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Responses to Referee #2

Li et al. proposed a connection between ozone pollution in central China and the stratospheric quasi biennial oscillation (QBO) under the warm phase of the El Niño–Southern Oscillation (ENSO). This topic is interesting and can be potentially important. The authors are commended for their effort to explore the related mechanisms for the proposed connection between QBO and surface ozone in China. The paper reads well. I recommend the paper to be accepted subject to revision that addresses the following points.

We thank the reviewer for all the insightful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

1. Lines 157-159, why? Do the emissions change more largely than in other years during the period?

Response:

The variations in O_3 concentrations are driven by a combination of changes in precursor emissions and the meteorological conditions. Anthropogenic emissions are the largest contributor to variation in tropospheric O_3 concentrations over multidecadal timescale, at least for several years (Fu and Tai, 2015; Cooper et al., 2012; Xu et al., 2008). On the interannual time scale, the variations in meteorological conditions have significant influences on surface O_3 concentrations (Ding et al., 2019; Li et al., 2020). Therefore, we compared the year-by-year variation in JJA O_3 concentrations in observations and BASE simulation rather than the trend, which is more related to the emission changes. The results show that GEOS-Chem could well capture the effect of meteorological parameters on O_3 concentrations over China.

The correlation coefficient between the observed and simulated 2018–2019 O_3 concentration changes is only 0.16, which may be partly attributed to the emission changes. The Clean Air Action Plan initiated in 2013 rapidly decreased pollutant emissions. However, ozone increased over the 2013–2017 period in the megacity clusters of eastern China (Lu et al., 2020). In 2018, Phase 2 of the Clean Air Action Plan was launched, which imposed new emission controls targeted at O_3 (Li et al., 2020). This may be one of the reasons for the significant decrease in O_3 concentration in 2019 compared to 2018 from observations (Fig. 1c). Ma et al. (2021) also noticed that MDA8 O_3 showed a decreasing trend in 2019 relative to 2018, which was opposite to that during 2013–2018.

In addition, Mousavinezhad et al. (2021) suggest that the meteorology in

2019 was favorable to the formation and the accumulation of O_3 in BTH, the YRD, and the PRD, by using MLR to separate the contributions from meteorology and precursor emissions to O_3 variations. This result is consistent with the increase of O_3 in 2019 in BASE simulation, implying a good performance of the model. In this study, the anthropogenic, biomass burning and natural emissions were fixed at 2017 levels to remove the impact of year-to-year emission changes. The spatial correlation coefficient between the observed and simulated O_3 concentrations was 0.87 in the year 2017, which also indicated that the GEOS-Chem model is credible in simulating O_3 concentration.

We have added a brief description in the manuscript:

The spatial correlation coefficients between the observed and modeled year-by-year changes in O_3 concentrations are about 0.5–0.6, except the 2018-to-2019 changes in O_3, which could be attributed to the influence of the changes in precursor emissions on the observed O_3 concentrations after Phase 2 of the Chinese Clean Air Action Plan launched in 2018 (Li et al., 2020).

2. Lines 161-171, Generally, the impact of climatic modes on some processes are investigated using monthly or seasonal climate indices with consideration of different lags, rather than the annual mean.

Response:

We apologize for not explicitly describing these indices. QBO and Nino 3.4 indices used in this study are defined as the average of the monthly indices during June, July, and August. We have added a more detailed interpretation as follows:

The QBO phases are determined by the zonal average of 30 hPa zonal wind over the equator (5°S–5°N) based on MERRA-2 reanalysis (Fig. 2a), with the averages during JJA used in this study.

The Niño 3.4 index averaged over JJA is used to characterize the warm and cold phases of SST anomaly over the eastern tropical Pacific in boreal summer, which is estimated as the SST anomalies over the Niño 3.4 region (5°S–5°N, 170°–120°W).

We have also performed the lag-correlation analysis. The results show that the regional correlation coefficient (r) between QBO index and surface O_3 in central China is 0.23 (p=0.16) during the whole 40-year period. The lag-correlations between the O_3 concentrations over central China and QBO index are even lower, with correlation coefficients of 0.10 (p=0.5) for a three-month of QBO index ahead of the O_3 concentrations and –0.14 (p=0.35) for a six-month of QBO index ahead of the O_3 concentrations.

We have summarized it in the manuscript as “The lag-correlation analysis is also performed but shows even weaker correlations.”

3. Line 258-265, For Figure 6, please explain why the spatial variation of the

differences in TCO in (a) and (b) are different?

Response:

The differences in JJA TCO between the selected QBOW year and QBOE year are based on the BASE simulation with emissions fixed at 2017 level, which is only influenced by meteorology fields. However, the differences of TCO from Aura OMI/MLS measurements are driven by a combination of emissions and meteorological conditions. This may be the main reason for the difference in spatial variation of TCO.

In addition, OMI/MLS tropospheric column ozone was determined daily by subtracting co-located MLS stratospheric column ozone (SCO) from OMI total column ozone each day, known as the tropospheric ozone residual method. This approach involved adjusting for calibration differences between the two instruments, which may cause the retrieval errors (Schoeberl et al., 2007; Liu et al., 2010; Ziemke et al., 2014). Meanwhile, MLS measurements are along-track only. A 2-D interpolation scheme is used to fill in data between orbital gaps to establish daily SCO maps (Ziemke et al., 2006). These factors are responsible for the differences between the GEOS-Chem model and satellites.

We have added the explanation in the manuscript as "However, it is also noted that the spatial variation of the differences in TCO varies between OMI/MLS and model simulation. It is partly because the emissions were fixed at the 2017 levels during model simulations. These potential biases in satellite retrievals also strongly contribute to the different spatial pattern (Schoeberl et al., 2007; Liu et al., 2010; Ziemke et al., 2006, 2014)."

4. Lines 276-278, Please elaborate more about this method, how is the 1% calculated?

Response:

Integrated process rate analysis has been widely conducted to assess the contribution of individual chemical or physical processes to the production and distribution of O_3 pollution per unit time in the study domain (Lou et al., 2015; Qu et al., 2021; Zhu et al., 2021). We have added this description in the manuscript.

According to the integrated process rate analysis, the net chemical production of O_3 from surface to the PBL is lower by –0.09 Gg d^{-1} over central China during QBOW compared to QBOE years (Table S1).

The calculation process is as follows:

0.09 / 7.53 ≈ 1%

Table S1. Net rate of change in O_3 mass (Tg $Season^{-1}$) of various processes from surface to the PBL over central China (92.5–112.5°E, 26–38°N) during the selected three QBOW years (1990, 1997, 2019) and QBOE years (1994, 2012, 2018) and their differences (QBOW-QBOE).

	Net chemical production	Horizontal advection	Diffusion and dry deposition	Vertical convection
QBOW	7.42	-0.35	-6.42	0.43
QBOE	7.53	-0.35	-6.31	0.34
Difference	-0.09	0.00	-0.11	0.09

5. Lines 287-289, an increase in the boundary height would actually dilute ozone concentrations in the surface. If so, how significant is this process?

Response:

The development of the planetary boundary layer can modulate the vertical extent of turbulent mixing, vertical diffusion and convective transport in the lower troposphere, which affects air pollutant concentrations (Guo et al., 2016; Duc et al., 2022). The lower PBLH may constrain vertical mixing and lead to the accumulation of air pollutants (Gao et al., 2015). The increased PBLH was conducive to enhance the atmosphere's ability to disperse particulate matters and improve PM2.5 air quality (Miao et al., 2018, Chen et al., 2020).

However, the sources of troposphere O_3 are complex, including downward transport of stratospheric ozone, photochemical reaction products of tropospheric nitrogen oxides (NOx) and volatile organic compounds (VOCs), long-range transport of O_3, which makes it difficult to study the influence of PBLH on near-surface O_3. Ma et al. (2021) showed that PBLH has a significant positive correlation with MDA8 O_3 over NCP region. They used numerical simulations with the National Center for Atmospheric Research Master Mechanism model to quantify the PBLH to the change in surface O_3 and found that the increase in PBLHs contribution about 18% to the increment in surface O_3. Gong et al. (2019) also reported a positive correlation between O_3 concentration and PBLH.

We have modified the biased expression in the revised manuscript as the following:

In addition, the increase in planetary boundary layer (PBL) height (Fig. 7d) favors the vertical mixing of air within the PBL and the O_3-enriched air above the PBL (Gong et al., 2019; Ma et al., 2021).

6. Lines 290-308, Table 1 is well done. It suggests that vertical transport is one of the causes for the proposed QBO-ozone relationship. I suggest that the authors provide a complete budget analysis that also includes other components, such as net chemical production and deposition, so it is convincing that vertical transport is the dominant factor.

Response:

As suggested by the reviewer, we summarize the process source/sink rates in Table S1. The major processes that influence O_3 concentrations include net chemical production, horizontal advection and vertical convention, diffusion and dry deposition. The role of each physical or chemical process can be quantified by the Integrated Process Rate analysis. However, we should note that the IPR values represent the instantaneous change in O_3 mass, which does not directly reflect the variation of O3 concentrations averaged over a long period.

7. Lines 326-332, Can these statements be supported by the surface measures?

Response:

Surface measurements can only tell us the increase or decrease in O_3 concentrations, but it is hard to identify which physical or chemical process dominates the O_3 change. That is also why we use the model to quantify the importance in individual processes. GEOS-Chem model contains the process analysis module which quantifies the contributions of individual physical and chemical processes to O_3 change. The major processes that influence O_3 concentrations include net chemical production, horizontal advection and vertical convention, diffusion and dry deposition. The role of each physical or chemical process can be quantified by the Integrated Process Rate (IPR) analysis. In this study, we used IPR analysis to identify the dominant role in enhancing the O_3 concentrations during QBOW years.

8. Overall, the analysis can be carried out more comprehensively. How can changes in stratospheric wind field in the tropics be connected to surface ozone in the middle latitudes? Are the two climatic modes equally important? Or one is more important? The authors can enhance their analysis and thus make their points more convincing in the revision.

Response:

The increase in near-surface O_3 over central China is mainly attributed to the anomalous downdraft in this study. The QBO of zonal winds is a prominent dynamical feature in the equatorial stratosphere. The QBO is driven by waves generated by convection in the troposphere and propagate upward into the middle and upper atmosphere (Schirber et al., 2015). The potential mechanisms of QBO and tropospheric interaction process have been extensively discussed (Giorgetta et al., 1999; Huang et al., 2012; Lee et al., 2019). Huangfu et al. (2021) suggest that the westerlies over the equator and the easterlies over the offequator form a cyclonic band, providing an upwelling force to the tropopause and affecting Pacific Wallker circulation.

According to the reconstructed records, Wang et al. (2021) show that the development of Walker circulation influences Asian Summer Monsoon strength.

Yuan et al. (2008) suggest that an anomalous reversed Wallker circulation leads to descending motion and hence suppressed convection in the western Pacific, which favors a later onset of the South China Sea summer monsoon. These results imply a significant influence of walker circulation on the climate of the mid-latitudes. Hence, we suspect that the vertical motion over China is obviously influenced by anomaly of walker circulation caused by QBO. The hypothetical mechanism should be proved by model sensitivity experiments. Although the physical mechanism for relationship remains elusive, we believe that our findings would be useful for future air pollution prediction and control. Meanwhile, we have added the discussion in the revised version of the manuscript, as follows:

"Also, it is assumed that the vertical motion over China is influenced by anomaly of Walker circulation caused by the QBO (Huangfu et al., 2021). Although the physical mechanism remains elusive, we believe that our findings would be useful for future air pollution prediction and control."

In the previous study, we investigated the impact of ENSO on summertime near-surface O_3 concentrations in China. The results show that simulated near-surface O_3 concentrations averaged over southern China (97.5–117.5°E, 20–32°N) present a positive correlation with ENSO index, with statistically significant correlation coefficient between O_3 and Niño 3.4 index of +0.55. Furthermore, our analysis suggests that the O_3 flux convergence associated with weakened southerlies is the primary cause of the increase in O_3 over southern China. And the increased O3 during El Nino years is mainly from domestic emissions.

In this study, QBO has a significant positive correlation with near-surface O_3 concentrations over central China (92.5°–112.5°E, 26°–38°N) when the sea surface temperature (SST) over the eastern tropical Pacific is warmer than normal, with a correlation coefficient of 0.53, but QBO has no significant effect on O_3 under the cold SST anomaly. Moreover, the O_3 increase over central China is mainly attributed to the anomalous downward transport of O_3 during the westerly phase of QBO when a warm SST anomaly occurs in the eastern tropical Pacific. When domestic anthropogenic emissions of O_3 precursors are turned off, JJA near-surface O_3 concentrations largely still increase across China.

Therefore, we find that ENSO has an impact on the near-surface O_3 over southern China due to weakened southerlies. However, QBO manifests its impacts on the O_3 over central China only during the warm SST phase, which is attributed to the anomalous downward transport of O_3. Because of the different regional and dominant mechanisms affected by QBO and ENSO over China, an ad hoc estimate of which modes are more important between ENSO and QBO is difficult. We assume that ENSO and QBO play a synergistic role in modulating O_3 pollution in China.

Minor points:

9. Title: this paper is focused on summer season. “summer” needs to be indicated in the title.

Response:

Thank you for the suggestion. We have changed the title to “Summertime ozone pollution in China affected by stratospheric quasi-biennial oscillation”.

10. L53, replace “it is” to “surface ozone is”.

Response:

Changed.

11. L67, “Yang et al. 2022”? which of “Yang et al. 2022”. Please indicate “Yang et al. 2022a” or “Yang et al., 2022b” throughout the manuscript.

Response:

Clarified.

12. L149-152, this may be the case for some regions of China. Ozone pollution can be serious in other regions of China.

Response:

Thank you for your comment. We have corrected it to “Considering that O_3 pollution is most critical during the boreal summer in many regions of China, only summer months (June-July-August, JJA) are examined in this study.”

13. L181, between 25° to 40°N, tropopause height decreases with altitudes largely from 100 to-250 hPa. Use a fix 150hPa can results in some biases.

Response:

Based on previous studies (Jing et al., 2006; Peiro et al., 2018), we used 150 hPa as an approximation of the tropopause level, which results in some biases in the calculation of TCO from the model simulation.

Meanwhile, we calculated the TCO using 100 and 250 hPa as approximate values of the tropopause in the following Figure. The difference in TCO between the selected QBOW (2019) and QBOE years (2012 and 2018) over central China is 2.7 and 2.2 DU, respectively, which is similar to the 2.5 DU around 150 hPa. Averaged over central China, the difference in TCO from satellite data is 2.8 DU. By comparison, we find that there is a slight bias in the values of the TCO calculated by choosing different heights as the tropopause, but this does not largely affect the qualitative results.

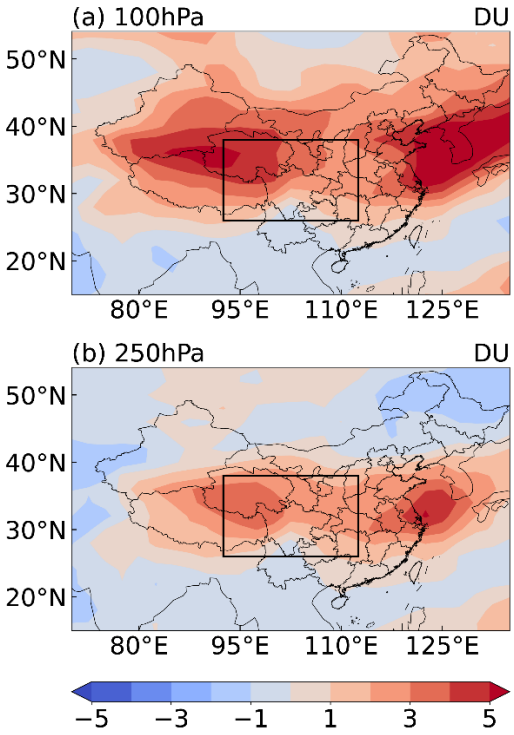

Figure A. Spatial distribution of JJA tropospheric column O_3 (TCO, DU) difference between the selected QBOW year (2019) and QBOE year (2012, 2018) based on Aura OMI/MLS (a) using 100 hPa and (b) 250 hPa as approximate values of the tropopause.

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