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Disentangling the chemistry and transport impacts of the Quasi-Biennial Oscillation on stratospheric ozone

Jinbo Xie[1], Qi Tang[1], Michael Prather[2], Jadwiga Richter[3], Shixuan Zhang[4]

[1]Lawrence Livermore National Laboratory, Livermore, CA, USA

[2]Department of Earth System Science, University of California, Irvine, CA, USA

[3]National Center for Atmospheric Research, Boulder, CO, USA

[4]Pacific Northwest National Laboratory, Richmond, CA, USA

Correspondence to: Jinbo Xie (xie7@llnl.gov)

Abstract

The quasi-biennial oscillation (QBO) in tropical winds perturbs stratospheric ozone throughout much of the atmosphere via changes in transport of ozone and other trace gases and via temperature changes that alter chemical processes. Here we separate the temperature-driven changes using the Department of Energy's Energy Exascale Earth System Model version 2 (E3SMv2) with linearized stratospheric ozone chemistry. E3SM produces a natural QBO cycle in winds, temperature, and ozone. Our analysis defines climatological QBO patterns of ozone for the period 1979-2020 using both nonlinear principal component analysis and monthly composites centered on QBO phase shift. As a climate model, E3SM cannot predict the timing of the phase shift, but it does match these climatological patterns. We develop an offline version of our stratospheric chemistry module to calculate the steady-state response of ozone to temperature and overhead ozone perturbations, assuming that other chemical families involved in ozone chemistry remain fixed. We find a clear demarcation: ozone perturbations in the upper stratosphere (above 20-hPa) are predicted by the steady-state response of the ozone column to the temperature changes; while those in the lower stratosphere show no temperature response and are presumably driven by circulation changes. These results are important for diagnosing model-model differences in the QBO-ozone responses for climate projections.

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1. Introduction

The Quasi-Biennial Oscillation (QBO) is the principal mode of dynamical variability in the tropical stratosphere, with impact on the circulation and greenhouse gases that extends from the tropical stratosphere into the troposphere. Its effect on ozone – the most important trace gas in the stratosphere – has been well studied (Reed 1964; Bowman, 1989; Wang et al., 2022). Despite being a robust research area for decades, assigning the pattern of ozone perturbations over the QBO cycle to specific processes is not easy due to the simultaneous temperature and transport changes (Plumb and Bell, 1982) and the photochemical linkages across most all reactive gases.

This study aims to provide a better understanding of what drives ozone variability over the QBO cycle. We develop a new index of the QBO phase from a nonlinear principal component analysis (NLPCA) of the tropical zonal winds that retains the observed asymmetric pattern and provides a more consistent measure of the phase throughout the cycle, not just when the zonal winds change sign. Second, we create phase-based composite diagrams to investigate the temporal evolution of ozone patterns, both observed and modeled. Our primary modeling tool is the Department of Energy (DOE) Energy Exascale Earth Model version 2 (E3SMv2, Golaz et al., 2022) with interactive stratospheric ozone (Linoz v2, McLinden et al., 2000), and secondarily we examine some QBO experiments from the National Center for Atmospheric Research (NCAR) Community Earth System Model (CESM). We find that QBO cycles in ozone can be attributed to temperature perturbations in the upper stratosphere (above 20-hPa) and mostly to circulation changes in the lower stratosphere over a wide range of latitudes. The observational data and ozone modeling are described in section 2. The NLPCA method is presented in section 3, followed by the description and use of the Linoz off-line chemistry model in section 4. The results are in section 5. The discussion and conclusion are in section 6.

2. The QBO

2.1 Overview

The QBO appears prominently as alternating easterly and westerly equatorial winds that propagate downward from the top (50 km) to the bottom (16 km) of the stratosphere with a period of about 28 months (Baldwin et al., 2001; Anstey and Shepherd, 2014; Coy et al., 2016). Associated with this propagation of the alternating equatorial winds, the QBO also modifies the vertical propagation of planetary waves and creates global changes in the Brewer-Dobson

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Circulation (BDC) (Holton and Tan, 1982; Watson and Gray, 2014; Zhang et al., 2020). Through perturbations to the BDC, the QBO has been identified as an important source of variability in the overall chemical composition of the tropical stratosphere (Randel et al., 1998; Shuckburgh et al. 2001; Park et al. 2017), and it reaches into the troposphere through stratosphere-troposphere exchange (STE) of ozone (Yang and Tung, 1995; Kinnersley and Tung, 1999) and nitrous oxide (Hamilton, 1989; Ruiz et al., 2021).

2.2 Ozone impacts

The QBO affects ozone through coupled transport and chemical processes, limiting our ability to ascribe the cause of ozone perturbations to specific processes. Baldwin et al. (2001) suggest that the dynamic impact of the QBO via direct transport of ozone accounts for most of the ozone variability. The primary mechanism being: maximum westerly winds correspond to warmer temperatures that result in diabatic cooling that slower tropical ascent of air parcels, with the opposite sense (more rapid ascent) for easterly winds. Tropical ozone has a steep, inverted gradient, 0.1 parts per million (ppm = micromol mol^{-1}) at 100 hPa peaking to 10 ppm at 10 hPa. In this region ozone values are below photochemical steady-state with production exceeding loss, and thus slower ascent rates lead to greater accumulation of ozone, including in total column ozone (TCO, Reed, 1964). This ozone anomaly is also impacted by vertical shifts in NOy (total reactive nitrogen reservoir), which photochemically destroys ozone (Chipperfield and Gray, 1992; Chipperfield et al., 1994, Politowicz and Hitchman, 1997; Jones et al., 1998). The QBO pattern in ozone reverses phase outside of the core tropics (15°S - 15°N), consistent with the return arm of the local equatorial QBO circulation (Holton et al., 1989; Gray and Dunkerton, 1990).

2.3. The QBO modeling initiative

Tropical stratospheric variability, in particular the QBO, has been poorly represented in climate models (Butchart et al., 2011; Butchart et al., 2018; Richter et al., 2020). The number of models with a naturally generated QBO was 0 in the third Coupled Model Intercomparison Project (CMIP3); it rose to 5 in CMIP5 and to 15 in CMIP6 (Richter et al., 2020). Even when models naturally produce a QBO-like variability, the amplitude and periods often fail to match the observed pattern. In the current Chemistry–Climate Model Initiative (CCMI), many of the CCMs forced a QBO signal by nudging the equatorial zonal wind (Morgenstern et al., 2017). Nudging of the winds winds is inherently unphysical and produces an anomalous BDC not found

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in the free-running versions of the same CCMs (Orbe et al., 2020). The World Climate Research Project (WCRP) Atmospheric Processes And their Role in Climate (APARC) started an QBO initiative (QBOi) in 2015 to improve CCM simulation of tropical variability (Butchart et al., 2018), and here we build on those experiments.

2.4. CCM models

The primary model for this study is E3SMv2. E3SM's atmospheric component (EAMv2) is run as a CCM with specified sea surface temperatures (SSTs) and has 72 vertical layers and a horizontal resolution of about 100 km. Following Richter et al. (2010), EAMv2 employs gravity wave (GW) parameterizations that include orographic GWs (McFarlane, 1987), convective GWs (Beres et al., 2004), and GWs generated by frontal systems (Charron and Manzini, 2002). Tunable parameters in the orographic and frontal GW parameterizations remain the same as in EAMv1 (Xie et al., 2018; Rasch et al., 2019). The tunable parameters in convective GWs were explored to produce a more realistic QBO in EAMv2 with a period around 27 months, much closer to observations (28 months) as compared to 16 months in EAMv1 (Richter et al., 2019). Nevertheless, the modeled QBO remains very weak in terms of amplitude. Stratospheric ozone in E3SMv2 is calculated interactively through transport and the chemical Linoz module (McLinden et al., 2000; Hsu and Prather, 2009) that was updated from the E3SM O3v1 to O3v2 module (Tang et al., 2021). Linoz v2 data tables are used to calculate the 24-hour-average ozone tendency (i.e., net production minus loss) from an adopted climatological mean state for key species (CH_4, H_2O, and NOy, Cly, Bry) and first-order Taylor series expansions about the local ozone, temperature, and overhead ozone column (see Eq. (3) in Sect. 5.1). The data tables are generated for each year assuming key chemical species (CH_4, H_2O, and NOy, Cly, Bry) follow monthly zonal-mean climatologies that scale with the slowly varying changes in tropospheric mean abundance of their source gases (e.g., N_2O, CFCs, halons, CH_4, tropopause H_2O). The Linoz model produces a reasonable stratospheric ozone climatology, including seasonal and interannual variability and the Antarctic ozone hole (Tang et al., 2021; Ruiz and Prather, 2022). The tropospheric chemical package for E3SMv2 (chemUCI) was not used and the lower boundary for Linoz was set to 30 ppb. Thus, none of the ozone column variability arises from tropospheric ozone chemistry. E3SMv2 diagnostics on the tendency of tropospheric ozone enable the geographically resolved stratosphere-troposphere exchange (STE) flux of ozone every time step (Hsu et al., 2005; Tang et al., 2013).

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The secondary model for this study is CESM2 (Emmons et al., 2020), using a modified version of the community atmosphere model (CAM) with 83 vertical levels (Randall et al., 2023; Isla et al., 2024), and also run as a CCM with specified sea surface temperatures (SSTs). CAM uses the finite-volume dynamical core with a nominal 1° horizontal resolution and with physics from the Whole Atmosphere Community Climate Model version 6 (WACCM6; Gettleman et al. 2019). The parameters for the convective GW momentum transport were tuned especially for this version to obtain a realistic, naturally generated QBO. The inline ozone calculation is replaced with a monthly mean 3D ozone climatology specified from a previous WACCM simulation. This ozone forcing is formed by merging WACCM simulations for historical (1850-2014) and future period (2015-2100). The ensemble mean of three historical WACCM simulations is used for the historical period while one future scenario run is used for future period. As the mean of free-running CCM simulations, this WACCM ozone climatology does not have any significant QBO-like variability, and thus it cannot trigger a QBO in the CCM (Butchart et al., 2023).

With these two different types of simulations, one with interactive ozone and one without, we must limit our analysis with the pair of models to examining the forced dynamical response (temperature, circulation), but will use the E3SM results to compare the modeled QBO-ozone response with observations.

2.5. Observed ozone and wind

For ozone, we derive the observed QBO signal from the monthly zonal mean total column ozone (TCO) using the Multi-Sensor Reanalysis version 2 data (MSRv2, R.J. van der A, et al. 2015). This latitude-by-month dataset initially covers the period 1979-2012 and later extended to 2020. For stratospheric profiles, we use the zonal monthly mean latitude-by-altitude from the Concentration Monthly Zonal Mean (CMZM) product (Sofieva et al., 2023). This altitude-by-month profile data covers the period 1985-2020. The vertical levels are converted to pressure levels inverting the pressure-altitude formula, $z^* = 16 \log_{10}(1000/P)$ km. We compared this ozone data with the overlapping period from the Microwave Limb Sounder (MLS) data (V5 Level 3: Schwartz et al., 2021) and found only small differences with regard to QBO patterns.

We use data from the ERA5 reanalysis produced by the European Center for Medium-Range Weather Forecast (ECMWF) Integrated Forecast System (Hersbach et al., 2020). The version we use has 137 hybrid sigma model levels from the surface to the model top at 0.01 hPa, and the

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horizontal resolution is about 31 km. We use monthly ERA5 data (wind, temperature, geopotential height) for the period 1979–2020 to analyze the QBO-related dynamical changes, and 6-hourly ERA5 tropical zonal wind (15°N-15°S) to nudge model simulations mentioned below. We use the 5°S-5°N tropical average zonal wind from ERA5 and simulations to determinate the QBO phase index. The combined station zonal wind data from Freie University of Berlin (Naujokat, 1986) for the period of 1979-2020 is also used in the NLPCA analysis (Fig. 1).

2.6 The QBOi simulations

We use a set of three experiments from our two models following the protocol for phase-2 of the QBOi (Butchart et al., 2018; Bushell et al., 2020; Richter et al., 2020):

(1) Exp1-ObsQBO (nudged): the zonal wind (i.e., u) in the tropical stratosphere is constrained to follow the observed QBO evolution by nudging it toward ERA5 reanalysis (Hitchcock et al. 2022). Thus, the stratospheric climate in the tropics is constrained.

(2) Exp1-AMIP (natural): the zonal wind in the tropical stratosphere evolves freely in each CCM being forced only by SSTs and trace-gas radiative heating; there is no nudging.

The nudging is applied to the zonal wind over the range 8 hPa-to-80 hPa and 15°S-to-15°N (Supplementary Fig. 1, nudging coefficient shown is for E3SMv2, that for CESM2 is similar). There is a slight difference in how the models were nudged: E3SMv2 is nudged to the 3-D ERA5 wind field, while CESM2 is nudged to a 2-D zonally-averaged ERA5 wind field. The nudging relaxation timescale is 5 days, which is expected to constrain the slowly evolving QBO winds.

3. NLPCA analysis of QBO phase

In diagnosing QBO-related changes to the dynamics and chemistry, we need to define the phases of successive QBOs, at least and align these phases over a 24-month period. Asymmetric and nonlinear features of the evolution of the QBO phase are found in many studies (Lindzen and Holton 1968; Holton and Lindzen, 1972; Giorgetta et al., 2002). The most obvious and sharp synchronization point is when the QBO west phase (QBOw, i.e. prevailing westerlies) transitions to the east phase (QBOe: prevailing easterlies) at some pressure level in the middle stratosphere (taken as 10 hPa here) (Naujokat et al., 1986; Pahlavan et al., 2021; Kang et al., 2022). The QBOe phase is typically longer (e.g., 63%, Bushell et al., 2019), with wind speeds about twice as strong as that of the QBOw (Naujokat et al.,1986; Kang et al., 2022). The problem with defining the QBO phase (index) simply as the absolute time difference relative to the synchronization

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point (e.g., Ruiz et al., 2021) is that the duration of different phases varies across successive QBOs.

Previous use of PCA-derived QBO indices (Hamilton and Hsieh, 2002; Lu et al., 2009) did not allow for this asymmetric and nonlinear behavior. Lu et al. (2009) noted that the reconstructed wind series from the PCA looked more sinusoidal in time than the actual winds, and thus the asymmetries between phases did not show up in the PCA-based indices. To address these issues, we use an NLPCA method that utilizes hierarchical-type neural network with an auto-associative architecture (Scholz et al. 2002). It is a nonlinear generalization of the standard PCA from straight lines to curves in the original data space, and natural extension to the PCA method by enforcing the nonlinear components to the same hierarchical order as in the standard PCA (Scholz et al., 2002). The NLPCA model described here has 5 layers with 3 hidden layers of neurons. The layers of the neural-network for NLPCA are in the sequence of input-encoding-bottleneck-decoding-output with the structure of n-(2k+2)-k-(2k+2)-n, where the n refers to dimension of input/output dataset and k is the number of dimensions for bottleneck layer. To achieve robustness, the NLPCA is applied to the tropical zonal wind data (5°S-5°N, 10-hPa to 70-hPa) for a set of k varying from 2 to 5, with 100 runs (different in random initialization weights) for each k. The optimal number of k is set as 5 as it gives the lowest root-mean-square-error between the input and output. It is shown that the first and second principal components (PC1 and PC2) of the NLPCA account for approximately 90% of the whole variance.

Following previous studies (Wallace et al., 1993; Hamilton and Hsieh, 2002; Lu et al., 2009), the QBO phase index ψ is calculated using PC1 and PC2 as follows:

$$\psi = arctan\,(v/u) \quad (-\pi \leq \psi \leq \pi\,), \quad (1)$$

where u and v are the time series of the PC1 and PC2, respectively. The positive/negative phase angle index ψ corresponds to QBOw/QBOe.

We compare the reconstructed zonal wind anomalies using NLPCA and PCA (Wallace et al., 1993) with the QBO cycle in the observation (Fig. 1). It is shown that the observed QBO transition corresponds to an abrupt downward propagation in QBOw and a slower downward transition in QBOe (indicated by clustering points in B to C to A on black triangular shape in Fig. 1a). The NLPCA captures large part of this sharp transition in QBOw while PCA underestimates it (indicated by points near C in Fig. 1a). This difference is also clearly shown in a typical QBO cycle of 1970. 9 – 1972.3 (Figs. 1b, 1b, and 1d, black arrow-sticks exhibits the

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downward propagation in QBOw) and the time series of NLPCA/PCA QBO phase (index) (Fig. S2).

While the NLPCA-derived QBO index is more realistic in following the atmospheric changes, it is impractical to map the NLPCA phases onto the monthly-mean model diagnostics. Thus, our QBO composites use simple monthly time steps about our best synchronization point, which from the NLPCA analysis we take to be the transition from easterlies to westerlies at when phase angle index ψ crosses 0 with negative values before and positive values after it. It is demonstrated that compared to QBO composites produced using the PCA-derived QBO index, that produced using the NLPCA-derived index show larger contrast in observed tropical zonal wind anomalies between QBOw/QBOe (Supplementary Figs. 3a and 3b) that is consistent with those described in previous literatures (Hamilton and Hsieh, 2002; Lu et al., 2009). This larger contrast between NLPCA and PCA in zonal wind anomalies is correspondent with the larger contrast in that of the total column ozone anomalies (Supplementary Figs. 3c and 3d).

4. Linoz calculation of the steady-state ozone

To examine the ozone response to the QBO we use the Linoz model, and the steady-state ozone is derived from Eq. 4 of Mclinden et al. (2000). The photochemical steady-state ozone mole fraction f_{ss} (parts per million, moles per mole of dry air) is expressed as follows:

$$f_{ss} = f_o \; + \; [\,(P-L)_o + \frac{\partial(P-L)}{\partial T}|_o\,(T-T_o) \; + \; \frac{\partial(P-L)}{\partial C_{O_3}}|_o\,(C_{O_3} - C_{O_3}^o)\,]\tau, \qquad (2)$$

The values f_o, T_o, and $C_{O_3}^o$ are the climatological values of local ozone, temperature, and overhead column ozone tables used to calculate the Linoz tendencies. $(P\text{–}L)_o$ is the ozone net production minus loss tendency and the partial derivatives are the sensitivity of the net production to temperature and overhead column ozone. All of these quantities are evaluated at the climatological values and tabulated by Linoz as a function of month, pressure altitude, and latitude. The effective lifetime of ozone, τ, is calculated from the Linoz tables as the negative reciprocal of the tabulated partial derivative of the production with respect to ozone, i.e., $\tau = -[\frac{\partial(P-L)}{\partial f}|_o\,]^{-1}$). A major assumption here is that the key chemical families (NOy, Cly, Bry) and long-lived reactive gases (N_2O, CH_4, H_2O) do not change from their climatological values used to generate the tables (Hsu and Prather, 2009). This steady-state calculation ignores transport tendencies and thus will be apply only where the photochemistry is rapid, $\tau < 100$ days.

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In application, we derive f_{SS} first locally from the T profile, and then calculate C_{O_3} to correct for the column ozone sensitivity. Note the calculation of C_{O_3} includes the column ozone based on the f_{ss} values from all the layers overhead plus a contribution from the local f_{SS} in that layer weighted by the air mass in the upper half of the layer. Thus, equation 2 becomes a linear algebraic equation involving f_{SS}. Fig. 2 shows this steady-state calculation (f_{SS}, T, τ) for January and July using ERA5 monthly mean temperature.

5. Impact of QBO on stratospheric ozone

Nudging the tropical zonal wind creates QBO-driven perturbations to the temperature and residual circulation that we can diagnose in both the E3SMv2 and CESM2 runs and compare with observations. For E3SMv2 with interactive ozone we are able to see the changes in ozone. This also applies to the simulations with an internally generated QBO.

We create a similar composite of the QBO cycle using E3SMv2/CESM2 following Ruiz et al., (2021) to see the full QBO cycle influence on stratospheric ozone. The time-composite is created for each month starting 14 months prior and extending to 14 months after the QBO transition for 1979-2020. The center is when the NLPCA-derived QBO phase angle index (see section 3) shifts from negative to positive (QBOe -> QBOw). We create the total column ozone (TCO), tropical ozone, and extratropical ozone composite as a function of QBO phase. For the TCO, we calculate the zonal-mean averages to produce the global map of composite. For tropical (15°S-15°N) and extratropical (30°S-60°S/30°N-60°N) ozone, the data is processed to produce vertical profiles of regional average ozone using latitudinal weight to produce the vertical profile composite. The CESM2 ozone composite is not shown since its ozone is prescribed. To further analyze the impact of QBO-induced circulation on ozone, the process is also repeated for temperature, zonal wind and steady state ozone (see section 4). We first analyze the impact of QBO on global TCO in section 5.1, and separately analyze impact on tropical and extratropical stratospheric ozone in section 5.2 and 5.3, followed by the overall performance in section 5.4.

5.1 Impact of QBO on global TCO

In this section, we examine the impact of QBO on ozone using TCO observations (MSRv2) and E3SMv2 model simulations. The TCO composites from E3SMv2 nudged and natural simulations are compared in Fig. 3.

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It is shown that the anomalous MSRv2 TCO exhibits a significant monopole-to-tripole pattern from QBOe to QBOw (Fig. 3a). The TCO pattern exhibits a monopole pattern of anomalous low during QBOe that gradually transits to tri-pole pattern of anomalous high in the tropics and low in the extratropic. The magnitude of the negative in QBOe (5 DU) is lower than the positive pattern (12 DU) in QBOw in the tropics, indicating asymmetric response of TCO to QBO in the tropics. The E3SMv2 nudged simulation is like MSRv2 in that it captures most of the monopole-to-tripole pattern within the tropics and extratropics with similar amplitudes (Fig. 3b), indicating the impact of nudged QBO on TCO is close to what observed. Internally generated QBO variability in E3SMv2 natural, on the other hand, only partly exhibits the patterns of MSRv2 (Fig. 3c) with weaker amplitude (nearly eight times weaker). This indicates the QBO-related signal is partly present in natural E3SMv2, and that nudging the tropical zonal wind contributes to the modulation and enhancement of this "QBO-driven" TCO variability.

5.2 Impact of QBO on tropical stratospheric ozone

In this section, we analyze the impact of QBO on tropical (15°S-15°N) stratospheric ozone concentration. The composites of ozone vertical profile (1-hpa to 100-hPa) from E3SMv2 nudged and natural simulations are compared with the CMZM satellite data (Fig. 4).

It is shown that the CMZM satellite ozone exhibits a double-peak vertical structure with large ozone variations between 1~20-hPa and 20~100-hPa (Fig. 4a). Both peaks shift in a sequence of negative-positive-negative from QBOe to QBOw, and the amplitude of the upper peak is smaller than that of the lower peak (Fig. 4a). The E3SMv2 nudged simulation captures parts of the double-peak structure (Fig. 4b). The E3SMv2 natural simulations, on the other hand, show similar double-peaked patterns but with smaller amplitude (3 times weaker) and shorter period (Fig. 4c). This may be because the period of internally generated QBO in E3SMv2 is ~21 years (Golaz et al., 2022). Overall, the E3SMv2 nudged simulation modifies the period and enhances the QBO response in ozone that is mostly consistent with the CMZM weaker above 20-hPa and stronger below 20-hPa.

As a coupled system, the QBO chemical and transport impacts on ozone are intertwined, making it difficult to diagnose which QBO impact is more important to the ozone differences between model and observation or among different models. Here we try to quantitatively separate these two terms with a new diagnostic tool, recognizing their time scale differences. We derive the steady state ozone (see Section 4 for details) for E3SM nudged and natural simulations

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(Figs. 5a and 5c). The steady state ozone for CESM2 is also derived (Figs. 5b and 5d). Although ozone is prescribed in CESM2, the steady state ozone for CESM2 shown here is the "would-be" temperature-ozone if CESM2 were to implement Linoz v2 as its diagnostic ozone module. The steady state ozone in both E3SMv2 and CESM2 nudged simulations show similar patterns of ozone peak above 20-hPa while weak response below 20-hPa (Figs. 5a and 5b). The steady state ozone of E3SMv2 and CESM2 natural simulations (Figs. 5c and 5d) partly resemble that of the nudged simulations except with weaker amplitude and different periods (shorter for E3SMv2 and longer for CESM2). This corresponds to their similar alternating temperature pattern phase shift in the tropics (Figs. 6b, 6c, 6d and 6e) and indicates that the QBO impacts ozone through temperature-sensitive, fast chemical reactions above 20-hPa. The prognostic ozone in E3SMv2 below 20-hPa corresponds to the alternating $\underline{w}^*$ shift patterns (Figs. 6g and 6i). This and the no response in steady state ozone indicates the prognostic ozone below 20-hPa in E3SMv2 is transport-driven. This demarcation of the QBO-induced ozone at 20-hPa may be due to the separation of ozone lifetime below/above 20-hPa (Reed et al., 1964). The ozone lifetime is relatively long compared with the dynamical process below this level while shortened considerably above it. The temperature affects ozone above 20-hPa through ozone destruction – colder/warmer anomalies slow/accelerate ozone destruction, leading to correspondent ozone increase/decrease (Wang et al., 2022); the transport effect of QBO-related wind modulates the temperature through thermal wind balance enhancing/lessening the upward motion in the tropics (Plumb and Bell, 1982; Baldwin et al., 2001; Ribera et al., 2004; Punge et al., 2009). This explains the apparent separation of transport- and chemistry-driven ozone changes above/below 20-hPa. It is also worth mentioning that the nudged CESM2 also produces similar temperature and $\underline{w}^*$ (Figs. 6c and 6h), it thus indicates that nudged CESM2 may produce similar prognostic ozone if it were to implement Linoz v2 as interactive ozone module. Overall, there are apparent demarcation of QBO impact on tropical stratospheric ozone (15°S-15°N) above/below 20-hPa in the nudged runs that can separately be explained by transport and chemical impact.

5.3 Impact of QBO on extratropical stratospheric ozone

We extend the analysis of the impact of QBO on ozone to the extratropical region in both hemispheres (30°N-60°N/30°S-60°S). Since in the nudged simulations the nudging is imposed only in the tropical regions, we can further examine the impact of nudged QBO in the extratropics where it is free running. Fig. 7 shows pressure-time cross-section of the extratropical

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(30°N-60°N/30°S-60°S) ozone concentration as a function of QBO phase for CMZM satellite ozone, E3SMv2 nudged, and E3SMv2 natural simulations. It is shown that nudged E3SMv2 simulations follow the similar positive-negative ozone phase shift in both hemispheres (Figs. 7b and 7e). The difference is that ozone is slightly stronger in QBOe while similar amplitude in QBOw. The natural E3SMv2 simulation does not reproduce the patterns of the nudged simulation for both hemispheres (Figs. 7c and 7f). This indicates that the nudged QBO is driving the phase shift of E3SMv2 ozone in both hemispheres' extratropic. For the natural simulations, the deficiency is likely due to the weak internally generated QBO in E3SMv2. Overall, the nudged E3SMv2 captures the QBO signal propagated outside of tropics and produces the extratropical ozone phase shift in both hemispheres. The natural simulation does not show the phase shift potentially due to weaker internally generated QBO.

In terms of the transport/chemical impact separation, we follow the analysis of Fig. 5 for E3SMv2 and CESM2 using the Linoz v2 model (Fig. 8). Like that of the analysis in the tropics, the chemical impact is stronger above 20-hPa for both E3SMv2 and CESM2 nudged simulations (Figs. 8a, 8b, 8e, and 8f), except the Southern Hemisphere (30°S-60°S) is overall noisier than that of the northern hemisphere (30°N-60°N). The natural simulations between the two models are different. The E3SMv2 natural simulations generally show consistent negative phase (Figs. 8c and 8g). The CESM2 natural simulations exhibits similar pattern to the nudged simulations in the northern hemisphere while that in the Southern Hemisphere is noisier (Figs. 8d and 8h). This noisier southern hemisphere steady state ozone above 20-hPa in the nudged simulations correspond to the noisier temperature for the two models (Figs. 9g and 9h), which may be largely affected by stronger and noisier southern polar vortex (Supplementary Figs. 5a and 5b) as also documented by other studies (Ribera et al., 2004). The intrusion of the polar vortex via events like stratospheric sudden warming (Butler et al., 2017) may have an impact on the QBO-ozone relationship in the extratropics. Below 20-hPa, the E3SMv2 nudged ozone corresponds to the $\underline{w}^*$ (Fig. 9l and 9q), indicating it's transport-driven.

In terms of the impact of QBO nudging in the extratropics, there are considerable differences between the two models especially in the Southern Hemisphere. It shown that CESM2's phase shift of temperature and $\underline{w}^*$ patterns (Figs. 9h and 9r) in the Southern Hemisphere is not as obvious as that shown in E3SM (Figs. 9g and 9q). Since this is outside the nudging region, it's complicated to differentiate the main impact factor. One reason may be the different ozone

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feedback between the two models – interactive ozone in E3SMv2 contributes to maintain the QBO-temperature structure (Butchart et al., 2023) while prescribed ozone in CESM2 does not; Another may be due to the overall 3-D nudging strategy in E3SMv2 that may provide more stringent constraint than the 2-D zonal mean nudging strategy that CESM2 adopted. Another interesting issue is that both nudged simulations can partly reproduce the observed ERA5 temperature and $\underline{w}^*$ patterns in the extratropic regions outside of the tropical nudging regions. This occurrence of the residual circulation consistent with the ERA5 in the extratropics indicates the validity of the nudging strategy for the QBOi protocol. Despite the fixed ozone, the CESM2 could still produce such circulation and temperature patterns that is consistent with ERA5 indicates the overall weak ozone feedback on formation of circulations both in the tropics and extratropics. These patterns and the steady state ozone analysis for the CESM2 nudged simulation also indicate that it may reproduce the prognostic ozone like E3SMv2 if it were to use Linoz-v2 as interactive ozone module under the QBOi nudging protocol.

5.4 Model performance in simulating QBO impact

In this sub-section, we examine the overall performance of E3SMv2 and CESM2 QBOi simulations in simulating the QBO-ozone relationship. We evaluate the pattern correlation and standard deviation of the area-weighted TCO pattern (60°S-60°N), vertically-weighted ozone concentration (15°S-15°N, 30°N-60°N, 30°S-60°S), zonal wind (15°S-15°N, 30°N-60°N, 30°S-60°S), temperature (15°S-15°N, 30°N-60°N, 30°S-60°S), and $\underline{w}^*$ (15°S-15°N, 30°N-60°N, 30°S-60°S). For ozone, only E3SMv2 results are shown since CESM2 has fixed ozone. The results are summarized in a Taylor diagram shown in Fig. 10. The observed pattern is plotted at the (1,0) reference point.

In terms of ozone (Fig. 10a), there are remarkable differences between the simulations. Overall, the E3SMSv2 nudged simulations perform the best, with the pattern correlation of all four variables over 0.8 while other simulations are below 0.5. This indicates nudging realistic QBO variability may increase the model performance in simulating ozone. In the extratropics, the E3SMv2 nudged simulation has good pattern correlations, but the amplitude is off by over 1.5 times. The results for temperature, zonal wind and $\underline{w}^*$ are similar with ozone in the tropics (Figs. 10b, 10c, and 10d). What's different is in the extratropics — both nudged E3SMv2/CESM2 temperature, zonal wind, and $\underline{w}^*$ show better performance in NH extratropics

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than in SH extratropics. This may be due to stronger polar vortices in SH and NH that disturb the QBO signal. Another difference is in the natural simulations – the tropical temperature (15°S-15°N) and zonal wind signals exhibit reasonable correlations of over 0.7 in zonal wind, over 0.5 in temperature. This indicates a discernable internally generated QBO signal in the E3SMv2/CESM2, although it's weaker and does not extend to the extratropics.

6. Discussion and Conclusion

6.1 Discussion

There are some interesting issues worth mentioning in this study. The first is the effect of nudging. It is shown that even in the extratropical regions where the QBO nudging is not imposed, the QBO impact on extratropical circulation is still apparent in the two models. In these QBOi simulations, the E3SMv2 employs a 3-D nudging strategy where the ERA5 3-D full field zonal wind is nudged to the model while CESM2 employs a 2-D nudging strategy. It may be recognized that the E3SMv2 posed a stronger nudging than CESM2, but both strategies were able to produce extratropical QBO-associated circulation outside of the nudging region, this demonstrates the overall effectiveness of the nudging strategy. Between the models, there are still minor differences. For example, the extratropical zonal wind and temperature in CESM2 are more scattered than that of E3SMv2. One reason may be the nudging strategy discussed above, another reason may be the interactive/non-interactive ozone in the model. In this QBOi simulation setup, E3SMv2 has the interactive ozone turned on, while CESM has only fixed ozone input. Thus, the QBO-ozone interaction in E3SMv2 may be more self-consistent than that in CESM2 – Studies have documented the impact of QBO-ozone interaction tend to maintain and the QBO-temperature structure and prolong its period (Hasebe et al., 1994; Shibata, 2021; Butchart et al., 2023).

Another noteworthy issue is the use of the offline Linoz v2 model to diagnose the dynamic and chemical impact of QBO on ozone. It is demonstrated here that the Linoz v2 is a simple but useful tool to diagnose and separate the dynamic/chemical impact of QBO on ozone. The results shown here are important for diagnosing model-model and model-observation differences in the QBO-ozone responses for climate simulations.

6.2 Conclusion

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In this study, we utilize the Linoz v2 model to separate the chemical and transport response of the QBO ozone impact in climate models. We derive a new QBO phase index using an NLPCA method, and utilize the index to form QBO cycle composites to analyze QBO-ozone relationship in observation and simulations produced under the QBOi protocol. By analyzing the simulations of two QBOi participant models (E3SMv2 and CESM2), it is shown that the nudged E3SMv2 simulation captures the monopole-to-tripole composite pattern in the observed TCO. The natural simulation partly reproduces the observed TCO pattern but with weaker amplitude and shorter period, indicating there is an internally generated QBO in E3SMv2 that is enhanced and prolonged by nudging in the E3SMv2 nudged simulations. Looking further into the vertical structure of the QBO-ozone relationship, it is shown that the E3SMv2 nudged simulations capture most of the double-peaked vertical structure in observed ozone data between 1~20-hPa and 20~100-hPa in the tropics but with weaker amplitudes in the extratropics. natural simulation only captures part of the structure with smaller amplitude, indicating the existence of internally generated QBO. This and the nudged simulations indicate that nudging enhanced the QBO amplitude and prolonged its period originally exists within E3SMv2.

Utilizing the Linoz v2, we separated the chemical and transport response of ozone in E3SMv2 nudged to QBO. It is shown that the two impacts have a rather clear demarcation on both tropical and extratropical ozone response below/above 20-hPa – chemistry impact correspondent to QBO-related temperature change dominates the response above 20-hPa linked to photochemical process, and transport impact related to QBO-related vertical motion dominates the response below 20-hPa. The results here are important for diagnosing model-model and model-observation differences in the QBO with free-running climate-change simulations, allowing us to separate temperature from circulation effects. In CESM2, the fixed ozone that is out-of-phase with the observed QBO variability seems to impose a weak constraint on the overall simulation. This indicates that using interactive ozone or not in the simulation does not significantly alter the results for QBO simulations, although the synchronization impact of QBO variability in observed ozone may need further examination (Butchart et al., 2023).

Stratospheric ozone is not only essential for protecting life on the Earth but also has important climate impacts. More and more studies reported the important role of ozone variations in modifying the stratospheric circulation and therefore influencing the surface climate (e.g. Xie et al., 2020). Since the QBO has relatively high predictability, considering its impacts

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on stratospheric ozone and subsequent atmospheric circulations may help improve the predictions of surface weather and climate (e.g., Li et al., 2023).

Despite the above studies, however, there are still caveats. Firstly, the current study makes use of only one model in QBOi that has interactive ozone feature. More models may be used in the future to examine the QBO-ozone relationship. The capability of the current version of O3v2 in E3SMv2 is limited due to the missing representation of the NO_x long-lived tracers. In the latest version of E3SMv3, the O3v2 is updated to include the impacts of these tracers, it would be interesting to see how these tracers interact with the current ozone calculations. These are to be assessed in future studies.

Data availability

The satellite data from the Copernicus Climate Change Service can be accessed at (https://cds.climate.copernicus.eu/cdsapp#!/dataset/satellite-ozone-v1?tab=form). The ERA5 data can be accessed at (https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-complete?tab=overview). The ChemDyg diagnostics can be accessed at (https://doi.org/10.5281/zenodo.11166488).

Author contribution

J.X., Q.T. designed the research; J.X. performed the E3SM simulations and wrote the manuscript. J.R. provided the CESM2 simulation. Q. T. and M.P.'s supervised the research and helped interpreting the results. All authors contributed to the scientific discussion and paper revision.

Competing interests

The authors declare that they have no conflict of interest.

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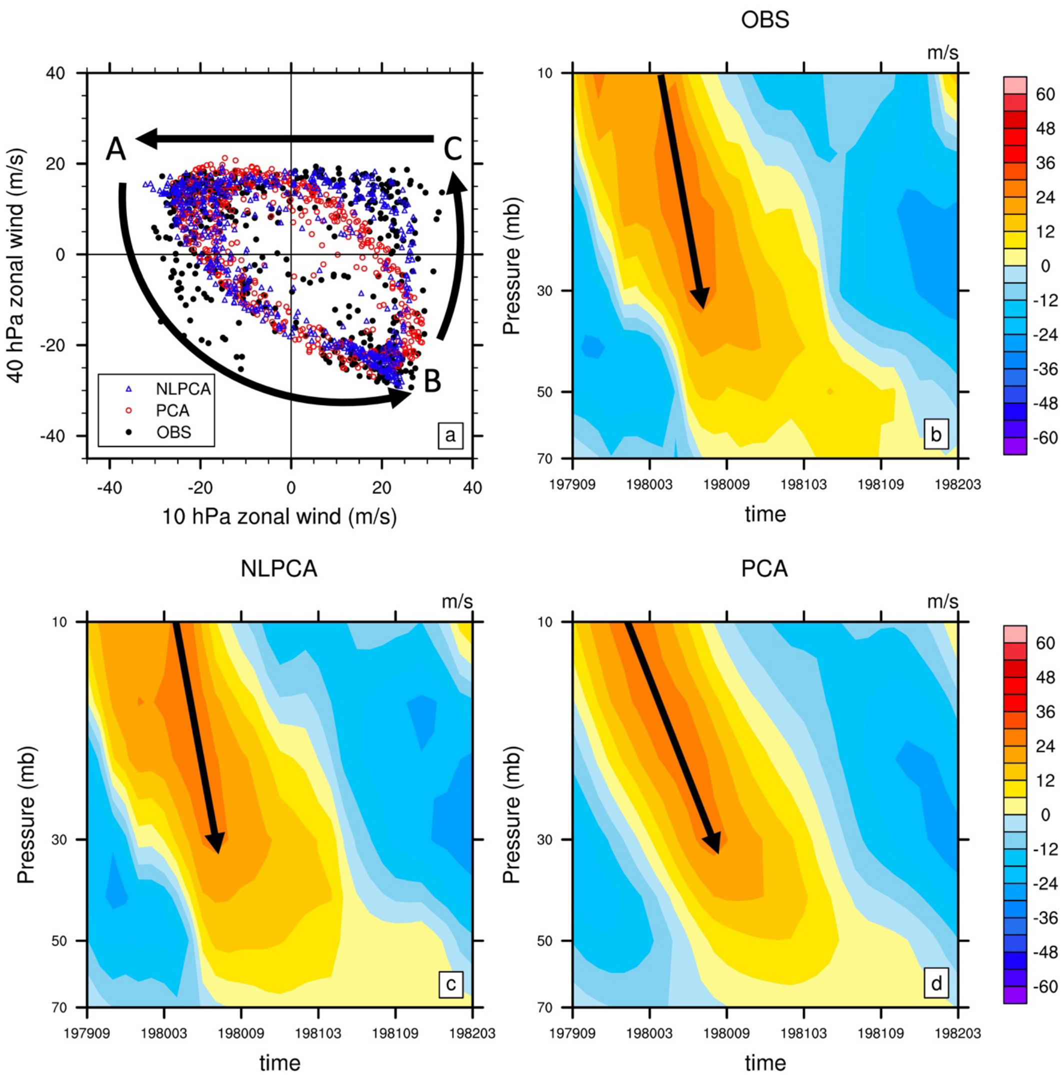

Fig. 1 (a) Scatter-plot of 1979-2020 anomalous monthly mean zonal wind (m/s) at 10-hPa vs 40-hPa for observation (black), NLPCA reconstruction (blue), and PCA reconstruction (red). Typical cycle of QBO from (b) Observational station data from University of Berlin, (c) NLPCA reconstruction, (d) PCA reconstruction.

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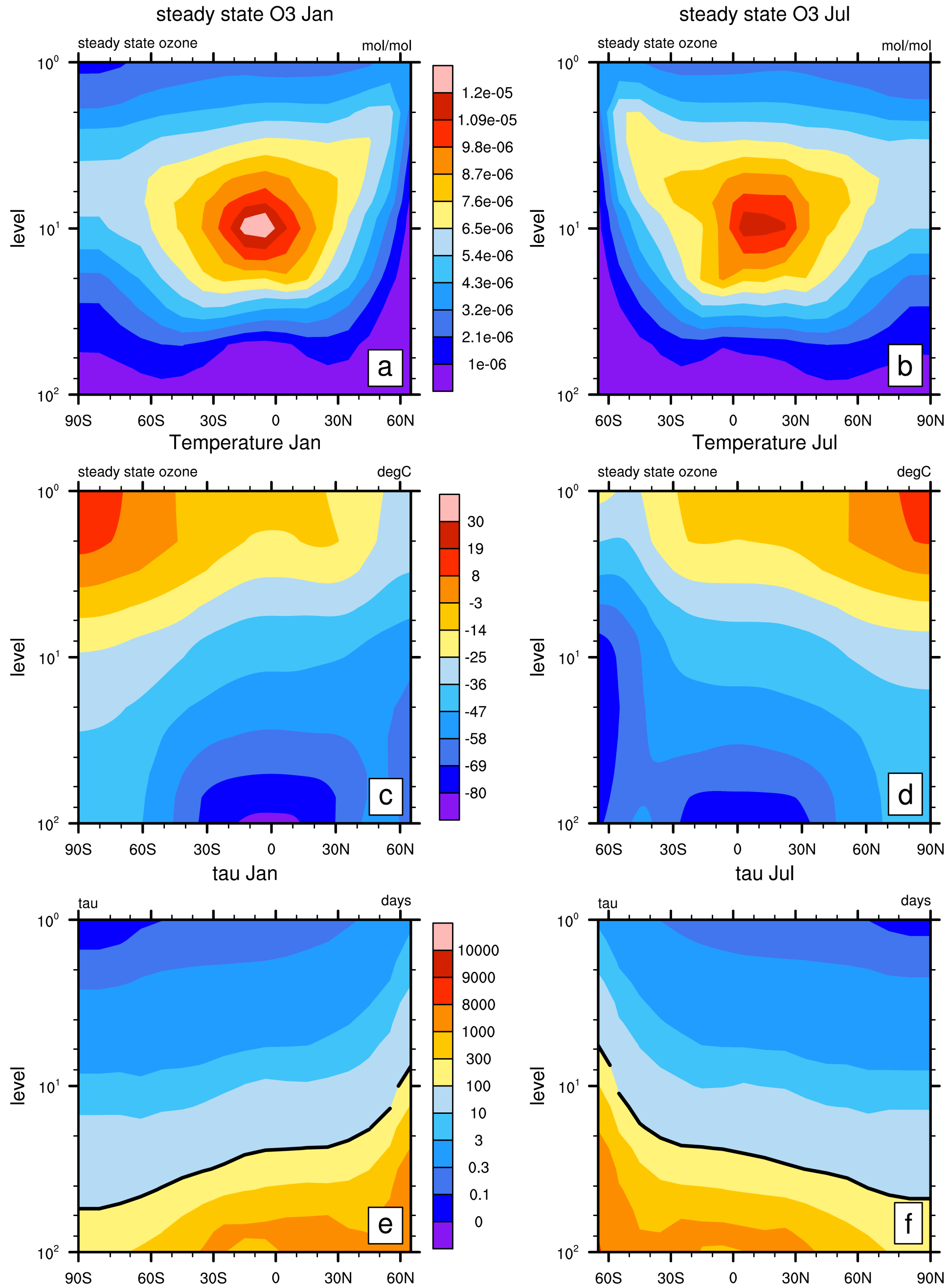

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Fig. 2 The (a, b) steady state ozone (mol/mol) derived using LINOZ on E3SMv2 temperature, (c, d) ERA5 temperature (°C), (e, f) photochemical relaxation time τ (days), and for January and July. The thick black line in (c, d) denotes the 300 value-line.

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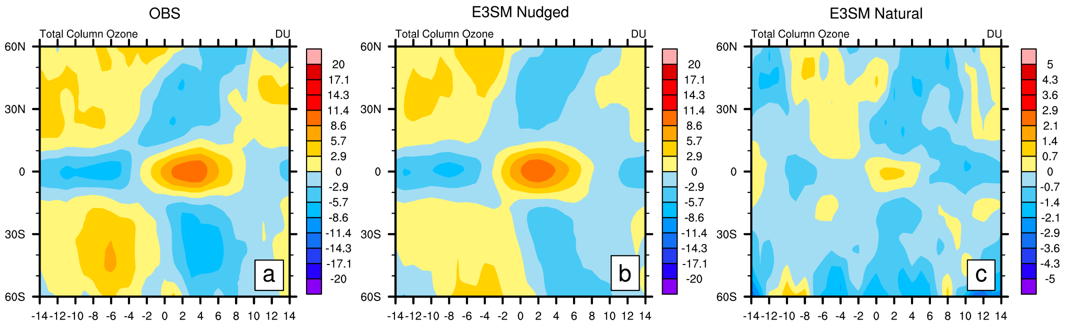

Fig. 3 Total column ozone (TCO, Dobson Unit) anomaly (relative to 1979-2020 mean) composites as function of QBO phase (determined by NLPCA QBO index) for (a) OBS (Multi-Sensor Reanalysis version 2), (b) E3SMv2 nudged simulation, (c) E3SMv2 natural simulation. 0 is centered on the month when QBO transits from QBOe to QBOw (determined by when current QBO index<0 and next QBO index>0). The QBO phase is determined by 5S-5N average of the zonal wind.

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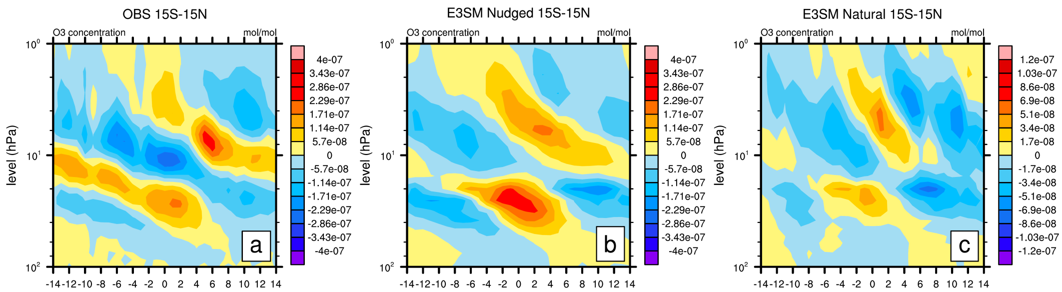

Fig. 4 Pressure-time cross-section of the tropical (15°S-15°N) 1979-2020 anomalous ozone concentration (mol/mol) as function of QBO phase for (a) OBS (Concentration Monthly Zonal Mean), (b) E3SMv2 nudged simulation, (c) E3SMv2 natural simulation. 0 is centered on the month when QBO transits from QBOe to QBOw (determined by when current QBO index<0 and next QBO index>0). The QBO phase is determined by 5S-5N average of the zonal wind.

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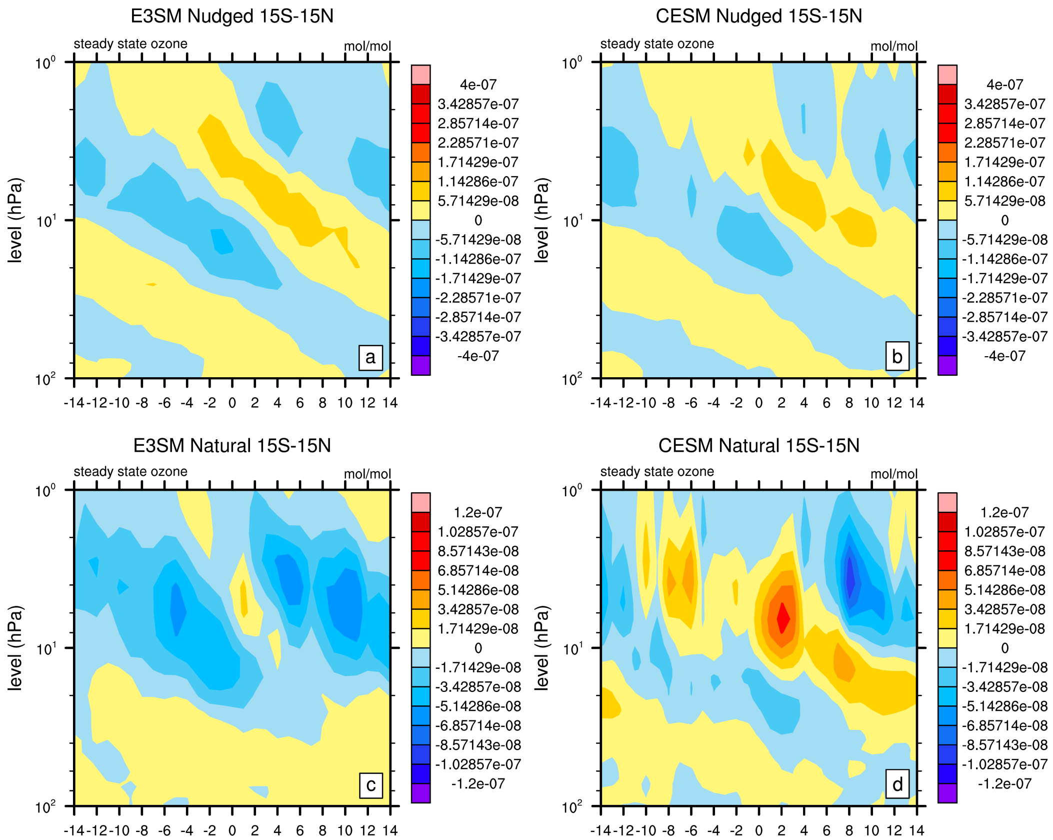

Fig. 5 Pressure-time cross-section of the tropical (15°S-15°N) 1979-2020 anomalous Linoz steady state ozone (mol/mol) as function of QBO phase for (a, c) E3SMv2 and (c, d) CESM2. 0 is centered on the month when QBO transits from QBOe to QBOw (determined by when current QBO index<0 and next QBO index>0). The QBO phase is determined by 5S-5N average of the zonal wind.

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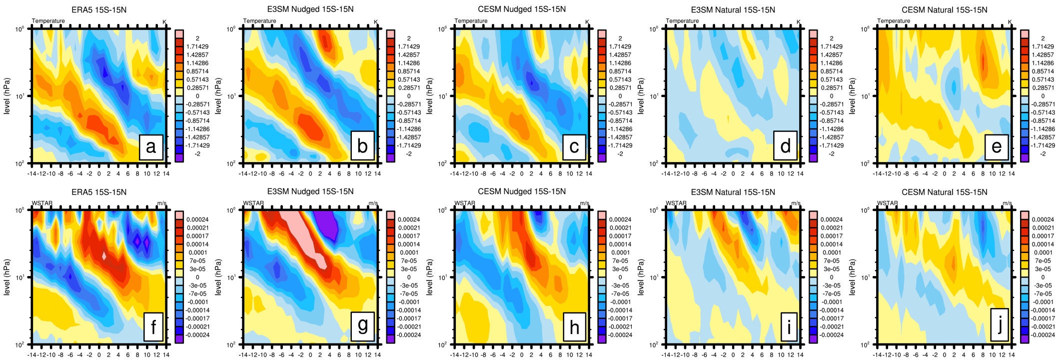

Fig. 6 Pressure-time cross-section of the tropical (15°S-15°N) 1979-2020 anomalous temperature (K) and $\underline{w}^*$ (Transformed Eulerian Mean residual vertical transport, m/s) as function of QBO phase for (a, f) ERA5, (b, d, g, i) E3SMv2, (c, e, QBOe, j) CESM2. 0 is centered on the month when QBO transits from QBOe to QBOw (determined by when current QBO index<0 and next QBO index>0). The QBO phase is determined by 5S-5N average of the zonal wind.

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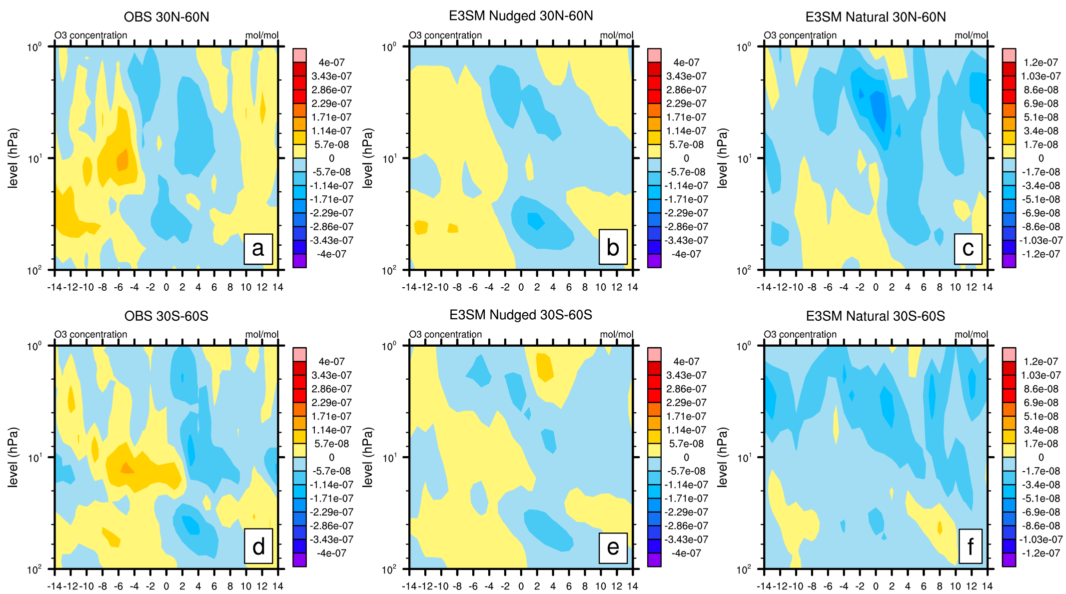

Fig. 7 Pressure-time cross-section of the extratropical (30°N-60°N/30°S-60°S) 1979-2020 anomalous ozone concentration (mol/mol) as function of QBO phase for (a, d) OBS (CMZM), (b, e) E3SMv2, (c, f) CESM2. 0 is centered on the month when QBO transits from QBOe to QBOw (determined by when current QBO index<0 and next QBO index>0). The QBO phase is determined by 5S-5N average of the zonal wind.

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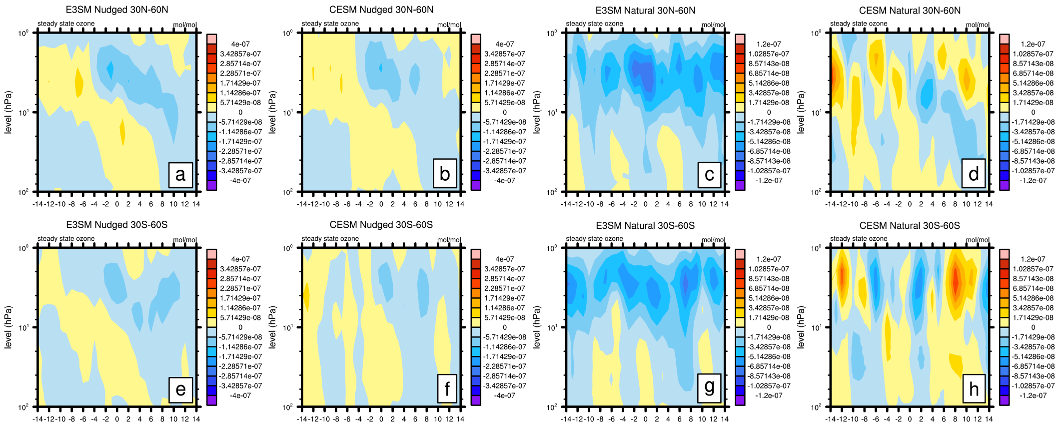

Fig. 8 Pressure-time cross-section of the extratropical (30°N-60°N/30°S-60°S) 1979-2020 anomalous steady state ozone (mol/mol) as function of QBO for (a, e) E3SMv2 nudged, (b, f) CESM2 nudged, (c, g) E3SMv2 natural, (d, QBOe) CESM2 natural. 0 is centered on the month when QBO transits from QBOe to QBOw (determined by when current QBO index<0 and next QBO index>0). The QBO phase is determined by 5S-5N average of the zonal wind.

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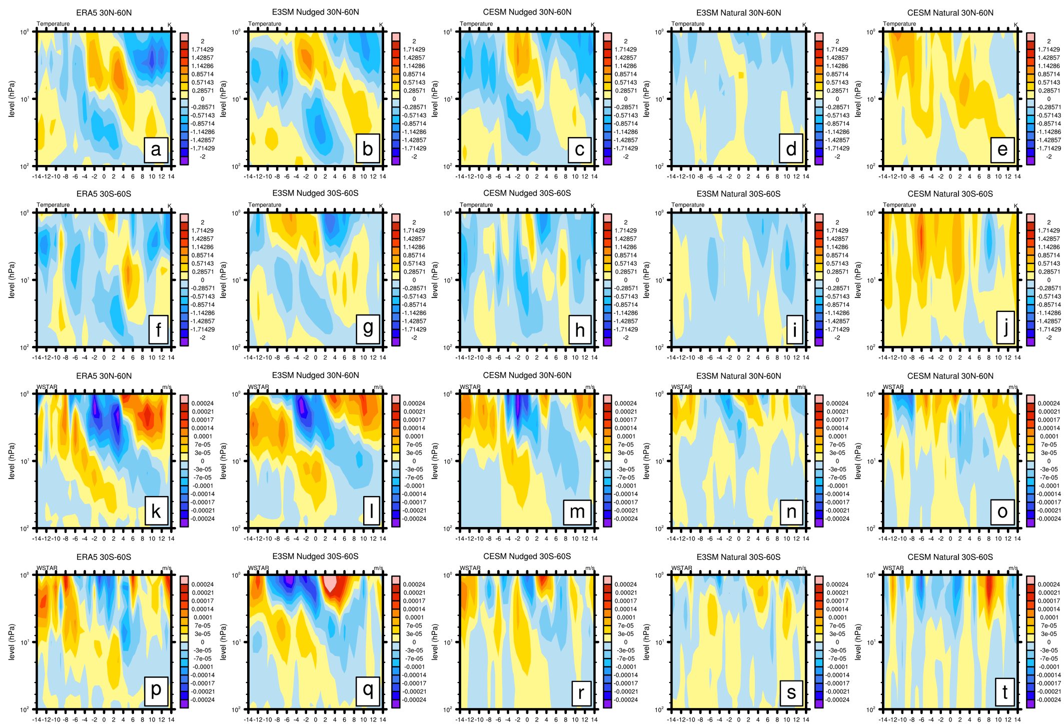

Fig. 9 Pressure-time cross-section of the extratropical (30°N-60°N/30°S-60°S) 1979-2020 temperature (K)/ $\underline{w}^*$ (m/s) as function of QBO for (a, f, k, p) ERA5, (b, g, l, q) E3SMv2 nudged, (c, QBOe , m, r) CESM2 nudged, (d, i, n, s) E3SMv2 natural, (e, j, o, t) CESM2 natural. CESM2. 0 is centered on the month when QBO transits from QBOe to QBOw (determined by when current QBO index<0 and next QBO index>0). The QBO phase is determined by 5S-5N average of the zonal wind.

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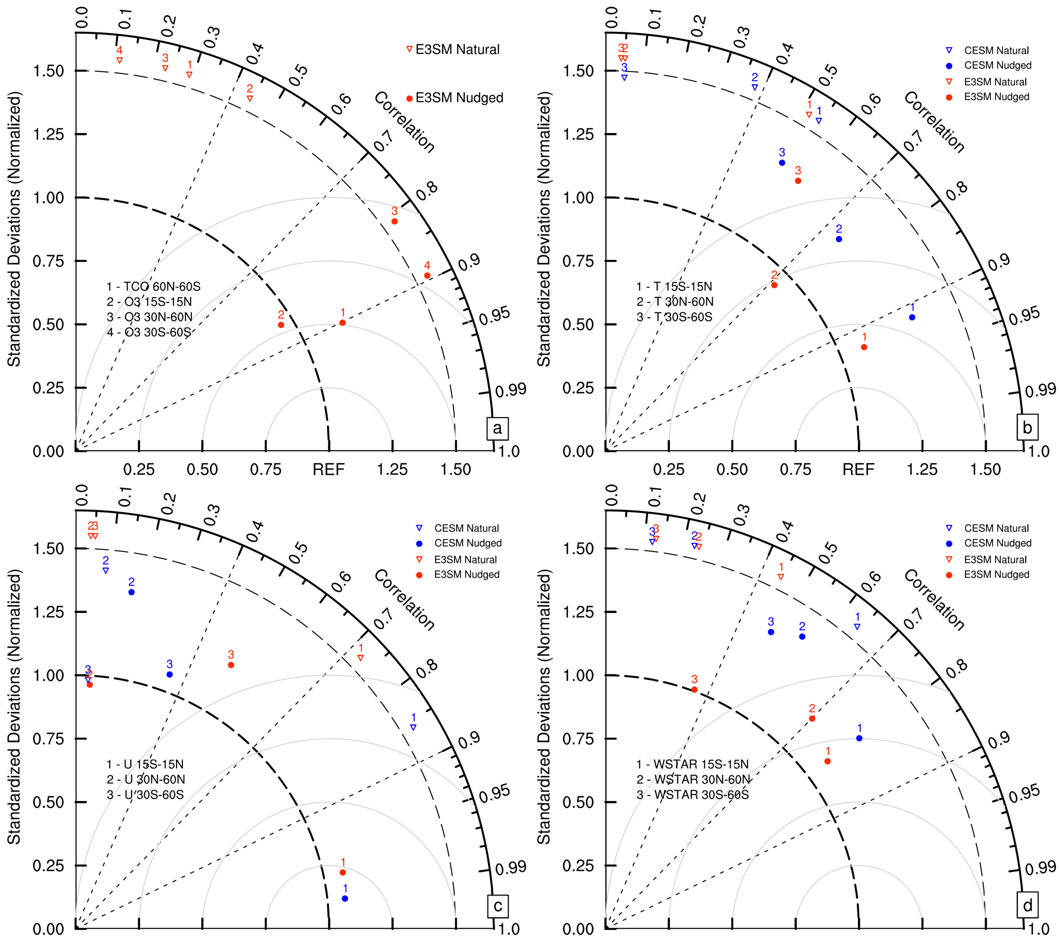

Fig. 10 Taylor diagram of the E3SMv2/CESM2 simulation for various datasets for 1979-2020. (a) The area-weighted total column ozone (60°S-60°N, DU) and pressure-time cross-sections of ozone concentration (15°S-15°N, 30°N-60°N, 30°S-60°S, mol/mol) anomalies with OBS (MSR and CMZM), respectively. (b) The area-weighted pressure-time cross-sections of temperature (15°S-15°N, 30°N-60°N, 30°S-60°S, K) anomalies with ERA5. (c) The area-weighted pressure-time cross-sections of zonal wind (15°S-15°N, 30°N-60°N, 30°S-60°S, m/s) anomalies with ERA5. For pattern correlations, the cross-sections are weighted by pressure layer thickness. On all Taylor diagrams, the model standard deviations are normalized by dividing the standard deviations of the reference.

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