

21 **Abstract**

22 In recent years, near-surface ozone (O₃) level has been rising fast in China,
23 with increasing damages to human health and ecosystems. In this study, the
24 impact of stratospheric quasi-biennial oscillation (QBO) on interannual
25 variations in summertime tropospheric O₃ over China is investigated based on
26 GEOS-Chem model simulations and satellite retrievals. QBO has a significant
27 positive correlation with near-surface O₃ concentrations over central China
28 (92.5°–112.5°E, 26°–38°N) when the sea surface temperature (SST) over the
29 eastern tropical Pacific is warmer than normal, with a correlation coefficient of
30 0.53, but QBO has no significant effect on O₃ under the cold SST anomaly.
31 Compared to the easterly phase of QBO, the near-surface O₃ concentrations
32 have an increase of up to 3 ppb (5% relative to the average) over central China
33 during its westerly phase under the warm SST anomaly. O₃ also increases
34 above the surface and up to the upper troposphere, with a maximum increase
35 of 2–3 ppb (3–5%) in 850–500 hPa over central China comparing westerly
36 phase to easterly phase. Process-based analysis and sensitivity simulations
37 suggest that the O₃ increase over central China is mainly attributed to the
38 anomalous downward transport of O₃ during the westerly phase of QBO when
39 a warm SST anomaly occurs in the eastern tropical Pacific, while the local
40 chemical reactions and horizontal transport processes partly offset the O₃
41 increase. This work suggests a potentially important role of QBO and the
42 related vertical transport process in affecting near-surface O₃ air quality, with
43 an indication for O₃ pollution prediction and prevention.

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50 **1. Introduction**

51 Ozone (O₃) is an important atmospheric trace gas. The presence of O₃ in
52 the stratosphere plays a crucial role in protecting the environment and humans
53 from UV light, but surface O₃ is detrimental to human health, ecosystem and
54 agricultural production within the troposphere (Wang et al., 2007; Nuvolone et
55 al., 2018; Zhao et al., 2020). Tropospheric O₃ is primarily produced by
56 photochemical reactions of nitrogen oxides (NO_x) and volatile organic
57 compounds (VOCs) (Wang et al., 2017). Apart from precursor emissions, the
58 temporal and spatial distribution of tropospheric O₃ is highly impacted by
59 meteorological conditions, among which low relative humidity (RH), cloud free,
60 strong solar radiation and high temperatures can lead to O₃ pollution by
61 enhancing its chemical production (Camalier et al., 2007; Porter et al., 2019;
62 Gong and Liao, 2019; Qu et al., 2021; Wang et al., 2022). The intrusion of
63 stratospheric O₃ into the troposphere is also one of the sources for near-surface
64 O₃ (Zeng et al., 2010; Wespes et al., 2017).

65 With the accelerated industrialization and urbanization in recent decades,
66 the air quality problem has become serious in China (Verstraeten et al., 2015;
67 Yang et al., 2022a). Although many environmental protection and control
68 measures have been implemented to prevent air pollution (Feng et al., 2019),
69 O₃ pollution is getting worse in China in recent years (Li et al., 2019; Gao et al.,
70 2022). Therefore, the factors causing O₃ pollution have been a research focus
71 in recent years. Many previous studies found that interannual variations in
72 large-scale circulations modulated the O₃ pollution in China (e.g., Yang et al.,
73 2014; Yin et al., 2017; Zhao and Wang, 2017). For example, Yang et al. (2014)
74 showed that summertime O₃ levels in China were positively correlated with the
75 strength of East Asian summer monsoon (EASM) associated with variations in
76 large-scale circulations, which led to an increase in O₃ concentration exceeding
77 6% in the strong EASM years relative to the weak ones.

78 El Niño–Southern Oscillation (ENSO) is the most pronounced mode of

79 internal variability of the Earth's climate system, which contains warm (El Niño)
80 and cold (La Niña) phases, describing anomalous warming and cooling of
81 surface waters, respectively, in the central-eastern tropical Pacific Ocean. It can
82 be responsible for global and regional oceanic and atmospheric pattern
83 anomalies, having significant impacts on wind, temperature and precipitation in
84 China (Zhou et al., 2007; Xu et al., 2007; Li et al., 2020; Zeng et al., 2021).
85 Recent studies have shown that the interannual variations in O₃ concentrations
86 over China are influenced by ENSO events (Jiang and Li, 2022; Li et al., 2022;
87 Yang et al., 2022b). Using the GEOS-Chem model, Yang et al. (2022b) showed
88 that summertime O₃ over southern China had a positive correlation with Niño
89 3.4 index. Near-surface O₃ concentrations increased by a maximum over 20%
90 in southern China during El Niño compared to La Niña, which was closely linked
91 to a weakened southerly over southern China that was conducive to the
92 accumulation of O₃. However, they also reported an unusual O₃ changes over
93 30°–40°N in China that could not be explained by the ENSO impact alone.

94 Quasi-Biennial Oscillation (QBO) is a major mode of variability of zonal
95 wind in the stratosphere characterized by alternating easterly and westerly, with
96 a period close to 28 months. The QBO is able to modulate large-scale vertical
97 and meridional circulations between tropics and subtropics (Punge et al., 2009),
98 which impacts the East Asian climate. For example, it is reported that southern
99 China sea summer monsoon is weakened during the QBO westerly phase due
100 to the associated anomalous Hadley-like circulation (Zheng et al., 2007). Kim
101 et al. (2020) revealed that there was stronger precipitation over East Asia with
102 a larger Madden-Julian oscillation (MJO) amplitude when the QBO is in easterly
103 phase, which is because MJO-induced vertical motion and moisture transport
104 is amplified by easterly QBO. Therefore, it is of interest to explore the influence
105 of QBO on interannual variations in summertime O₃ pollution over China and
106 their connections during anomalous modes of sea surface temperature (SST)
107 over the eastern tropical Pacific, as well as the mechanisms involved.

108 In the present study, the impact of QBO on O₃ variations in China is
109 examined based on GEOS-Chem simulations over 1981–2020, together with
110 satellite retrievals. The paper is organized as follows: In Section 2, we describe
111 the model, numerical experiments, the reanalysis datasets, the indices used in
112 this study and satellite retrieval data. The connection between QBO and
113 tropospheric O₃ in China and the possible mechanisms are explored in Section
114 3. Conclusions and discussion are given in Section 4.

115 **2. Methods**

116 **2.1. GEOS-Chem model simulations**

117 GEOS-Chem is a global three-dimensional chemical transport model with
118 comprehensive chemistry mechanisms of O₃-NO_x-hydrocarbon-aerosol
119 involved in the model (Zhai et al., 2021). In this study, we apply the GEOS-
120 Chem version 12.9.3 to simulate O₃ from 1981 to 2020. The horizontal grid of
121 the model is 2° × 2.5° (latitude × longitude) with 47 vertical levels above the
122 surface. Stratospheric O₃ is calculated based on the linearized chemistry
123 mechanism (McLinden et al., 2000). Meteorological fields driving the GEOS-
124 Chem simulations are from the Modern-Era Retrospective analysis for
125 Research and Applications version 2 (MERRA-2) (Gelaro et al., 2017),
126 produced by the NASA's Global Modeling and Assimilation Office.

127 Anthropogenic emissions in China are obtained from the Multi-resolution
128 Emission Inventory for China (MEIC) (Zheng et al., 2018). Anthropogenic
129 emissions outside China are adopted from the Community Emissions Data
130 System (CEDS) version 20210205 (Hoesly et al., 2018). Biogenic emissions
131 employ the Model of Emissions of Gases and Aerosols from Nature (MEGAN)
132 version 2.1 (Guenther et al., 2012). Global biomass burning emissions are from
133 the Global Fire Emissions Database version 4 (GFED4) (van der Werf et al.,
134 2017). Soil NO_x emissions are estimated in a soil parameterization scheme
135 (Hudman et al., 2012). Lightning-produced NO_x emissions are estimated in the
136 model based on Murray et al. (2012) and Ott et al. (2010).

137 The GEOS-Chem simulations are performed to assess the impact of QBO
138 on interannual variation of O₃ covering the period of 1981–2020, following a 6-
139 month model spin-up. In order to minimize the impact of interannual variations
140 in emissions on the modeled O₃ concentrations, the anthropogenic, biogenic
141 burning and natural emissions of O₃ precursors are all fixed at their 2017 levels
142 in the base simulation (BASE). The BASE simulation is analyzed to quantify the
143 impact of QBO on O₃, unless stated otherwise.

144 A sensitivity simulation (NO_CHN) is conducted with a different emission
145 configuration than BASE, aiming to investigate the impact of domestic
146 emissions in China on tropospheric O₃ during QBO events. Different from BASE,
147 anthropogenic emissions of NO_x, CO and VOCs in China are turned off in
148 NO_CHN. Considering that O₃ pollution is most critical during the boreal
149 summer in many regions of China, only summer months (June-July-August,
150 JJA) are examined in this study. Time-varying meteorological fields follow those
151 from MERRA-2 during all simulations.

152 Figure 1 compares the year-by-year changes in JJA O₃ concentrations in
153 observations and BASE simulation. GEOS-Chem can roughly capture the
154 interannual variation in surface O₃ concentrations in China during 2016–2020.
155 The spatial correlation coefficients between the observed and modeled year-
156 by-year changes in O₃ concentrations are about 0.5–0.6, except the 2018-to-
157 2019 changes in O₃, which could be attributed to the influence of the changes
158 in precursor emissions on the observed O₃ concentrations after Phase 2 of the
159 Chinese Clean Air Action Plan launched in 2018 (Li et al., 2020).

160 **2.2. QBO and Niño 3.4 indices**

161 The QBO phases are determined by the zonal average of 30 hPa zonal
162 wind over the equator (5°S–5°N) based on MERRA-2 reanalysis (Fig. 2a), with
163 the averages during JJA used in this study. Positive values denote westerly
164 QBO phase (QBOW), while negative values denote easterly QBO phase
165 (QBOE).

166 The Niño 3.4 index averaged over JJA is used to characterize the warm
167 and cold phases of SST anomaly over the eastern tropical Pacific in boreal
168 summer, which is estimated as the SST anomalies over the Niño 3.4 region
169 (5°S – 5°N , 170° – 120°W) (Fig. 2b). Positive (negative) Niño 3.4 index indicates
170 a warm (cold) phase when SST in eastern tropical Pacific is higher (lower) than
171 the climatological mean (1981–2020). The 40 years can be divided into the
172 warm and cold phases of the JJA SST anomalies over the eastern tropical
173 Pacific according to Niño 3.4 index.

174 QBO and Niño 3.4 indices calculated in this study using MERRA-2
175 reanalysis are highly correlated with those derived from NCEP/NCAR
176 reanalysis and HadISST1, with correlation coefficients of 0.97 and 0.98,
177 respectively. It suggests that the QBO and the eastern tropical Pacific SST
178 anomaly are well represented in the GEOS-Chem simulations, which is
179 important for appropriately quantifying impacts of QBO on the interannual
180 variations in O_3 variations over China.

181 **2.3 Satellite data**

182 The monthly mean tropospheric column O_3 (TCO) data from Ozone
183 Monitoring Instrument/Microwave Limb Sounder (OMI/MLS) on board the Aura
184 satellite since 2004 are used to verify the modeled impact of QBO on O_3
185 pollution in China. The grid resolution of OMI/MLS data is 1.25° longitude \times 1.0°
186 latitude, covering the measurement area between 60°S and 60°N . TCO is
187 calculated by subtracting MLS stratospheric column O_3 from OMI total column
188 O_3 (Ziemke et al., 2011). The tropopause height is calculated according to 2 K
189 km^{-1} lapse rate, which generally locates around 150 hPa in mid-latitudes (Jing
190 et al., 2006; Peiro et al., 2018). In this study, we used 150 hPa as an
191 approximation of the tropopause level for the calculation of TCO from the model
192 simulation, although it may lead to a small bias in the magnitude of TCO.

193 **3. Result**

194 **3.1. Impact of QBO on tropospheric O_3 in China**

195 To illustrate the effects of QBO on summertime near-surface O₃ over China,
196 the spatial distribution of the correlation coefficients between the JJA O₃
197 concentrations and concurrent QBO index is presented in Fig. 3a. It shows that
198 the correlation coefficients between QBO index and surface O₃ are insignificant
199 over most regions of China, except for part of Qinghai province, which means
200 that the single impact of QBO events cannot significantly affect O₃ pollution in
201 China. The lag-correlation analysis is also performed but shows even weaker
202 correlations. Previous studies have shown that the impact of QBO can be
203 compounded with ENSO (Sun et al., 2019; Xue et al., 2015). Motivated by these
204 studies, we further examine the relationships between QBO and summertime
205 O₃ in the warm/cold phases of SST anomalies of the eastern tropical Pacific.
206 Note that the correlation coefficient between QBO index and Niño 3.4 index is
207 only 0.09, indicating that there is no direct linear relationship between QBO and
208 ENSO, which has also been reported in previous studies (Christiansen et al.,
209 2016; Sun et al., 2019).

210 The influences of QBO on O₃ under different SST anomalies over the
211 eastern tropical Pacific are quite different (Fig. 3b and 3c). During years under
212 the warm SST phase, significant correlations between JJA near-surface O₃
213 concentrations and QBO index are located over the latitudinal band of 25°–
214 40°N in China. In central China (92.5°–112.5°E, 26°–38°N), the correlation
215 coefficient between the regionally averaged O₃ concentration and QBO index
216 under the warm phase is 0.53, which is much higher than 0.23 during the whole
217 40-year period. However, under the cold ENSO phase, there is no significant
218 correlation over China, with a regional correlation coefficient of –0.06. These
219 results suggest that QBO may have a remarkable effect on tropospheric O₃
220 over central China during the warm anomaly of the eastern tropical Pacific SST,
221 while it has little impact on O₃ in China during years with cold SST anomalies.
222 Once there is a coincidence of QBO and warm SST anomaly in eastern
223 tropical Pacific, the combined effects could worsen the O₃ pollution over China.

224 Therefore, the three strongest QBOW (1990, 1997 and 2019) and QBOE (1994,
225 2012 and 2018) years under the warm phase of SST anomaly during the past
226 four decades are chosen to further quantify the influence of QBO on O₃ pollution
227 in China.

228 Figure 4 presents JJA O₃ anomalies in the selected QBOW and QBOE
229 years relative to the climatological mean (1981–2020). Under the combined
230 influence of QBOW and warm SST anomaly, positive O₃ concentration
231 anomalies are observed over central and southern China. In contrast, the
232 surface O₃ concentration increases over southern China while it decreases in
233 central China during QBOE years. The increases in O₃ levels over southern
234 China under warm SST anomaly in both QBOW and QBOE years are due to
235 the positive correlation between Niño 3.4 index and tropospheric O₃
236 concentrations in southern China. Previous studies have reported that O₃
237 concentrations increased over southern China during El Niño years, which is
238 related to O₃ convergence due to weakened southerlies (Yang et al., 2022b; Li
239 et al., 2022). The different characteristics of O₃ changes in central China
240 highlight the role of QBO in affecting the distribution of O₃ over China under
241 warm SST anomalies of the eastern tropical Pacific.

242 Figure 5 presents the spatial distribution near the surface and pressure–
243 longitude cross-sections of absolute and percentage differences between
244 QBOW and QBOE in O₃ concentrations over China under the warm SST
245 anomaly. Compared with QBOE years, positive O₃ concentration anomalies are
246 located between 25°N and 40°N over China during QBOW, especially over
247 central China where the maximum anomaly exceeds 3 ppb (parts per billion)
248 (or 5% relative to the climatological average). The differences are higher than
249 the standard deviation during the analyzed period, suggesting that the
250 differences are significant. The simulated O₃ pollution enhancement is also
251 shown in the vertical distribution of the zonal mean (26°–38°N) composite
252 differences (Fig. 5b, d). For QBOW years, increased O₃ occurred in the whole

253 troposphere, with the maximum increase of 2–3 ppb (3–5%) between 850 hPa
254 and 500 hPa over central China, indicating a high probability of enhanced O₃
255 pollution during QBOW relative to QBOE. O₃ concentrations also increase in
256 the coastal area of eastern China, which is mainly due to the decreases in O₃
257 concentrations in the selected QBOE years relative to the climatological mean,
258 as the O₃ concentrations only slightly increase in the QBOW years. The
259 correlation between O₃ and QBO index over this region is not as strong as that
260 over central China, which indicates that the anomalous increase in O₃ over the
261 coastal area of eastern China may not be a typical feature of the QBO impact
262 and will not be discussed hereafter.

263 The modeled difference in summertime tropospheric O₃ between the
264 QBOW and QBOE years can also be observed from satellite (Fig. 6). The
265 OMI/MLS retrieved TCO are higher in QBOW than QBOE years between 25°–
266 35°N in China, which is in accordance with the model results. Averaged over
267 central China, the difference in TCO between the selected QBOW (2019) and
268 QBOE years (2012 and 2018) from satellite data is 2.8 DU, similar to the 2.5
269 DU from model simulation. Both model simulations and satellite retrievals
270 suggest that the QBO can significantly influence tropospheric O₃ in China. Also,
271 the simulated change in TCO is also higher than the standard deviation (1.4
272 DU). However, it is also noted that the spatial variation of the differences in TCO
273 varies between OMI/MLS and model simulation. It is partly because the
274 emissions were fixed at the 2017 levels during model simulations. This potential
275 biases in satellite retrievals also strongly contribute to the different spatial
276 pattern (Schoeberl et al., 2007; Liu et al., 2010; Ziemke et al., 2006, 2014).

277 **3.2. Mechanism of the QBO impacts on O₃ in China**

278 Composite differences of relevant meteorological variables between the
279 selected QBOW and QBOE years are shown in Fig. 7 to illustrate mechanisms
280 of the QBO impacts on O₃ in China. During QBOW years under warm SST
281 anomaly, the decrease in cloud fraction (Fig. 7g) allows more solar radiation to

282 reach the surface (Fig. 7h) and the RH also decreases over central China (Fig.
283 7e), relative to QBOE years. These changes in meteorological parameters tend
284 to increase the photochemical production of O₃. However, the air temperature
285 significantly decreases in the lower (Fig. 7i) and mid-troposphere (Fig. 7f) in
286 QBOW years compared to QBOE years, which suppresses the O₃ production.
287 Integrated process rate analysis has been widely conducted to assess the
288 contribution of individual chemical or physical processes to the production and
289 distribution of O₃ pollution per unit time in the study domain (Lou et al., 2015;
290 Qu et al., 2021; Zhu et al., 2021). The combined effect of the changes in these
291 meteorological parameters leads to a reduction in net O₃ chemical production
292 by about 1% over central China in QBOW compared to QBOE years based on
293 an integrated process rate analysis (Table S1). Therefore, the chemical
294 production change is not the major process causing the O₃ pollution
295 deterioration during QBOW years under the warm SST anomaly.

296 Compared with QBOE years, anomalous northwesterly winds at 850 hPa
297 occurred over central China during the QBOW years, located at the east edge
298 of an anomalous high over western China (Fig. 7a). Under the influence of this
299 anomalous high, the anomalous downdraft throughout the troposphere over
300 central China (Fig. 7c) can reduce the vertical transport of O₃ to the upper
301 troposphere, which leads to an O₃ accumulation in the lower and mid-
302 troposphere. In addition, the increase in planetary boundary layer (PBL) height
303 (Fig. 7d) favors the vertical mixing of air within the PBL and the O₃-enriched air
304 above the PBL (Ma et al., 2021).

305 Considering the effect of winds on O₃ transport, the horizontal JJA O₃ mass
306 fluxes from the surface to 850 hPa and the vertical mass flux at 850 hPa over
307 central China are calculated and summarized in Table 1. Due to an anomalous
308 northwesterly, the outflow transport of O₃ from the north boundary of central
309 China is reduced by 1.11 Tg during QBOW years relative to QBOE years.
310 However, through the east boundary of central China, an inflow transport of O₃

311 is reduced by 1.35 Tg, which overwhelms the gain from the reduced northward
312 transport. The O₃ flux changes through the west and south boundaries are
313 relatively small and almost offset each other. The overall changes in the
314 horizontal transport result in a decrease in O₃ mass by 0.29 Tg from surface to
315 850 hPa in QBOW relative to QBOE years, suggesting that the horizontal
316 advection change is also not the primary process causing the enhanced O₃
317 pollution.

318 The anomalous downdraft over central China weakens the upward mixing
319 of high lower-tropospheric O₃ concentrations and causes an anomalous
320 downward transport of O₃ by 0.59 Tg at 850 hPa, contributing to the increase
321 in surface O₃ concentrations. Therefore, the impact of the QBO under warm
322 SST anomaly on the distribution of tropospheric O₃ over central China is mainly
323 via changes in the vertical motion.

324 **3.3. Role of China domestic anthropogenic emission**

325 Comparison of the O₃ anomaly in BASE and NO_CHN identifies the impact
326 of China domestic emissions on O₃ concentrations. When domestic
327 anthropogenic emissions of O₃ precursors are turned off, JJA mean near-
328 surface O₃ concentrations largely increase across China, especially between
329 30°–40°N, with maximum increases exceeding 5 ppb during QBOW compared
330 to QBOE years (Fig. 8a). Averaged over central China, the anomalous increase
331 in near-surface O₃ concentration is 3.0 ppb in NO_CHN, even higher than that
332 (1.7 ppb) in BASE simulation. It results from that the reduction in the net export
333 of horizontal mass flux of O₃ due to the removal of domestic emissions (Table
334 S2) leads to a more significant increase in O₃ over central China in the
335 NO_CHN experiment.

336 Figure 8b shows the simulated vertical distribution of O₃ concentration
337 difference between the selected QBOW and QBOE years from the NO_CHN
338 experiment. The positive O₃ anomaly in the troposphere is similar to that from
339 the BASE experiment, but the increases are mainly between 95°E and 115°E

340 from the surface to 500 hPa over central China. These results suggest that the
341 vertical transport process dominates the increase in summertime tropospheric
342 O₃ concentrations over central China during QBOW under warm SST anomaly
343 of the eastern tropical Pacific. In the NO_CHN experiment, the reduction in the
344 O₃ horizontal export results in a more significant increase of O₃ concentration
345 during QBOW compared to QBOE years.

346 **4. Conclusion and discussion**

347 Based on GEOS-Chem model simulations over 1981–2020, we
348 investigate the impacts of different QBO events on the surface and tropospheric
349 O₃ over China. Although only weak correlations are found between JJA mean
350 near-surface O₃ concentrations and QBO index over China, their positive
351 correlation is significant in years with warm SST anomalies over the eastern
352 tropical Pacific. Averaged over central China (92.5°–112.5°E, 26°–38°N), the
353 correlation coefficient between the regional near-surface O₃ concentration and
354 QBO index during the warm ENSO phase is 0.53. It suggests that the co-
355 occurrence of the westerly phase of QBO and warm SST anomalies over the
356 eastern tropical Pacific would exacerbate summertime O₃ pollution in China.
357 Compared with QBOE years, near-surface O₃ concentrations increase by up to
358 3 ppb (5% relative to the average) across China during QBOW, especially over
359 central China, and the increase in O₃ extends from the surface to the upper
360 troposphere, especially between 850 hPa and 500 hPa.

361 A combined effect of changes in meteorological conditions (i.e., less cloud,
362 higher RH, and lower temperature) leads to a slightly lower net O₃ chemical
363 production rate in QBOW years than in QBOE years. Central China is
364 influenced by anomalous northwesterlies during QBOW, which weakens O₃
365 import from the east boundary and the export from north boundary of central
366 China, leading to a net O₃ export of 0.29 Tg during QBOW, compared to QBOE
367 years, from surface to 850 hPa. However, change in the vertical transport is the
368 main process causing O₃ concentration increases in QBOW years. An

369 anomalous downdraft leads to the O₃ mass increase of 0.59 Tg below 850 hPa
370 by suppressing vertical mixing and promoting O₃ accumulation in the lower
371 troposphere. The sensitivity experiment with China domestic anthropogenic
372 emissions of O₃ precursors turned off shows a greater increase of O₃ (3.0 ppb)
373 than that in the default simulation (1.7 ppb). It indicates that the O₃ increase
374 over central China during QBOW years under the warm SST anomaly is mainly
375 due to the anomalous vertical transport, while a decrease in local chemical
376 production partly offsets the O₃ increases in central China. Moreover, the
377 positive anomaly of TCO based on GEOS-Chem model simulation is consistent
378 with the satellite retrieval from the OMI/MLS.

379 This study explores the effect of QBO on tropospheric O₃ over China and
380 the underlying mechanisms during the warm SST anomalies of the eastern
381 tropical Pacific, which can improve the understanding of causes of O₃ pollution
382 over China. For climatological average, prevailing easterly winds at 30 hPa
383 dominate the equator, accompanied by the upward motion over central China
384 within the troposphere. During QBOW years, the prevailing winds reverse to
385 westerlies, which may induce the anomalous downward motion over central
386 China. However, the dynamical mechanism of how the stratospheric QBO
387 drives changes in the vertical motion and circulation patterns in China along
388 with the SST anomaly over the eastern tropical Pacific is out of the scope of
389 this study and merits further investigation. Nevertheless, the QBO index is
390 positively correlated with the vertical velocity throughout the troposphere over
391 China, especially between 100°E and 110°E (Fig. 9), where the lower
392 tropospheric O₃ increases the most in the NO_CHN experiment during QBOW
393 years under the warm SST anomaly. These positive correlations demonstrate
394 that the weakened (strengthened) upward motion increase (decrease)
395 tropospheric O₃ concentrations during QBOW (QBOE) years, confirming that
396 changes in the vertical transport driven by QBO events play an important role
397 in modulating summertime O₃ pollution over China. The phenomenon of

398 changes in tropospheric O₃ between different QBO phases is also verified by
399 satellite retrievals. Compared with cold conditions, stratospheric QBO forcing is
400 strengthened due to the increase of tropospheric temperature and changes of
401 analytical and parametric waves under warm SST anomalies of the eastern
402 tropical Pacific, which causes a faster downward propagation in QBO (Taguchi,
403 2010; Schirber et al., 2015; Geller et al., 2016; Zheng et al., 2007). This may
404 explain why the correlation coefficient between the O₃ and QBO indices is
405 insignificant, but shows a significant correlation during warm SST anomalies of
406 the eastern tropical Pacific. The mechanisms deserve further investigation in
407 future studies. Also, it is assumed that the vertical motion over China is
408 influenced by anomaly of Walker circulation caused by the QBO (Huangfu et
409 al., 2021). Although the physical mechanism remains elusive, we believe that
410 our findings would be useful for future air pollution prediction and control.
411

412 **Author contributions.** YY designed the research; ML performed simulations
413 and analyzed the data. All authors including HW, LH, PW, and HL discussed
414 the results and wrote the paper.

415

416 **Code and data availability.** The GEOS-Chem model is available at
417 <https://zenodo.org/record/3974569#.YTD81NMzagR> (last access: 1 July 2022).
418 MERRA-2 reanalysis data can be downloaded at
419 <https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/> (last access: 1 July 2022).
420 The monthly mean tropospheric O₃ data from OMI/MLS is downloaded from
421 https://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html (last
422 access: 1 July 2022). Our model results are available at
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438

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440 **References**

441

442 Camalier, L., Cox, W. M., and Dolwick, P.: The effects of meteorology on
443 ozone in urban areas and their use in assessing ozone trends, *Atmos.*
444 *Environ.*, 41, 7127–7137, <https://doi.org/10.1016/j.atmosenv.2007.04.061>,
445 2007.

446

447 Christiansen, B., Yang, S., and Madsen, M. S.: Do strong warm ENSO events
448 control the phase of the stratospheric QBO?, *Geophys. Res. Lett.*, 43, 10,489-
449 10,495, <https://doi.org/10.1002/2016gl070751>, 2016.

450

451 Feng, Y., Ning, M., Lei, Y., Sun, Y., Liu, W., and Wang, J.: Defending blue sky
452 in China: Effectiveness of the “Air Pollution Prevention and Control Action
453 Plan” on air quality improvements from 2013 to 2017., *J. Environ. Manage.*,
454 252, 109603–109603, <https://doi.org/10.1016/j.jenvman.2019.109603>, 2019.

455

456 Gao, J., Yang, Y., Wang, H., Wang, P., Li, H., Li, M., Ren, L., Yue, X., and
457 Liao, H.: Fast climate responses to emission reductions in aerosol and ozone
458 precursors in China during 2013–2017, *Atmos. Chem. Phys.*, 22, 7131–7142,
459 <https://doi.org/10.5194/acp-22-7131-2022>, 2022.

460

461 Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L.,
462 Randles, C. A., Darmenov, A., Bosilovich, M. G., Reichle, R., Wargan, K., Coy,
463 L., Cullather, R., Draper, C., Akella, S., Buchard, V., Conaty, A., da Silva, A.
464 M., Gu, W., Kim, G.-K., Koster, R., Lucchesi, R., Merkova, D., Nielsen, J. E.,
465 Partyka, G., Pawson, S., Putman, W., Rienecker, M., Schubert, S. D.,
466 Sienkiewicz, M., and Zhao, B.: The Modern-Era Retrospective Analysis for
467 Research and Applications, Version 2 (MERRA-2), *J. Clim.*, 30, 5419–5454,
468 <https://doi.org/10.1175/JCLI-D-16-0758.1>, 2017.

469

470 Gong, C. and Liao, H.: A typical weather pattern for ozone pollution events in
471 North China, *Atmos. Chem. Phys.*, 19, 13725–13740,
472 <https://doi.org/10.5194/acp-19-13725-2019>, 2019.

473

474 Geller, M. A., Zhou, T., and Yuan, W.: The QBO, gravity waves forced by
475 tropical convection, and ENSO, *J. Geophys. Res. Atmos.*, 121, 8886–8895,
476 <https://doi.org/10.1002/2015JD024125>, 2016.

477

478 Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T.,
479 Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and
480 Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated
481 framework for modeling biogenic emissions, *Geosci. Model Dev.*, 5, 1471–
482 1492, <https://doi.org/10.5194/gmd-5-1471-2012>, 2012.

483

484 Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G.,
485 Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C.,
486 Dawidowski, L., Kholod, N., Kurokawa, J., Li, M., Liu, L., Lu, Z., Moura, M. C.
487 P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic
488 emissions of reactive gases and aerosols from the Community Emissions
489 Data System (CEDS), *Geosci. Model Dev.*, 11, 369–408,
490 <https://doi.org/10.5194/gmd-11-369-2018>, 2018.

491
492 Huangfu, J., Tang, Y., Ma, T., Chen, W., and Wang, L.: Influence of the QBO
493 on tropical convection and its impact on tropical cyclone activity over the
494 western North Pacific, *Clim. Dyn.*, 57, 657–669,
495 <https://doi.org/10.1007/s00382-021-05731-x>, 2021.

496
497 Hudman, R. C., Moore, N. E., Mebust, A. K., Martin, R. V., Russell, A. R.,
498 Valin, L. C., and Cohen, R. C.: Steps towards a mechanistic model of global
499 soil nitric oxide emissions: implementation and space based-constraints,
500 *Atmos. Chem. Phys.*, 12, 7779–7795, [https://doi.org/10.5194/acp-12-7779-](https://doi.org/10.5194/acp-12-7779-2012)
501 2012, 2012.

502
503 Jiang, Z. and Li, J.: Impact of eastern and central Pacific El Niño on lower
504 tropospheric ozone in China, *Atmos. Chem. Phys.*, 22, 7273–7285,
505 <https://doi.org/10.5194/acp-22-7273-2022>, 2022.

506
507 Jing, P., Cunnold, D., Choi, Y., and Wang, Y.: Summertime tropospheric ozone
508 columns from Aura OMI/MLS measurements versus regional model results
509 over the United States, *Geophys. Res. Lett.*, 33, L17817,
510 <https://doi.org/10.1029/2006GL026473>, 2006

511
512 Kim, H., Son, S.W., and Yoo, C.: QBO modulation of the MJO-related
513 precipitation in East Asia, *J. Geophys. Res.*, 125, e2019JD031929,
514 <https://doi.org/10.1029/2019JD031929>, 2020.

515
516 Li, H., Fan, K., He, S., Liu, Y., Yuan, X., and Wang, H.: Intensified Impacts of
517 Central Pacific ENSO on the Reversal of December and January Surface Air
518 Temperature Anomaly over China since 1997, *J. Clim.*, 34, 1601–1618,
519 <https://doi.org/10.1175/jcli-d-20-0048.1>, 2020.

520
521 Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q., and Bates, K.:
522 Anthropogenic drivers of 2013–2017 trends in summer surface ozone in
523 China, *P. Natl. Acad. Sci. USA*, 116, 422–427,
524 <https://doi.org/10.1073/pnas.1812168116>, 2019.

525
526 Li, K., Jacob, D. J., Shen, L., Lu, X., De Smedt, I., and Liao, H.: Increases in
527 surface ozone pollution in China from 2013 to 2019: anthropogenic and

528 meteorological influences, *Atmos. Chem. Phys.*, 20, 11423–11433,
529 <https://doi.org/10.5194/acp-20-11423-2020>, 2020.

530

531 Li, M., Yang, Y., Wang, P., Ji, D., and Liao, H.: Impacts of strong El Niño on
532 summertime near-surface ozone over China, *Atmos. Ocean. Sci. Lett.*,
533 100193, <https://doi.org/10.1016/j.aosl.2022.100193>, 2022.

534

535 Liu, X., Bhartia, P. K., Chance, K., Spurr, R. J. D., and Kurosu, T. P.: Ozone
536 profile retrievals from the Ozone Monitoring Instrument, *Atmos. Chem. Phys.*,
537 10, 2521–2537, <https://doi.org/10.5194/acp-10-2521-2010>, 2010.

538

539 Lou, S., Liao, H., Yang, Y., and Mu, Q.: Simulation of the interannual
540 variations of tropospheric ozone over China: Roles of variations in
541 meteorological parameters and anthropogenic emissions, *Atmos. Environ.*,
542 122, 839–851, <https://doi.org/10.1016/j.atmosenv.2015.08.081>, 2015.

543

544 Ma, X., Huang, J., Zhao, T., Liu, C., Zhao, K., Xing, J., and Xiao, W.: Rapid
545 increase in summer surface ozone over the North China Plain during 2013–
546 2019: a side effect of particulate matter reduction control?, *Atmos. Chem.*
547 *Phys.*, 21, 1–16, <https://doi.org/10.5194/acp-21-1-2021>, 2021.

548

549 McLinden, C. A., Olsen, S. C., Hannegan, B., Wild, O., Prather, M. J., and
550 Sundet, J.: Stratospheric ozone in 3-D models: A simple chemistry and the
551 cross-tropopause flux, *J. Geophys. Res.*, 105, 14653–14665,
552 <https://doi.org/10.1029/2000JD900124>, 2000.

553

554 Murray, L. T., Jacob, D. J., Logan, J. A., Hudman, R. C., and Koshak, W. J.:
555 Optimized regional and interannual variability of lightning in a global chemical
556 transport model constrained by LIS/OTD satellite data, *J. Geophys. Res.*, 117,
557 <https://doi.org/10.1029/2012JD017934>, 2012.

558

559 Nuvolone, D., Petri, D., and Voller, F.: The effects of ozone on human health,
560 *Environ. Sci. Pollut. R.*, 25, 8074–8088, <https://doi.org/10.1007/s11356-017-9239-3>, 2018.

561

562

563 Ott, L. E., Pickering, K. E., Stenchikov, G. L., Allen, D. J., DeCaria, A. J.,
564 Ridley, B., Lin, R.-F., Lang, S., and Tao, W. K.: Production of lightning NO_x
565 and its vertical distribution calculated from three-dimensional cloud-scale
566 chemical transport model simulations, *J. Geophys. Res.*, 115, D04301,
567 <https://doi.org/10.1029/2009JD011880>, 2010.

568

569 Peiro, H., Emili, E., Cariolle, D., Barret, B., and Le Flochmoën, E.: Multi-year
570 assimilation of IASI and MLS ozone retrievals: variability of tropospheric

571 ozone over the tropics in response to ENSO, *Atmos. Chem. Phys.*, 18, 6939–
572 6958, <https://doi.org/10.5194/acp-18-6939-2018>, 2018.

573

574 Porter, W. C. and Heald, C. L.: The mechanisms and meteorological drivers of
575 the summertime ozone–temperature relationship, *Atmos. Chem. Phys.*, 19,
576 13367–13381, <https://doi.org/10.5194/acp-19-13367-2019>, 2019.

577

578 Punge, H. J., Konopka, P., Giorgetta, M. A., and Müller, R.: Effects of the
579 quasi-biennial oscillation on low-latitude transport in the stratosphere derived
580 from trajectory calculations, *J. Geophys. Res.*, 114, D03102,
581 <https://doi.org/10.1029/2008JD010518>, 2009.

582

583 Qu, K., Wang, X., Yan, Y., Shen, J., Xiao, T., Dong, H., Zeng, L., and Zhang,
584 Y.: A comparative study to reveal the influence of typhoons on the transport,
585 production and accumulation of O₃ in the Pearl River Delta, China, *Atmos.*
586 *Chem. Phys.*, 21, 11593–11612, <https://doi.org/10.5194/acp-21-11593-2021>,
587 2021.

588

589 Schirber, S.: Influence of ENSO on the QBO: Results from an ensemble of
590 idealized simulations, *J. Geophys. Res. Atmos.*, 120, 1109–1122,
591 <https://doi.org/10.1002/2014JD022460>, 2015.

592

593 Schoeberl, M. R., Ziemke, J. R., Bojkov, B., Livesey, N., Duncan, B., Strahan,
594 S., Froidevaux, L., Kulawik, S., Bhartia, P. K., Chandra, S., Levelt, P. F., Witte,
595 J. C., Thompson, A. M., Cuevas, E., Redondas, A., Tarasick, D. W., Davies, J.,
596 Bodeker, G., Hansen, G., Johnson, B. J., Oltmans, S. J., Vömel, H., Allaart,
597 M., Kelder, H., Newchurch, M., Godin-Beekmann, S., Ancellet, G., Claude, H.,
598 Andersen, S. B., Kyrö, E., Parrondos, M., Yela, M., Zabolocki, G., Moore, D.,
599 Dier, H., von der Gathen, P., Viatte, P., Stübi, R., Calpini, B., Skrivankova, P.,
600 Dorokhov, V., de Backer, H., Schmidlin, F. J., Coetzee, G., Fujiwara, M.,
601 Thouret, V., Posny, F., Morris, G., Merrill, J., Leong, C. P., Koenig-Langlo, G.,
602 and Joseph, E.: A trajectory-based estimate of the tropospheric ozone column
603 using the residual method, *J. Geophys. Res.*, 112, D24S49,
604 <https://doi.org/10.1029/2007JD008773>, 2007.

605

606 Sun, L., Wang, H., and Liu, F.: Combined effect of the QBO and ENSO on the
607 MJO, *Atmos. Ocean. Sci. Lett.*, 12, 170–176,
608 <https://doi.org/10.1080/16742834.2019.1588064>, 2019.

609

610 Taguchi, M.: Observed connection of the stratospheric quasi-biennial
611 oscillation with El Niño–Southern Oscillation in radiosonde data, *J. Geophys.*
612 *Res.*, 115, <https://doi.org/10.1029/2010jd014325>, 2010.

613

614 van der Werf, G. R., Randerson, J. T., Giglio, L., van Leeuwen, T. T., Chen, Y.,
615 Rogers, B. M., Mu, M., van Marle, M. J. E., Morton, D. C., Collatz, G. J.,
616 Yokelson, R. J., and Kasibhatla, P. S.: Global fire emissions estimates during
617 1997–2016, *Earth Syst. Sci. Data*, 9, 697–720, [https://doi.org/10.5194/essd-9-](https://doi.org/10.5194/essd-9-697-2017)
618 697-2017, 2017.

619

620 Verstraeten, W. W., Neu, J. L., Williams, J. E., Bowman, K. W., Worden, J. R.,
621 and Boersma, K. F.: Rapid increases in tropospheric ozone production and ex
622 port from China, *Nat. Geosci.*, 8, 690–695, <https://doi.org/10.1038/ngeo2493>,
623 2015.

624

625 Wang, P., Yang, Y., Li, H., Chen, L., Dang, R., Xue, D., Li, B., Tang, J., Leung,
626 L. R., and Liao, H.: North China Plain as a hot spot of ozone pollution
627 exacerbated by extreme high temperatures, *Atmos. Chem. Phys.*, 22, 4705–
628 4719, <https://doi.org/10.5194/acp-22-4705-2022>, 2022.

629

630 Wang, T., Xue, L., Brimblecombe, P., Lam, Y. F., Li, L., and Zhang, L.: Ozone
631 pollution in China: A review of concentrations, meteorological influences,
632 chemical precursors, and effects, *Sci. Total Environ.*, 575, 1582–1596,
633 <https://doi.org/10.1016/j.scitotenv.2016.10.081>, 2017.

634

635 Wang, X., Manning, W. J., Feng, Z., and Zhu, Y.-G.: Ground-level ozone in
636 China: Distribution and effects on crop yields, *Environ. Pollut.*, 147, 394–400,
637 <https://doi.org/10.1016/j.envpol.2006.05.006>, 2007.

638

639 Wespes, C., Hurtmans, D., Clerbaux, C., and Coheur, P.-F.: O₃ variability in
640 the troposphere as observed by IASI over 2008–2016: Contribution of
641 atmospheric chemistry and dynamics, *J. Geophys. Res. Atmos.*, 122, 2429–
642 2451, <https://doi.org/10.1002/2016jd025875>, 2017.

643

644 Xu, Z. X., Li, J. Y., Takeuchi, K., and Ishidaira, H.: Long-term trend of
645 precipitation in China and its association with the El Niño–southern oscillation,
646 *Hydrol. Process.*, 21, 61–71, <https://doi.org/10.1002/hyp.6180>, 2007.

647

648 Xue, X., Chen, W., Chen, S., and Zhou, D.: Modulation of the connection
649 between boreal winter ENSO and the South Asian high in the following
650 summer by the stratospheric quasi-biennial oscillation, *J. Geophys. Res.*
651 *Atmos.*, 120, 7393–7411, <https://doi.org/10.1002/2015JD023260>, 2015.

652

653 Yang, Y., Ren, L., Wu, M., Wang, H., Song, F., Leung, L. R., Hao, X., Li, J.,
654 Chen, L., Li, H., Zeng, L., Zhou, Y., Wang, P., Liao, H., Wang, J., and Zhou,
655 Z.-Q.: Abrupt emissions reductions during COVID-19 contributed to record
656 summer rainfall in China, *Nat. Commun.*, 13, 959,
657 <https://doi.org/10.1038/s41467-022-28537-9>, 2022a.

658

659 Yang, Y., Li, M., Wang, H., Li, H., Wang, P., Li, K., Gao, M., and Liao, H.:
660 ENSO modulation of summertime tropospheric ozone over China, *Environ.*
661 *Res. Lett.*, 17, 034020, <https://doi.org/10.1088/1748-9326/ac54cd>, 2022b.

662

663 Yang, Y., Liao, H., and Li, J.: Impacts of the East Asian summer monsoon on
664 interannual variations of summertime surface-layer ozone concentrations over
665 China, *Atmos. Chem. Phys.*, 14, 6867–6879, [https://doi.org/10.5194/acp-14-](https://doi.org/10.5194/acp-14-6867-2014)
666 6867-2014, 2014.

667

668 Yin, P., Chen, R., Wang, L., Meng, X., Liu, C., Niu, Y., Lin, Z., Liu, Y., Liu, J.,
669 Qi, J., You, J., Zhou, M., and Kan, H.: Ambient Ozone Pollution and Daily
670 Mortality: A Nationwide Study in 272 Chinese Cities, *Environ. Health*
671 *Perspect.*, 125, 117006, <https://doi.org/10.1289/EHP1849>, 2017.

672

673 Zeng, G., Morgenstern, O., Braesicke, P., and Pyle, J. A.: Impact of
674 stratospheric ozone recovery on tropospheric ozone and its budget, *Geophys.*
675 *Res. Lett.*, 37, <https://doi.org/10.1029/2010gl042812>, 2010.

676

677 Zeng, G., Morgenstern, O., Shiona, H., Thomas, A. J., Querel, R., and Nichol,
678 S. E.: Attribution of recent ozone changes in the Southern Hemisphere mid-
679 latitudes using statistical analysis and chemistry–climate model simulations,
680 *Atmos. Chem. Phys.*, 17, 10495–10513, [https://doi.org/10.5194/acp-17-](https://doi.org/10.5194/acp-17-10495-2017)
681 10495-2017, 2017.

682

683 Zeng, L., Yang, Y., Wang, H., Wang, J., Li, J., Ren, L., Li, H., Zhou, Y., Wang,
684 P., and Liao, H.: Intensified modulation of winter aerosol pollution in China by
685 El Niño with short duration, *Atmos. Chem. Phys.*, 21, 10745–10761,
686 <https://doi.org/10.5194/acp-21-10745-2021>, 2021.

687

688 Zhai, S., Jacob, D. J., Brewer, J. F., Li, K., Moch, J. M., Kim, J., Lee, S., Lim,
689 H., Lee, H. C., Kuk, S. K., Park, R. J., Jeong, J. I., Wang, X., Liu, P., Luo, G.,
690 Yu, F., Meng, J., Martin, R. V., Travis, K. R., Hair, J. W., Anderson, B. E.,
691 Dibb, J. E., Jimenez, J. L., Campuzano-Jost, P., Nault, B. A., Woo, J.-H., Kim,
692 Y., Zhang, Q., and Liao, H.: Relating geostationary satellite measurements of
693 aerosol optical depth (AOD) over East Asia to fine particulate matter (PM
694 2.5): insights from the KORUS-AQ aircraft campaign and GEOS-Chem model
695 simulations, *Atmos. Chem. Phys.*, 21, 16775–16791,
696 <https://doi.org/10.5194/acp-21-16775-2021>, 2021.

697

698 Zhao, H., Zheng, Y., Zhang, Y., Li, T., and Li, T.: Evaluating the effects of
699 surface O₃ on three main food crops across China during 2015-2018.,
700 *Environ. Pollut.*, 258, 113794–113794,
701 <https://doi.org/10.1016/j.envpol.2019.113794>, 2020.

702
703 Zhao, Z. and Wang, Y.: Influence of the West Pacific subtropical high on
704 surface ozone daily variability in summertime over eastern China, *Atmos.*
705 *Environ.*, 170, 197–204, <https://doi.org/10.1016/j.atmosenv.2017.09.024>,
706 2017.

707
708 Zheng, B., Gu, D., Lin, A., and Li, C.: Dynamical mechanism of the
709 stratospheric quasi-biennial oscillation impact on the South China Sea
710 Summer Monsoon, *Sci. China Earth Sci.*, 50, 1424–1432,
711 <https://doi.org/10.1007/s11430-007-0075-z>, 2007.

712
713 Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L.,
714 Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends
715 in China's anthropogenic emissions since 2010 as the consequence of clean
716 air actions, *Atmos. Chem. Phys.*, 18, 14095–14111,
717 <https://doi.org/10.5194/acp-18-14095-2018>, 2018.

718
719 Zhou, W. and Chan, J. C.: ENSO and the South China Sea summer monsoon
720 onset, *Int. J. Climatol.*, 27, 157–167, <https://doi.org/10.1002/joc.1380>, 2007.

721
722 Zhu, J., Chen, L., Liao, H., Yang, H., Yang, Y., and Yue, X.: Enhanced PM_{2.5}
723 Decreases and O₃ Increases in China During COVID-19 Lockdown by
724 Aerosol-Radiation Feedback, *Geophys. Res. Lett.*, 48, e2020GL090260,
725 <https://doi.org/10.1029/2020gl090260>, 2021.

726
727 Ziemke, J. R., Chandra, S., Duncan, B. N., Froidevaux, L., Bhartia, P. K.,
728 Levelt, P. F., and Waters, J. W.: Tropospheric ozone determined from Aura
729 OMI and MLS: Evaluation of measurements and comparison with the Global
730 Modeling Initiative's Chemical Transport Model, *J. Geophys. Res.*, 111,
731 D19303, <https://doi.org/10.1029/2006JD007089>, 2006.

732
733 Ziemke, J. R., Chandra, S., Labow, G. J., Bhartia, P. K., Froidevaux, L., and
734 Witte, J. C.: A global climatology of tropospheric and stratospheric ozone
735 derived from Aura OMI and MLS measurements, *Atmos. Chem. Phys.*, 11,
736 9237–9251, <https://doi.org/10.5194/acp-11-9237-2011>, 2011.

737
738 Ziemke, J. R., Olsen, M. A., Witte, J. C., Douglass, A. R., Strahan, S. E.,
739 Wargan, K., Liu, X., Schoeberl, M. R., Yang, K., Kaplan, T. B., Pawson, S.,
740 Duncan, B. N., Newman, P. A., Bhartia, P. K., and Heney, M. K.: Assessment
741 and applications of NASA ozone data products derived from Aura OMI/MLS
742 satellite measurements in context of the GMI chemical transport model, *J.*
743 *Geophys. Res. Atmos.*, 119, 5671–5699,
744 <https://doi.org/10.1002/2013JD020914>, 2014.

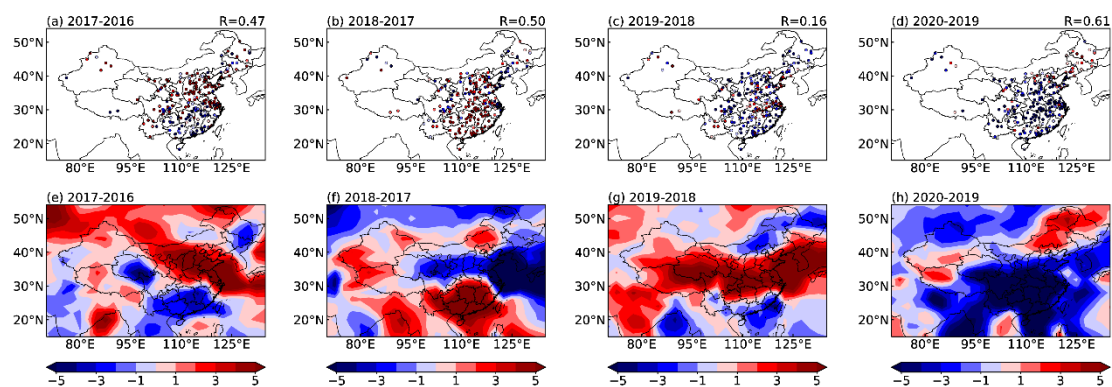
745 **Table 1.** The horizontal mass flux (Tg) of JJA O₃ from the surface to 850 hPa
 746 and the vertical mass flux (Tg) at 850 hPa over central China (92.5°–112.5°E,
 747 26°–38°N). The values are averaged over the selected three QBOW years
 748 (1990, 1997 and 2019) and QBOE (1994, 2012 and 2018) years and their
 749 differences (QBOW-QBOE). Positive values indicate incoming fluxes and
 750 negative values indicate outgoing fluxes.

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	QBOW	QBOE	Difference
Horizontal mass flux			
East	1.46	2.81	-1.35
West	0.92	0.74	0.18
North	-0.06	-1.17	1.11
South	3.60	3.83	-0.23
Vertical mass flux			
Top	-5.68	-6.27	0.59

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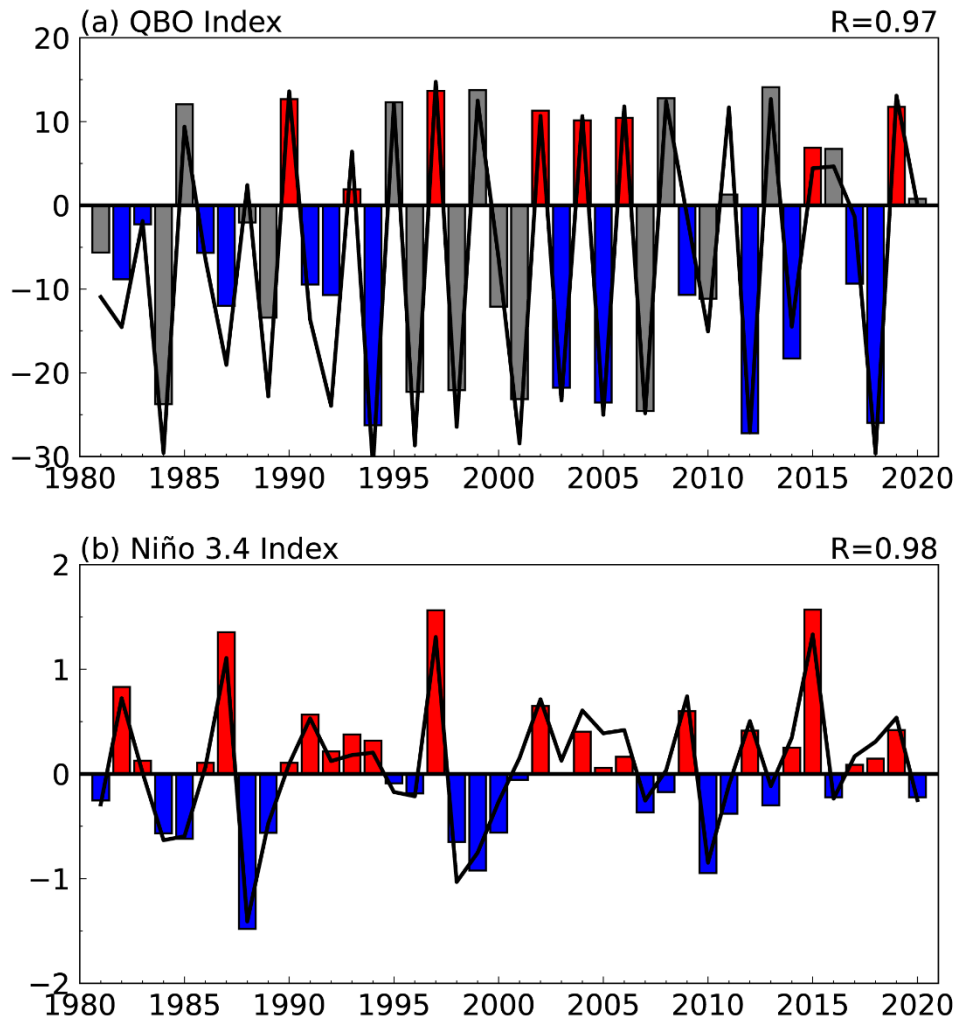


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755 **Figure 1.** Spatial distributions of year-by-year changes in the (a-d) observed
 756 and (e-h) modeled JJA O₃ concentrations (ppbv) during 2016–2020. The O₃
 757 observations are obtained from the China National Environmental Monitoring
 758 Centre (CNEMC). Spatial correlation coefficients between simulations and
 759 observations are shown at the top right corner of panel a-d.

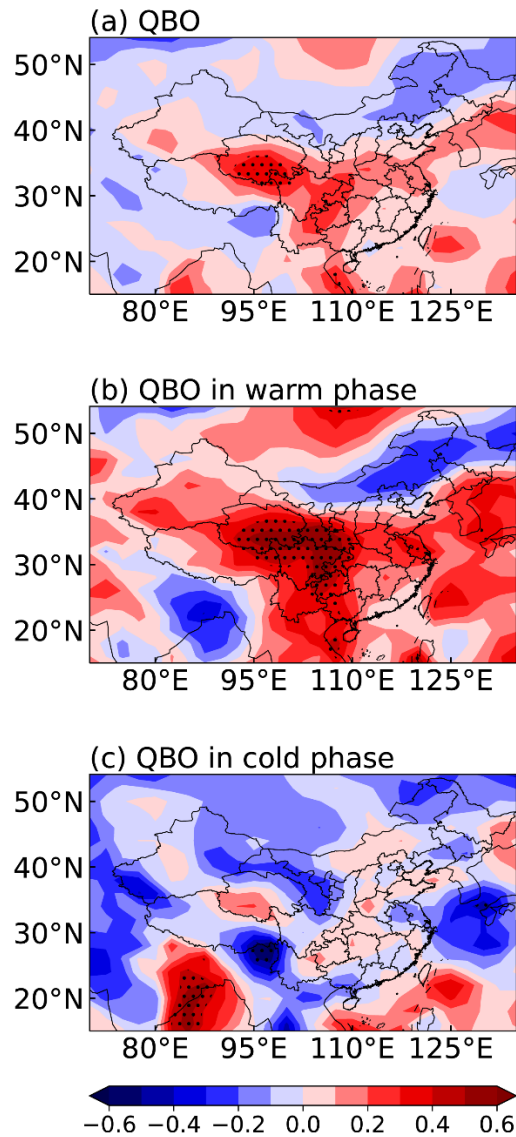
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Figure 2. Time series of the JJA mean (a) QBO index (m s^{-1}) and (b) Niño 3.4 index ($^{\circ}\text{C}$) over 1981–2020. Warm phase of SST anomalies over the eastern tropical Pacific includes 22 years (1982, 1983, 1986, 1987, 1990, 1991, 1992, 1993, 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.



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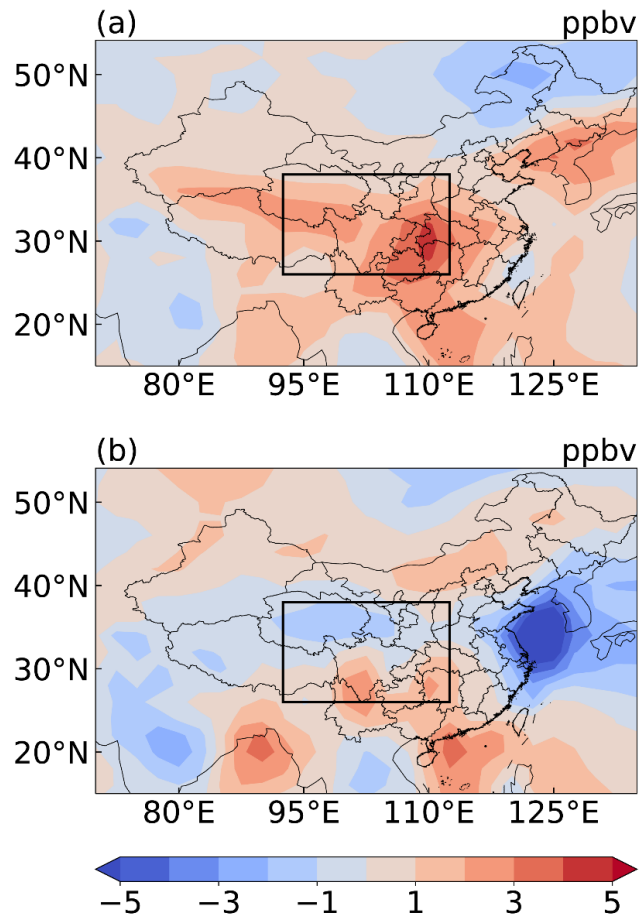
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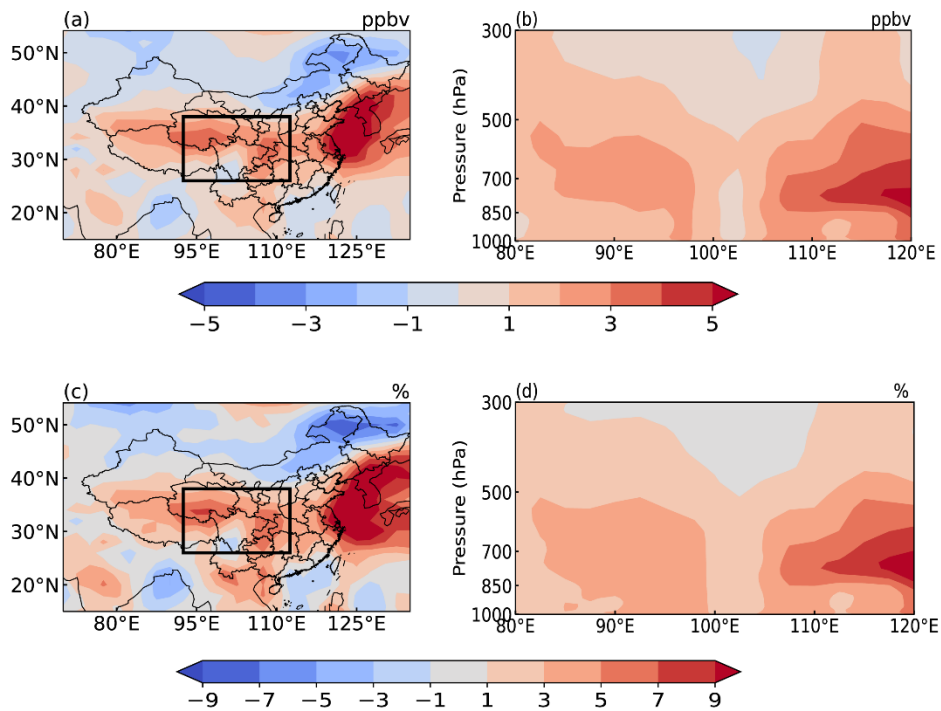
Figure 3. (a) Spatial distribution of the correlation coefficients between JJA near-surface O₃ concentrations and QBO index over 1981–2020. (b) and (c) are the same as (a), but during years having positive (22 years) and negative (18 years) SST anomalies over the eastern tropical Pacific, respectively. The stippled areas indicate statistical significance at the 90% confidence level.



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784 **Figure 4.** Spatial distribution of JJA surface O₃ concentration anomalies of (a)
 785 the selected three QBOW years (1990, 1997 and 2019), (b) the selected three
 786 QBOE years (1994, 2012 and 2018), respectively, relative to the
 787 climatological average (1981–2020).

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Figure 5. Spatial distributions of (a) absolute (ppbv) and (c) percentage (%) differences relative to the climatological mean (1981–2020) in JJA near-surface O₃ concentrations between the selected three QBOW years (1990, 1997 and 2019) and QBOE years (1994, 2012 and 2018) (QBOW–QBOE).

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The pressure–longitude cross sections averaged over the latitudes of 26°–38°N show (b) absolute (ppbv) and (d) percentage (%) differences relative to the climatological mean in JJA O₃ concentrations between the selected three QBOW years and QBOE years.

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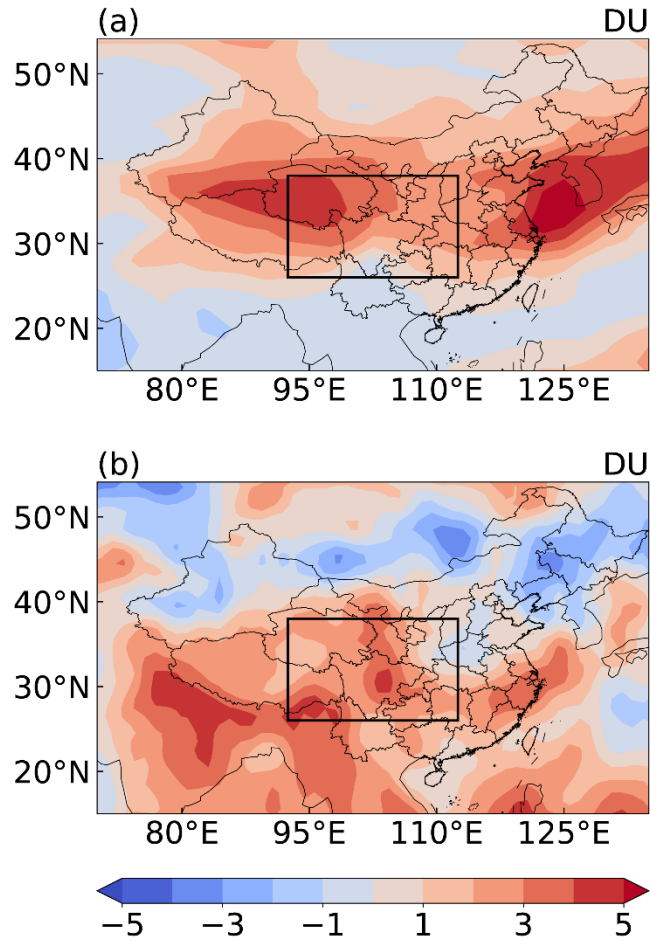
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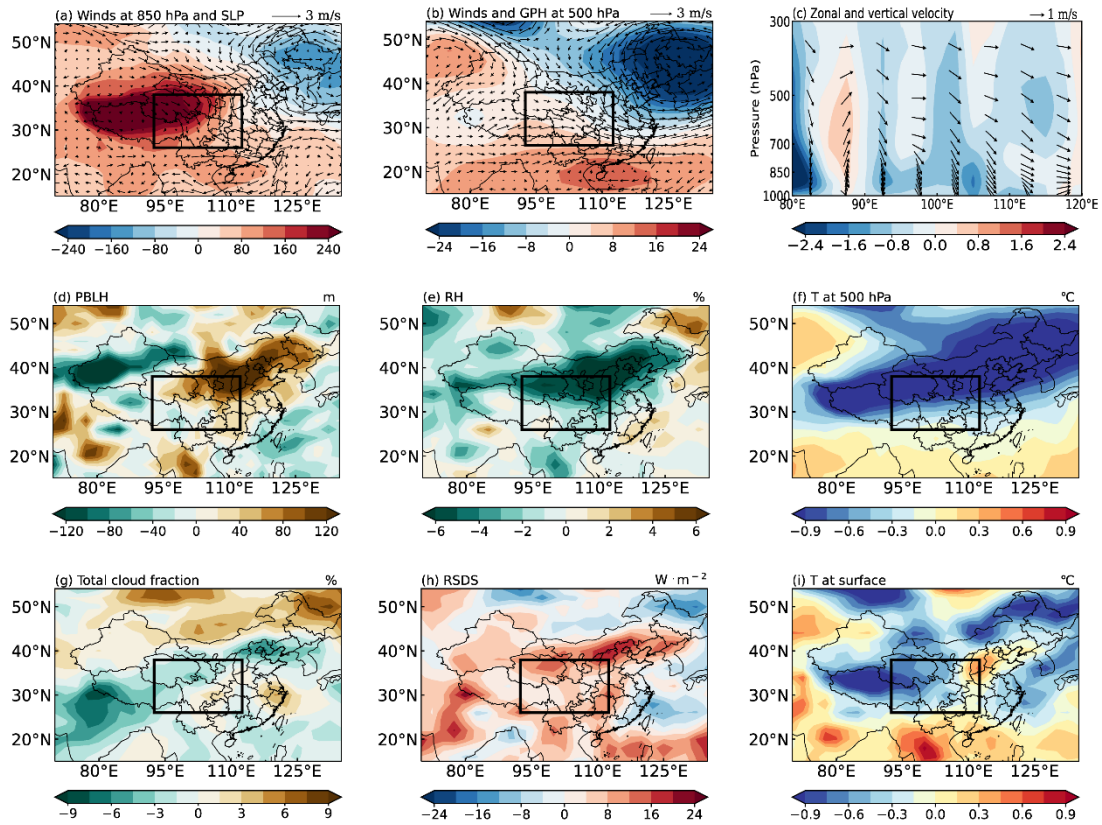


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802 **Figure 6.** Spatial distribution of JJA tropospheric column O₃ (TCO, DU)
 803 difference between the selected QBOW year (2019) and QBOE year (2012,
 804 2018) based on (a) GEOS-Chem simulations and (b) Aura OMI/MLS.

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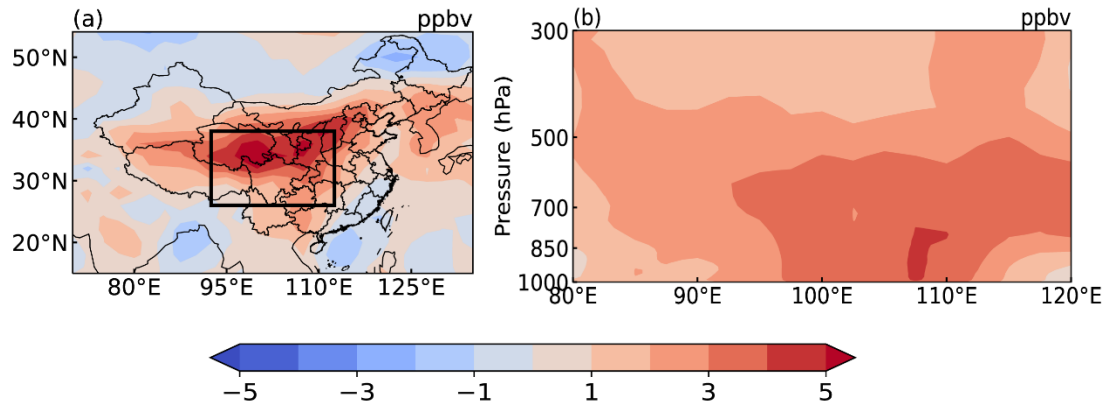


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808 **Figure 7.** Composite differences in the spatial distribution of JJA mean (a)
 809 wind fields (m s^{-1} , vector) at 850 hPa and sea level pressure (SLP, Pa,
 810 contour), (b) wind fields (m s^{-1} , vector) and geopotential height (GPH, m,
 811 contour) at 500 hPa, (d) planetary boundary layer height (PBLH, m), (e)
 812 relative humidity (RH, %) at the surface, (f) air temperature (T , $^{\circ}\text{C}$) at 500 hPa,
 813 (g) total cloud fraction (%), (h) downwelling shortwave radiation at the surface
 814 (RSDS, W m^{-2}), and (i) surface air temperature (T , $^{\circ}\text{C}$) between three QBOW
 815 years (1990, 1997 and 2019) and QBOE years (1994, 2012 and 2018)
 816 (QBOW–QBOE). In (c) the differences in JJA mean zonal wind (m s^{-1} , vector)
 817 and vertical velocity (OMEGA, Pa s^{-1} , vector and contour) multiplied by a
 818 factor of -100 , averaged over 26° – 38°N between three QBOW years (1990,
 819 1997 and 2019) and QBOE years (1994, 2012 and 2018) (QBOW–QBOE).
 820 The solid black boxes mark central China (92.5° – 112.5°E , 26° – 38°N).

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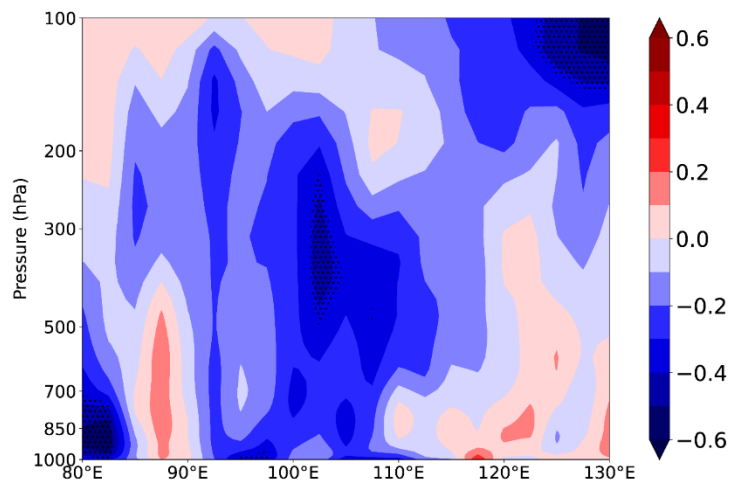


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824 **Figure 8.** (a) Spatial distribution of differences in JJA near-surface O₃
 825 concentrations (ppbv) and, (b) the pressure–longitude cross sections
 826 averaged over the latitudes of 26°–38°N of differences in JJA O₃
 827 concentrations (ppbv) between three QBOW years (1990, 1997 and 2019)
 828 and QBOE years (1994, 2012 and 2018) (QBOW–QBOE) from the simulation
 829 that has the China anthropogenic emissions of O₃ precursors turned off
 830 (NO_CHN). The solid black box in a marks central China.

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Figure 9. Pressure-longitude distribution of the correlation coefficients between QBO index and vertical velocity (OMEGA, multiplied by a factor of -100) in JJA averaged over 26° – 38° N for years with warm SST anomaly. The stippled areas indicate statistical significance at the 90% confidence level.