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Responses to Referee #1

Stratospheric quasi-biennial oscillation (QBO) is an important climate mode that not only modulates the variability in tropical climate system, but also has potential influence globally and may further lead to possible impacts on air quality. Li et al. examined the effects of QBO on interannual variabilities of tropospheric ozone over China mainly by correlation analysis with the help of GEOS-Chem simulations. It is quite an interesting and novel topic. The manuscript is well-organized and easy to follow. I suggest it to be published after addressing my comments below.

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 62-64: The 'downward transport of stratospheric ozone into troposphere' is usually named by 'stratospheric ozone intrusion'. I suggest authors using the proper terminology here.

Response:

Thank you for the suggestion. We have revised the description as "The intrusion of stratospheric O_3 into the troposphere is also one of the sources for near-surface O_3 (Zeng et al., 2010; Wespes et al., 2017)."

2. Lines 69: O3 pollution is getting worse in recent years, not decades. At least the two references did not show trends over 10 years.

Response:

Revised as suggested. "O_3 pollution is getting worse in China in recent years (Li et al., 2019; Gao et al., 2022)."

3. Line 153: If the anthropogenic emissions are fixed in 2017, then why use this simulation to evaluate simulated O3 interannual variability? I think changes in anthropogenic emissions could significantly influence O3 year-by-year. Also, I am very confused by the last sentence in this paragraph. Did anthropogenic emissions significantly change between 2018 and 2019?

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 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).

4. Lines 150-151: O3 concentrations are accounted during summertime, but QBO and Niño 3.4 indices are calculated with annual climate data (If my understanding is right). Will such mismatch significantly influence the results?

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).

5. Lines 174-175: The correlation coefficient numbers should be exhibited in one of the Tables or Figures. The same issue also shows in GEOS-Chem process analysis (lines 277, 294-299 and 305). I strongly suggest authors to add one or several figures to show the differences in O3 budget between QBOW and QBOE.

Response:

We are grateful for the suggestion. The correlation coefficient numbers are added in the top right of each panel of Fig. 2.

The horizontal and vertical O_3 mass fluxes are discussed from line 290 to 308, which have been shown in Table 1.

As suggested by the reviewer, we also 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 (IPR) 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 O_3 concentrations averaged over a long period.

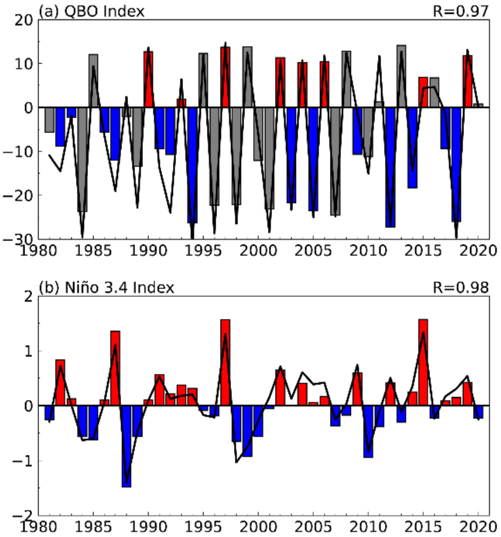

Figure 2. Time series of the JJA mean (a) QBO index (m s^{-1}) and (b) Niño 3.4

index (℃) over 1981–2020. Warm phase of SST anomalies over the eastern tropical Pacific includes 22 years (1982, 1983,1986, 1987, 1990, 1991, 1992, 1993, 1994, 1997, 2002, 2003, 2004, 2005, 2006, 2009, 2012, 2014, 2015, 2017, 2018, 2019) and cold phase includes 18 years (1981, 1984, 1985, 1988, 1989, 1995, 1996, 1998, 1999, 2000, 2001, 2007, 2008, 2010, 2011, 2013, 2016, 2020). Colored bars in (a) indicate years with Niño 3.4 index above zero. The black solid lines represent the indices based on MERRA-2 reanalysis. Bars are QBO index from NCEP/NCEP reanalysis in (a) and Niño 3.4 index from HadISST1 in (b). The correlation coefficients of the indices between MERRA-2 and the NCEP/NCEP reanalysis and between MERRA-2 and HadISST1 are shown in the top right of panels.

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

6. Lines 195-197: Although authors did many analyses in this study to show that O3 differences over China can be significant only when ENSO and QBO were considered together, I still wonder if such insignificant correlations between O3 and QBO were led by some time-lag effects since the tropical QBO signal may need some time to influence China (although I'm not sure about the exact period…). Could authors try some lag-correlation analysis to examine this hypothesis?

Response:

According to the reviewer's suggestion, we have 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."

7. Lines 287-289: I don't think changes in boundary layer height could influence vertical O3 transports between lower and upper troposphere. In addition, I suggest authors to clarify the exact levels of the vertical transport. I guess it is mainly in the free troposphere, since 850 hPa -500hPa is higher than boundary layer, but much lower than stratosphere. If so, I believe such downward transport also cannot be considered as a stratospheric ozone intrusion.

Response:

Many studies suggested that 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 (Reddy et al., 2012; Gao et al., 2015; Guo et al., 2016; Miao et al., 2018; Gong et al., 2019; Chen et al., 2020; Ma et al., 2021; Duc et al., 2022). Therefore, it is necessary to consider changes in PBLH when analyzing the meteorology impacts on atmospheric pollutants. But we agree with the reviewer that the description about the "between lower and upper troposphere" is incorrect. We have revised it as "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)."

We are grateful for the reviewer's suggestion and have added clarification regarding the levels of the vertical transport (300 hPa) in the manuscript:

Line 289-292: Under the influence of this anomalous high, the anomalous downdraft throughout the troposphere over central China (Fig. 7c) can reduce the vertical transport of O_3 to the upper troposphere, which leads to an O_3 accumulation in the lower and mid-troposphere.

We agree with the reviewer that the stratospheric O_3 intrusion may not be the major factor causing O_3 increase over central China. Compared with the QBOE years, O_3 increases above the surface and up to the upper troposphere, with a maximum increase between 850 and 500 hPa over central China during the QBOW. The increase in O_3 is mainly due to the constrained upward mixing of the tropospheric O_3 under the influence of the anomalous downdraft from about 300 hPa to the surface. Therefore, we focus on the effect of vertical transport in the troposphere rather than stratospheric ozone intrusion in this manuscript.

8. Lines 317-321: It is interesting that QBO may have higher influences on O3 in China without anthropogenic emission. Could authors add more explanations here? And I am confused by the statement that this finding is consistent with the significant roles of vertical transport. Does the vertical transport become higher in NO_CHN compared to BASE?

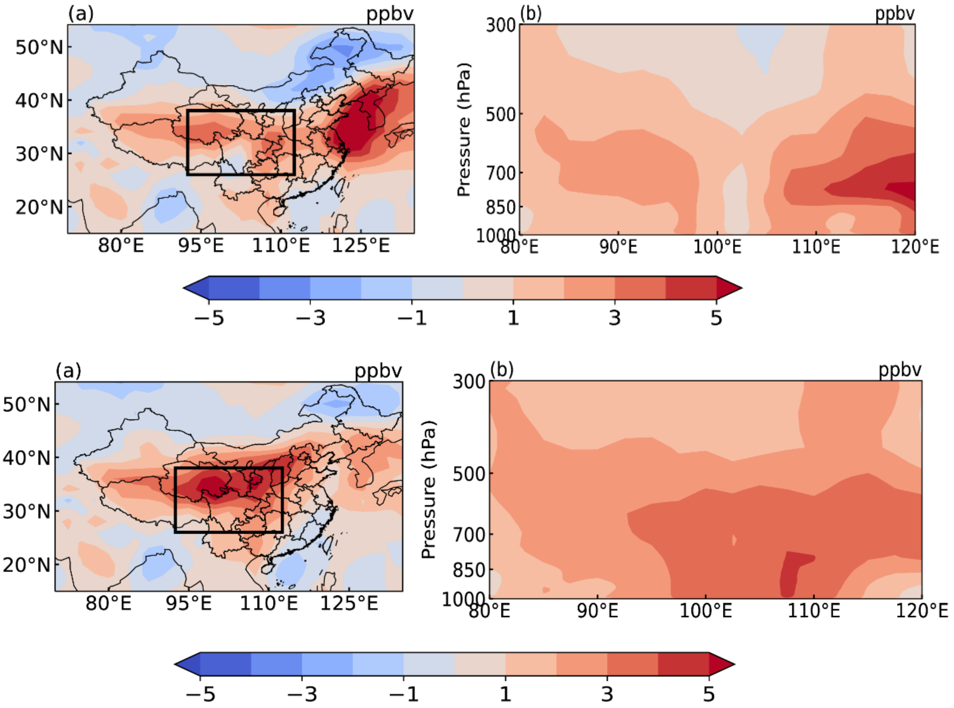

Figure A. (a) Spatial distribution of differences in JJA near-surface O_3 concentrations (ppbv) between three QBOW years (1990, 1997 and 2019) and QBOE years (1994, 2012 and 2018) (QBOW–QBOE) from the (a) BASE and (b) NO_CHN simulations.

Response:

The increase in O_3 near the surface is mainly related to the vertical transport. However, Central China is also affected by an anomalous westerly wind during the QBOW, resulting in a net O_3 export of 0.29 Tg from surface to 850 hPa during the QBOW, compared to the QBOE years (Table 1). The net decrease in horizontal transport partly offsets the increase in vertical transport. And the O_3 near surface is largely contributed by the domestic anthropogenic emissions. When domestic anthropogenic emissions of O_3 precursors are turned off, the net export of O_3 horizontal mass flux is only 0.02 Tg (Table S2) and thus the offsetting effect disappears in NO_CHN simulation. Therefore, the O_3 increase is stronger in NO_CHN than BASE.

We have provided the mass flux for NO_CHN in Table S2 and revised the description as follows:

Averaged over central China, the anomalous increase in near-surface O_3 concentration is 3.0 ppb in NO_CHN, even higher than that (1.7 ppb) in BASE simulation. It results from that the reduction in the net export of horizontal mass flux of O_3 due to the removal of domestic emissions (Table S2) leads to a more significant increase in O_3 over central China in the NO_CHN experiment.

Table S2. The horizontal and vertical mass flux (Tg) of JJA O_3 concentration from the surface to 850 hPa over central China (92.5–112.5°E, 26–38°N) based on NO_CHN simulation. The values are averaged over the selected three QBOW years (1994, 2012, 2018) and QBOE years (1990, 1997, 2019) and their differences (QBOW-QBOE). Positive values indicate incoming fluxes and negative values indicate outgoing fluxes.

	QBOW	QBOE	Difference
		Horizontal mass flux	
East	0.44	1.15	-0.71
West	0.95	0.80	0.15
North	0.77	0.09	0.68
South	3.20	3.34	-0.14
		Vertical mass flux	
Top	-4.11	-4.49	0.38

9. Lines 329-332: Could authors provide data or numbers to support this statement?

Response:

According to the comment 8, the horizontal transport of O_3 is not favorable for O_3 accumulation in QBOW compared to QBOE years. When domestic anthropogenic emissions of O_3 precursors are turned off, the effect of horizontal transport is weakened, resulting in a more significant increase in the NO_CHN experiment. The original statement is incorrect. We have revised the description in the manuscript:

In the NO_CHN experiment, the reduction in the O_3 horizontal export results in a more significant increase of O_3 concentration during QBOW compared to QBOE years.

10. Lines 366-373: I suggest to increase some discussion in the dynamic mechanism, although it may slightly beyond the scope of this study. At least one significant question needs to be answered: If the related upward-downward motion transition between QBOE and QBOW are important for O3 in China, why the correlation coefficient between O3 and QBO index is insignificant? What are possible roles of ENSO in influencing meteorological factors in China between QBOW and QBOE years? I believe further discussion depending on data analysis or literature is necessary.

Response:

Thank you for the suggestion. Based on the radiosonde observations, Taguchi (2010) reported a faster QBO downward propagation rate during El Niño conditions, especially westerly QBO phase. Schirber (2015) further analyzed the mechanisms of changes in QBO downward propagation due to

ENSO. Due to the increase in tropospheric temperature under El Niño conditions compared with LA conditions, tropospheric wave activity increases, which strengthens stratospheric QBO forcing. They found that the changes in QBO properties during ENSO were driven by analytical and parametric waves (Christiansen et al., 2016). During El Niño condition, the weaker underlying jet and the increase forcing due to waves cause a faster downward propagation in QBOW. Geller et al. (2016) hypothesized that the more widespread deep convection that occurs in connection with El Niño lead to greater zonally averaged GWMFs, in turn, leading to more rapid descent of QBO westerlies and easterlies. The QBO induced residual circulation propagates downwards, affecting the tropopause and upper troposphere (Zheng et al., 2007). Therefore, we argue that the QBO downlink propagation rate can represent the extent of QBO penetration into the troposphere, implying that a faster propagation rate reflects a more significant impact of QBO on the troposphere. We speculate that this may be the reason why the correlation coefficient between the O_3 and QBO indices is insignificant, but shows a significant correlation during El Niño.

Meanwhile, we have added the discussion in the revised version of the manuscript, as follows:

Compared with cold conditions, stratospheric QBO forcing is strengthened due to the increase of tropospheric temperature and changes of analytical and parametric waves under warm SST anomalies of the eastern tropical Pacific, which causes a faster downward propagation in QBO (Taguchi, 2010; Schirber et al., 2015; Geller et al., 2016; Zheng et al., 2007). This may explain why the correlation coefficient between the O_3 and QBO indices is insignificant, but shows a significant correlation during warm SST anomalies of the eastern tropical Pacific. The mechanisms deserve further investigation in future studies.

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