Southern Hemisphere. In addition, the ray
13 stops at about 60°S, which possibly implies an upward propagation of the wave at this
14 location.

15 The correlation coefficients between the ST_MSEA index and 300-hPa
16 geopotential height variations from the ERA-Interim reanalysis across the four
17 seasons are shown in Figure 6. The positive and negative centers of correlation
18 coefficients represent the teleconnection patterns. The teleconnection patterns in
19 summer and autumn (Fig. 6b and c) are in good agreement with the ray paths (Fig. 5b
20 and c). In summer, two clear wave train paths appear over the marginal seas of East
21 Asia with one moving westwards to South America and reflecting to the middle to
22 high latitudes of the Southern Hemisphere, and the second reflecting directly into the
23 Southern Hemisphere (Fig. 6b). In autumn, the first path is very distinct (Fig. 6c), i.e.,
24 the negative correlation coefficient over the Indian Ocean is small, which suggests
25 that most of the waves do not propagate westwards. The second path also remains



1 evident. These two teleconnection pathways of the wave trains in summer and autumn
2 (Figs. 5 and 6) are discussed in detail by Zhao et al., (2016), who refer to them as the
3 North Australia–Southern Hemisphere and South Africa–Southern Hemisphere
4 pathways, respectively. In spring and winter, the above two teleconnection patterns
5 don't exist (Fig. 6a and d).

6 Figure 7a shows the correlation coefficients between the ST_MSEA index and
7 stratospheric ozone variations, which indicate that warm (cold) SST anomalies over
8 the East Asian marginal seas are associated with a decrease (increase) in southern high
9 latitude lower stratospheric ozone. Bodeker and Scourfield (1995), Shindell et al.
10 (1997a, 1997b), and Huck et al. (2005) have shown that interannual differences in the
11 severity of southern high latitude lower stratospheric ozone depletion are related to
12 Southern Hemisphere planetary wave activity. All of the above analysis illustrates that
13 the SST anomalies over the marginal seas of East Asia are a possible main source of
14 this planetary wave activity.

15 Figure 7b shows that ST_MSEA is positively correlated with zonal wind around
16 60°S, where is the boundary of the southern polar vortex in summer and autumn,
17 while Figs. 7c indicate that ST_MSEA is negatively correlated with temperature. The
18 correlations shown in Figs 3, 5, 6, and 7 can be used to establish a hypothesis of
19 chemical process for the connection between SST variations over the marginal seas of
20 East Asia and southern high latitude lower stratospheric ozone as follows: 1. The
21 warm (cold) SST anomalies over the marginal seas (Fig. 3) depress (enhance)
22 planetary wave activity in the middle to high latitudes of the Southern Hemisphere
23 (Figs 5 and 6). 2. The anomalous propagation of planetary waves into the stratosphere
24 and dissipation of ultra-long Rossby waves in the stratosphere strengthen/cool
25 (weaken/warm) the southern polar vortex (Fig. 7b and c). 3. A cooler (warmer) polar



1 vortex allows more (less) PSCs and active chlorine to form. 4. Consequently, southern
2 high latitude lower stratospheric ozone decreases (increases) (Fig. 7a).

3 However, it needs to point out that Antarctic polar vortex temperature is deeply
4 below the threshold for heterogeneous chemistry, so that a warming (cooling) in the
5 center of Antarctic polar vortex will have very little impact on Antarctic ozone by
6 affecting heterogeneous chemistry (Tilmes et al. 2006; Kirner et al. 2015). It seems to
7 challenge the above hypothesis.

8 Fig. 7c shows that the center of the correlation confidences locates near 60 °S. It
9 means that the center of stratospheric temperature changes caused by SST changes in
10 the East Asia Marginal Seas locates near 60 °S but not near 90 °S. Temperature change
11 near 60 °S maybe more effectively affects southern high latitude lower stratospheric
12 ozone than that near 90 °S since the background temperature in the lower stratosphere
13 near 60 °S would be higher than that near 90 °S. The chemical process maybe has a
14 certain contribution on the southern high latitude lower stratospheric ozone changes
15 caused by SST changes in the East Asia Marginal Seas.

16 We also found that the SST changes in the East Asia Marginal Seas are positively
17 correlated with southern high latitude stratospheric meridional wind (Fig. 7d),
18 suggesting a stronger (weaker) zonal circulation (Fig. 7b) related to the SST changes
19 impeding (promoting) transport of ozone from the middle latitude stratosphere to high
20 latitude stratosphere. Note that this correlation is the strongest in autumn but not in
21 summer when the south polar vortex is too stable that doesn't allow ozone rich air
22 into the vortex. Fig. 7d implies a dynamical contribution on the southern high latitude
23 lower stratospheric ozone changes caused by SST changes in the East Asia Marginal
24 Seas.

25 It is noteworthy that warm (cold) SST anomalies are generally thought to



1 increase (suppress) planetary wave activity via strengthening (weakening) convection
2 (Xie et al., 2008; Shu et al., 2010; Hu et al., 2014). However, this study shows that
3 warm (cold) SST anomalies over the marginal seas of East Asia suppress (increase)
4 planetary wave activity. This may be the warm (cold) SST anomalies over the
5 marginal seas in summer and autumn are equal to weaken (enhance) sea–land contrast
6 along the coastline of East Asia. This results in weaker (stronger) convection, which
7 suppresses (increases) planetary wave activity.

8

9 **4. Simulating the effect of SST changes in the marginal seas of East Asia on** 10 **southern high latitude lower stratospheric ozone**

11 We performed three time-slice simulations with WACCM4 to validate the mechanism
12 described in Section 3. The monthly mean climatologies of surface emissions used in
13 the model were obtained from the A1B emissions scenario developed by the
14 Intergovernmental Panel on Climate Change (IPCC), and averaged over the period
15 1979–2015. QBO signals with a 28-month fixed cycle are included in WACCM4 as
16 an external forcing for zonal wind. The SST forcing used in the first time-slice
17 experiment (S1, the control experiment) was the 12-month climatology cycle
18 averaged over the period 1979–2015 and based on the HadISST dataset. S2 was a
19 sensitivity experiment and was the same as S1 except that warm anomalies (as in Fig.
20 3b) were added to the SST in the marginal seas of East Asia (5 °S–35 °N and 100–
21 140 °E). The third experiment, S3, was the same as S2, but with cold SST anomalies
22 (as in Fig. 3c). Detailed descriptions of experiments S1–S3 are provided in Table 2.

23 Figure 8 first shows the southern high latitude lower stratospheric ozone
24 anomalies forced by warm and cold SST anomalies over the marginal seas of East
25 Asia. It can be seen that the warm SST anomalies indeed cause ozone decrease in the



1 southern high latitude lower stratosphere (Fig. 8a) and cold SST anomalies results in
2 ozone increase (Fig. 8b). The simulations support the results shown from the
3 statistical analysis in Section 3.

4 Figure 9 shows the E–P flux vectors and divergence anomalies in the
5 stratosphere caused by SST anomalies over the marginal seas of East Asia. Analysis
6 of changes in the E–P flux (Eliassen and Palm 1961; Andrews et al. 1987) is often
7 used as a diagnostic for planetary wave propagation from the troposphere to the
8 stratosphere (Edmon et al., 1980). During periods of warm (cold) SST over the
9 marginal seas of East Asia, a decrease (increase) in upward wave flux entering the
10 stratosphere accompanied by stronger (weaker) convergence of the E–P flux in the
11 stratosphere at middle to high latitudes of the Southern Hemispheres (ca. 60°S) is
12 evident (Fig. 9a and c). The anomalous wave flux entering the stratosphere around
13 60°S confirms the result in Fig. 5, which shows that the wave rays terminate at about
14 60°S.

15 Many previous studies have demonstrated a strongly negative correlation
16 between upward propagating wave activity and the intensity of the stratospheric polar
17 vortex, with an anomalously negative and positive upward wave flux alongside a
18 stronger and weaker polar vortex, respectively (Christiansen 2001; Polvani and
19 Waugh 2004; Li and Lau 2013). During periods of warm (cold) SST over the marginal
20 seas of East Asia, the anomalous downward (upward) E–P flux, and larger (smaller)
21 E–P flux divergence at middle to high latitudes (ca. 60°S) in the Southern Hemisphere
22 (Fig. 9a and c), imply suppressed (active) wave activity in the stratosphere, which
23 induces a strengthened (weakened) circulation at southern polar vortex edge (Fig. 9b
24 and d). Finally, the cold (warm) polar vortex (Fig. 10a and c) allows more (less)



1 PSCs/active chlorine (Fig. 10b and d) to form. This is one process through which SST
2 variations over the marginal seas of East Asia causes southern high latitude lower
3 stratospheric ozone changes. The other process is that the strengthened (weakened)
4 southern polar vortex impedes (promotes) air exchange between middle and high
5 latitude stratosphere (Figure 11), and further decreases (increases) southern high
6 latitude lower stratospheric ozone levels.

7 As a result of human activity, the amount of Antarctic stratospheric ozone has
8 decreased remarkably over recent decades (Solomon 1990, 1999; Ravishankara et al.,
9 1994, 2009). At the same time, SST over the marginal seas of East Asia has followed
10 an increasing trend, but superimposed on strong interannual variations (Zheng et al.,
11 2014). Figure 12 shows the correlation coefficients between southern high latitude
12 lower stratospheric ozone and SST in which the SST and southern high latitude lower
13 stratospheric ozone variations have not been detrended as that in Fig. 2. Comparing
14 Fig. 12 with Fig. 2, we can see that the negative correlation coefficients over the
15 marginal seas of East Asia become larger in Fig. 12, implying a contribution of
16 warmer SST in the marginal seas of East Asia to decline trend of southern high
17 latitude lower stratospheric ozone.

18 We used ensemble transient experiments to estimate the contribution of SST
19 variations in the marginal seas of East Asia to southern high latitude lower
20 stratospheric ozone changes. The transient experiments incorporated the following
21 natural and anthropogenic external forcings for the period 1955–2005: observed SST
22 from the HadISST dataset, surface emissions from the IPCC A1B emissions scenario,
23 spectrally resolved solar variability (Lean et al., 2005), volcanic aerosols (from the
24 Stratospheric Processes and their Role in Climate (SPARC) Chemistry–Climate
25 Model Validation (CCMVal) REF-B2 scenario recommendations), and nudged QBO



1 (the time series in CESM is determined from the observed climatology). The first
2 transient experiment, T1, was the historical experiment covering the period 1955–
3 2005 (Marsh et al., 2013). The second transient experiment, T2, was the same as T1
4 except that the SST in the marginal seas of East Asia (5°S–35°N and 100–140°E) for
5 the period 1955–2005 was replaced by the 12-month cycle of climatology averaged
6 over the same period. This means that in T2, the SST over the marginal seas of East
7 Asia had only a seasonal cycle, but no trend and no interannual variability. T3 was the
8 same as T2, but used a slightly different initial condition as an ensemble experiment.
9 Detailed descriptions of runs T1–T3 are provided in Table 3.

10 Figure 13a and b shows the southern high latitude lower stratospheric ozone
11 variations over the period 1955–2005 from T1 and the ensemble experiments
12 $((T2+T3)/2)$. The southern high latitude lower stratospheric ozone variations caused
13 by the SST variability over the marginal seas of East Asia can be obtained by
14 subtracting simulated ozone in the ensemble experiments $((T2+T3)/2)$ from the
15 ozone in T1 (Fig. 13c). There are evident differences in southern high latitude lower
16 stratospheric ozone variations between T1 and the ensemble experiments
17 $((T2+T3)/2)$. This illustrates that the SST variability over the marginal seas of East
18 Asia (Fig. 13d) does have a significant effect on southern high latitude lower
19 stratospheric ozone over the past five decades (Fig. 13c). The correlation coefficient
20 between the two lines in Fig. 13c and d is 0.29 which is significant at 99% confident
21 level. A further analysis reveals that the linear trend of ozone variations over the
22 region 200–50 hPa and 60–90°S from T1 (Trend1, Fig. 3a) is -1.2×10^{-3} ppmv/month,
23 and from $(T1 - (T2+T3)/2)$ (Trend2, Fig. 3c) is -0.204×10^{-3} ppmv/month. See Table 4.
24 It implies that the increasing linear trend in SST over the marginal seas of East Asia
25 can contribute approximately 17% of the declining trend in southern high latitude



1 lower stratospheric ozone from 1955–2005 ($\text{Trend2} / \text{Trend1} \times 100\%$).

2

3 **5. Conclusions and Summary**

4 In this study, the connection between SST and the southern high latitude lower
5 stratospheric ozone variations at the interannual time scale is examined. We found that
6 SST over the marginal seas of East Asia can significantly modulate the interannual
7 variability of southern high latitude lower stratospheric ozone and the processes
8 involved in this modulation are related to anomalous planetary wave activity induced
9 by SST variations over the marginal seas of East Asia. The planetary waves
10 originating from the marginal seas can propagate to the middle and high latitudes of
11 the Southern Hemisphere in summer and autumn via the North Australia–Southern
12 Hemisphere and South Africa–Southern Hemisphere pathways. The anomalous
13 propagation and dissipation of ultra-long Rossby waves in the stratosphere
14 strengthens/cool (weakens/warms) the southern polar vortex, which allows more
15 (less) active chlorine to form and deplete more (less) ozone on one hand. On the other
16 hand, a stronger (weaker) polar vortex impedes (promotes) the transport of middle
17 latitude ozone to high latitudes and further decreases (increases) southern high
18 latitude lower stratospheric ozone. The above results and analysis are based on
19 observations but are also supported by time-slice experiments conducted using the
20 CESM.

21 Our transient model simulations further demonstrated that SST variations over
22 the marginal seas of East Asia not only modulate the interannual variability of
23 southern high latitude lower stratospheric ozone, but also contribute to southern high
24 latitude lower stratospheric ozone trend over the past five decades. Our analysis
25 reveals that the trend of increasing SST over the marginal seas of East Asia may have



1 contributed approximately 17% to the decreasing trend of southern high latitude lower
2 stratospheric ozone over the past five decades.

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9



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- 7 stratosphere, *J. Geophys. Res.*, 117, D04104, doi:10.1029/2011JD016690, 2012.



- 1 Table 1. Warm and cold SST events in the marginal seas of East Asia from 1979 to 2015 analyzed
 2 in this paper using the ST_MSEA index (Fig. 3a).

Warm Events*	Cold Events*
JUN1983–NOV1983	SEP1979–MAY1980
MAY1987–NOV1988	OCT1981–NOV1982
NOV1997–MAR2000	MAY1985–MAY1986
MAR2002–AUG2003	AUG1992–MAY1993
AUG2008–FEB2009	JUL2004–DEC2004
MAY2010–NOV2010	FEB2011–SEP2011
	MAY2012–APR2012
	NOV2014–SEP2015

- 3 *Following the definition of ENSO events (Trenberth 1997), we propose a threshold of ± 0.15 ,
 4 which is equal to the standard deviation of the ST_MSEA series, as the indicator of warm and cold
 5 events.



1 **Table 2.** Experiments S1–S3.

Experiments ^{*1}	Descriptions
S1	Time-slice run using case F_2000_WACCM in CESM. The SST is the 12-month cycle climatology mean for the period 1979–2015 based on HadISST dataset (Rayner et al., 2003); the monthly mean climatologies of surface emissions used in the model are obtained from the A1B emissions scenario developed by the IPCC, averaged over the period 1979–2015. QBO phase signals with a 28-month fixed cycle are included in WACCM4 as an external forcing for zonal wind.
S2	Same as S1, except that the SST in the marginal seas of East Asia (5°S–35°N and 100–140°E) adds warm SST anomalies (as Fig. 3b).
S3	Same as S1, except that the SST in the marginal seas of East Asia (5°S–35°N and 100–140°E) adds cold SST anomalies (as Fig. 3c).

2 ^{*1}Each experiment is run for 53 years, with the first 3 years excluded as a spin-up period. The
 3 remaining 50 years are used for the analysis.

1 **Table 3.** Experiments T1–T3.

Experiments ^{*1}	Descriptions
T1	Transient run using case F_1955-2005_WACCM_CN in CESM. SST forcing based on HadISST dataset, surface emissions are obtained from the A1B emissions scenario developed by the IPCC, spectrally resolved solar variability (Lean et al., 2005), volcanic aerosols (from the SPARC CCMVal REF-B2 scenario recommendations), nudged QBO (the time series in CESM is determined from the observed climatology).
T2	Same as T1, except that the SST in the marginal seas of East Asia (5°S–35°N and 100–140°E) between 1955 and 2005 is replaced by the 12 months cycle of climatology averaged for the period 1955–2005.
T3	Same as T2, but with slightly different initial condition ^{*2}

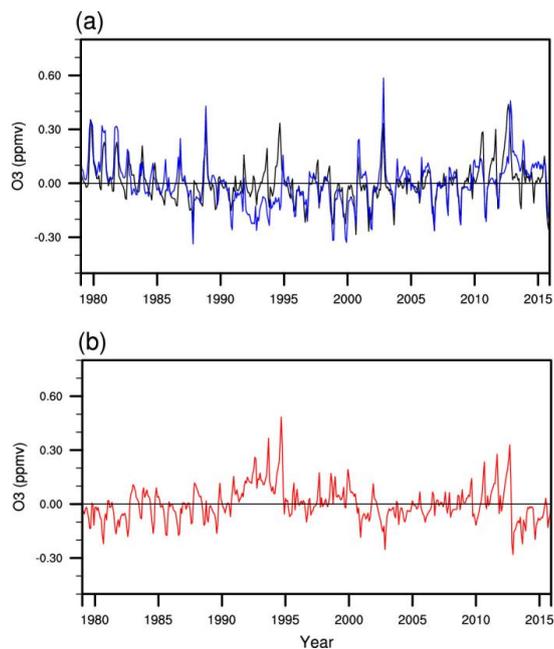
2 ^{*1}Integration period is 1955–2005 for T1–T3.

3 ^{*2}The parameter <pertlim> is used to produce different initial conditions in the CESM model,
 4 which produces an initial temperature perturbation. The magnitude was about e^{-14} .

5

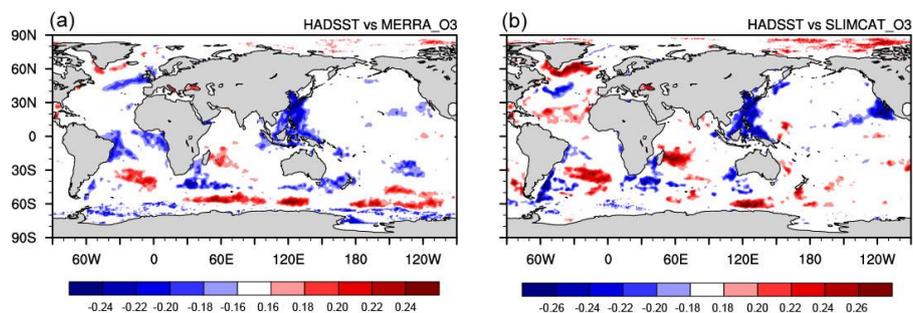
6 Table 4. Linear trends of ozone variations over the region 200–50 hPa and 60–90°S from
 7 experiments with (T1) and without SST (T2 +T3) variations in the East Asia Marginal Seas (T1–3
 8 see Table 3).

Experiments	Values
Linear trend of ozone variations over the region 200–50 hPa and 60–90°S from T1 (Trend1)	$-1.2 \times e^{-3}$ ppmv/month
Linear trend of ozone variations over the region 200–50 hPa and 60–90°S from (T1 – (T2+T3)/2) (Trend2)	$-0.204 \times e^{-3}$ ppmv/month

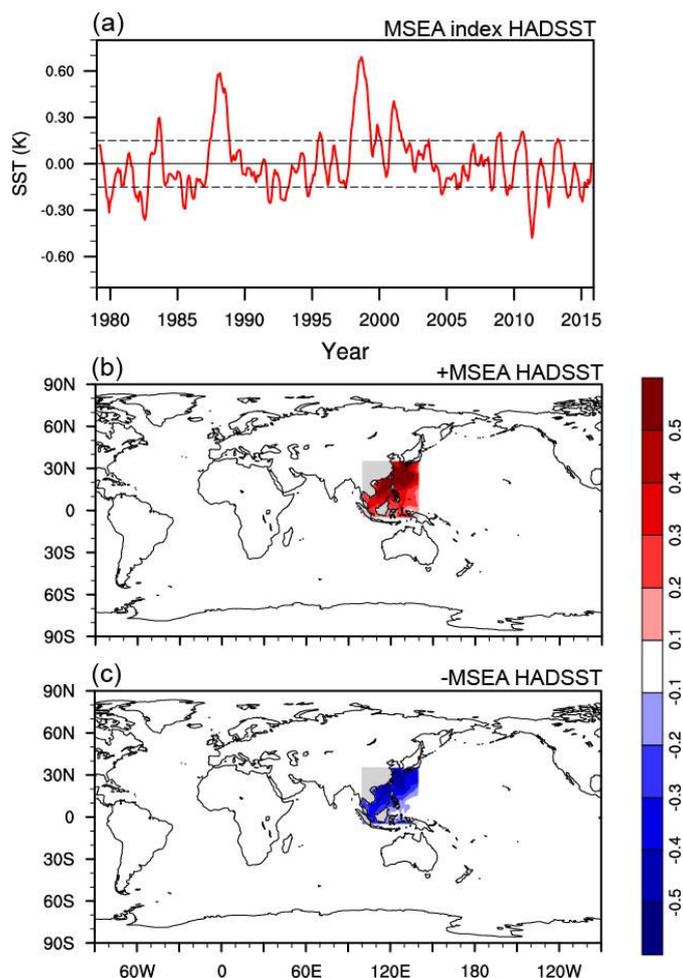


1

2 **Figure 1.** (a) Time series of southern high latitude lower stratospheric ozone variations averaged
3 over the region 60–90 S at 200–50 hPa from the MERRA2 (black line), and SLIMCAT monthly
4 ozone (blue line) datasets. (b) The difference between MERRA2 and SLIMCAT ozone.

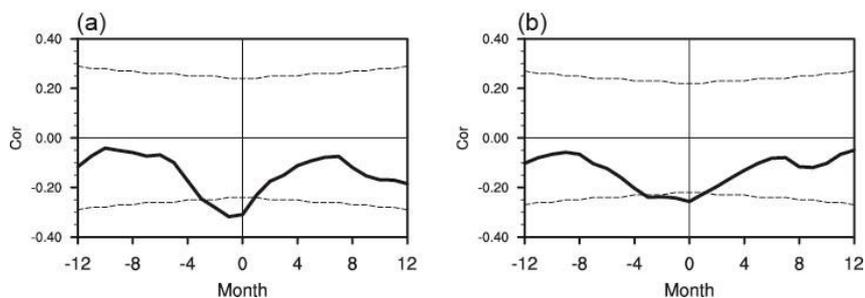


1
2 **Figure 2.** Correlation coefficients between southern high latitude lower stratospheric ozone
3 variations and SST (from HadISST) between 1979 and 2015. Southern high latitude lower
4 stratospheric ozone variations are averaged over the region 60–90 °S at 200–50 hPa. (a) Ozone
5 from MERRA2. (b) Ozone from SLIMCAT. Only the significant correlations are colored;
6 statistical significance was calculated using the two-tailed Student's t -test and the N^{eff} of DOF (see
7 section 2). The seasonal cycles and linear trends were removed prior to calculating the correlation
8 coefficients.



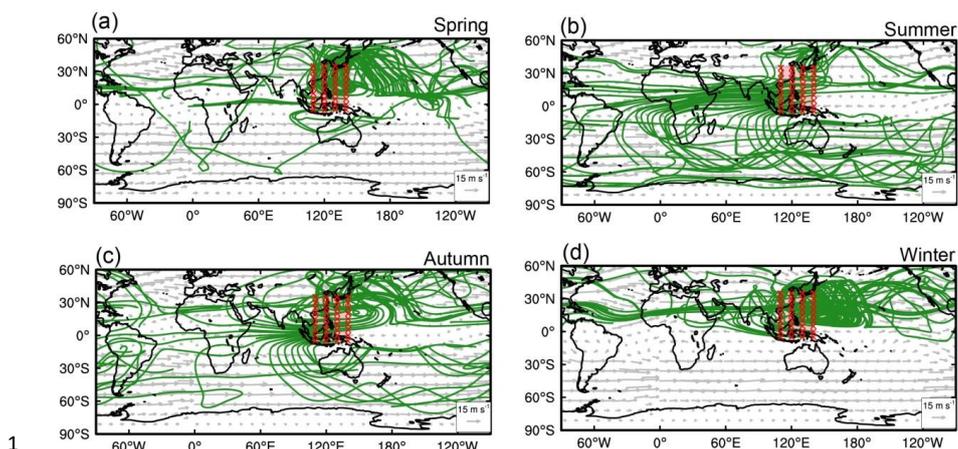
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2 **Figure 3.** (a) SST variations in the marginal seas of East Asia defined using the ST_MSEA index
3 that was calculated by averaging SST over the region from 5°S–35°N at 100°E–140°E (from
4 HadISST), and then removing the seasonal cycle and linear trend. The dashed lines indicate the
5 thresholds for definition of warm and cold events. (b) and (c) show the composite warm and cold
6 SST anomalies, respectively, for the events listed in Table 1.



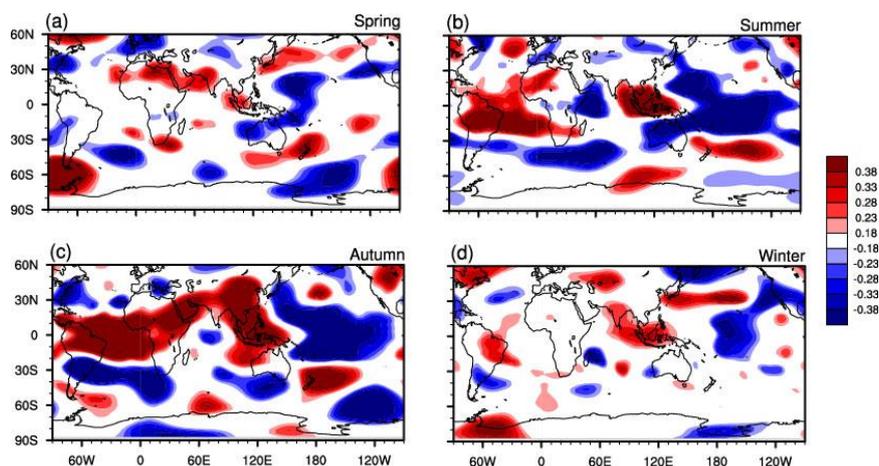
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2 **Figure 4.** Lead-lag correlations between the ST_MSEA index and southern high latitude lower
3 stratospheric ozone variations between 1979 and 2015; Southern high latitude lower stratospheric
4 ozone variations are averaged over the region 60–90 °S at 200–50 hPa from the (a) MERRA2 and
5 (b) SLIMCAT datasets. Negative months on the x -axis refer to ST_MSEA leading southern high
6 latitude lower stratospheric ozone variations, and positive months refer to the southern high
7 latitude lower stratospheric ozone variations leading ST_MSEA. Dotted lines indicate the 90%
8 confidence level; lead-lag correlations exceeding the dotted lines are statistically significant. The
9 statistical significance of the lead-lag correlation between two auto-correlated time series was
10 calculated using the two-tailed Student's t -test and the N^{eff} of DOF (see section 2).



1
2 **Figure 5.** Ray paths (green lines) at 300 hPa in (a) spring, (b) summer, (c) autumn, and (d) winter.
3 Red points denote wave sources in the marginal seas of East Asia (5°S–35°N, 100°E–140°E). The
4 wavenumbers along these rays are in the range 1–5. The grey vectors indicate climatological
5 flows.

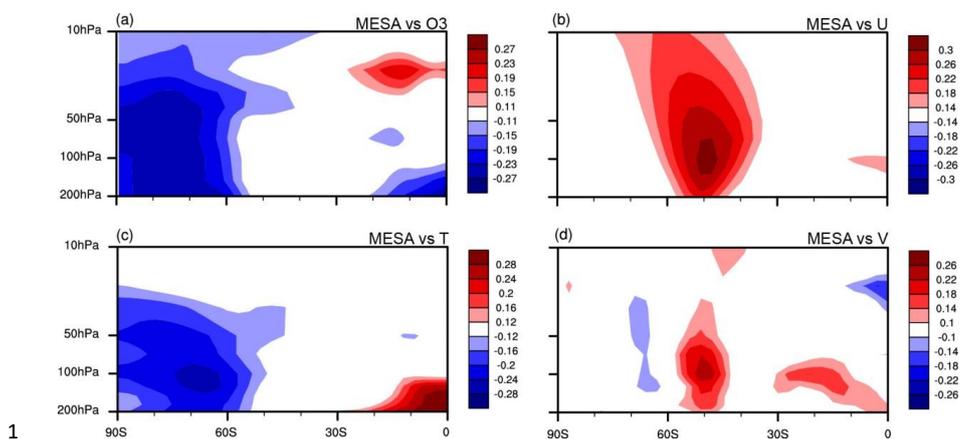
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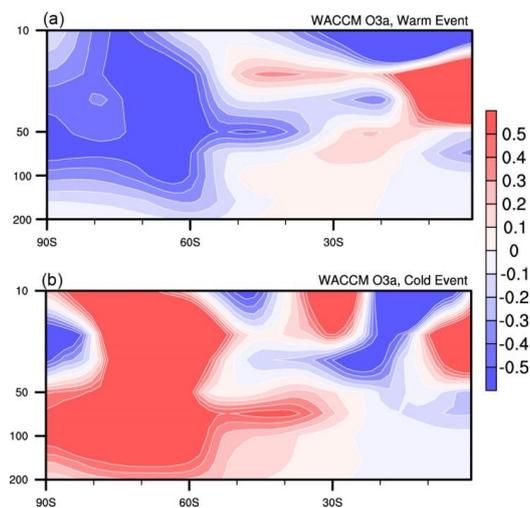
2 **Figure 6.** Correlation coefficients between the ST_MSEA index and 300-hPa geopotential
3 from the ERA-Interim reanalysis in (a) spring, (b) summer, (c) autumn, and (d) winter between
4 1979 and 2015. Only significant correlations are colored. The seasonal cycles and linear trends
5 were removed before calculating the correlation coefficients.

6

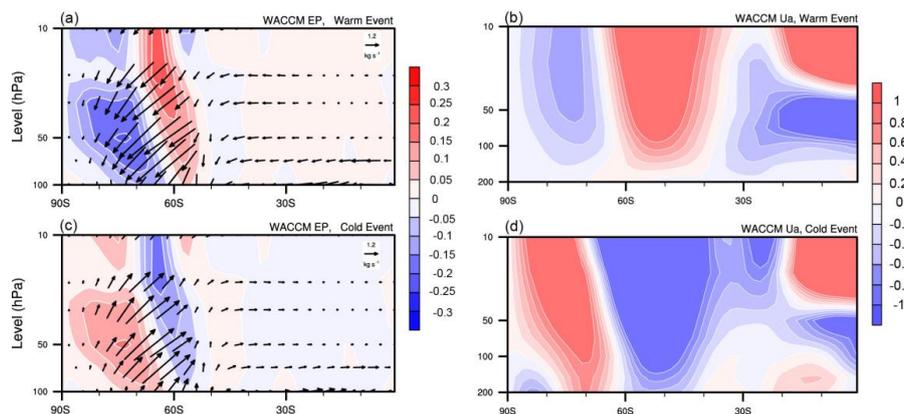


1
2 **Figure 7.** Correlation coefficients between ST_MSEA and (a) zonally averaged ozone, (b) zonal
3 wind, (c) temperature, and (d) meridional wind. Wind and temperature from ERA-Interim
4 reanalysis data; ozone from MERRA2. Only significant correlations are colored. The seasonal
5 cycles and linear trends were removed before calculating the correlation coefficients.

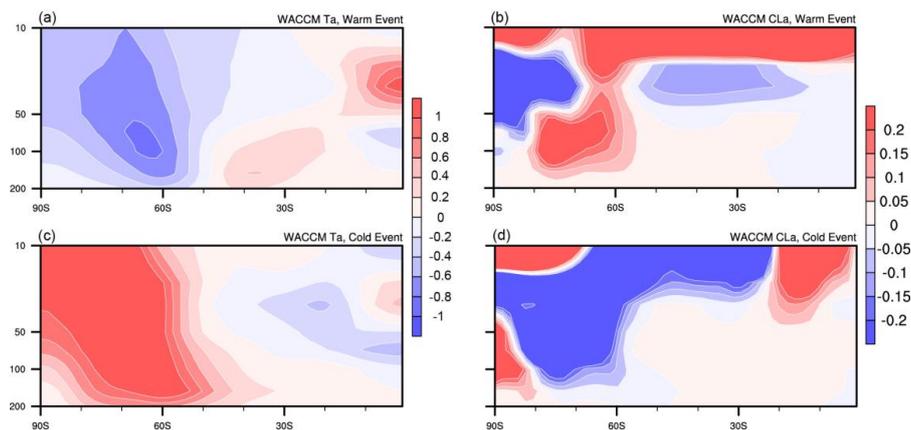
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1
2 **Figure 8.** Zonal mean differences in ozone (ppmv) between WACCM simulations (a) S2 and S1,
3 and (b) S3 and S1.



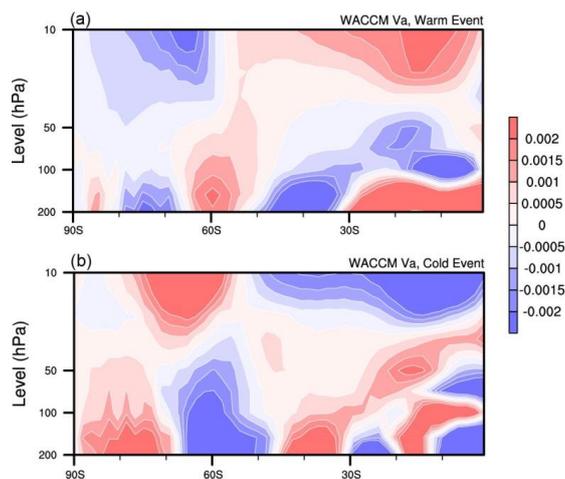
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2 **Figure 9.** Differences in E–P flux vectors (black arrows) and divergence (color shading) between
3 (a) S2 and S1, and (c) S3 and S1. Units for the horizontal and vertical vector directions are 10^7 and
4 10^5 kg s^{-1} , respectively. (b) and (d), as (a) and (c), but for zonal wind (m s^{-1}).



1
2 **Figure 10.** Zonal mean difference in temperature (K) between (a) S2 and S1, and (c) S3 and S1. (b)
3 and (d), as (a) and (c), but for active chlorine (ppbv).



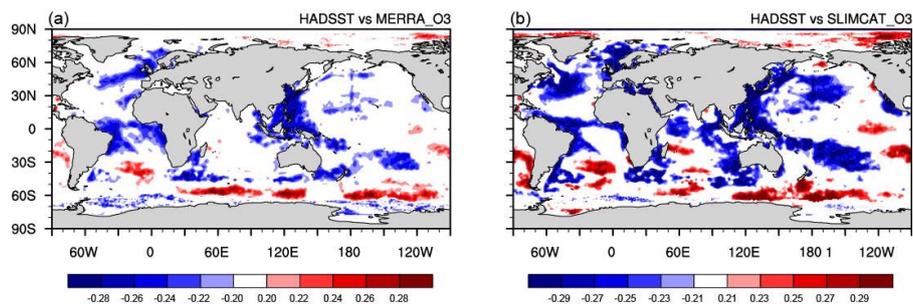
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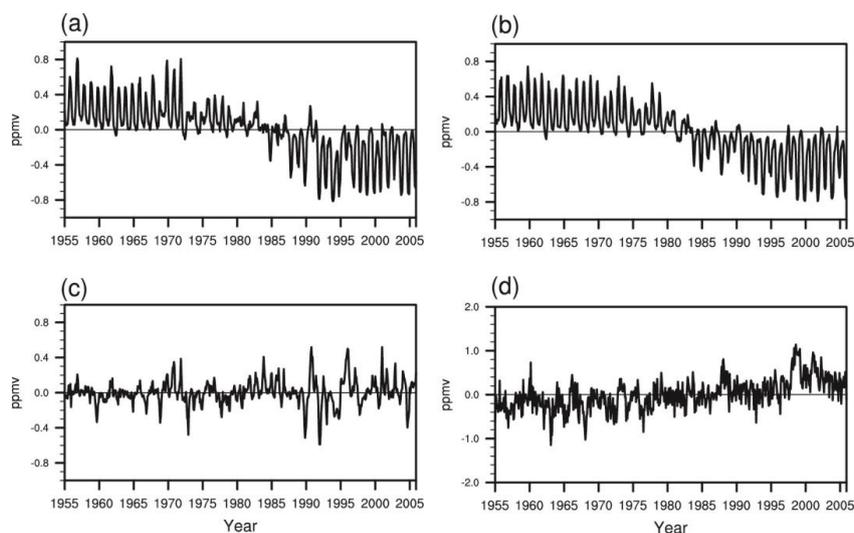
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3 **Figure 11.** Zonal mean difference in meridional wind (m s^{-1}) between (a) S2 and S1, and (b) S3

4 and S1.



- 1
- 2 **Figure 12.** As Fig. 2, but with only the seasonal cycle removed before calculating the correlation
- 3 coefficients.



1

2 **Figure 13.** The southern high latitude lower stratospheric ozone variations averaged over the
3 region 60–90 °S at 200–50 hPa from T1 (a) and (T2+T3)/2 (b). (c) The difference in southern high
4 latitude lower stratospheric ozone variations between T1 and (T2+T3)/2. (d) SST variations ($\times -1$)
5 in the marginal seas of East Asia (5 °S–35 °N, 100 °E–140 °E) based on the HadISST data. All
6 values are removed the seasonal cycle.