Responses to Referees

Manuscript number: acp-2016-1053

Title: The Relationship between Lower Stratospheric Ozone in the Southern High Latitude and Sea Surface Temperature in the East Asia Marginal Seas

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Summary of changes for Referees and the Editor

We sincerely thank the reviewers for their important comments and the editor for their kind assistance with our manuscript. The major revisions are summarized as follows:

1. In the revised paper, we focus on investigating the relationship between lower stratospheric ozone in the southern high latitudes and sea surface temperature in the East Asian Marginal Seas **only** in austral spring. The title of the manuscript has been changed to "The Relationship between Lower Stratospheric Ozone in the Southern High Latitudes and Sea Surface Temperature in the East Asian Marginal Seas in Austral Spring".

2. Using two kinds of satellite ozone data (SWOOSH and GOZCARDS) to investigate the relationship between lower stratospheric ozone in the southern high latitudes and sea surface temperature in the East Asian Marginal Seas. The results are in good agreement with those from the MERRA2 reanalysis and SLIMCAT output. This improves the confidence level of the ozone–SST relationship in our study.

3. The significance of the results from the WACCM4 outputs is tested.

Response to Referee 1

This paper explores a possible linkage between the SST over East Asia Marginal Seas and the lower stratospheric ozone concentration over the Antarctica. The authors illustrated the correlation between the two from Reanalysis data and observation constrained model simulations. They explained it by the SST-excite planetary waves that propagate across the equator and influence the stratosphere, and validated the mechanism by simulations of CESM. What the authors proposed is a novel mechanism for the coupling between the stratosphere and the surface. However, I have some concerns about the robustness of their results, and hope the authors can address them.

Response: Thanks to the reviewer for sparing time to go through the manuscript, highlighting very important issues and providing helpful comments and valuable suggestions to improve the manuscript. We have revised the manuscript carefully according to the reviewer's comments and suggestions. The detailed point-to-point responses to the reviewer's comments are listed as follows.

Main comments:

1. *The ozone data. The ozone data employed in this study is from MERRA2 reanalysis and the TOMCAT/SLIMCAT simulations. While both data are constrained by observations in some way, they are not observations. Especially over southern high latitudes where direct observations are relatively sparse, the observational constrains would be quite weak. Since the study is build on the weak-but-significant correlation between the stratospheric ozone and the SST, it is important to establish the existence of such correlation and make sure it is not due to some artifact such as model biases. Satellite observations of ozone is widely used in previous studies, which the authors claimed to have used in the abstract but is not even mentioned in the main text. I understand that most satellite observations of ozone has no coverage during polar night, but the largest variation of stratospheric ozone over Antarctica usually occurs in austral spring when photo-chemistry is active and the dynamical coupling with the troposphere is strong, and the satellite does cover this season. Another possibility is to use the ozone-sonde observations. Antarctic stations such as South Pole and Syowa maintain ozone-sonde observations back to 1960s. If the authors can show consistent results from these more direct observations, even if the correlations may not be as strong, it would greatly improve the confidence level for the proposed ozone-SST relation.*

Response: Thanks for this important comment. In the revised paper, we have added two types of satellite ozone data to investigate the relationship between lower stratospheric ozone in the southern high latitudes and sea surface temperature in the East Asian Marginal Seas. One is the Global OZone Chemistry And Related trace gas

Data records for the Stratosphere (GOZCARDS) (Froidevaux et al. 2015) and the other is the Stratospheric Water and OzOne Satellite Homogenized (SWOOSH) ozone satellite data (Davis et al. 2016). The zonal mean satellite-based GOZCARDS is produced from high quality data from past missions (e.g., SAGE, HALOE data) as well as ongoing missions (ACE-FTS and Aura MLS). Its meridional resolution is 10° with 25 pressure levels from the surface up to 0.1 hPa. The zonal mean SWOOSH dataset is a merged record of stratospheric ozone and water vapor measurements taken by a number of limb sounding and solar occultation satellites (SAGE-II/III, UARS HALOE, UARS MLS, and Aura MLS instruments). Its meridional resolution is 2.5° with 31 pressure levels from 300 to 1 hPa.

Figure R1 shows the correlation coefficients between southern high latitude lower stratospheric ozone variations from the four ozone datasets and SST from HadISST in austral spring. It is apparent that the regions of significant correlation are generally different for the four ozone datasets except for the East Asian Marginal Seas; i.e., 5°S–35°N, 100°E–140°E, where the most significant correlations between Antarctic stratospheric ozone variations and SST are seen in all four ozone datasets. This result improves the confidence level of the ozone–SST relationship in our study.

Figure R1 is Figure 2 in the revised paper.

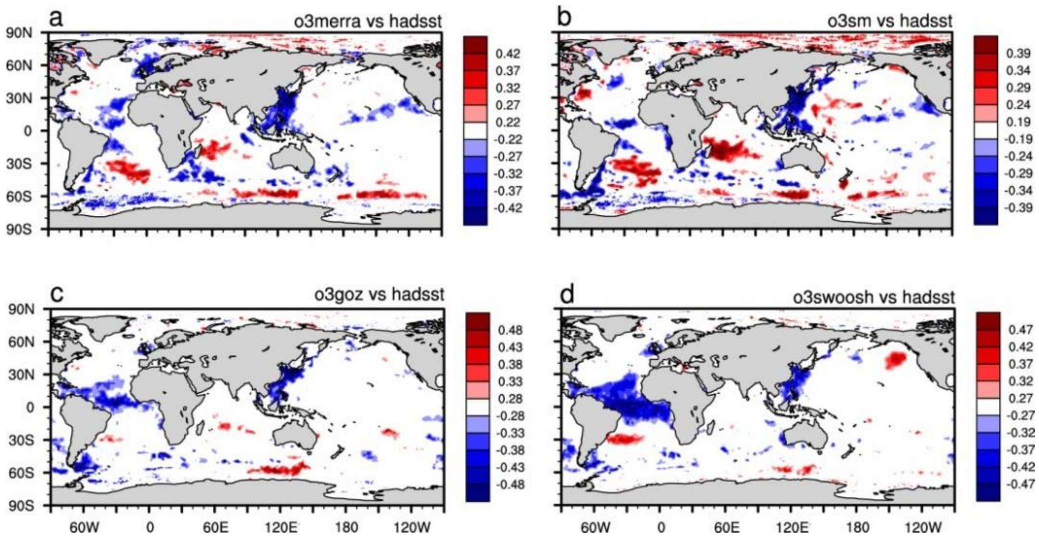

Figure R1. Correlation coefficients between southern high latitude lower stratospheric ozone variations and SST from HadISST in austral spring. The ozone variations are averaged over the region 60–90°S at 200–50 hPa in austral spring. (a) Ozone from MERRA2 and (b) ozone from SLIMCAT for 1979–2015. (c) Ozone from GOZCARDS for 1979–2012. (d) Ozone from SWOOSH for 1984–2015. Only regions with statistical significance above the 95% confidence level are colored; statistical significance is calculated using the two-tailed Student's *t*-test and the N^{eff} of DOF. The seasonal cycles and linear trends were removed prior to calculating the correlation coefficients.

References:

Davis S M et al. 2016, The Stratospheric Water and Ozone Satellite Homogenized (SWOOSH) database: A long-term database for climate studies, *Earth Syst. Sci. Data*, **8**, 461–490.

Froidevaux L et al. 2015, Global OZone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS): methodology and sample results with a focus on HCl, H2O, and O3, *Atmos. Chem. Phys.* **15(18)**, 10471–10507.

2. *The authors showed results from time-slice and transient simulations and the quantify the contribution to ozone trends from SST warming based on these simulations. It is comforting to see these model simulations to be consistent with reanalysis, but the authors have not taken the full advantage of model simulations to establish the robustness of the results. Given that the SST-related stratospheric signal is usually small compared to the random internal variations, one may wonder if some of the signal appears just by chance. For example, the time series in Fig. 13(c) probably does not have a statistically significant trend judging by eyes. For the time-slice simulations, it should be straightforward to quantify the statistical significance. For the transient simulations, since the authors have two ensemble member (T2 and T3), it would be nice to show how different are the two member. If the difference between these two member is larger than the signal (T1-(T2+T3)/2), then the T1-(T2+T3)/2 may contain considerable contribution from random noise besides the SST over East Asia Marginal Seas, then the 17% contribution to ozone depletion trends may also be questionable.*

Response: Thanks for this comment. In the revised paper, we have calculated the significance of the results from WACCM4 simulations.

For the trend of the time series in Fig. 13c, the corresponding Table 4 in the original manuscript has been revised as Table R1 below. Note that the significance of the trends has been calculated.

Table R1. Linear trends of ozone variations over the region 200–50 hPa and 60–90°S from 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)	-1.2×10^{-3} ppmv/month**
Linear trend of ozone variations over the region 200–50 hPa and 60–90°S from (T1 – (T2+T3)/2) (Trend2)	-0.204×10^{-3} ppmv/month*

**: the trend is significant at 99% confident level. *: the trend is significant at 95% confident level. The calculation of the statistical significance of the trend uses the two-tailed Student's *t*-test.

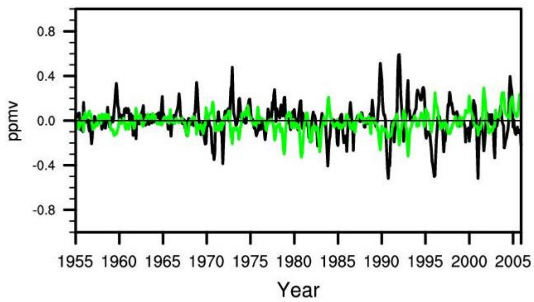

Figure R2. The differences in southern high latitude lower stratospheric ozone variations between T2 and T3 (green) and between T1 and (T2+T3)/2) (black), averaged over the region 60–90°S at 200–50 hPa.

Figure R2 shows the differences in southern high latitude lower stratospheric ozone variations between T2 and T3 and between T1 and (T2+T3)/2). It is evident that the difference between T2 and T3 is much smaller than that between T1 and (T2+T3)/2). This means that the difference between T1 and (T2+T3)/2) is not random but mainly originates from the SST forcing over the East Asian Marginal Seas.

For the results from the time-slice simulations, the statistical significance of the simulated anomalies is also calculated using the two-tailed Student's *t*-test. Figures 8–11 in the original manuscript have been replotted as Figures R3–R6 below.

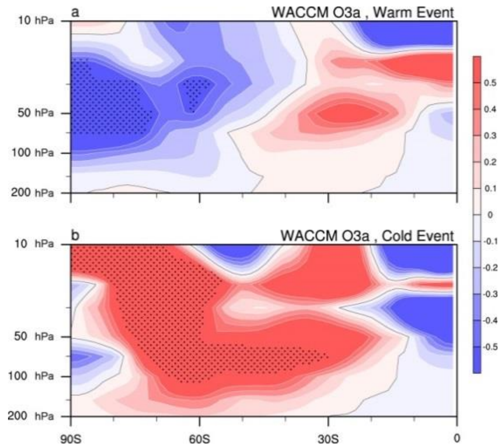

Figure R3. Zonal mean differences in ozone (ppmv) in austral spring between WACCM simulations (a) S2 and S1, and (b) S3 and S1. Statistical significance above 95% confident level is stippled. Statistical significance of the simulated anomalies is calculated using the two-tailed Student's t-test.

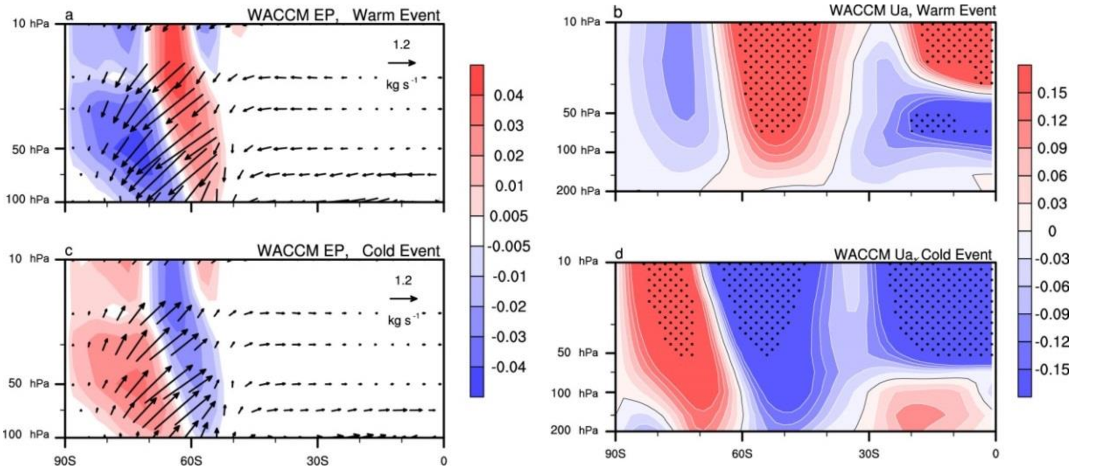

Figure R4. Differences in E–P flux vectors (black arrows) and divergence (color shading) in austral spring between (a) S2 and S1, and (c) S3 and S1. Units for the horizontal and vertical vector directions are 10^7 and 10^5 kg s^{-1}, respectively. (b) and (d), as (a) and (c), but for zonal wind (m s^{-1}). Statistical significance above 95% confident level is stippled.

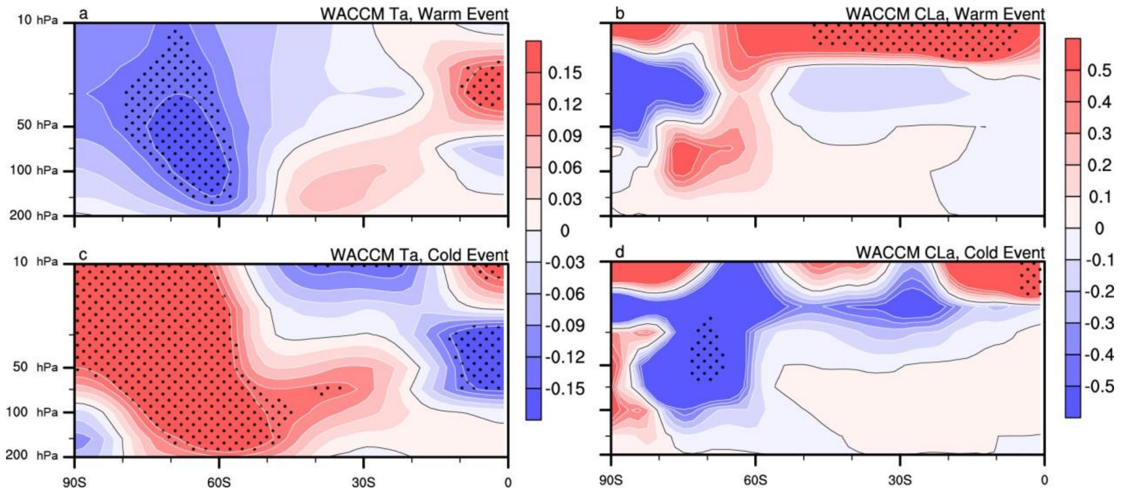

Figure R5. Zonal mean difference in temperature (K) in austral spring between (a) S2 and S1, and (c) S3 and S1. (b) and (d), as (a) and (c), but for active chlorine (ppbv). Statistical significance above 95% confident level is stippled.

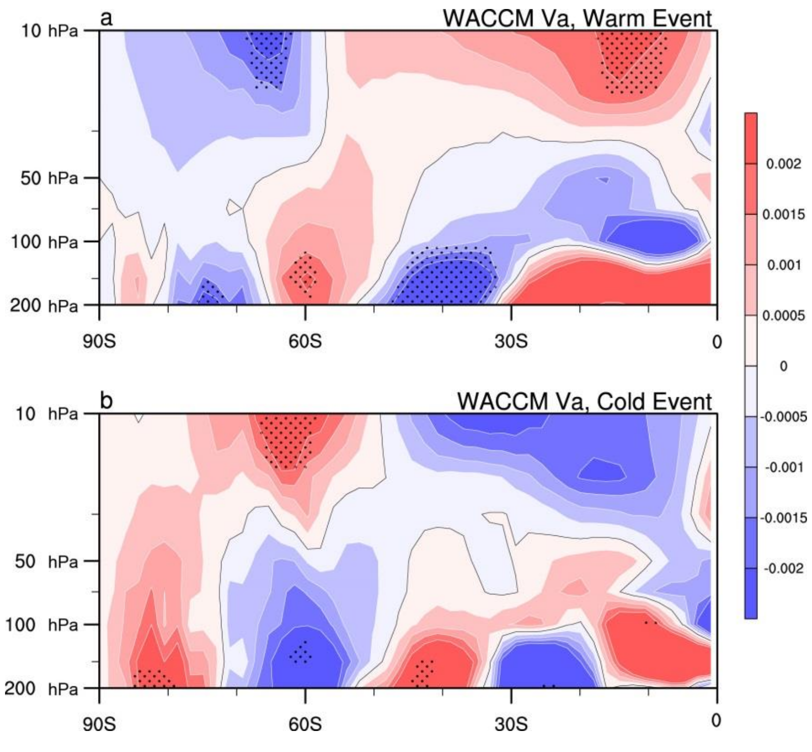

Figure R6. Zonal mean difference in meridional wind (m s^{-1}) in austral spring between (a) S2 and S1, and (b) S3 and S1. Statistical significance above 95% confident level is stippled.

Minor points:

1. *It is not clear whether you are referring to the boreal or austral seasons throughout the paper since both hemispheres are involved in the paper. It is important to clarify this, because if the authors mean austral seasons, then the mechanisms do* *not work. The cross-equator propagation of anomalous planetary waves only occurs in austral summer and autumn, but that is the season when Southern Hemisphere stratosphere is dominated by easterlies and the prohibits vertical wave propagation into the stratosphere.*

Response: Thanks for the comment. When we discuss the mechanism of cross-equatorial propagation of anomalous planetary waves, the seasons are boreal summer and autumn. In the revised paper, we have unified the description of the season in the whole manuscript. Sorry for the confusion.

2. *P6 L23: "Rieder et al. 2014, Zhang et al. 2015b" The authors cited these references to support the argument that MERRA2 ozone compares well with satellite observations. However, Rieder et al. did not even mention satellite observations. Zhang et al only compared data over China, which is not the region of interest in this paper.*

Response: Thanks for the comment. We have deleted these two references and added Wargan et al. (2017) in the revised paper. In addition, we have replotted Figure 1, which now includes the comparison of MERRA2 ozone with satellite ozone observations. Please see Figure R7a below. The time series of southern high latitude lower stratospheric ozone variations from MERRA2 is significantly correlated with that from GOZCARDS ozone (r = 0.54) and SWOOSH ozone (r = 0.50). In austral spring, these correlation coefficients increase to 0.72 and 0.77, respectively (Figure R7b). This illustrates that the variations of southern high latitude lower stratospheric ozone from MERRA2 compare well with those from satellite observations.

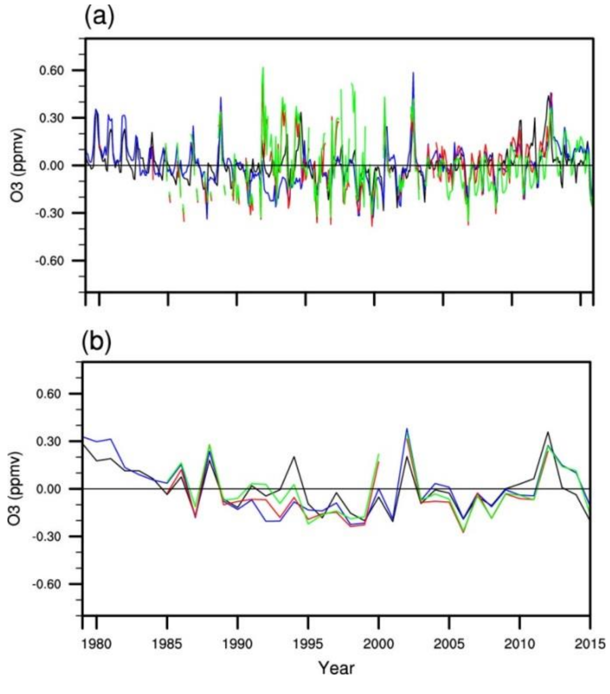

Figure R7. (a) Time series of southern high latitude lower stratospheric ozone variations averaged over the region 60–90°S at 200–50 hPa from the MERRA2 (black line), SLIMCAT (blue line), GOZCARDS (red line) and SWOOSH (green line) ozone datasets. (b) Same as (a), but only for austral spring. The seasonal cycles and linear trends are removed.

Reference:

Wargan K., G. Labow, S. Frith, S. Pawson, N. Livesey, and G. Partyka. 2017. Evaluation of the Ozone Fields in NASA's MERRA-2 Reanalysis. J. Climate., DOI: http://dx.doi.org/10.1175/JCLI-D-16-0699.1

3. *P7 L7-9 "Figure 1 shows . . . past five decades" What do you mean by "ozone variations" here? Do you mean anomalies (with seasonal cycle removed)? Also, there is only 36 years not five decades.*

Response: Thanks for the careful check. Yes, the "ozone variations" are calculated by removing the seasonal cycles and linear trends. And, the "in the past five decades" to "in the past three decades".

4. *P7 L11-13: The authors claimed the difference between MERRA2 and SLIMCAT ozone is small, but from the Fig. 1, the magnitudes of the difference is comparable to the magnitudes of ozone anomalies itself. Also, as the author stated later, "the regions of significant correlation are generally different for the two ozone datasets", which is another proof that the difference between the two datasets is not small.*

Response: Thanks for the comment. The differences between MERRA2 and SLIMCAT ozone in some periods are not small. We have deleted the relevant sentences in the revised paper. Since the variations of the southern high latitude lower stratospheric ozone from these two datasets in the past three decades are highly correlated, the differences between that two dataset should have no significant influence on our result.

5. *P11 L12-14: "In addition, .. this location" From the Fig. 5, it looks like most wave rays reaching 60S do not stop there, but refract to lower latitudes.*

Response: Thanks for the comment. The sentence has been rewritten as:

"These rays can reach about 60°S and can then be refracted to lower latitudes, which implies that the pathway of upward propagation of tropospheric waves from the marginal seas of East Asia possibly extends to 60°S."

6. *P13 L18: "zonal circulation" I think you mean meridional circulation or zonal-mean circulation. Note that the meridional circulation in the stratosphere is better described by the Transformed Eulerian Mean (TEM) velocity rather than the conventional meridional wind, because the TEM formula includes the contribution from eddies, which is often important in the stratosphere.*

Response: Thanks for the comment. On page 13 line 18, this is really "zonal circulation". We attempt to demonstrate that a stronger (weaker) southern high latitude lower stratospheric zonal circulation, which is related to the SST changes in the East Asian Marginal Seas, impedes (promotes) southern high latitude lower stratospheric meridional circulation. This is equivalent to stronger (weaker) zonal circulation impeding (promoting) transport of ozone from the middle latitude stratosphere to the high latitude stratosphere.

In Figure 7d, we used conventional meridional wind to calculate the correlation between meridional wind and SST variations. In the revised paper, we have used the transformed Eulerian-mean (TEM) meridional wind (v^*), which is given by Edmon et al. [1980]:

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

where θ is the potential temperature, v is meridional wind and subscript p denotes derivative with pressure p. The overbar denotes the zonal mean and the prime denotes deviations from the zonal mean value.

7. *P14 L4-7: "This may be . . . wave activity." This discussion on how SST warming over East Asia Marginal Seas leads to a weaker wave activity in the Southern Hemisphere stratosphere is very puzzling. Can you support your arguments with more evidence, such as observations of convective activity?*

Response: Thanks for this important comment. The discussion is indeed puzzling. Figure R8 shows the correlation between SST changes in the East Asian Marginal Seas and OLR (outgoing longwave radiation) in four seasons. It is found that the correlation coefficients are the largest over East Asian Marginal Seas only in austral spring. This corresponds to the relationship we found between SST changes in the East Asian Marginal Seas and southern high latitude lower stratospheric ozone being very strong in austral spring.

However, it should be pointed out that the negative correlation coefficient between SST changes and OLR over the East Asian Marginal Seas does not support our argument for SST warming (cooling) over the East Asian Marginal Seas creating less (more) convection due to weakened (enhanced) sea–land contrast along the coastline of East Asia in austral spring. We have deleted this paragraph in the revised paper.

It is found that there is enhanced E-P flux from lower latitudes to southern high latitudes in the SST warming event over the East Asian Marginal Seas (Figure R9a). However, this increased EP flux does not propagate upward into the stratosphere but downward to lower levels, and *vice versa* for the SST cooling event. Fig. R9 explains why SST warming (cooling) over the East Asian Marginal Seas leads to a weaker (stronger) wave activity in the Southern Hemisphere stratosphere.

This explanation and Figures R8 and R9 have been added in the revised paper.

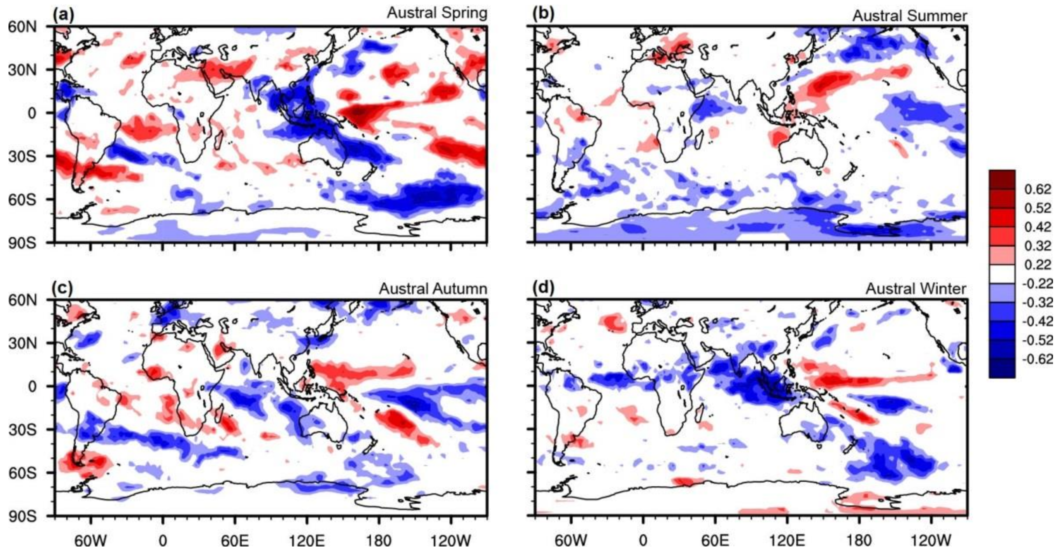

Figure R8. Correlation coefficients between the ST_MSEA index and OLR from NOAA in (a) austral spring, (b) austral summer, (c) austral autumn, and (d) austral winter between 1975 and 2013. Only significant correlations are colored. The seasonal cycles and linear trends were removed before calculating the correlation coefficients.

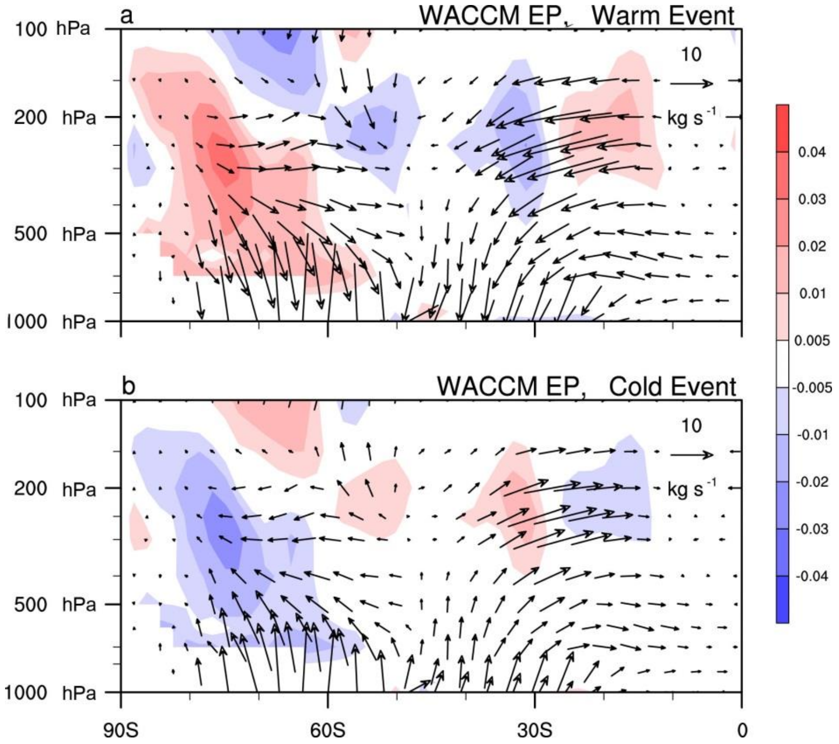

Figure R9. Differences in E–P flux vectors (black arrows) and divergence (color shading) at 1000 to 100 hPa between (a) S2 and S1, and (c) S3 and S1. Units for the horizontal and vertical vectors are 10^7 and 10^5 kg s^{-1}, respectively. Details of experiments S1–3 are given in Table 2 in the manuscript.

8. *Figure 9. Can you also show the EP flux anomalies in the troposphere? Does the tropospheric EP flux anomalies support your proposed mechanism?*

Response: Please see Figure R9 in above **Response 7**.

9. *Figure 13 caption: "SST variations (x-1)" I don't think you timed SST variations by -1 in the figure, as it shows increasing and warming trends. Also the unit for panel (d) should be K.*

Response: Thanks for the careful check. The Figure 13 has been replotted in the revised paper.

Response to Referee 2

In this study, the authors observed a correlation between sea surface temperature over East Asian Marginal Seas and ozone in the southern high latitude lower stratosphere (the targeted region) from assimilated MERRA data and a model simulation (SLIMCAT). Using the WACCM model (WACCM4) and a defined temperature index from the HadISST data, they separated warm and cold events over the East Asian Marginal Seas and found distinct differences between the two groups of the events in ozone concentrations in targeted region. The model simulation further reveals large differences,generally in the opposite directions, between the warm and clod events in dynamic and chemical conditions that modulate transport and formation/depletion of stratospheric ozone. Finally, the impact of such a connection on the ozone trend in the targeted region was quantified with a series of numerical experiments using WACCM4. The authors attributed 17% of decreasing ozone trend in the targeted region to increasing SST over the marginal seas of East Asia. The authors proposed a hypothesis that establishes the connection between SST variation over the marginal seas of East Asia and ozone in the targeted region (P12-13). The paper is well written in articulating the connection and explaining the hypothesis. The proposed connection is novel and important. I suggest that the authors use some additional datasets to confirm or adjust their proposal. These data include ozonesonde data (there are about 7 stations over the Antarctic region during different periods), the TOST data (the Trajectory-mapped Ozonesonde dataset for the Stratosphere and Troposphere), and satellite data (although satellite data quality usually decreases with latitude). For the long-term ozone trend over the targeted region, the ozone concentrations from MERRA, SLIMCAT should be compared with the WACCM simulation. More importantly, all simulated or assimilated data can be compared with observations so the estimated 17% contribution of increasing SST over the marginal seas of East Asia to the ozone trend in the targeted region can be confirmed or refined.

Response: We thank the reviewer for the positive evaluation of our study and we sincerely appreciate the reviewer’s very helpful comments, which have helped us to greatly improve our paper. We have revised the manuscript carefully according to the reviewer’s comments and suggestions.

According to the referee’s comment, the major revision is using the observed ozone to confirm the relationship between SST changes in the East Asian Marginal Seas and southern high latitude lower stratospheric ozone. Since the satellite ozone observations cover wider range compared with Ozonesonde data, we have added two types of satellite ozone data to investigate the relationship between the lower stratospheric ozone in the southern high latitudes and the sea surface temperature in the East Asian Marginal Seas. One is the Global OZone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS) (Froidevaux et al. 2015) and the other is the Stratospheric Water and OzOne Satellite Homogenized (SWOOSH) ozone satellite data (Davis et al. 2016). The zonal mean satellite-based GOZCARDS is produced from high quality data from past missions (e.g., SAGE, HALOE data) as well as ongoing missions (ACE-FTS and Aura MLS). Its meridional resolution is 10° with 25 pressure levels from the surface up to 0.1 hPa. The zonal mean SWOOSH dataset is a merged record of stratospheric ozone and water vapor measurements taken by a number of limb sounding and solar occultation satellites (SAGE-II/III, UARS HALOE, UARS MLS, and Aura MLS instruments). Its meridional resolution is 2.5° with 31 pressure levels from 300 to 1 hPa.

Figure RR1 shows the correlation coefficients between southern high latitude lower stratospheric ozone variations from the four ozone datasets and SST from HadISST in austral spring. It is apparent from Figure RR1 that the regions of significant correlation are generally different for the four ozone datasets except for the East Asian Marginal Seas; i.e., 5°S–35°N, 100°E–140°E, where the most significant correlations between Antarctic stratospheric ozone variations and SST are seen in all four ozone datasets. This result improves the confidence level of the ozone–SST relationship in our study.

Figure RR1 is Figure 2 in the revised paper.

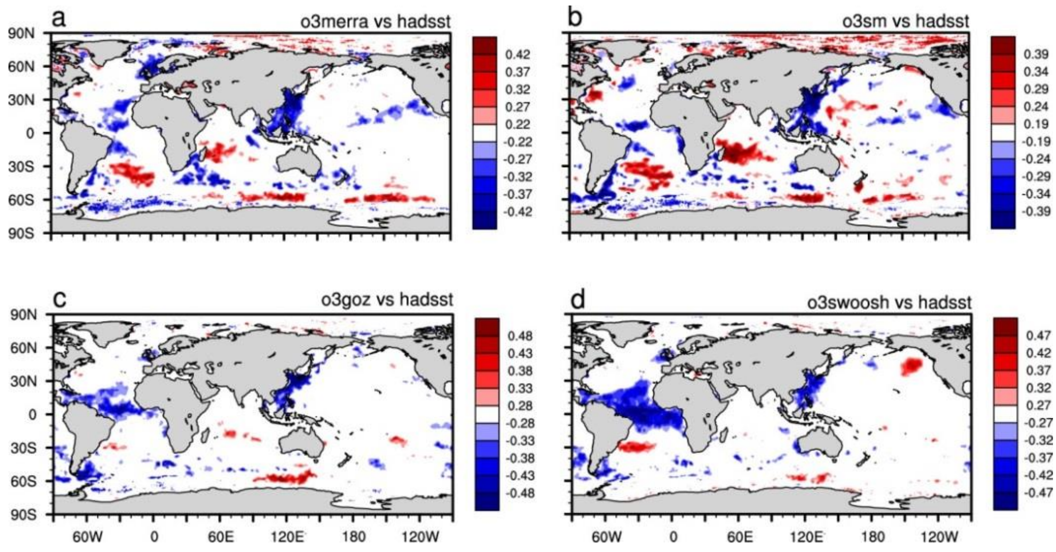

Figure RR1. Correlation coefficients between southern high latitude lower stratospheric ozone variations and SST from HadISST in austral spring. The ozone variations are averaged over the region 60–90°S at 200–50 hPa in austral spring. (a) Ozone from MERRA2 and (b) ozone from SLIMCAT for 1979–2015. (c) Ozone from GOZCARDS for 1979–2012. (d) Ozone from SWOOSH for 1984–2015. Only regions with statistical significance above the 95% confidence level are colored; statistical significance is calculated using the two-tailed Student's *t*-test and the N^{eff} of DOF. The seasonal cycles and linear trends were removed prior to calculating the correlation coefficients.

References:

Davis S M et al. 2016, The Stratospheric Water and Ozone Satellite Homogenized (SWOOSH) database: A long-term database for climate studies, *Earth Syst. Sci. Data*, **8**, 461–490.
Froidevaux L et al. 2015, Global OZone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS): methodology and sample results with a focus on HCl, H2O, and O3, *Atmos. Chem. Phys.* **15(18)**, 10471–10507.

The authors also looked into the seasonal variation of the proposed connection in some aspects (P13, L 20-22, Figures 5-6). Does removing the seasonal cycle enhance or smooth the signal of this connection? As some lags appear in the MERRA data (Figure 4), will the connection be more significant if the authors use monthly data with consideration of the lags?

Response: Thanks for the comment. In the analysis of Figure 6 of the original manuscript, the seasonal cycle had been removed. In line with the comment, Figure 6 has been replotted as Figure RR2. It is found that there is no obvious difference between the new figure and Figure 6.

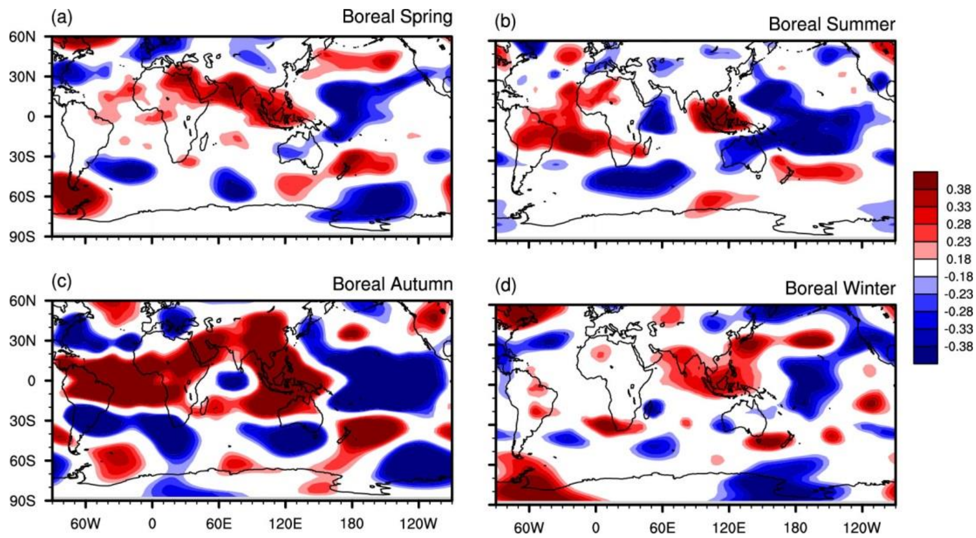

Figure RR2. Correlation coefficients between the ST_MSEA index and 300-hPa geopotential height in the four seasons. When calculating the correlation coefficient in one season, the ST_MSEA index is averaged over the first two months and the geopotential height averaged over the last two months. For example, when calculating the correlation coefficient in spring, the ST_MSEA index uses the average for March and April and geopotential height uses the average for April and May.

Specific

1. *P2, L2, no satellite data are directly used in this study.*

Response: Satellite datasets are added in the revised paper. Thanks.

2. *P3, L2, in this and other places (e.g., P7, L8-9, P16, L7-9), the authors stated similar sentences like "Ozone variations over recent decades exhibit a strong* *decreasing trend. . .". This may be the case for the Antarctic region, which is the focus of this study. It is not necessarily the case for other regions. In the Northern Hemisphere, stratospheric ozone recovery has been observed since the late 1990s after the Montreal Protocol and its amendments, although some surprising declines in ozone there were observed in recent years. So, please be specific.*

Response: Thanks. We have revised these sentences as following:

"Ozone variations over recent decades exhibit not only strong trends, ……"

The relevant text in other places in the manuscript are also modified accordingly.

3. *P6, L4, what is the horizontal resolution for MERRA2 data used in this study, 2_x2.5_? How about SLIMCAT?*

Response: The horizontal resolution (lon × lat) for MERRA2 ozone is $1.25^{o} \times 1.25^{o}$and for SLIMCAT ozone is $5.625^{o} \times 5.5^{o}$. It has been clarified in the revised paper.

4. *P8, L4-6, what is the significant level? 90%? 95%?*

Response: It is at 95% confidence level. This has been stated in the captions of corresponding figures.

5. *P8, L17, add "phi (using the Greek letter) is latitude".*

Response: Thanks. Added.

6. *P13, L20-22, not shown?*

Response: Yes, it is not shown. It is stated in the revised paper.

7. *P14, L4-6, please rephrase the sentence.*

Response: According to referee 1's comment. This sentence has been deleted in the revised paper. The discussion on how SST warming over East Asian Marginal Seas leads to a weaker wave activity in the Southern Hemisphere stratosphere please see Page 16 Line 22 to Page 17 Line 8 in the revised paper.

8. *P11, L11, use "further support" to replace "validate".*

Response: Replaced. Thanks.

9. *P18, L19, "observation"? No observation data are directly shown in this paper. The MERRA data may not be taken as "observation".*

Response: The corresponding sentence has revised to as below, the “observation” changed to “statistical analysis”.

“The above results are based on statistical analysis but are also supported by time-slice experiments conducted using the CESM.”

10. *Indicate whether boreal or austral seasons (including months) are referred earlier in the paper.*

Response: In the revised paper, we have unified the description of the season in the whole manuscript. Sorry for the confusion.

11. *Figure 1, is the ozone variation the same as or different from ozone anomaly? Is this normalized ozone anomaly? Are the seasonal cycle and trend removed? If not, add a trend to the figure. Indicate if the trend is significant. The variation is not straightly downward. A slight increase in ozone appears during 2010-2015.*

Response: The ozone anomaly variations in Figure 1 have the seasonal cycle and trend removed. This is why the ozone anomaly variations do not uniformly decrease before 2000. As we mainly focus on investigating the interannual variability of the ozone in the paper, we removed the long-tern linear trend of the ozone time series. The above information has been clarified in all relevant figures in the revised paper.

12. *Figures 2, 6, and 10, what is the significant level?*

Response: It is at 95% confidence level. This has been stated in the captions of corresponding figures.

13. *Figure 4, the label for the x-axis is Lag (month). Is the long-term trend removed fromthe data?*

Response: Yes, the long-term trend is removed. This information has been stated in all relevant figures in the revised paper.

14. *Figure 6, indicate these are boreal or austral spring, summer, autumn and winter, including months. Should the seasonal cycles be removed?*

Response: In the revised paper, we have unified the description of the season in the whole manuscript. Sorry for the confusion. Yes, the seasonal cycle is removed.

15. *Figures 7-11, the y-axis is not in the same format. Some have no unit, and some no label.*

Response: All the figures are corrected. Thanks very much for the carefully check.

16. *Figure 9, the annotation for the arrow is too small to see clearly.*

Response: The annotation has been enlarged. Thanks.

17. *Figure 13, the unit for SST variation should be K as shown in an earlier version. Also, please provide label for the y-axes. Is the ozone variation the same as Figure 1? Or the ozone trend in Figure 1 is removed? Why are they different?*

Response: Thanks for the comment. We are sorry for the confusion in Figure 13. We have made the caption of Figure 13 clearer in the revised paper.

The unit for SST has been modified to "K". The ozone variations in Figure 13 are different from those in Figure 1. In Figure 1, the ozone variations come from MERRA2 and SLIMCAT ozone from 1979 to 2015. The ozone in Figure 13 is the output from the WACCM4 transient experiments from 1955 to 2005. As we explained in **Response 11**, the ozone variations in Figure 1 have the linear trend removed to investigate the interannual variability. Figure 13 is shown to investigate the trend, so the ozone trend is not removed.

18. *There are a few inconsistency in the reference format. For example, some capitalize each word, some not.*

Response: The reference format are checked and corrected. Thanks.

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

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Abstract

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

1. Introduction

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

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

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

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

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

Over recent decades, SST in the East Asian Marginal Seas has exhibited an increasing trend with strong interannual variations (Zheng et al., 2014). Zhao et al. (2015, 2016) pointed out that Rossby waves generated by variations in the SST of the South China Sea can cross the equator and propagate towards to southern middle to high latitudes in austral spring. It is likely that the Rossby waves generated by SST changes in austral spring in the vicinity of the East Asian Marginal Seas can cross the equator to the Southern Hemisphere and regulate austral spring ozone levels in the southern high latitude stratosphere via their influence on the southern stratospheric circulation. Therefore, it is worthwhile to examine the potential connections between SST variations over the East Asian Marginal Seas and southern high latitude lower stratospheric ozone variations. The remainder of the paper is organized as follows. The data, method and model used are introduced and briefly described in section 2. Section 3 analyzes the connection between the East Asian Marginal Seas and southern high latitude lower stratospheric ozone. Section 4 presents and discusses the simulations of the connection. Finally, the results are summarized and conclusions drawn in section 5.

2. Data, Model, and Methods

The ozone data used in this study is obtained from the NASA Modern Era Retrospective Analysis for Research and Applications (MERRA) dataset version 2 (Rienecker et al., 2011), TOMCAT/SLIMCAT 3-D model simulations (Chipperfield, 2006), Global OZone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS) ozone satellite data (Froidevaux et al. 2015) and The Stratospheric Water and OzOne Satellite Homogenized (SWOOSH) ozone satellite data (Davis et al. 2016). The MERRA2 data (lon $\times$ lat: $1.25^{o} \times 1.25^{o}$) has 42 pressure levels from the surface up to 0.1 hPa. The vertical resolution of MERRA2 is ~1–2 km in the UTLS and 2–4 km in the middle and upper stratosphere. MERRA2 is assimilated by the Goddard Earth Observing System Model, Version 5 (GEOS-5) with ozone from the Solar Backscattered Ultra Violet (SBUV) radiometers from October 1978 to October 2004, and thereafter from the Ozone Monitoring Instrument (OMI) and AURA Microwave Limb Sounder (MLS) (Bosilovich et al., 2015). The MERRA2 reanalysis ozone data compares well with satellite ozone observations (Wargan et al., 2017) and shows a better representation of the QBO and stratospheric ozone compared to MERRA1 (Coy et al., 2016). In the present study, the ozone field (lon × lat: 5.625^{o} × 5.5^{o}) simulated by a 3D offline chemical transport model, SLIMCAT (Feng et al., 2007, 2011), is also used. The simulation performed in this study is driven by horizontal winds and temperatures from meteorological analyses of the ERA-Interim data provided by European Centre for Medium-Range Weather Forecasts (ECMWF) (Dee et al., 2011). The vertical advection in the model is calculated from the divergence of the horizontal mass flux (Chipperfield, 2006), and chemical tracers are advected by the conservation of second-order moments (Prather, 1986). The zonal mean satellite-based GOZCARDS is produced from high quality data from past missions (e.g., SAGE, HALOE data) as well as ongoing missions (ACE-FTS and Aura MLS). Its meridional resolution is 10° with 25 pressure levels from the surface up to 0.1 hPa. The zonal mean SWOOSH dataset is a merged record of stratospheric ozone and water vapor measurements taken by a number of limb sounding and solar occultation satellites (SAGE-II/III, UARS HALOE, UARS MLS, and Aura MLS instruments). Its meridional resolution is 2.5° with 31 pressure levels from 300 to 1 hPa.

Figure 1 shows the ozone variations averaged over the region 200–50 hPa and 60–90°S, where the variability and trend of ozone concentration is most pronounced in the Southern Hemisphere (Austin and Wilson, 2006; Solomon 1990, 1999; Ravishankara et al., 1994, 2009), from the four datasets. The ozone variations from two of the four datasets are significantly correlated (Fig. 1a, the correlation coefficients aren't shown). In austral spring, the ozone variations from the four datasets are in better agreement with each other (Fig. 1b).

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

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

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

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

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

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

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

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

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

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

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

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

where θ is the potential temperature, v is meridional wind and subscript p denotes derivative with pressure p. The overbar denotes the zonal mean and the prime denotes deviations from the zonal mean value.

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

Figure 2a shows the correlation coefficients between SST and southern high latitude lower stratospheric ozone variations in austral spring between 1979 and 2015 using ozone data from the MERRA2 dataset and SST from HadISST dataset. Ozone from SLIMCAT simulations, GOZCARDS and SWOOSH datasets were further used to confirm the robustness of the correlations (Fig. 2b–d). The regions of significant correlation are generally different for the four ozone datasets except for the East Asian Marginal Seas; i.e., 5°S–35°N, 100°E–140°E, where the most significant correlations between Antarctic stratospheric ozone variations and SST are seen in the four datasets. Figure 2 implies an interannual connection between SST in the East Asian Marginal Seas and southern high latitude lower stratospheric ozone variations in austral spring. Figure 2 also indicates that SST variations in austral spring associated with ENSO are not the main factor controlling the interannual variability of southern high latitude lower stratospheric ozone.

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

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

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

Figure 4 shows the ray paths of waves generated by the SST anomalies over the region 5°S–35°N, 100°E–140°E, at 300 hPa in four seasons. The wavenumbers along these rays are between 1 and 5. The wave ray paths represent the climate teleconnections, i.e., the propagation of stationary waves in realistic flows. The calculation of the wave ray paths and application of the barotropic model is described in detail by Li et al. (2015) and Zhao et al. (2015). We found that the Rossby waves generated by SST anomalies in the marginal seas of East Asia could indeed propagate to the middle to high latitudes of the Southern Hemisphere in austral spring and winter (Fig. 4a and d), but not in austral summer and autumn (Fig. 4b and c) because the Rossby waves motivated by the low-latitude SST anomalies move mostly northwards in austral summer and autumn. Meanwhile, we must note that the propagating paths of those waves in austral spring and winter aren't totally same (Fig. 4a and d). In austral spring, the path of rays originates over the marginal seas of East Asia reflects directly into the southern Indian Ocean and reaches the Southern Hemisphere (Fig. 4a). In austral winter (Fig. 4d), the rays follow the austral spring path to the Southern Hemisphere. In addition, the second path of rays originates over the marginal seas of East Asia, crosses the Indian Ocean to arrive over tropical Africa or even South America, and then reflects equatorward to the middle to high latitudes of the Southern Hemisphere. These rays can reach about 60 °S and then be refracted to lower latitudes, implying that the pathway of upward propagation of tropospheric waves from the marginal seas of East Asia possibly extends to 60 °S.

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

Figs. 4 and 5 show the pathways of the wave trains generated by the SST anomalies over the marginal seas of East Asia in four seasons. Figure 6 shows the relationship between the SST anomalies and outgoing longwave radiation (ORL) which represents the relationship between the SST anomalies and generated wave activity anomalies to same extent. It is found that the correlation coefficients over the marginal seas of East Asia are the largest in the austral spring compared with other seasons. It implies that the wave activity anomalies caused by the SST anomalies over the marginal seas of East Asia are very strong in austral spring. Figs. 4, 5 and 6 illustrate the possibility of the SST anomalies over the marginal seas of East Asia influencing on the wave activity at southern high latitudes. Bodeker and Scourfield (1995), Shindell et al. (1997a, 1997b), and Huck et al. (2005) have shown that interannual differences in the severity of southern high latitude lower stratospheric ozone depletion are related to Southern Hemisphere planetary wave activity. All of the above analysis illustrates that the SST anomalies over the marginal seas of East Asia are a possible main source of this planetary wave activity.

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

However, it needs to point out that Antarctic polar vortex temperature is deeply below the threshold for heterogeneous chemistry, so that a warming (cooling) in the center of Antarctic polar vortex will have very little impact on Antarctic ozone by affecting heterogeneous chemistry (Tilmes et al. 2006; Kirner et al. 2015). It seems to challenge the above hypothesis. Fig. 7c shows that the center of the correlation coefficients locates near 60°S. It suggests that the center of stratospheric temperature changes caused by SST changes in the East Asian Marginal Seas locates near 60°S but not near 90°S. Temperature changes near 60°S may have more effective effects on southern high latitude lower stratospheric ozone than that near 90°S since the background temperature in the lower stratosphere near 60°S would be higher than that near 90°S. The chemical process maybe has a contribution to the southern high latitude lower stratospheric ozone changes caused by SST changes in the East Asian Marginal Seas.

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

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

We performed three time-slice simulations with WACCM4 to further support the mechanism described in Section3. The monthly mean climatologies of surface emissions used in the model were obtained from the A1B emissions scenario developed by the Intergovernmental Panel on Climate Change (IPCC), and averaged over the period 1979–2015. QBO signals with a 28-month fixed cycle are included in WACCM4 as an external forcing for zonal wind. The SST forcing used in the first time-slice experiment (S1, the control experiment) was the 12-month climatology cycle averaged over the period 1979–2015 and based on the HadISST dataset. S2 was a sensitivity experiment and was the same as S1 except that warm anomalies (as in Fig. 3b) were added to the SST in the marginal seas of East Asia (5°S–35°N and 100–140°E). The third experiment, S3, was the same as S2, but with cold SST anomalies (as in Fig. 3c). Detailed descriptions of experiments S1–S3 are provided in Table 2.

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

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

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

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

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

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

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

We used ensemble transient experiments to estimate the contribution of SST variations in the marginal seas of East Asia to southern high latitude lower stratospheric ozone changes. The transient experiments incorporated the following natural and anthropogenic external forcings for the period 1955–2005: observed SST from the HadISST dataset, surface emissions from the IPCC A1B emissions scenario, spectrally resolved solar variability (Lean et al., 2005), volcanic aerosols (from the Stratospheric Processes and their Role in Climate (SPARC) Chemistry–Climate Model Validation (CCMVal) REF-B2 scenario recommendations), and nudged QBO (the time series in CESM is determined from the observed climatology). The first transient experiment, T1, was the historical experiment covering the period 1955–2005 (Marsh et al., 2013). The second transient experiment, T2, was the same as T1 except that the SST in the marginal seas of East Asia (5 °S–35 °N and 100–140 °E) for the period 1955–2005 was replaced by the 12-month cycle of climatology averaged over the same period. This means that in T2, the SST over the marginal seas of East Asia had only a seasonal cycle, but no trend and no interannual variability. T3 was the same as T2, but used a slightly different initial condition as an ensemble experiment. Detailed descriptions of runs T1–T3 are provided in Table 3.

The southern high latitude lower stratospheric ozone variations caused by the SST variability over the marginal seas of East Asia can be obtained by subtracting simulated ozone in the ensemble experiments ((T2+T3)/2)) from the ozone in T1 (Figure 14, black line). There are evident differences in southern high latitude lower stratospheric ozone variations between T1 and the ensemble experiments ((T2+T3)/2)). This illustrates that the SST variability over the marginal seas of East Asia (Fig. 14, red line) does have a significant effect on southern high latitude lower stratospheric ozone over the past five decades (Fig. 14, black line). The correlation coefficient between the two time series is 0.29 which is significant at 95% confidence level. A further analysis reveals that the linear trend of ozone variations over the region 200–50 hPa and 60–90 °S from T1 (Trend1) is -1.2×10^{-3} ppmv/month, and from (T1 – (T2+T3)/2) (Trend2, Fig. 14, black line) is -0.204×10^{-3} ppmv/month. See Table 4. It implies that the increasing linear trend in SST over the marginal seas of East Asia can contribute approximately 17% of the declining trend in southern high latitude lower stratospheric ozone from 1955–2005 (Trend2 / Trend1 × 100%).

6. Conclusions and Summary

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

Our transient model simulations further demonstrated that SST variations over the marginal seas of East Asia not only modulate the interannual variability of southern high latitude lower stratospheric ozone, but also contribute to southern high latitude lower stratospheric ozone trend over the past five decades. Our analysis reveals that the trend of increasing SST over the marginal seas of East Asia may have contributed approximately 17% to the decreasing trend of southern high latitude lower stratospheric ozone over the past five decades.

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Table 1. Warm and cold SST events in the marginal seas of East Asia in austral spring during the 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

*Following the definition of ENSO events (Trenberth 1997), we propose a threshold of ±0.2, which is equal to the standard deviation of the ST_MSEAI series, as the indicator of warm and cold events.

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).

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

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]

[*1] Integration period is 1955–2005 for T1–T3.

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

Table 4. Linear trends of ozone variations over the region 200–50 hPa and 60–90°S from 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)	-1.2×10^{-3} ppmv/month**
Linear trend of ozone variations over the region 200–50 hPa and 60–90°S from (T1 – (T2+T3)/2) (Trend2)	-0.204×10^{-3} ppmv/month*

**: the trend is significant at 99% confidence level. *: the trend is significant at 95% confidence level. The calculation of the statistical significance of the trend uses the two-tailed Student's *t*-test.

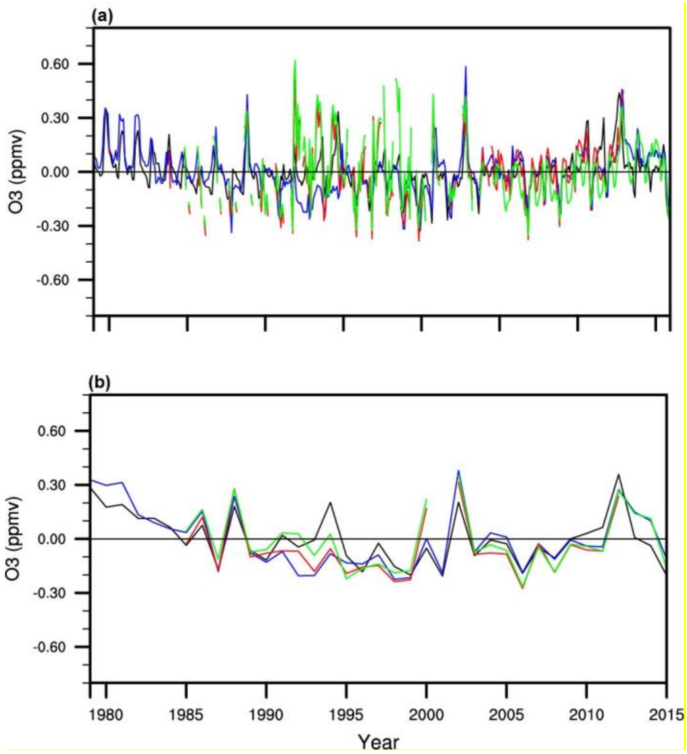

Figure 1. (a) Time series of southern high latitude lower stratospheric ozone variations averaged over the region 60–90°S at 200–50 hPa from the MERRA2 (black line), SLIMCAT (blue line), GOZCARDS (red line) and SWOOSH (green line) ozone datasets. (b), Same as (a), but only for austral spring. Ozone variations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

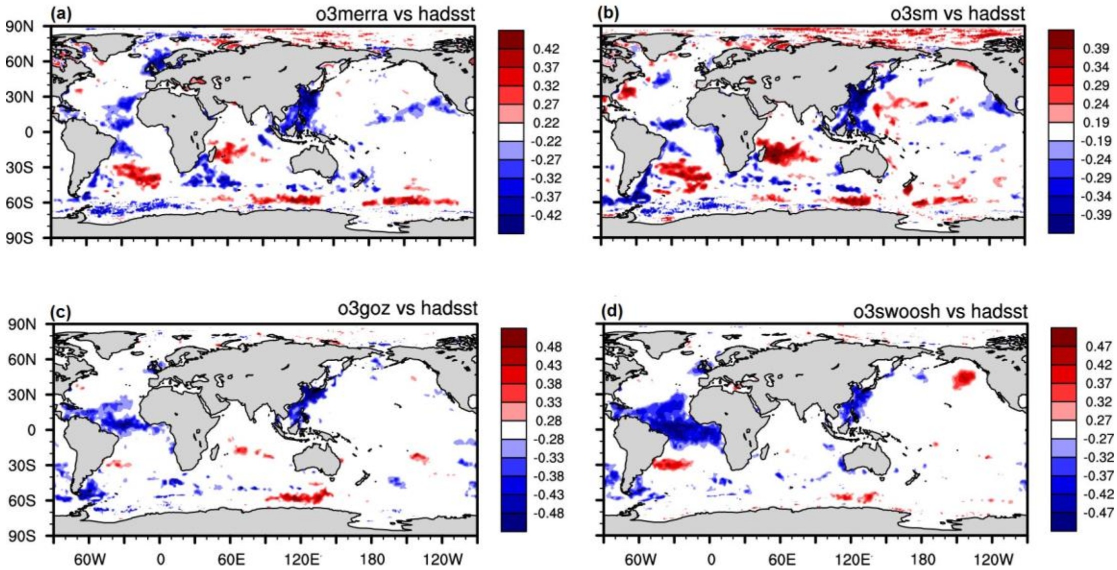

Figure 2. Correlation coefficients between southern high latitude lower stratospheric ozone variations and SST from HadISST in austral spring. Southern high latitude lower stratospheric ozone variations are averaged over the region 60–90 °S at 200–50 hPa in austral spring. (a) Ozone from MERRA2 and (b)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

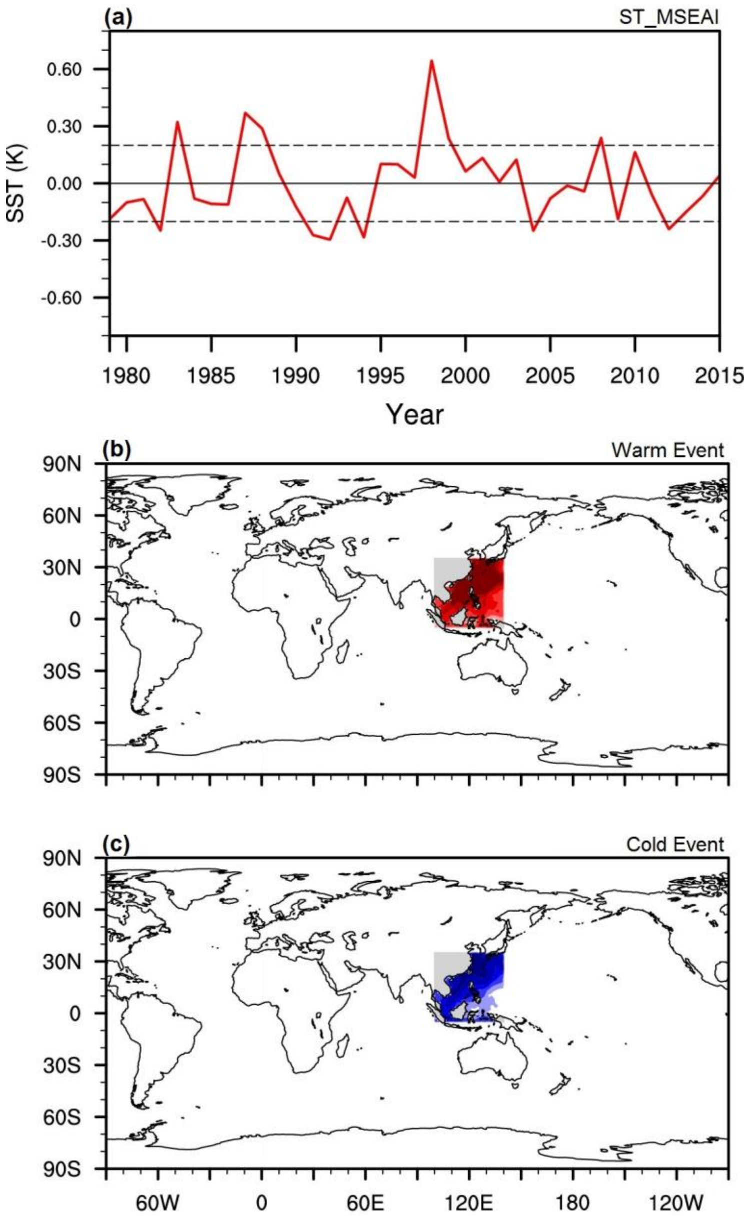

Figure 3. (a) SST variations in the marginal seas of East Asia in austral spring defined using the ST_MSEA index (ST_MSEAI) that was calculated by averaging SST over the region from 5°S–35°N at 100°E–140°E (from HadISST), and then removing seasonal cycles and linear trend. The dashed lines indicate the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

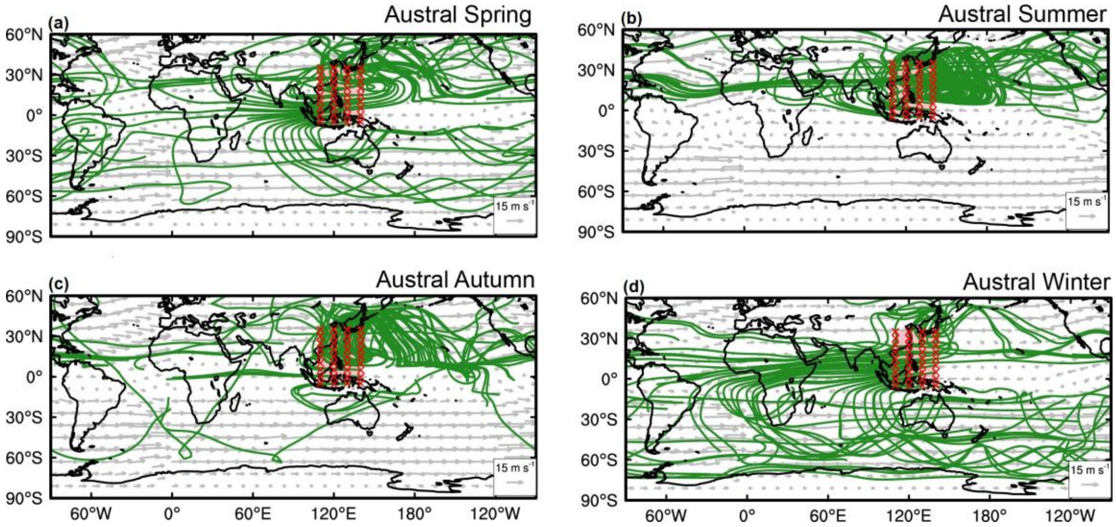

Figure 4. Ray paths (green lines) at 300 hPa in (a) austral spring, (b) austral summer, (c) austral autumn, and (d) austral winter. Red points denote wave sources in the marginal seas of East Asia (5°S–35°N, 100°E–140°E). The wavenumbers along these rays are in the range 1–5. The grey vectors indicate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

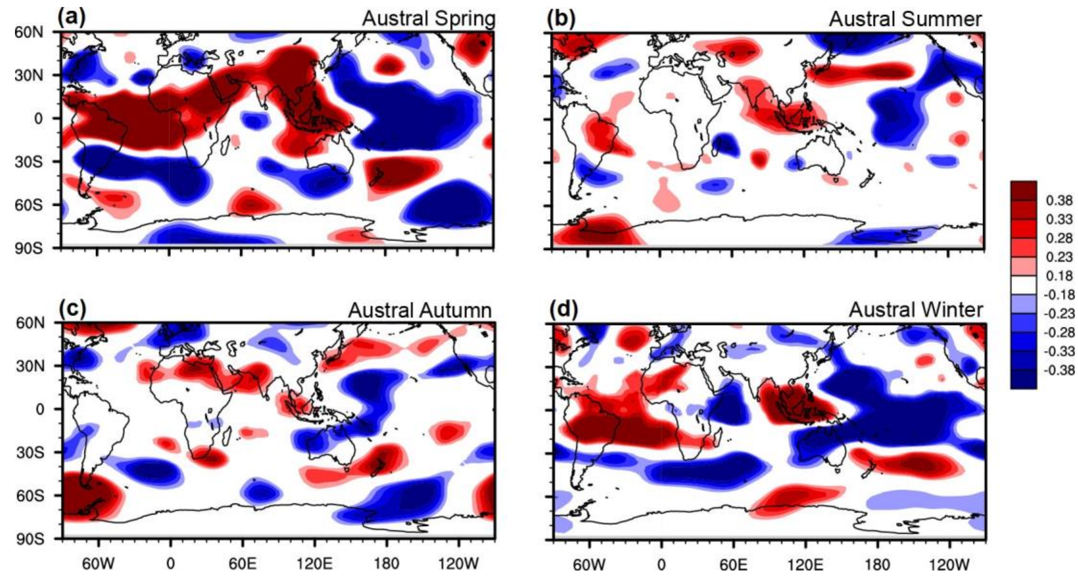

Figure 5. Correlation coefficients between the ST_MSEAI and 300-hPa geopotential height from the ERA-Interim reanalysis in (a) austral spring, (b) austral summer, (c) austral autumn, and (d) austral winter between 1979 and 2015. Only statistical significance above 95% confidence level is colored. The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

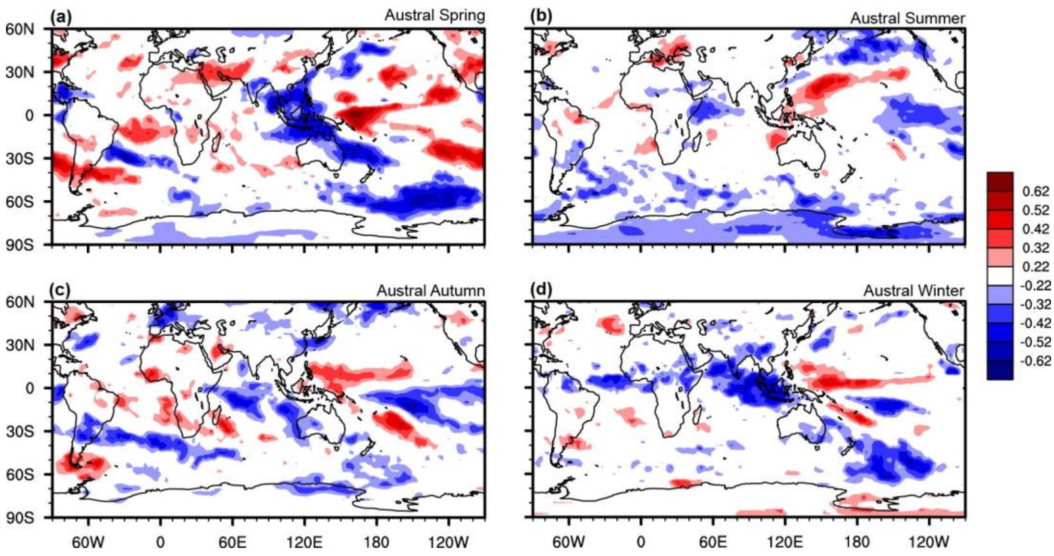

Figure 6. Same as Figure 5, but between the ST_MSEAI and Outgoing longwave radiation from NOAA.

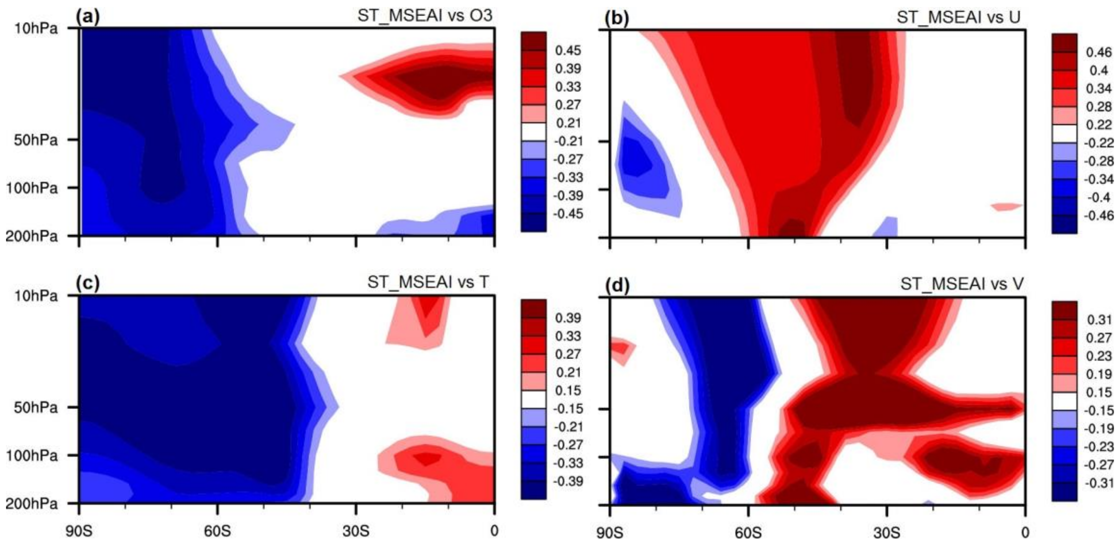

Figure 7. Correlation coefficients between ST_MSEAI and (a) zonally averaged ozone, (b) zonal wind, (c) temperature, and (d) meridional wind in austral spring. Wind and temperature from ERA-Interim reanalysis data; ozone from MERRA2. Only statistical significance above 95% confidence level is colored.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

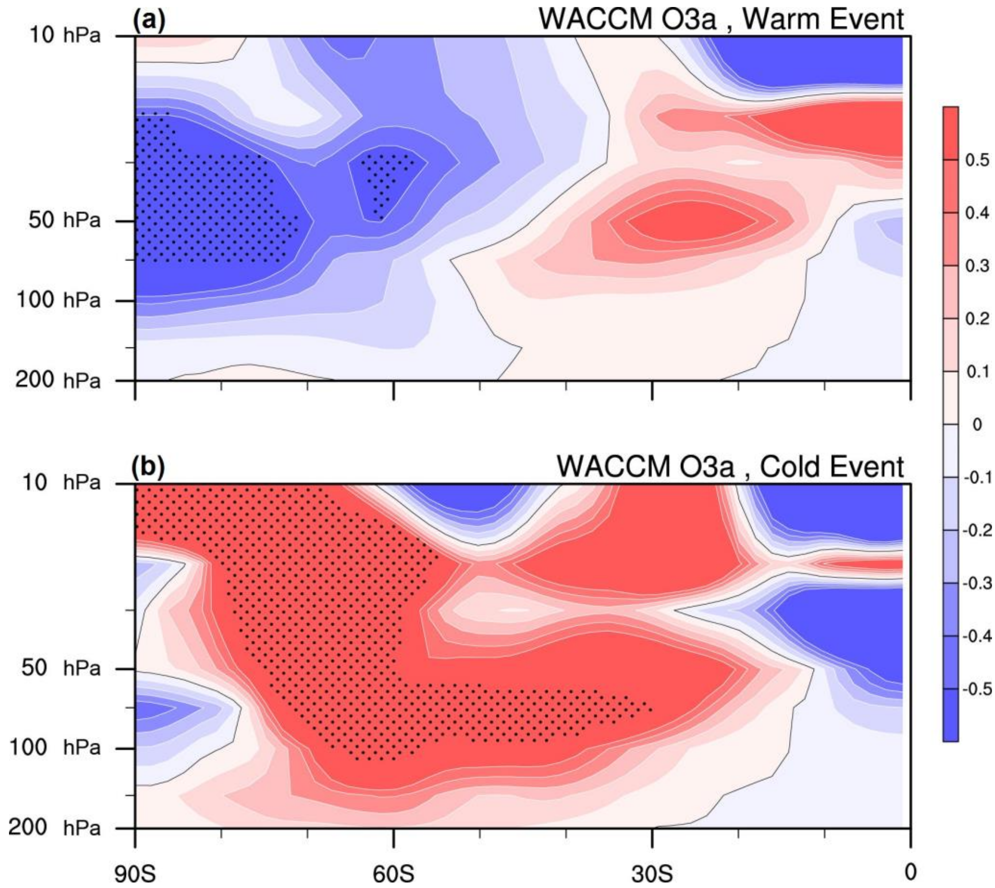

Figure 8. Zonal mean differences in ozone (ppmv) in austral spring between WACCM simulations (a) S2 and S1, and (b) S3 and S1. Statistical significance above 95% confidence level is stippled. Statistical significance of the simulated anomalies is calculated using the two-tailed Student's t-test.

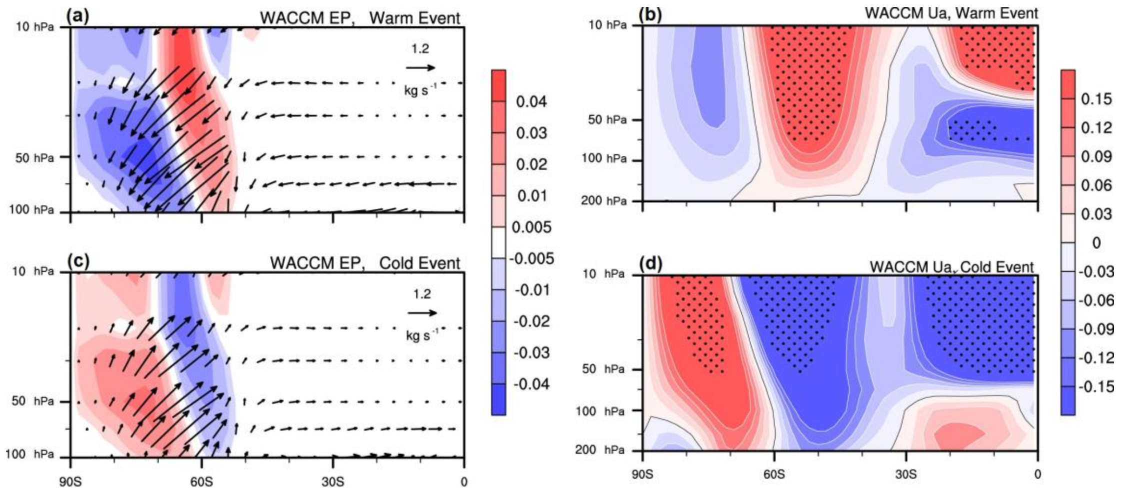

Figure 9. Differences in E–P flux vectors (black arrows) and divergence (color shading) in austral spring between (a) S2 and S1, and (c) S3 and S1. Units for the horizontal and vertical vector directions are 10^7 and 10^5 kg s^{-1}, respectively. (b) and (d), as (a) and (c), but for zonal wind (m s^{-1}).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

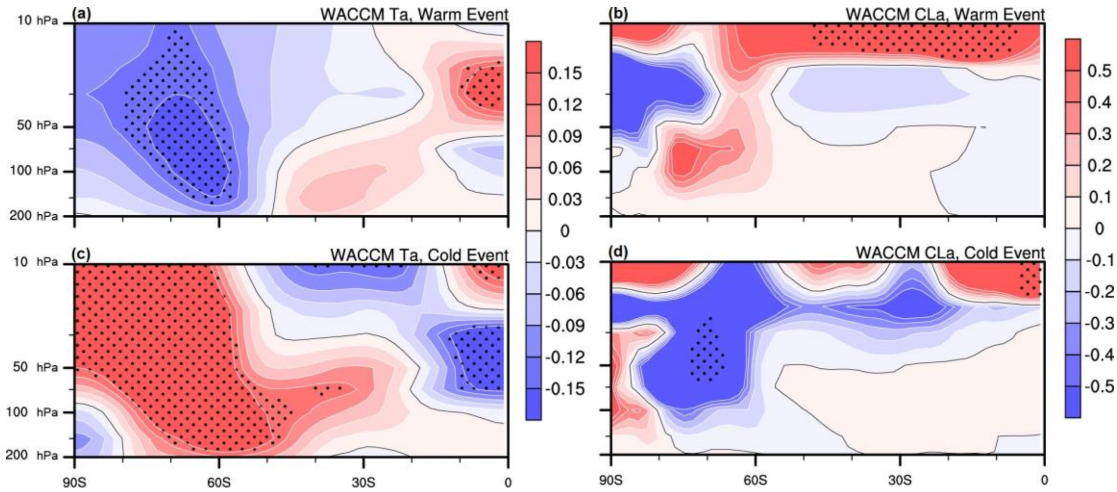

Figure 10. Zonal mean difference in temperature (K) in austral spring between (a) S2 and S1, and (c) S3 and S1. (b) and (d), as (a) and (c), but for active chlorine (ppbv). Statistical significance above 95% confidence level is stippled.

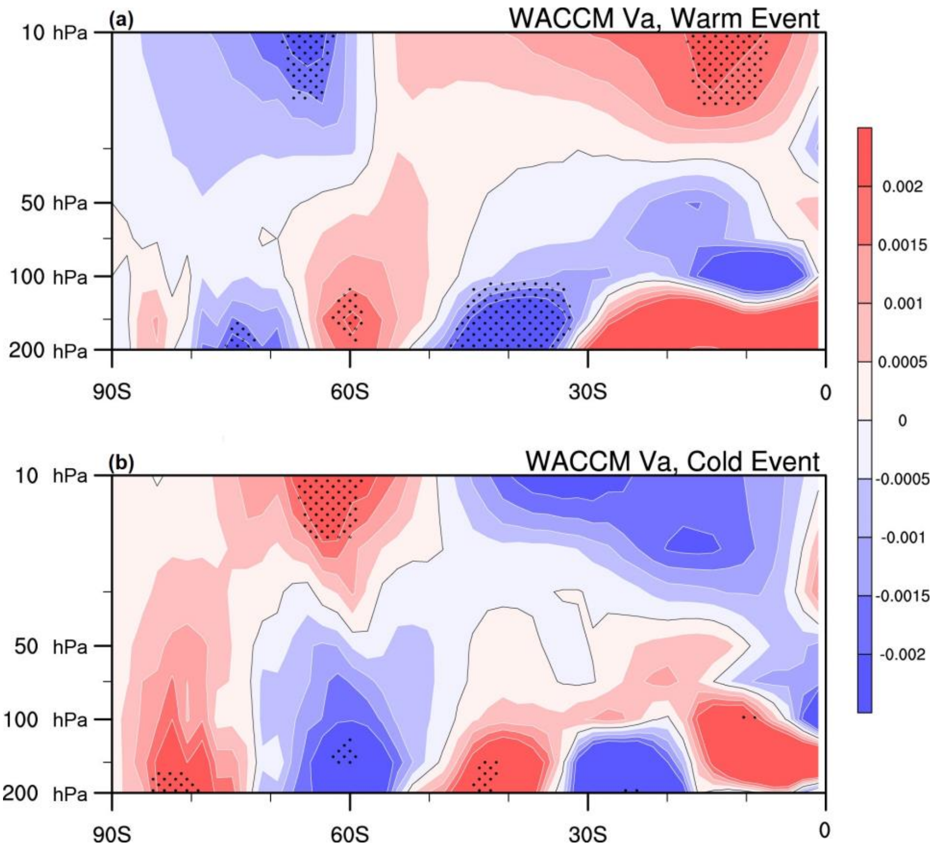

Figure 11. Zonal mean difference in TEM meridional wind (m s^{-1}) in austral spring between (a) S2 and S1, and (b) S3 and S1. Statistical significance above 95% confidence level is stippled.

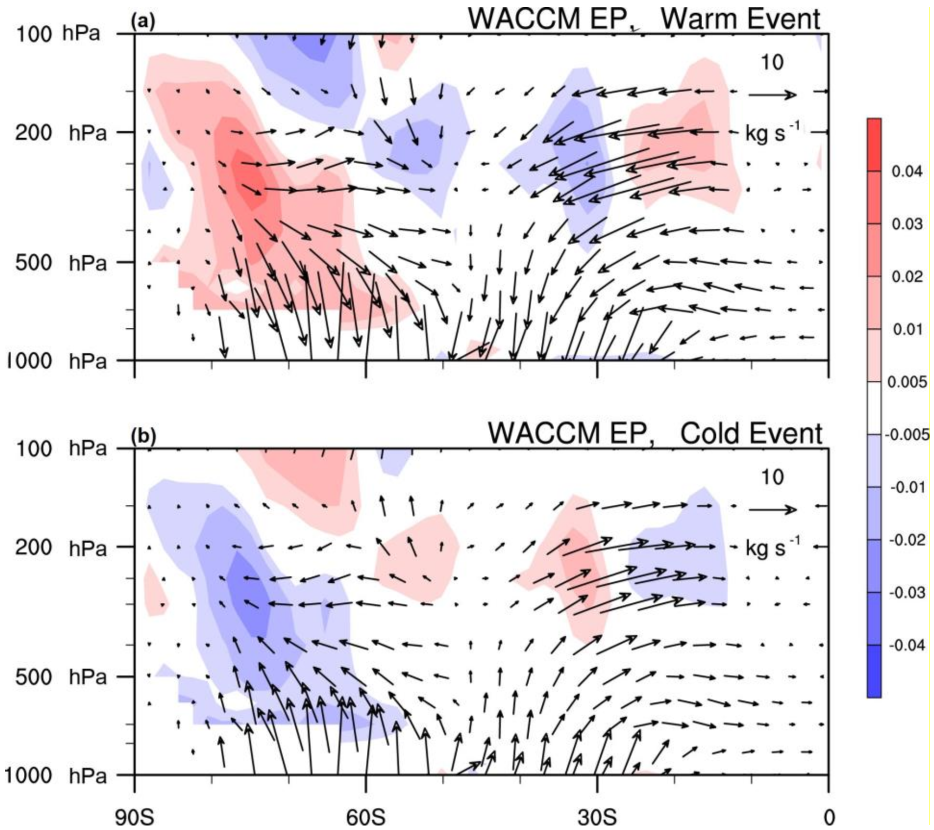

Figure 12. Same as Figure 9a and c, but for 1000 hPa to 100 hPa.

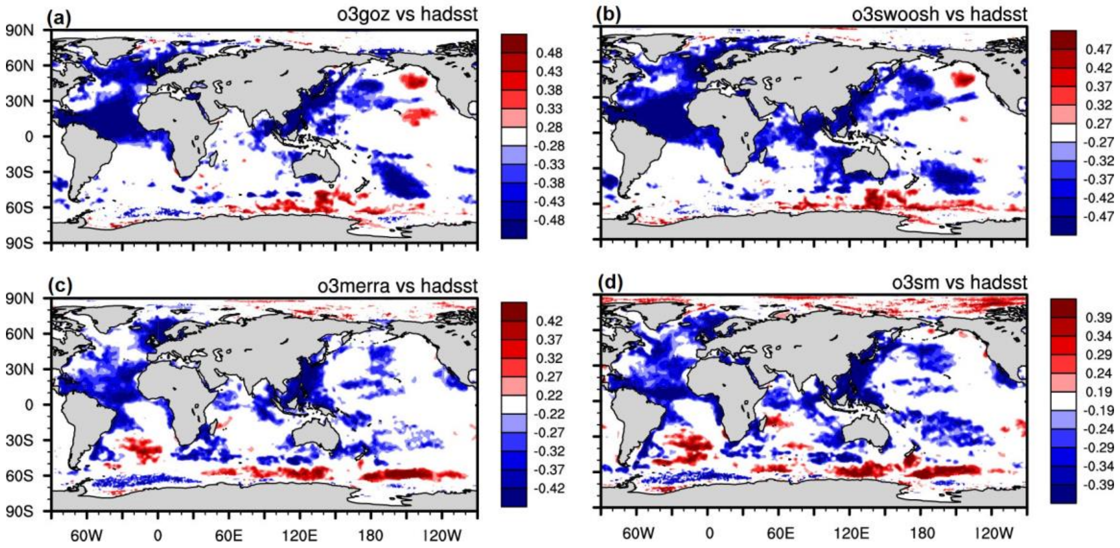

Figure 13. As Fig. 2, but with only the seasonal cycle removed before calculating the correlation coefficients.

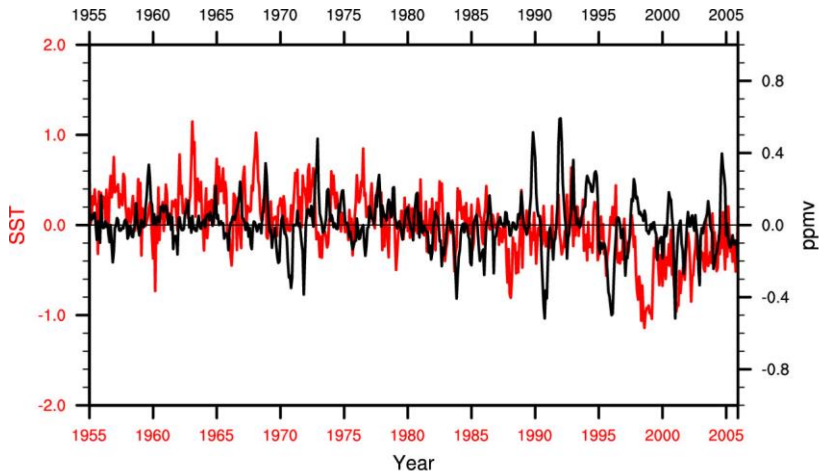

Figure 14. The difference in southern high latitude lower stratospheric ozone variations between T1 and (T2+T3)/2) (black line) and SST variations (×–1) in the marginal seas of East Asia (5 °S–35 °N, 100 °E–140 °E) based on the HadISST data (red line). The seasonal cycle is removed from two time series.