

1 **The Relationship between Lower Stratospheric Ozone in the**  
2 **Southern High Latitudes and Sea Surface Temperature in**  
3 **the East Asian Marginal Seas in Austral Spring**

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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 latitudes in austral spring. It is found  
5 that the SST variations across the East Asian Marginal Seas (5 °S–35 °N, 100 °E–140 °E)  
6 rather than the tropical eastern Pacific Ocean, where ENSO occurs, have the most  
7 significant correlation with the southern high latitude lower stratospheric ozone  
8 changes in austral spring. Further analysis reveals that planetary waves originating  
9 over the marginal seas in austral spring can propagate towards to southern middle to  
10 high latitudes via teleconnection pathway. The anomalous propagation and dissipation  
11 of 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 strong trends, forced by  
3 changes in ozone-depleting substances superimposed on a changing climate, but also  
4 interannual variability influenced by various external and internal climate forcings  
5 (e.g. Manney et al. 1994; Müller et al., 1994, 2005; Weiss et al., 2001; Hadjinicolaou  
6 et al., 2002; Tian and Chipperfield, 2005; Austin et al., 2006, 2010; Eyring et al., 2010;  
7 Liu et al., 2011, 2013; Douglass et al., 2014). Ozone variations can change the amount  
8 of harmful solar ultraviolet rays reaching the Earth's surface (Kerr and McElroy, 1993)  
9 and even influence climate (Forster and Shine, 1997; Thompson et al., 2011; Li et al.,  
10 2016; Xie et al., 2016). Therefore, clarifying the processes that are responsible for  
11 ozone variability is crucial for understanding how global climate interacts with ozone  
12 variations (Austin et al., 2006; Hess and Lamarque, 2007; Frossard et al., 2013;  
13 Rieder et al., 2013). Many previous studies have analyzed the ozone variability  
14 caused by external processes such as volcanic aerosols (e.g. Hofmann and Oltmans,  
15 1993; Rozanov et al., 2002; Dhomse et al., 2015) and the solar cycle (e.g. Chandra  
16 and McPeters, 1994; Rozanov et al., 2005; Dhomse et al., 2016) and these studies  
17 showed that volcanic aerosols and solar variations can result in considerable short-  
18 and long-term variations in ozone levels. Ozone variations can also be caused by  
19 changes in the surface climate (Zhang et al., 2014). Other studies have reported the  
20 effects of internal climate variability on ozone, including El Niño–Southern  
21 Oscillation (ENSO; Ziemke and Chandra, 1999; Cagnazzo et al., 2009; Randel et al.,  
22 2009; Xie et al., 2014a, 2014b; Zhang et al., 2015a, 2015b), Madden–Julian  
23 Oscillation (MJO; Fujiwara et al., 1998; Tian et al., 2007; Liu et al., 2009; Weare,  
24 2010; Li et al., 2012), Arctic Oscillation (AO) or North Atlantic Oscillation (NAO;  
25 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 in austral spring (Farman et al., 1985) over the  
6 Antarctic has been shown to have an important impact on the Southern Hemisphere  
7 climate (Shindell and Schmidt, 2004; Son et al., 2008, 2009, 2010, Perlwitz et al.,  
8 2008; Feldstein, 2011, Kang et al., 2011, Polvani et al., 2011; Thompson et al., 2011;  
9 Cagnazzo et al., 2013; Keeble et al., 2014; Previdi and Polvani, 2014). Although the  
10 principal mechanisms responsible for the formation of the ozone hole are well  
11 understood (e.g., Solomon, 1990, 1999; Ravishankara et al., 1994, 2009), the factors  
12 or processes that generate interannual variations in ozone levels in the southern high  
13 latitude stratosphere remain under debate. Among various factors, the QBO has been  
14 reported to have a significant impact on the interannual variations of the Antarctic  
15 ozone (Garcia and Solomon, 1987; Lait et al., 1989; Mancini et al., 1991; Gray and  
16 Ruth, 1993; Bodeker and Scourfield, 1995; Shindell et al., 1997a). The September to  
17 March levels of ozone over the Antarctic is also marginally correlated with the  
18 wintertime mean eddy heat flux (Weber et al., 2003). Heat transport induced by  
19 upward propagating planetary waves warms the polar vortex (Schoeberl and  
20 Hartmann, 1991), which reduces the occurrence of polar stratospheric clouds (PSCs),  
21 a key prerequisite for the heterogeneous chemistry that depletes Antarctic ozone.  
22 Subsequent efforts to understand Antarctic ozone variations during individual years  
23 have considered planetary wave activity, which account for much of the interannual  
24 variations of ozone levels over the Northern Hemisphere (Hadjinicolaou et al., 1997;  
25 Fusco and Salby, 1999; Salby and Callaghan, 2004, 2007a, 2007b; Hadjinicolaou and

1 Pyle, 2004). Studies based on measurements (Bodeker and Scourfield, 1995),  
2 modeling (Shindell et al., 1997a, 1997b), and reanalysis data (Huck et al., 2005) have  
3 shown 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 a  
14 correspondence to the trend of temperature in the southern polar stratosphere (Grassi  
15 et al., 2005, 2006; Hu and Fu, 2009; Li et al., 2010; Clem et al., 2016). Although  
16 ENSO 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 exhibited 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 propagate towards to southern middle to  
25 high latitudes in austral spring. It is likely that the Rossby waves generated by SST

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

12

## 13 **2. Data, Model, and Methods**

14 The ozone data used in this study is obtained from the NASA Modern Era  
15 Retrospective Analysis for Research and Applications (MERRA) dataset version 2  
16 (Rienecker et al., 2011), TOMCAT/SLIMCAT 3-D model simulations (Chipperfield,  
17 2006), Global OZone Chemistry And Related trace gas Data records for the  
18 Stratosphere (GOZCARDS) ozone satellite data (Froidevaux et al. 2015) and The  
19 Stratospheric Water and OzOne Satellite Homogenized (SWOOSH) ozone satellite  
20 data (Davis et al. 2016). The MERRA2 data ( $\text{lon} \times \text{lat}: 1.25^\circ \times 1.25^\circ$ ) has 42 pressure  
21 levels from the surface up to 0.1 hPa. The vertical resolution of MERRA2 is  $\sim 1\text{--}2$  km  
22 in the UTLS and  $2\text{--}4$  km in the middle and upper stratosphere. MERRA2 is  
23 assimilated by the Goddard Earth Observing System Model, Version 5 (GEOS-5) with  
24 ozone from the Solar Backscattered Ultra Violet (SBUV) radiometers from October  
25 1978 to October 2004, and thereafter from the Ozone Monitoring Instrument (OMI)

1 and AURA Microwave Limb Sounder (MLS) (Bosilovich et al., 2015). The MERRA2  
2 reanalysis ozone data compares well with satellite ozone observations (Wargan et al.,  
3 2017) and shows a better representation of the QBO and stratospheric ozone  
4 compared to MERRA1 (Coy et al., 2016). In the present study, the ozone field (lon  $\times$   
5 lat:  $5.625^\circ \times 5.5^\circ$ ) simulated by a 3D offline chemical transport model, SLIMCAT  
6 (Feng et al., 2007, 2011), is also used. The simulation performed in this study is  
7 driven by horizontal winds and temperatures from meteorological analyses of the  
8 ERA-Interim data provided by European Centre for Medium-Range Weather  
9 Forecasts (ECMWF) (Dee et al., 2011). The vertical advection in the model is  
10 calculated from the divergence of the horizontal mass flux (Chipperfield, 2006), and  
11 chemical tracers are advected by the conservation of second-order moments (Prather,  
12 1986). The zonal mean satellite-based GOZCARDS is produced from high quality  
13 data from past missions (e.g., SAGE, HALOE data) as well as ongoing missions  
14 (ACE-FTS and Aura MLS). Its meridional resolution is  $10^\circ$  with 25 pressure levels  
15 from the surface up to 0.1 hPa. The zonal mean SWOOSH dataset is a merged record  
16 of stratospheric ozone and water vapor measurements taken by a number of limb  
17 sounding and solar occultation satellites (SAGE-II/III, UARS HALOE, UARS MLS,  
18 and Aura MLS instruments). Its meridional resolution is  $2.5^\circ$  with 31 pressure levels  
19 from 300 to 1 hPa.

20 Figure 1 shows the ozone variations averaged over the region 200–50 hPa and  
21 60–90  $^\circ$ S, where the variability and trend of ozone concentration is most pronounced  
22 in the Southern Hemisphere (Austin and Wilson, 2006; Solomon 1990, 1999;  
23 Ravishankara et al., 1994, 2009), from the four datasets. The ozone variations from  
24 two of the four datasets are significantly correlated (Fig. 1a, the correlation  
25 coefficients aren't shown). In austral spring, the ozone variations from the four

1 datasets are in better agreement with each other (Fig. 1b).

2 SST is obtained from HadISST dataset compiled by the UK Met Office Hadley  
3 Centre for Climate Prediction and Research (Rayner et al., 2003). Geopotential height,  
4 zonal wind, and temperature fields are obtained from the ECMWF ERA-Interim  
5 dataset.

6 We also use version 4 of the Whole Atmosphere Community Climate Model  
7 (WACCM4) in this study since WACCM has been shown to have a good performance  
8 in simulating the stratospheric circulation, temperature and ozone variations (Garcia  
9 et al. 2007). WACCM4 is part of the Community Earth System Model (CESM)  
10 framework developed by the National Center for Atmospheric Research (NCAR).  
11 WACCM4 uses a finite-volume dynamical core, with 66 vertical levels extending  
12 from the ground to  $4.5 \times 10^{-6}$  hPa (145 km geometric altitude), and a vertical  
13 resolution of 1.1–1.4 km in the tropical tropopause layer and the lower stratosphere  
14 (below a height of 30 km). The simulations presented in this paper are performed at a  
15 horizontal resolution of  $1.9^\circ \times 2.5^\circ$  and with interactive chemistry (Garcia et al.,  
16 2007). More details regarding WACCM4 are provided in Marsh et al. (2013).

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

$$21 \quad \frac{1}{N^{\text{eff}}} \approx \frac{1}{N} + \frac{2}{N} \sum_{j=1}^N \frac{N-j}{N} \rho_{XX}(j) \rho_{YY}(j)$$

22 where  $N$  is the sample size, and  $\rho_{XX}$  and  $\rho_{YY}$  are the autocorrelations of two  
23 sampled time series,  $X$  and  $Y$ , respectively, at time lag  $j$ .

24 We use the formulae given by Andrews et al. (1987) to calculate the



1 quasi-geostrophic 2D Eliassen–Palm (E–P) flux. The meridional ( $F_y$ ) and vertical ( $F_z$ )  
 2 components of the E–P flux, and the E–P flux divergence  $D_F$ , are expressed as:

$$\begin{aligned}
 3 \quad F_y &= -\rho_0 a \cos \varphi \overline{\varphi u'v'} \\
 4 \quad F_z &= -\rho_0 a \cos \varphi \frac{Rf}{HN^2} \overline{v'T'} \\
 5 \quad D_F &= \frac{\nabla \cdot F}{\rho_0 a \cos \varphi} = \frac{\partial(F_y \cos \varphi)/a \cos \varphi \partial \varphi + \partial F_z / \partial z}{\rho_0 a \cos \varphi},
 \end{aligned}$$

6 where  $\rho_0$  is the air density,  $\varphi$  is the latitude,  $a$  is the radius of the Earth,  $R$   
 7 is the gas constant,  $f$  is the Coriolis parameter,  $H$  is the atmospheric scale height (7  
 8 km),  $u$  and  $v$  are the zonal and meridional wind components, respectively, and  $T$   
 9 is the temperature; the overbar denotes the zonal mean, and the prime symbol denotes  
 10 departures from the zonal mean.

11 The Transformed Eulerian Mean (TEM) meridional wind ( $v^*$ ) which is given by  
 12 Edmon et al. (1980):

$$13 \quad v^* = \bar{v} - [(\overline{v'\theta'}) / \bar{\theta}_p]_p$$

14 where  $\theta$  is the potential temperature,  $v$  is meridional wind and subscript  $p$   
 15 denotes derivative with pressure  $p$ . The overbar denotes the zonal mean and the  
 16 prime denotes deviations from the zonal mean value.

17

### 18 **3. The connection between the East Asian Marginal Seas and southern high** 19 **latitude lower stratospheric ozone in austral spring**

20 Figure 2a shows the correlation coefficients between SST and southern high latitude  
 21 lower stratospheric ozone variations in austral spring between 1979 and 2015 using  
 22 ozone data from the MERRA2 dataset and SST from HadISST dataset. Ozone from  
 23 SLIMCAT simulations, GOZCARDS and SWOOSH datasets were further used to

1 confirm the robustness of the correlations (Fig. 2b–d). The regions of significant  
2 correlation are generally different for the four ozone datasets except for the East Asian  
3 Marginal Seas; i.e., 5°S–35°N, 100°E–140°E, where the most significant correlations  
4 between Antarctic stratospheric ozone variations and SST are seen in the four datasets.  
5 Figure 2 implies an interannual connection between SST in the East Asian Marginal  
6 Seas and southern high latitude lower stratospheric ozone variations in austral spring.  
7 Figure 2 also indicates that SST variations in austral spring associated with ENSO are  
8 not the main factor controlling the interannual variability of southern high latitude  
9 lower stratospheric ozone.

10 Through the interannual connection in Fig. 2 possibly caused by the influence of  
11 lower latitude SST on the south high latitude stratosphere, south high latitude  
12 stratospheric ozone has also been shown to affect tropical climate (Son et al., 2008;  
13 Kang et al., 2011; Thompson et al., 2011). Thus, it is first necessary to confirm the  
14 causality of this connection. To investigate the SST variations across the marginal  
15 seas of East Asia, we first define an austral spring SST index over the region with the  
16 most significant correlations in Fig. 2, i.e., the ST\_MSEA index (ST\_MSEAI). This  
17 index is a time series that represents SST variations across the marginal seas of East  
18 Asia in austral spring (Figure 3a). It is calculated by averaging the SST variations in  
19 austral spring in the region from 5°S–35°N at 100°E–140°E, and then removing the  
20 seasonal cycle and linear trend. Fig. 3b and c show the composite warm and cold SST  
21 anomalies for the events that occurred in the marginal seas of East Asia in austral  
22 spring between 1979 and 2015 (see Table 1).

23 It is well known that the SST changes in the eastern Pacific, the Indo-Pacific  
24 warm pool, and the Atlantic can significantly influence the northern polar stratosphere  
25 (Calvo et al., 2004, 2009; Hoerling et al., 2001, 2004; Cagnazzo et al., 2009; Hu and

1 Fu, 2009; Hu and Pan, 2009; Li et al., 2010; Hurwitz et al., 2011a, b; Lin et al., 2012;  
2 Zubiaurre and Calvo, 2012; Xie et al., 2012; Li and Chen, 2014). SST variations in  
3 some regions can excite Rossby wave trains and those waves can propagate into the  
4 northern middle and high latitude stratosphere (Gettelman et al., 2001; Sassi et al.,  
5 2004; Manzini et al., 2006; Garc á-Herrera et al., 2006; Taguchi and Hartmann, 2006;  
6 Garfinkel and Hartmann, 2007, 2008; Free and Seidel, 2009). The mechanism that  
7 allows SST variations in the East Asian Marginal Seas to affect the southern high  
8 latitude stratosphere is also possibly related to tropospheric wave propagation from  
9 northern lower latitude to southern middle and high latitudes.

10 Figure 4 shows the ray paths of waves generated by the SST anomalies over the  
11 region 5 °S–35 °N, 100 °E–140 °E, at 300 hPa in four seasons. The wavenumbers along  
12 these rays are between 1 and 5. The wave ray paths represent the climate  
13 teleconnections, i.e., the propagation of stationary waves in realistic flows. The  
14 calculation of the wave ray paths and application of the barotropic model is described  
15 in detail by Li et al. (2015) and Zhao et al. (2015). We found that the Rossby waves  
16 generated by SST anomalies in the marginal seas of East Asia could indeed propagate  
17 to the middle to high latitudes of the Southern Hemisphere in austral spring and  
18 winter (Fig. 4a and d), but not in austral summer and autumn (Fig. 4b and c) because  
19 the Rossby waves motivated by the low-latitude SST anomalies move mostly  
20 northwards in austral summer and autumn. Meanwhile, we must note that the  
21 propagating paths of those waves in austral spring and winter aren't totally same (Fig.  
22 4a and d). In austral spring, the path of rays originates over the marginal seas of East  
23 Asia reflects directly into the southern Indian Ocean and reaches the Southern  
24 Hemisphere (Fig. 4a). In austral winter (Fig. 4d), the rays follow the austral spring  
25 path to the Southern Hemisphere. In addition, the second path of rays originates over

1 the marginal seas of East Asia, crosses the Indian Ocean to arrive over tropical Africa  
2 or even South America, and then reflects equatorward to the middle to high latitudes  
3 of the Southern Hemisphere. These rays can reach about 60 °S and then be refracted to  
4 lower latitudes, implying that the pathway of upward propagation of tropospheric  
5 waves from the marginal seas of East Asia possibly extends to 60 °S.

6 The correlation coefficients between the ST\_MSEAI and 300-hPa geopotential  
7 height variations from the ERA-Interim reanalysis across the four seasons are shown  
8 in Figure 5. The positive and negative centers of correlation coefficients represent the  
9 teleconnection patterns. The teleconnection patterns in austral spring and winter (Fig.  
10 5a and d) are in good agreement with the ray paths (Fig. 4a and d). In austral spring, a  
11 wave train path appears over the marginal seas of East Asia and reflects directly into  
12 the Southern Hemisphere (Fig. 5a). In austral winter, two clear wave train paths  
13 appear with one moving westwards to South America and reflecting to the middle to  
14 high latitudes of the Southern Hemisphere and the second reflecting to the middle to  
15 high latitudes of the Southern Hemisphere. These two teleconnection pathways of the  
16 wave trains in austral spring and winter (Figs. 4 and 5) are discussed in detail by Zhao  
17 et al., (2016), who refer to them as the North Australia–Southern Hemisphere and  
18 South Africa–Southern Hemisphere pathways, respectively. In austral summer and  
19 autumn, the above two teleconnection patterns don't exist (Fig. 5b and c).

20 Figs. 4 and 5 show the pathways of the wave trains generated by the SST  
21 anomalies over the marginal seas of East Asia in four seasons. Figure 6 shows the  
22 relationship between the SST anomalies and outgoing longwave radiation (ORL)  
23 which represents the relationship between the SST anomalies and generated wave  
24 activity anomalies to some extent. It is found that the correlation coefficients over the  
25 marginal seas of East Asia are the largest in the austral spring compared with other

1 seasons. It implies that the wave activity anomalies caused by the SST anomalies over  
2 the marginal seas of East Asia are very strong in austral spring. Figs. 4, 5 and 6  
3 illustrate the possibility of the SST anomalies over the marginal seas of East Asia  
4 influencing on the wave activity at southern high latitudes. Bodeker and Scourfield  
5 (1995), Shindell et al. (1997a, 1997b), and Huck et al. (2005) have shown that  
6 interannual differences in the severity of southern high latitude lower stratospheric  
7 ozone depletion are related to Southern Hemisphere planetary wave activity. All of the  
8 above analysis illustrates that the SST anomalies over the marginal seas of East Asia  
9 are a possible main source of this planetary wave activity.

10 Figure 7a shows the correlation coefficients between the ST\_MSEAI and  
11 stratospheric ozone variations in austral spring, which indicate that warm (cold) SST  
12 anomalies over the East Asian Marginal Seas are associated with a decrease (increase)  
13 in southern high latitude lower stratospheric ozone in austral spring. Figure 7b shows  
14 that ST\_MSEAI is positively correlated with zonal wind around 60°S, where is the  
15 climatological location of the boundary of the southern polar vortex in austral spring,  
16 while Figs. 7c indicate that ST\_MSEAI is negatively correlated with the zonal mean  
17 temperature. The correlations shown in Figs 3, 4, 5, and 7 can be used to establish a  
18 hypothesis of chemical process for the connection between SST variations over the  
19 marginal seas of East Asia and southern high latitude lower stratospheric ozone in  
20 austral spring as follows: 1. The warm (cold) SST anomalies over the marginal seas of  
21 East Asia (Fig. 3) depress (enhance) planetary wave activity in the middle to high  
22 latitudes of the Southern Hemisphere (Figs 4 and 5); 2. The anomalous propagation of  
23 planetary waves into the stratosphere and dissipation of ultra-long Rossby waves in  
24 the stratosphere strengthen/cool (weaken/warm) the southern polar vortex (Fig. 7b  
25 and c); 3. A cooler (warmer) polar vortex allows more (less) PSCs and active chlorine

1 to form. 4. Consequently, southern high latitude lower stratospheric ozone decreases  
2 (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. Fig. 7c shows that the center of the correlation  
8 coefficients locates near 60 °S. It suggests that the center of stratospheric temperature  
9 changes caused by SST changes in the East Asian Marginal Seas locates near 60 °S but  
10 not near 90 °S. Temperature changes near 60 °S may have more effective effects on  
11 southern high latitude lower stratospheric ozone than that near 90 °S since the  
12 background temperature in the lower stratosphere near 60 °S would be higher than that  
13 near 90 °S. The chemical process maybe has a contribution to the southern high  
14 latitude lower stratospheric ozone changes caused by SST changes in the East Asian  
15 Marginal Seas.

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

25

#### 1 **4. Simulating the effect of SST changes in the marginal seas of East Asia on** 2 **southern high latitude lower stratospheric ozone**

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

15 Figure 8 first shows the southern high latitude lower stratospheric ozone  
16 anomalies in austral spring forced by warm and cold SST anomalies over the marginal  
17 seas of East Asia. It can be seen that the warm SST anomalies indeed cause ozone  
18 decreases in the southern high latitude lower stratosphere (Fig. 8a) and cold SST  
19 anomalies results in ozone increases (Fig. 8b). The simulations support the results  
20 shown from the statistical analysis in Section 3.

21 Figure 9 shows the E–P flux vectors and divergence anomalies in the  
22 stratosphere in austral spring caused by SST anomalies over the marginal seas of East  
23 Asia. Analysis of changes in the E–P flux (Eliassen and Palm 1961; Andrews et al.  
24 1987) is often used as a diagnostic for planetary wave propagation from the  
25 troposphere to the stratosphere (Edmon et al., 1980). During periods of warm (cold)

1 SST over the marginal seas of East Asia, a decrease (increase) in upward wave flux  
2 entering the stratosphere accompanied by stronger (weaker) convergence of the E–P  
3 flux in the stratosphere at middle to high latitudes of the Southern Hemispheres (ca.  
4 60°S) is evident (Fig. 9a and c). The anomalous wave flux entering the stratosphere  
5 around 60°S confirms the result in Figs. 4 and 5, which shows that the wave rays can  
6 reach about 60°S.

7 Many previous studies have demonstrated a strongly negative correlation  
8 between upward propagating wave activity and the intensity of the stratospheric polar  
9 vortex, with an anomalously negative and positive upward wave flux alongside a  
10 stronger and weaker polar vortex, respectively (Christiansen 2001; Polvani and  
11 Waugh 2004; Li and Lau 2013). During periods of warm (cold) SST over the marginal  
12 seas of East Asia, the anomalous downward (upward) E–P flux, and larger (smaller)  
13 E–P flux divergence at middle to high latitudes (ca. 60°S) in the Southern Hemisphere  
14 (Fig. 9a and c) imply suppressed (active) wave activity in the stratosphere, which  
15 induces a strengthened (weakened) circulation at southern polar vortex edge (Fig. 9b  
16 and d). Finally, the cold (warm) polar vortex (Fig. 10a and c) allows more (less)  
17 PSCs/active chlorine (Fig. 10b and d) to form. This is one process through which SST  
18 variations over the marginal seas of East Asia causes southern high latitude lower  
19 stratospheric ozone changes. The other process is that the strengthened (weakened)  
20 southern polar vortex impedes (promotes) air exchange between middle and high  
21 latitude stratosphere (Figure 11), and further decreases (increases) southern high  
22 latitude lower stratospheric ozone levels.

23 It is noteworthy that warm (cold) SST anomalies are generally thought to  
24 increase (suppress) planetary wave activity via strengthening (weakening) convection



1 (Xie et al., 2008; Shu et al., 2010; Hu et al., 2014). However, this study shows that  
2 warm (cold) SST anomalies over the marginal seas of East Asia suppress (increase)  
3 planetary wave activity in the southern high latitude stratosphere. Indeed, it is found  
4 that there is an enhanced of the E-P flux from lower latitudes to southern high  
5 latitudes in the SST warming event over the East Asian Marginal Seas (Figure 12a).  
6 However, this increased EP flux does not propagate upward into the stratosphere but  
7 downward to lower levels, and *vice versa* for the SST cooling event (Fig. 12b). Fig.  
8 12 explains why SST warming (cooling) over the East Asian Marginal Seas leads to a  
9 weaker (stronger) wave activity in the Southern Hemisphere stratosphere.

10 As a result of human activity, the amount of Antarctic stratospheric ozone has  
11 decreased remarkably from 1950 to 2000 (Solomon 1990, 1999; Ravishankara et al.,  
12 1994, 2009). At the same time, the SST over the marginal seas of East Asia has  
13 followed an increasing trend, but superimposed on strong interannual variations  
14 (Zheng et al., 2014). Figure 13 shows the correlation coefficients between southern  
15 high latitude lower stratospheric ozone and SST in which the SST and southern high  
16 latitude lower stratospheric ozone variations have not been detrended as that in Fig. 2.  
17 Comparing Fig. 13 with Fig. 2, we can see that the negative correlation coefficients  
18 over the marginal seas of East Asia become larger in Fig. 13, implying a contribution  
19 of warmer SST in the marginal seas of East Asia to the decline trend of southern high  
20 latitude lower stratospheric ozone.

21 We used ensemble transient experiments to estimate the contribution of SST  
22 variations in the marginal seas of East Asia to southern high latitude lower  
23 stratospheric ozone changes. The transient experiments incorporated the following  
24 natural and anthropogenic external forcings for the period 1955–2005: observed SST  
25 from the HadISST dataset, surface emissions from the IPCC A1B emissions scenario,

1 spectrally resolved solar variability (Lean et al., 2005), volcanic aerosols (from the  
2 Stratospheric Processes and their Role in Climate (SPARC) Chemistry–Climate  
3 Model Validation (CCMVal) REF-B2 scenario recommendations), and nudged QBO  
4 (the time series in CESM is determined from the observed climatology). The first  
5 transient experiment, T1, was the historical experiment covering the period 1955–  
6 2005 (Marsh et al., 2013). The second transient experiment, T2, was the same as T1  
7 except that the SST in the marginal seas of East Asia (5 °S–35 °N and 100–140 °E) for  
8 the period 1955–2005 was replaced by the 12-month cycle of climatology averaged  
9 over the same period. This means that in T2, the SST over the marginal seas of East  
10 Asia had only a seasonal cycle, but no trend and no interannual variability. T3 was the  
11 same as T2, but used a slightly different initial condition as an ensemble experiment.  
12 Detailed descriptions of runs T1–T3 are provided in Table 3.

13 The southern high latitude lower stratospheric ozone variations caused by the  
14 SST variability over the marginal seas of East Asia can be obtained by subtracting  
15 simulated ozone in the ensemble experiments ((T2+T3)/2)) from the ozone in T1  
16 (Figure 14, black line). There are evident differences in southern high latitude lower  
17 stratospheric ozone variations between T1 and the ensemble experiments  
18 ((T2+T3)/2)). This illustrates that the SST variability over the marginal seas of East  
19 Asia (Fig. 14, red line) does have a significant effect on southern high latitude lower  
20 stratospheric ozone over the past five decades (Fig. 14, black line). The correlation  
21 coefficient between the two time series is 0.29 which is significant at 95% confidence  
22 level. A further analysis reveals that the linear trend of ozone variations over the  
23 region 200–50 hPa and 60–90 °S from T1 (Trend1) is  $-1.2 \times 10^{-3}$  ppmv/month, and  
24 from (T1 – (T2+T3)/2) (Trend2, Fig. 14, black line) is  $-0.204 \times 10^{-3}$  ppmv/month.  
25 See Table 4. It implies that the increasing linear trend in SST over the marginal seas

1 of East Asia can contribute approximately 17% of the declining trend in southern high  
2 latitude lower stratospheric ozone from 1955–2005 ( $\text{Trend}_2 / \text{Trend}_1 \times 100\%$ ).

3

#### 4 **6. Conclusions and Summary**

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

3

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9

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1 Table 1. Warm and cold SST events in the marginal seas of East Asia in austral spring during the  
2 period from 1979 to 2015 analyzed in this paper using the ST\_MSEAI (Fig. 3a).

Warm Events*	Cold Events*
1983	1982
1987	1991
1988	1992
1998	1994
1999	2004
2008	2012

3 \*Following the definition of ENSO events (Trenberth 1997), we propose a threshold of  $\pm 0.2$ ,  
4 which is equal to the standard deviation of the ST\_MSEAI series, as the indicator of warm and  
5 cold events.

1 **Table 2.** Experiments S1–S3.

Experiments <sup>*1</sup>	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 <sup>\*1</sup>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 <sup>*1</sup>	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 <sup>*2</sup>

2 <sup>\*1</sup>Integration period is 1955–2005 for T1–T3.

3 <sup>\*2</sup>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  $e^{-14}$ .

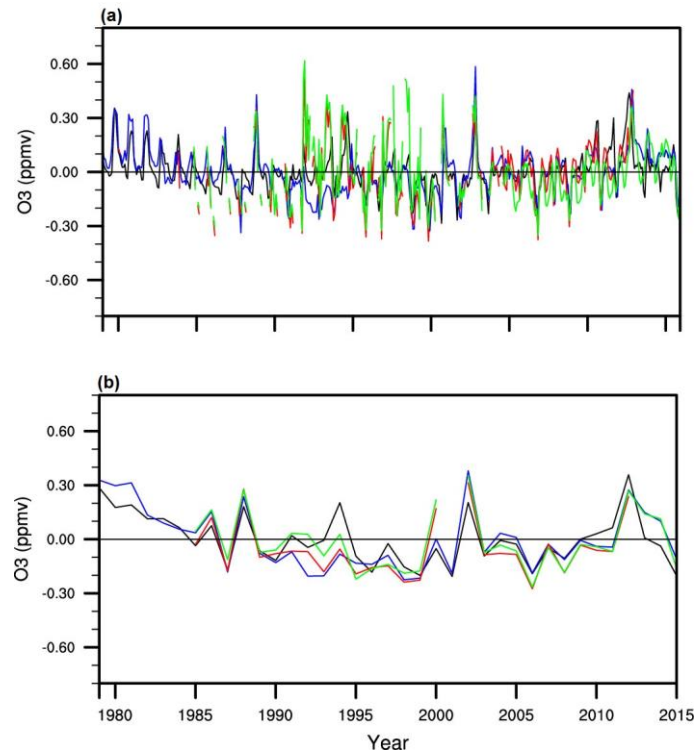
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1 Table 4. Linear trends of ozone variations over the region 200–50 hPa and 60–90 °S from  
 2 experiments with (T1) and without SST (T2 +T3) variations in the East Asian Marginal Seas (T1–  
 3 see Table 3).

Experiments	Values
Linear trend of ozone variations over the region 200–50 hPa and 60–90 °S from T1 (Trend1)	$-1.2 \times 10^{-3}$ ppmv/month <sup>**</sup>
Linear trend of ozone variations over the region 200–50 hPa and 60–90 °S from (T1 – (T2+T3)/2) (Trend2)	$-0.204 \times 10^{-3}$ ppmv/month <sup>*</sup>

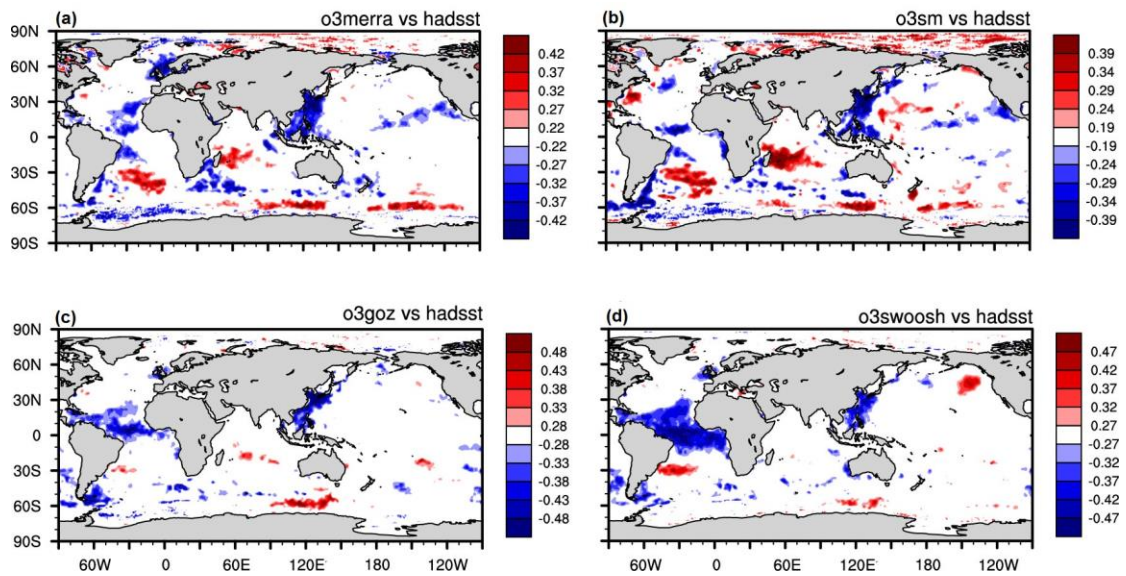
4 <sup>\*\*</sup>: the trend is significant at 99% confidence level. <sup>\*</sup>: the trend is significant at 95% confidence  
 5 level. The calculation of the statistical significance of the trend uses the two-tailed Student's *t*-test.

6



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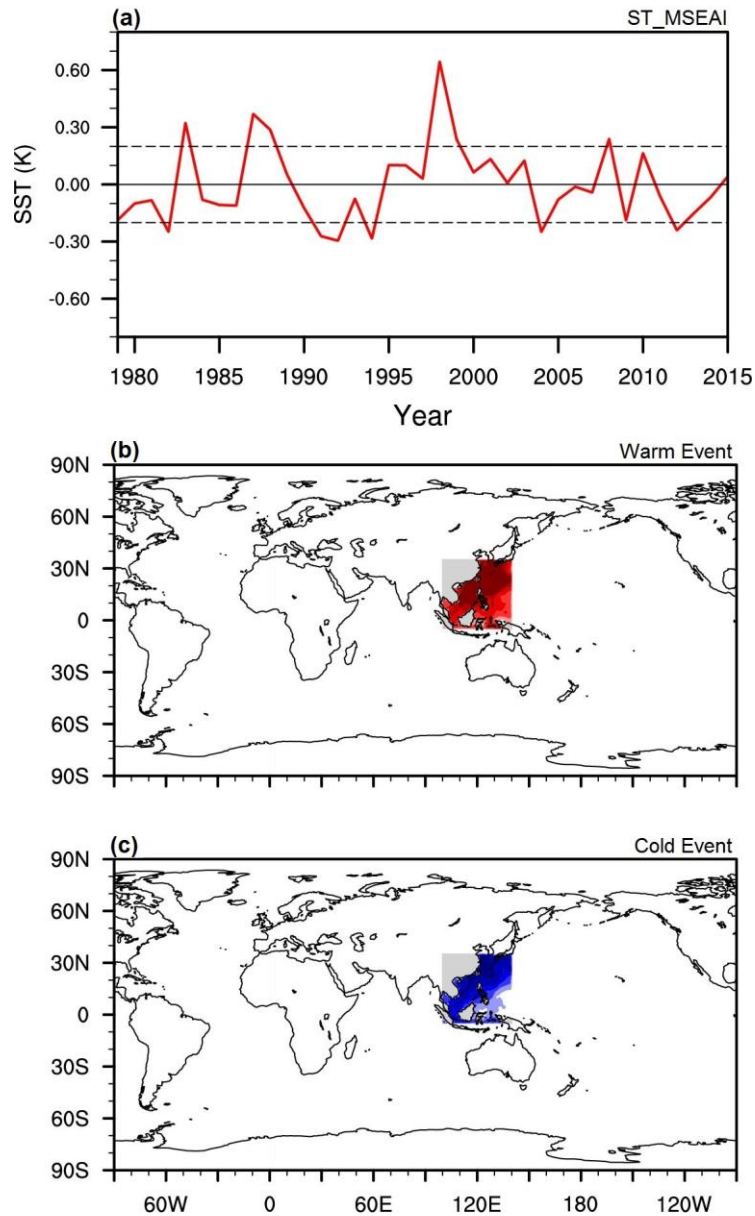
2 **Figure 1.** (a) Time series of southern high latitude lower stratospheric ozone variations averaged  
3 over the region 60–90 S at 200–50 hPa from the MERRA2 (black line), SLIMCAT (blue line),  
4 GOZCARDS (red line) and SWOOSH (green line) ozone datasets. (b), Same as (a), but only for  
5 austral spring. Ozone variations are calculated by removing the seasonal cycles and linear trends.



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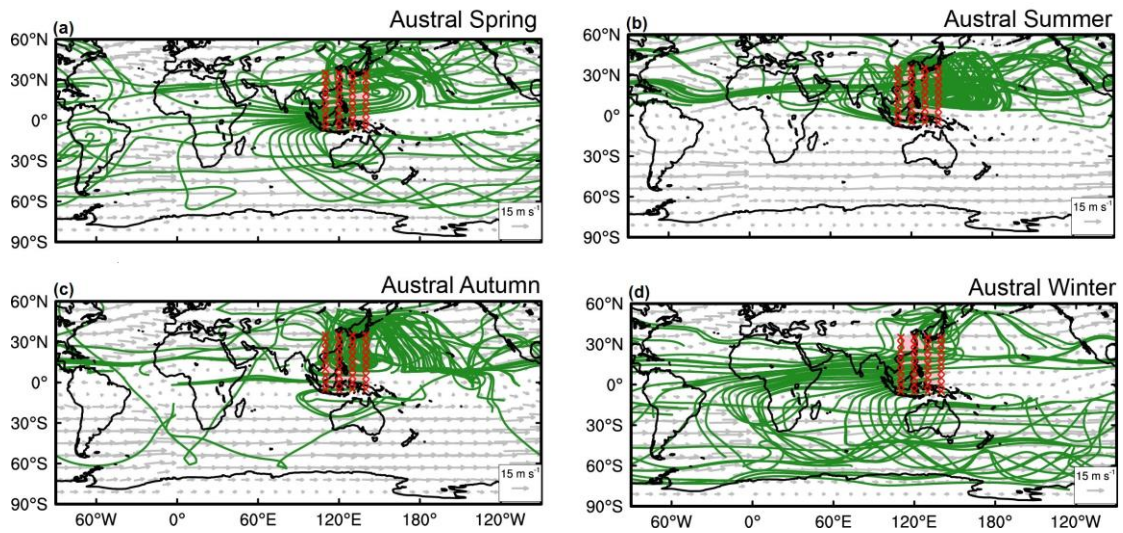
2 **Figure 2.** Correlation coefficients between southern high latitude lower stratospheric ozone  
 3 variations and SST from HadISST in austral spring. Southern high latitude lower stratospheric  
 4 ozone variations are averaged over the region 60–90 °S at 200–50 hPa in austral spring. (a) Ozone  
 5 from MERRA2 and (b) Ozone from SLIMCAT for period 1979–2015. (c) Ozone from  
 6 GOZCARDS for period 1979–2012. (d) Ozone from SWOOSH for period 1984–2015. Only  
 7 statistical significance above 95% confidence level is colored; statistical significance was  
 8 calculated using the two-tailed Student’s  $t$ -test and the  $N^{\text{eff}}$  of DOF. The seasonal cycles and linear  
 9 trends were removed prior to calculating the correlation coefficients.





1

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

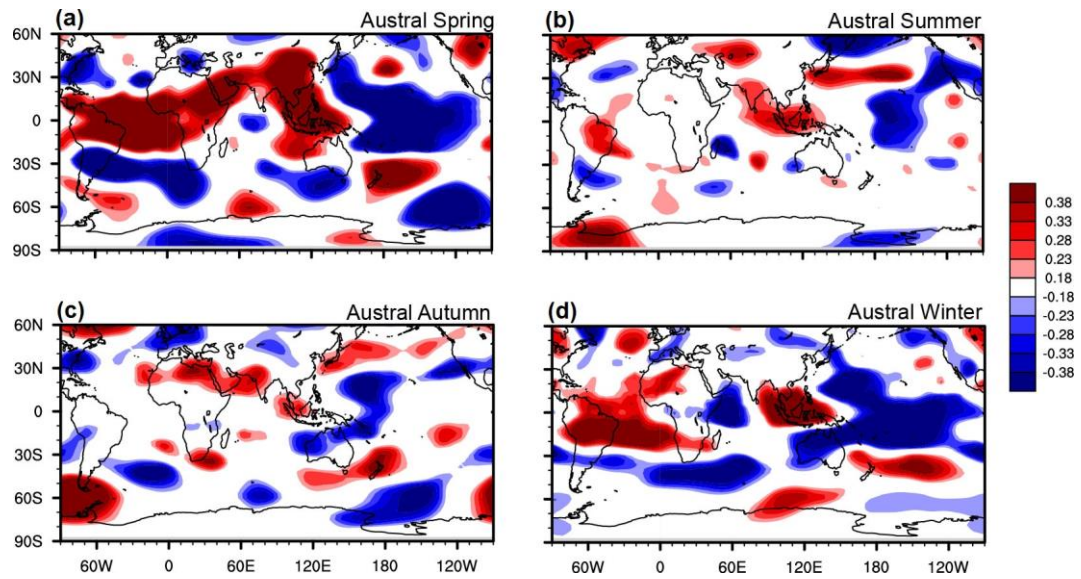


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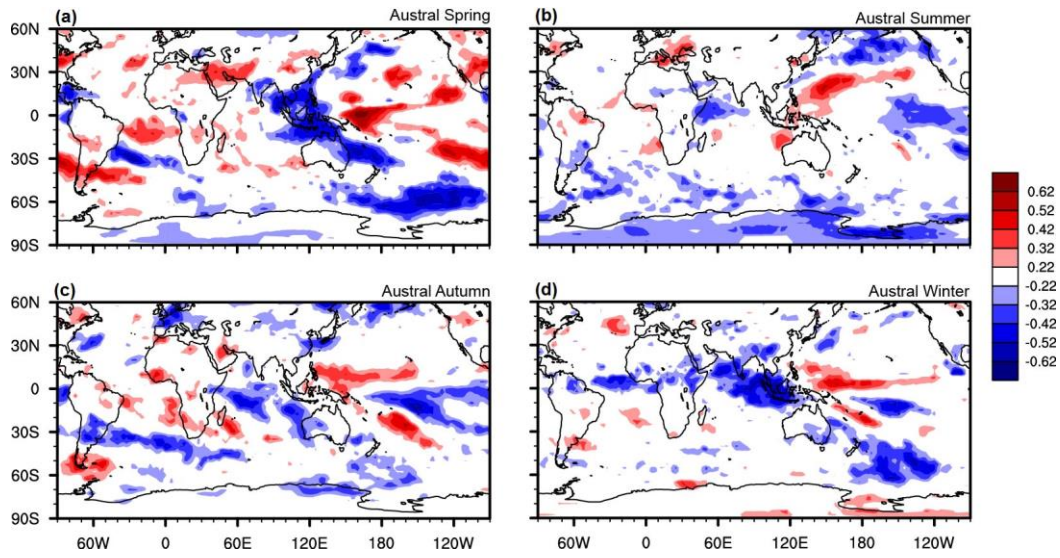
**Figure 4.** Ray paths (green lines) at 300 hPa in (a) austral spring, (b) austral summer, (c) austral autumn, and (d) austral winter. Red points denote wave sources in the marginal seas of East Asia (5°S–35°N, 100°E–140°E). The wavenumbers along these rays are in the range 1–5. The grey vectors indicate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

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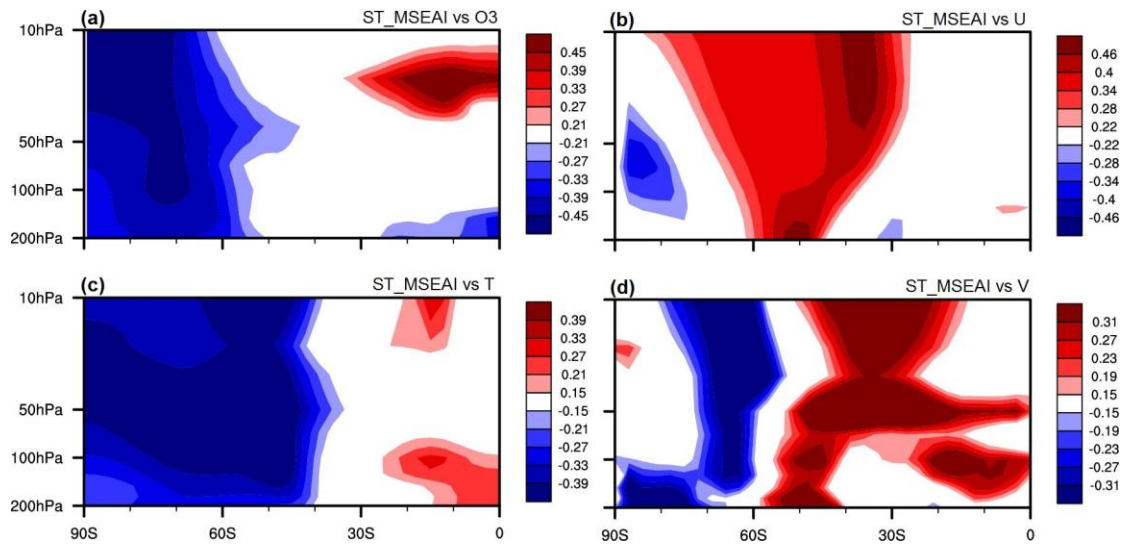
**Figure 5.** Correlation coefficients between the ST\_MSEAI and 300-hPa geopotential height from the ERA-Interim reanalysis in (a) austral spring, (b) austral summer, (c) austral autumn, and (d) austral winter between 1979 and 2015. Only statistical significance above 95% confidence level is colored. The seasonal cycles and linear trends were removed before calculating the correlation coefficients.



1

2 **Figure 6.** Same as Figure 5, but between the ST\_MSEAI and Outgoing longwave radiation from  
 3 NOAA.

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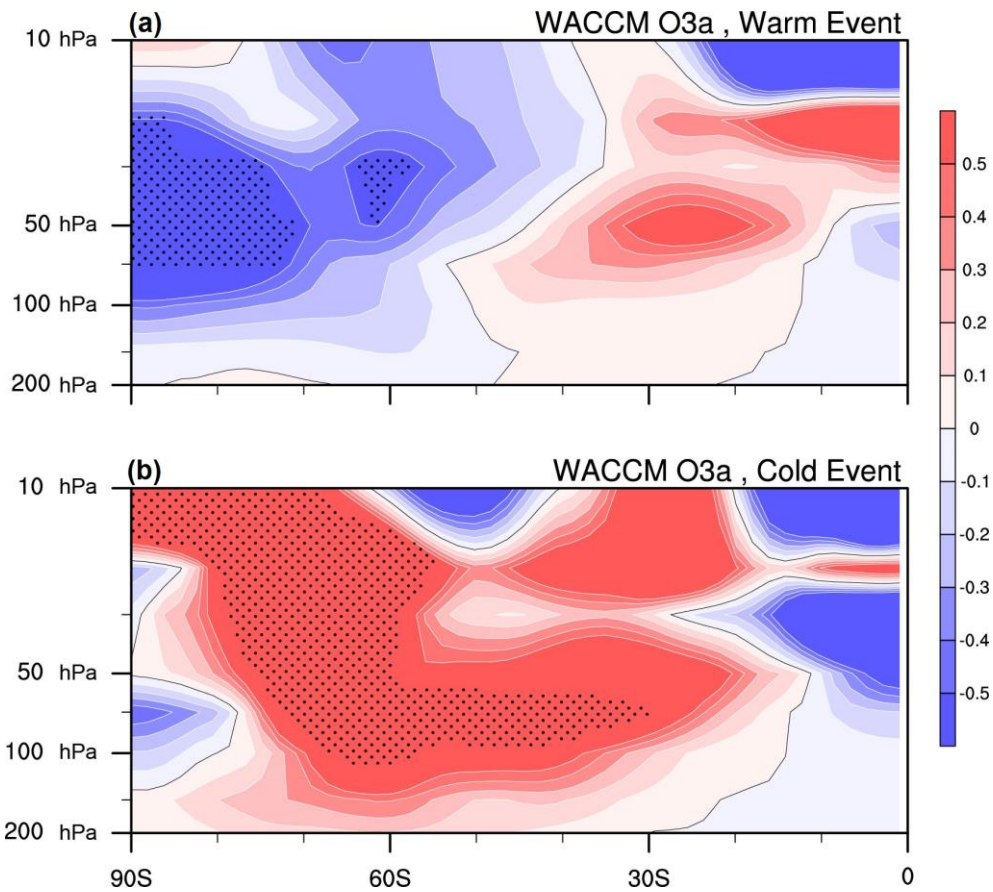


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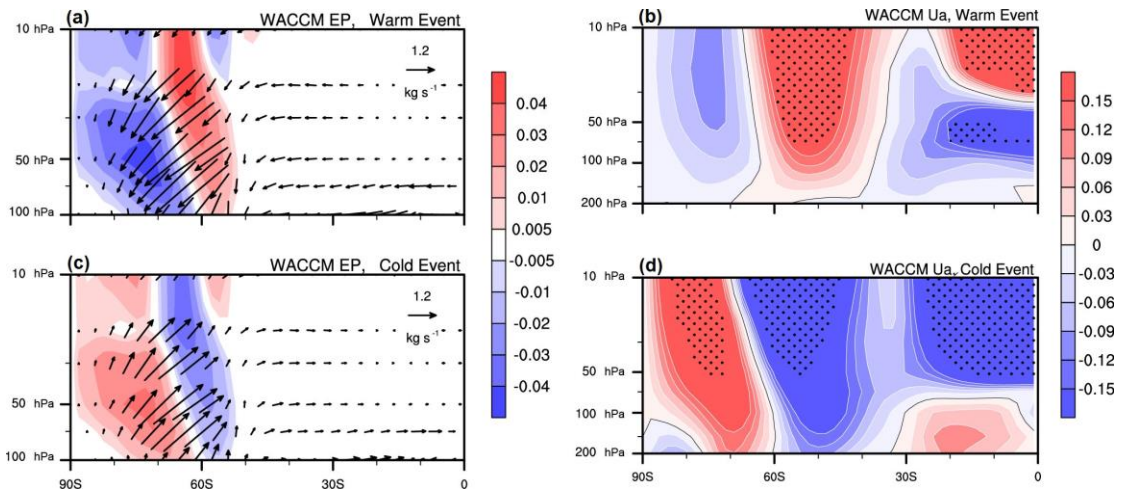
2 **Figure 7.** Correlation coefficients between ST\_MSEAI and (a) zonally averaged ozone, (b) zonal  
 3 wind, (c) temperature, and (d) meridional wind in austral spring. Wind and temperature from  
 4 ERA-Interim reanalysis data; ozone from MERRA2. Only statistical significance above 95%  
 5 confidence level is colored. The seasonal cycles and linear trends were removed before calculating  
 6 the correlation coefficients.

7



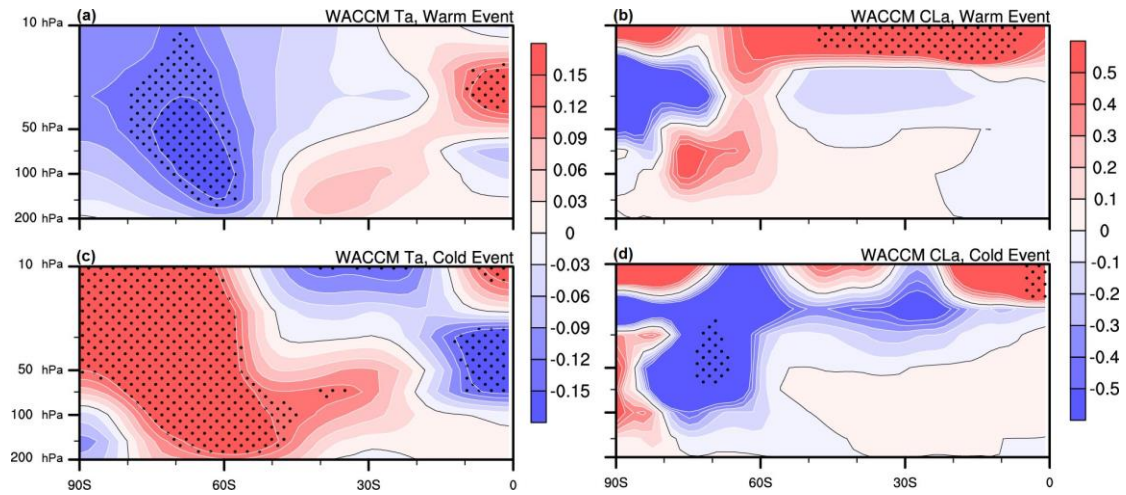


1  
 2 **Figure 8.** Zonal mean differences in ozone (ppmv) in austral spring between WACCM simulations  
 3 (a) S2 and S1, and (b) S3 and S1. Statistical significance above 95% confidence level is stippled.  
 4 Statistical significance of the simulated anomalies is calculated using the two-tailed Student's  
 5 t-test.



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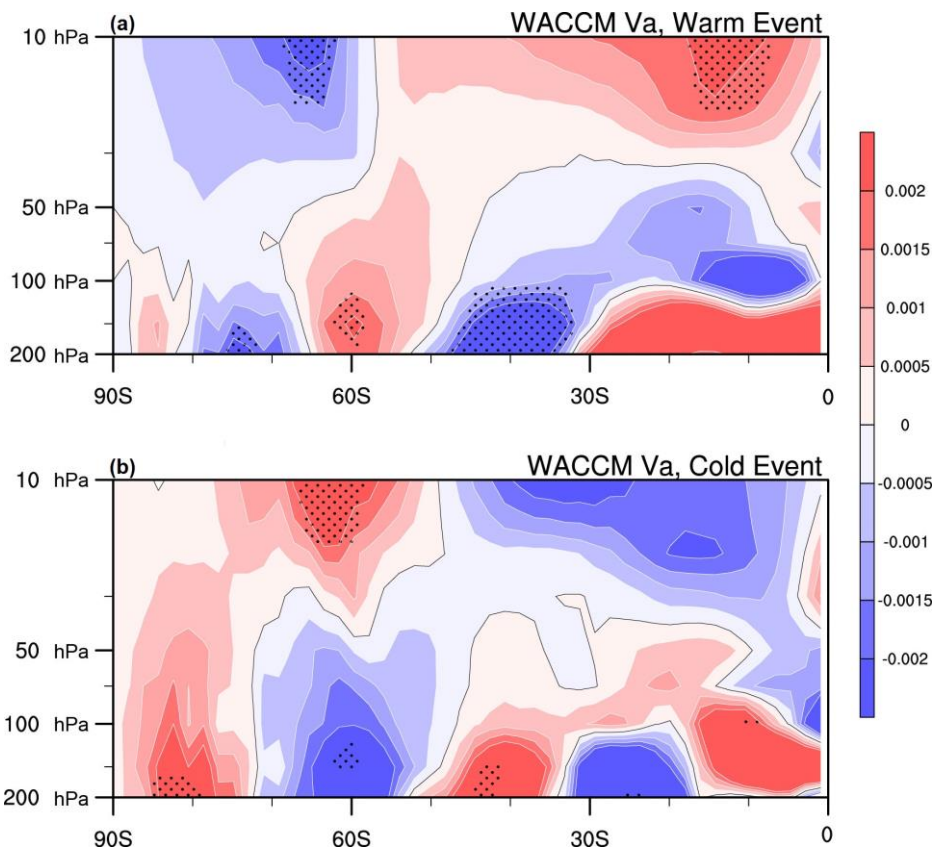
2 **Figure 9.** Differences in E–P flux vectors (black arrows) and divergence (color shading) in austral  
 3 spring between (a) S2 and S1, and (c) S3 and S1. Units for the horizontal and vertical vector  
 4 directions are  $10^7$  and  $10^5$   $\text{kg s}^{-1}$ , respectively. (b) and (d), as (a) and (c), but for zonal wind ( $\text{m s}^{-1}$ ).  
 5 Statistical significance above 95% confidence level is stippled.



1  
 2 **Figure 10.** Zonal mean difference in temperature (K) in austral spring between (a) S2 and S1, and  
 3 (c) S3 and S1. (b) and (d), as (a) and (c), but for active chlorine (ppbv). Statistical significance  
 4 above 95% confidence level is stippled.

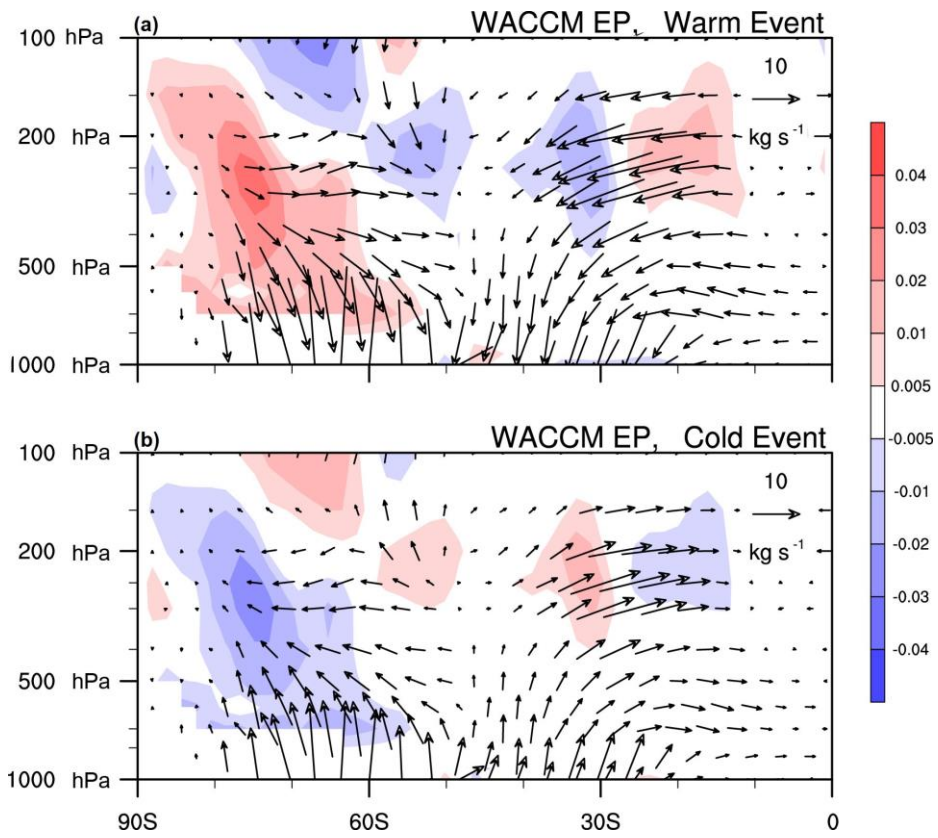


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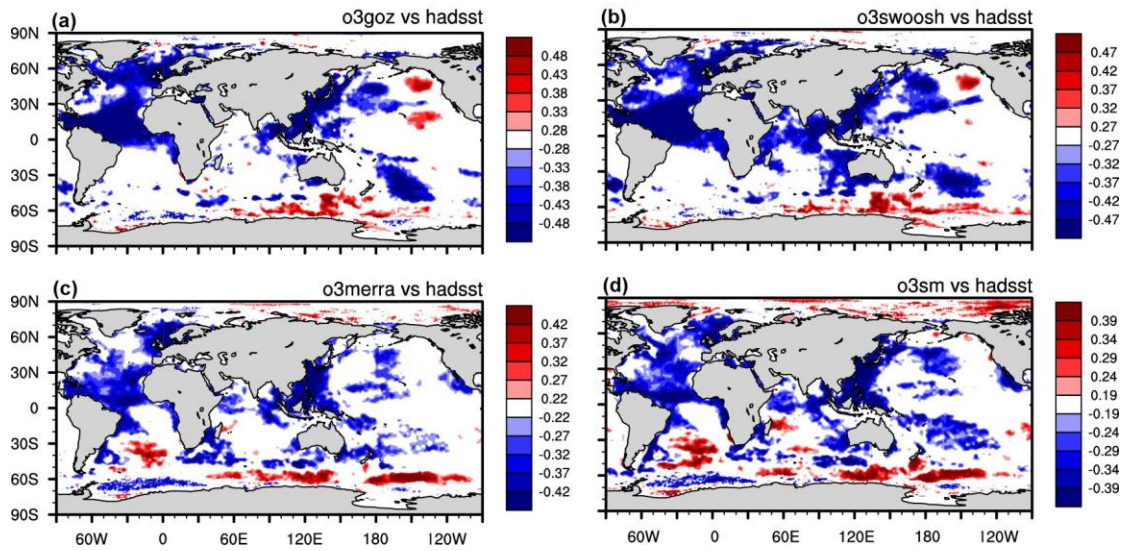
3 **Figure 11.** Zonal mean difference in TEM meridional wind ( $\text{m s}^{-1}$ ) in austral spring between (a)  
4 S2 and S1, and (b) S3 and S1. Statistical significance above 95% confidence level is stippled.



1

2 **Figure 12.** Same as Figure 9a and c, but for 1000 hPa to 100 hPa.

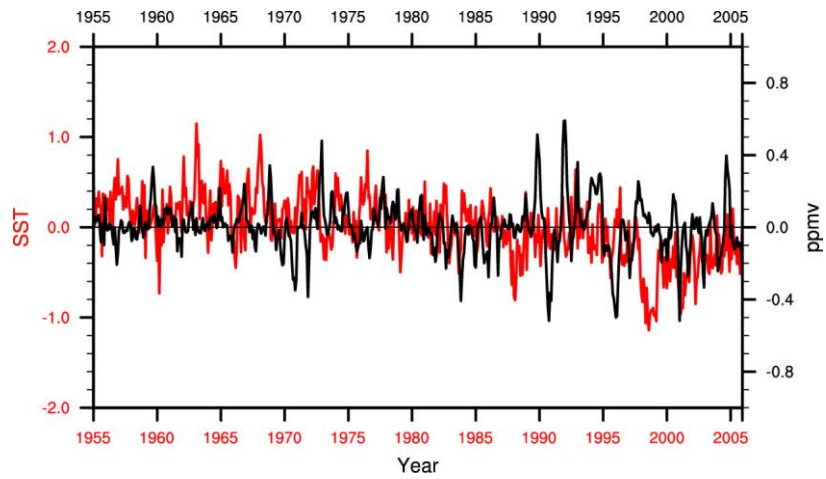
3



1

2 **Figure 13.** As Fig. 2, but with only the seasonal cycle removed before calculating the correlation

3 coefficients.



1

2 **Figure 14.** The difference in southern high latitude lower stratospheric ozone variations between  
 3 T1 and  $(T2+T3)/2$  (black line) and SST variations ( $\times -1$ ) in the marginal seas of East Asia ( $5^{\circ}\text{S}$ –  
 4  $35^{\circ}\text{N}$ ,  $100^{\circ}\text{E}$ – $140^{\circ}\text{E}$ ) based on the HadISST data (red line). The seasonal cycle is removed from  
 5 two time series.