1	Enhanced upward motion through the troposphere over the tropical
2	western Pacific and its implications for the transport of trace gases
3	from the troposphere to the stratosphere
4	Kai Qie 1, Wuke Wang2, Wenshou Tian 1*, Rui Huang1, Mian Xu1, Tao Wang1,
5	Yifeng Peng <sup>1</sup>
6	
7	<sup>1</sup> College of Atmospheric Sciences, Lanzhou University, Lanzhou 730000, China
8	<sup>2</sup> Department of Atmospheric Science, China University of Geosciences, Wuhan
9	430074, China
10	
11	
12	*Corresponding author: Wenshou Tian (wstian@lzu.edu.cn)
13	

#### Abstract

14

15

16

17

18

19

20

21

22

23

24

25

26

27

28

29

30

31

32

33

34

35

The tropical western Pacific (TWP) is a preferential area of air uplifting from the surface to the upper troposphere. A significantly intensified upward motion through the troposphere over the TWP in the boreal wintertime (November to March of the next year, NDJFM) has been detected using multiple reanalysis datasets. The upward motion over the TWP is intensified at rates of 8.0±3.1% decade-1 and 3.6±3.3% decade-1 in NDJFM at 150 hPa from 1958 to 2017 using JRA55 and ERA5 reanalysis datasets, while the MERRA2 reanalysis data show a 7.5±7.1% decade-1 intensified upward motion for the period 1980-2017. Model simulations using the Whole Atmosphere Community Climate Model, version 4 (WACCM4) suggest that warming global sea surface temperatures (SSTs), particularly SSTs over the eastern maritime continent and tropical western Pacific, play a dominant role in the intensification of the upward motion by strengthening the Pacific Walker circulation and enhancing the deep convection over the TWP. Using CO as a tropospheric tracer, the WACCM4 simulations show that an increase of CO at a rate of 0.4 ppbv decade-1 at the layer 150-70 hPa in the tropics is mainly resulted from the global SST warming and the subsequent enhanced upward motion over the TWP in the troposphere and strengthened tropical upwelling of Brewer-Dobson (BD) circulation in the lower stratosphere. This implies that more tropospheric trace gases and aerosols from both natural maritime source and outflow from polluted air from South Asia may enter the stratosphere through the TWP region and affect the stratospheric chemistry and climate.

- **Keywords**: Upward motion; Troposphere-to-stratosphere transport; Tropical western
- Pacific; Trend; Sea surface temperature

### 1 Introduction

39

40

41

42

43

44

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

The tropical western Pacific (TWP) is a critical region for tropical and global climate (e.g., Webster et al., 1996; Hu et al., 2020). It has the largest area of warm sea surface temperature (exceeding 28 °C) which fuels intense and massive deep convection and thus is the largest source of latent heat and water vapor into the atmosphere (Webster and Lukas, 1992). The TWP region is also the most important source of tropospheric air entering the stratosphere due to the strong upward motion and deep convection over this region (e.g., Fueglistaler et al., 2004; Pan et al., 2016). Through the TWP region, tropospheric trace gases, e.g., the natural maritime bromine-containing substances and outflow from anthropogenic emissions from South Asia, are lifted to the upper troposphere and lower stratosphere (UTLS) by the strong upward motion and the deep convection and subsequently into the stratosphere by the large-scale upwelling (e.g., Levine et al., 2007, 2008; Navarro et al., 2015), which affect the ozone concentration and other chemical processes in the stratosphere (e.g., Feng et al., 2007; Sinnhuber et al., 2009). At the same time, the TWP region has the lowest cold-point tropopause temperature (CPTT) over the globe and plays an important role in controlling the water vapor concentration in the stratosphere (e.g., Fueglistaler et al., 2009; Newell and Gould-Steward, 1981; Pan et al., 2016; Randel and Jensen, 2013). The TWP is an important region for tropospheric trace gases being transported from the troposphere to the stratosphere, and therefore influencing the stratospheric chemistry (e.g., Fueglistaler et al., 2004; Levine et al., 2007; Krüger et al., 2008; Pan et al., 2016).

62

63

64

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

The TWP was thought to be the main pathway of the troposphere-to-stratosphere transport. A concept of "stratospheric fountain" was proposed by Newell and Gould-Steward (1981), which suggested that the poor-water vapor air in the stratosphere stems mainly from the TWP region. However, following studies using the observational and reanalysis data showed that there is subsidence at the near-tropopause level over the maritime continent, which is named as the "stratospheric drain" (Gettelman et al., 2000; Sherwood, 2000; Fueglistaler et al., 2004). Further studies verified that the large-scale transport from the tropical tropopause layer (TTL) to the stratosphere is dominated by the upward branch of the Brewer-Dobson (BD) circulation (Brewer, 1949; Dobson, 1956; Holton et al., 1995) while the local upwelling may play a minor role (e.g., Levine et al., 2007; Fueglistaler et al., 2009; Schoeberl et al., 2018). Though the vertical transport from TTL to the lower stratosphere is dominated by the BD circulation, numerous studies confirmed that the TWP region is an important pathway of the surface air entering the TTL (Fueglistaler et al., 2004; Levine et al., 2007; Krüger et al., 2008; Haines and Esler, 2014). Based on a trajectory model, Fueglistaler et al. (2004) pointed out that approximately 80% of the trajectories ascending into the stratosphere from the TTL are originated from the TWP region. Bergman et al. (2012) suggested that the tropospheric air over the TWP enters the stratosphere mainly in boreal winter, while less air over the TWP could be transported into the stratosphere during boreal summer. Other studies also found that the TWP

region is an important source of the tropospheric trace gases in the TTL (e.g., Newton et al., 2018; Pan et al., 2016; Wales et al., 2018), even the polluted air from East Asia could be transported rapidly to Southeast Asia by meridional winds and subsequently be elevated to the tropical upper troposphere by the strong upward motion and the deep convection (Ashfold et al., 2015). Hence, the strength of the upward motion over the TWP region during boreal winter is a key feature for understanding the variations of trace gases in the TTL and therefore important for stratospheric chemistry and climate.

The strength of the TWP upward motion is closely related to atmospheric circulation and deep convection. The ascending branch of the Pacific Walker circulation and the strong deep convection over the TWP allow rapid transport from the surface to the upper troposphere (Hosking et al., 2012). In association with global warming, atmospheric circulation, deep convection as well as the boundary conditions (e.g., sea surface temperature; SST) have been changed. For example, the Hadley cell has been extended to the subtropics and the Walker circulation over the Pacific has been shifted westward over the past decades (e.g., Lu et al., 2007; Garfinkel et al., 2015; Ma and Zhou, 2016). At the same time, SSTs over most of areas are getting warmer (Cane et al., 1997; Deser et al., 2010), which modulates the deep convection and atmospheric wave activities in the troposphere and then lead to changes of atmospheric circulations from the troposphere and the stratosphere (e.g., Garfinkel et al., 2013; Xie et al., 2012, 2014a; Wang et al., 2015; Hu et al., 2016; Lu et al., 2020). However, how the strength of the upward motion in the lower TTL over the TWP

region has been changed over the past decades remains unclear. In this study, we investigate the long-term trend of the upward motion over the TWP using JRA55, ERA5, and MERRA2 reanalysis datasets and different WACCM4 simulations as described in Section 2. The implication of the changes in the upward motion over the TWP to the transport of trace gases from the surface to the UTLS will be discussed in Section 3.

#### 2 Data and method

105

106

107

108

109

110

111

112

113

114

115

116

117

118

119

120

121

122

123

124

125

126

Reanalysis data. To investigate the long-term trend of the upward motion over the TWP through the troposphere, three most recent reanalysis datasets, including JRA55 from the Japan Meteorological Agency (JMA), ERA5 from the European Centre for Medium Range Weather Forecasting (ECMWF) and MERRA2 from the National Aeronautics and Space Administration/Global Modeling and Assimilation Office (NASA/GMAO) are used in this study. The JRA55 is produced by a four dimensional variational (4D-Var) data assimilation system and provide 6-hourly data (Harada et al. 2016). It has a vertical resolution of 60 levels from the surface to 0.1 hPa and a high horizontal resolution (T319). It covers the period from 1958 to present, are interpolated to the standard pressure levels and 1.25°×1.25° horizontal mesh. More details could be found in Kobayashi et al., 2015. The ERA5 reanalysis is the newest generation product from the ECMWF (Hersbach et al., 2020). The ERA5 data are based on the Integrated Forecasting System (IFS) Cy41r2, which includes the improved model physics, core dynamics and data assimilation. The ERA5 has hourly output, a horizontal resolution of 31 km, and a vertical resolution of 137 levels

extending into the middle atmosphere (0.01 hPa) and covers the period from 1979 to present. The ERA5 data also extend back to 1958, which is coinciding with the time that radiosonde observations in the Arctic became more systematic and regular. It should be noted that the ERA5 data suffer from a bias during 2000-2006, and are replaced by the ERA5.1 data in this period here. The MERRA2 data are also used, which are only accessible after 1980 (Gelaro et al., 2017). The MERRA2 data are produced by NASA/GMAO using Goddard Earth Observing System model (GEOS), which have 3-hourly temporal resolution, 72 vertical levels up to 0.01 hPa, and a horizontal resolution of 0.5°×0.625°. Although the horizontal and vertical resolution of MERRA2 data are similar to MERRA data, the MERRA2 data represent UTLS processes better (Gelaro et al., 2017). The monthly mean air temperature, horizontal wind fields and vertical velocity at different pressure levels are extracted from the three Reanalysis datasets. In the present study, we mainly focus on the upward motion over TWP region in NDJFM, which is defined as 20°S-10°N, 100°E-180° due to the strong upward motion (Fig. 1) and significantly increasing trends of the upward motion (Fig. 2) over there.

127

128

129

130

131

132

133

134

135

136

137

138

139

140

141

142

143

144

145

146

147

148

A special caution is needed because of the limitations of reanalysis data. The reanalysis datasets assimilate observational data based on the ground- and space-based remote sensing platforms to provide more realistic data products. However, previous studies suggested that there are still uncertainties in the reanalysis data (e.g., Simmons et al., 2014; Long et al., 2017; Uma et al., 2021). The accuracy of the vertical velocity in reanalysis data sets has been evaluated by the Reanalysis

Intercomparison Project (Fujiwara et al., 2017), which is initiated by the Stratosphere-troposphere Processes And their Role in Climate (SPARC). Results of a comparison between the radar observed data and the reanalysis data indicate that the updrafts in the UTLS are captured well near the TWP even though there are still large biases in the reanalysis datasets and the updrafts from the JRA55 data are stronger than those from the ERA5 and MERRA2 data (Uma et al. 2021). Additionally, discontinuities in the reanalysis data due to different observing systems (for example, transition from TOVS to ATOVS) may still exist (e.g., Long et al., 2017), which could lead to uncertainties in the long-term trend of a certain meteorological field. Hitchcock (2019) suggested that the reanalysis uncertainty is larger in the radiosonde era (after 1958) than in the satellite era (after 1979), but the radiosonde era is of equivalent value to the satellite era because the dynamical uncertainty dominates in the both eras. The data in the radiosonde era (1958-1978) used in the present study may induce uncertainties in our results. Therefore, we discuss the trends for both the periods of 1958-2017 and 1980-2017. In addition, we combine three most recent reanalysis datasets (JRA55, ERA5, and MERRA2) to obtain relatively robust results. Observed CO data. CO is used as a tropospheric tracer in this study to indicate the vertical transport from the near-surface to the upper troposphere and the lower stratosphere. The CO data used in the present study are from space-borne Microwave Limb Sounder (MLS; Livesey et al., 2015) observation and Measurements Of

149

150

151

152

153

154

155

156

157

158

159

160

161

162

163

164

165

166

167

168

169

170

Pollution In The Troposphere instrument (MOPITT; Deeter et al., 2019). MLS is

carried by Aura, which has a sun-synchronous orbit at 705 km with a 16-day repeat

cycle. MLS observations are made from 82°S to 82°N and cover the period from 2005 to the present. MLS provides the CO data from the upper troposphere to the mesosphere. MLS CO v4 level1 data used in the present study are processed using the recommended procedures (Livesey et al., 2015) and interpolated into a 5°×5° horizontal mesh. MOPITT CO data are also used for comparison. MOPITT instrument is aboard on the Terra satellite permitting retrievals of CO vertical profiles using both thermal-infrared and near-infrared measurements and has a field of view of 22 km×22 km. The Terra satellite was launched in 1999 with a 705 km sun-synchronous orbit. MOPITT provides the CO data from the surface to the upper troposphere during the period of 2000/03 to the present. Here, we use the daytime only MOPITT v8 level3 CO data. For comparison, we focus on the CO concentrations in MLS and MOPITT data at similar level (215 hPa in MLS data and 200 hPa in MOPITT data, respectively).

SST and outgoing longwave radiation (OLR) data. SST data are used in this study to investigate the relationship between the upward motion and SSTs. The SST data are from the HadISST dataset (1°×1° horizontal mesh) during 1958-2018 (Rayner et al., 2003). OLR is often utilized to reflect the deep convection in the tropics. The OLR data are extracted from NOAA Interpolated OLR dataset on a 2.5°×2.5° horizontal mesh during 1974/11-2018/03 (Liebmann and Smith, 1996).

**Model simulations.** A series of model simulations with the Whole Atmosphere Community Climate Model version 4 (WACCM4) are performed to find out the main impact factors of the trend of the upward motion over the TWP. The WACCM4 is a

chemical-climate model with a horizontal resolution of 1.9°×2.5° (Marsh et al., 2013). The WACCM4 has vertical 66 levels from the surface to 145 km with vertical resolution of approximately 1 km in the UTLS, which is numerously used to investigate the transport of the trace gases from the troposphere to the stratosphere (e.g., Randel et al., 2010; Xie et al., 2014b; Minganti et al., 2020). A hindcast simulation (Control simulation) is performed with observed greenhouse gases, solar irradiances, and prescribed SSTs (HadISST dataset is used) during 1955-2018. A single-factor controlling simulation (Fixsst simulation) is done for the same period with the same forcings, except that the global SSTs are fixed to the climatological mean values during 1955-2018 (long-term mean for each calendar month during 1955-2018).

To figure out the impact of the warming SST over the TWP region on the intensification of the upward motion over the TWP region, a couple of time-slice simulations (R1 and R2) are also integrated for 33 years. The SSTs over the eastern maritime continent and tropical western Pacific (20°S-20°N, 120°E-160°E) in the boreal wintertime (November to March of the next year, NDJFM) in R1 are prescribed as the climatological mean SSTs during 1998-2017, while the SSTs over other regions are fixed as the climatological mean SSTs during 1958-2017. The SSTs in R2 are the same as the SSTs in R1 except that the SSTs over the region (20°S-20°N, 120°E-160°E) in NDJFM are prescribed as the climatological mean SSTs during 1958-1977. Since the SSTs over the eastern maritime continent and tropical western Pacific (20°S-20°N, 120°E-160°E) show significantly warming trends, the SSTs

- during 1998-2017 are higher than the SSTs during 1958-1977 (approximately 0.5 K).
- Hence, the difference between R1 and R2 reflects the impact of the warmed SSTs
- over the eastern maritime continent and tropical western Pacific (20°S-20°N,
- 218 120°E-160°E) on the atmospheric circulation. The first 3 years of the numeric
- simulations are not used in the present study to provide a spin-up.
- Transformed Eulerian Mean (TEM) Calculation. To diagnose the changes in
- 221 the BD circulation, the meridional and vertical velocities of the BD circulation are
- calculated by the TEM equations (Andrews and Mcintyre, 1976):

223 
$$v^* = \overline{v} - \frac{1}{\rho} \left( \frac{\rho \overline{v' \theta'}}{\overline{\theta_z}} \right)_z$$

224 
$$w^* = \overline{w} + \frac{1}{a \cos \varphi} \left( \cos \varphi \frac{\overline{v' \theta'}}{\overline{\theta_z}} \right)_{\varphi}$$

- Where  $v^*$  and  $w^*$  denote the meridional and vertical velocities of the BD circulation;
- the overbar represents the zonal mean; the prime denotes the deviation from the zonal
- mean;  $\theta$ , a,  $\varphi$ , and  $\rho$  indicate the potential temperature, the radius of the earth,
- the latitude, and the standard density.
- Linear trends and the significance test. The linear trends are estimated using a
- 230 simple least square regression method. The significances of the correlation
- coefficients, mean differences, and trends are determined via a two-tail Student's t-test.
- The confidence interval of trend is calculated using the following equation (Shirley et

233 al., 2004): 
$$\left(b - t_{1-\frac{\alpha}{2}}(n-2)\sigma_b, b + t_{1-\frac{\alpha}{2}}(n-2)\sigma\right)$$

where b is the estimated slope,  $\sigma$  denotes the standard error of the slope, and

235  $t_{\frac{1-\alpha}{2}}(n-2)$  represents the value of t-distribution with the degree of freedom equal to

236 n-2.  $\alpha$  is the two-tailed confidence level.  $\sigma$  is calculated as:  $\sigma = b\sqrt{\frac{\frac{1}{r^2} - 1}{n - 2}}$ .

#### 3 Results

237

238

239

240

241

242

243

244

245

246

247

248

249

250

251

252

253

254

255

#### 3.1 Enhanced upward motion over the TWP

According to previous studies, the lapse-rate tropopause is a good proxy to separate the tropospheric and the stratospheric dynamic behavior (vertical motion dominated and horizontal mixing dominated, respectively) over the TWP (Pan et al., 2019). Since the lapse-rate tropopause over the TWP in the boreal winter is near 100 hPa (not shown), we utilize the vertical velocity at 150 hPa to reflect the vertical transport in the upper troposphere. Figure 1 shows mean values of the vertical velocity at 150 hPa for each month averaged over 60 years from 1958 to 2017. The TWP region at the UTLS level has strong upward motion due to the frequent intense deep convection and the Pacific Walker circulation. It is noteworthy that there is strong upward motion at 150 hPa in NDJFM over the TWP, while the upward motion in other months shifts northward corresponding to the Asia summer monsoon. This is consistent with previous studies (Newell and Gould-Steward, 1981; Bergman et al., 2012). Therefore, we mainly focus on the changes in the upward motion in NDJFM, which is more important to the transport of air over the TWP from the lower troposphere to the TTL compared to the summer months (as shown in Fig. 1) and subsequently to the lower stratosphere. As seen in Figs. 1a-c and 1k-l, the upward motion (w) at 150 hPa is most evident over the region 20°S-10°N, 100°E-180°, which

is used to indicate the TWP in the following analysis. The climatological mean 150 hPa vertical velocity (*w*) in NDJFM in ERA5 during 1958-2017 and MERRA2 during 1980-2017 are also given in Supplementary Fig. 1. Comparing with the 150 hPa *w* in NDJFM using JRA5, the 150 hPa *w* in ERA5 and MERRA2 data shows larger values (maximum larger than 1.5 m s<sup>-1</sup>) over the land areas but smaller values (minimum less than -0.4 m s<sup>-1</sup>) over the marine area. Notably, the 150 hPa *w* shows no subsidence over the maritime continent, while there is descending motion over the maritime continent at 100 hPa (Supplementary Fig. 2), which is referred to the "stratospheric drain" (Gettleman et al., 2000; Sherwood, 2000).

Figure 2 displays the linear trends of *w* in the upper (150 hPa), middle (500 hPa) and lower (700 hPa) troposphere in NDJFM from 1958 to 2017 using JRA55, ERA5, and MERRA2 reanalysis datasets. The 150 hPa *w* increased significantly over most areas of the TWP during 1958-2017 (Fig. 2). At the same time, the upward motion over the TWP in the lower and middle troposphere also mainly shows positive trends (Figs. 2d and g). This indicates that the upward motion over the TWP is increasing through the troposphere from 1958 to 2017. Such an enhancement of the upward motion over the TWP is evident in all three reanalysis datasets used here (JRA55, ERA5, and MERRA2), although there are also some differences between the three reanalysis datasets. For example, the trends of the horizontal winds in the upper troposphere in MERRA2 (Fig. 2c) are larger than those in JRA55 and ERA5 (Figs. 2a and b). There are negative trends of vertical velocity in JRA55 and ERA5 while

these differences are mainly due to the different time periods which are used to calculate the linear trends in JRA55 (1958-2017), ERA5 (1958-2017) and MERRA2 (1980-2017). Supplementary Fig. 3 gives the trends of *w* and horizontal winds in NDJFM during 1980-2017 derived from JRA55, ERA5, and MERRA2 data, which shows insignificant differences between these reanalysis datasets. The trend patterns of the horizontal winds in JRA55, ERA5, and MERRA2 are consistent with each other (Supplementary Fig. 3). For the trends of vertical velocity, significant positive trends over the TWP region can be noted in the JRA55, ERA5, and MERRA2 datasets, although the trends in ERA5 are slightly weaker than those in JRA55 and MERRA2 (Fig. 2 and Supplementary Fig. 3). Comparing to the negative trends of the vertical velocity over the central Pacific in JRA55 and ERA5, the negative trends in MERRA2 extend more northward (Supplementary Fig. 3).

The time series of the upward motion intensity over the TWP from different datasets are given in Fig. 3. The intensity of the upward motion over the TWP used in Fig. 3 is simply defined as the area-averaged upward mass flux at a specific level, and the standardized intensity is calculated as the intensity divided by the standard deviation of the intensity at the corresponding level. The intensity of the upward motion over the TWP at 150 hPa increased significantly in NDJFM during last decades, which can be confirmed by all the three reanalysis datasets (Fig. 3). The intensity of the upward motion over the TWP at 150 hPa increased  $3.0\pm1.2\times10^8$  kg s<sup>-1</sup> decade<sup>-1</sup> ( $8.0\pm3.1\%$  decade<sup>-1</sup>),  $1.3\pm1.2\times10^8$  kg s<sup>-1</sup> decade<sup>-1</sup> ( $3.6\pm3.3\%$  decade<sup>-1</sup>), and  $3.0\pm2.8\times10^8$  kg s<sup>-1</sup> decade<sup>-1</sup> ( $7.5\pm7.1\%$  decade<sup>-1</sup>) in JRA55, ERA5, and MERRA2 data,

respectively. As shown in Figs. 3b and c, the intensity of the upward motion at 500 hPa and 700 hPa in JRA55 and the intensity of the upward motion at 500 hPa in ERA5 over the TWP also increased significantly at 95% confidence level  $(4.6 \pm 2.6 \times 10^8 \ kg \ s^{\text{-}1} \ decade^{\text{-}1}, \ 2.9 \pm 1.7 \times 10^8 \ kg \ s^{\text{-}1} \ decade^{\text{-}1}, \ and \ 2.5 \pm 2.5 \times 10^8 \ kg \ s^{\text{-}1}$ decade<sup>-1</sup>, respectively). The increasing trends of the intensity of the upward motion at 700 hPa in ERA5 and at 500 hPa and 700 hPa in MERRA2 are significant at the 90% confidence level at rates of 1.9±1.6×10<sup>8</sup> kg s<sup>-1</sup> decade<sup>-1</sup>, 5.4±5.3×10<sup>8</sup> kg s<sup>-1</sup> decade<sup>-1</sup> and  $3.9\pm3.8\times10^8$  kg s<sup>-1</sup> decade<sup>-1</sup>, respectively. This suggests a comprehensive enhancement of vertical velocity through the whole troposphere, which is evident from the surface to 100 hPa (Supplementary Fig. 4). It can also be inferred that the upward motions over the TWP increased at different rates during the past decades due to the difference between JRA55, ERA5, and MERRA2 data. Hence, caution is suggested when investigating the trend of the upward motion over the TWP using the reanalysis data. While the trace gases in the TTL are modulated by the upward motion and subsequent vertical transport (e.g., Garfinkal et al., 2013; Xie et al., 2014b), such a strengthening of the upward motion over the TWP may lead to more tropospheric trace gases in the TTL. The changes in the atmospheric circulation at the UTLS level in the tropics are

300

301

302

303

304

305

306

307

308

309

310

311

312

313

314

315

316

317

318

319

320

321

closely related to the changes in the tropical deep convection and SSTs (e.g., Levine et al., 2008; Garfinkal et al., 2013; Xie et al., 2020). Here, the trends of observed OLR provided by NOAA (see Section 2) in NDJFM during 1974-2017 are shown in Fig. 4a. Though the time period of the observed OLR data is shorter than the time period we

analyzed, the changes in OLR could partly reflect the changes in the deep convection during 1958-2017. The OLR shows significantly negative trends over the TWP which indicates intensified deep convection over the TWP. The OLR trend pattern is very similar to the trend pattern of the 150 hPa w (Figs. 2a-c), which indicates that the increasing trends of 150 hPa w are closely related to the intensified deep convection over the TWP. The intensified deep convection not only lead to the strengthened upward motion in the UTLS (Highwood and Hoskins, 1998; Ryu and Lee, 2010), but also result in the decreased temperature near the tropopause which plays a dominant role in modulating the lower stratospheric water vapor concentration (e.g., Hu et al., 2016; Wang et al., 2016). Corresponding to the enhanced deep convection over the TWP, the CPTT derived from JRA55 data (see Fig. 4b) shows significantly decreasing trends over the TWP in NDJFM during 1958-2017, which is consistent with Xie et al., (2014a). However, negative trends are also found in other regions in low and mid-latitudes, except over the central and east Pacific. It should be noted that the CPTT from different reanalysis datasets may show different trends even for the satellite period (Tegtmeier et al., 2020). Additionally, the JRA55 data before 1978 may also lead to uncertainties in the CPTT trends. Caution is needed when discussing the trends of CPTT from reanalysis datasets. The changes in the deep convection over the tropical Pacific may be related to

322

323

324

325

326

327

328

329

330

331

332

333

334

335

336

337

338

339

340

341

342

343

The changes in the deep convection over the tropical Pacific may be related to the changes in the Pacific Walker circulation. The Pacific Walker circulation shows a significant intensification over the past decades (e.g., Meng et al., 2012; L'Heureux et al., 2013; McGregor et al., 2014). The vertical velocity at 500 hPa and 150 hPa shows

significantly positive trends over the TWP in NDJFM during 1958-2017 (Fig. 2). Meanwhile, the lower tropospheric zonal wind shows easterly trends over the tropical Pacific, while the upper tropospheric zonal wind shows westerly trends over the tropical Pacific, which suggests a strengthened Pacific Walker circulation and is consistent with previous studies (Hu et al., 2016; Ma and Zhou, 2016).

344

345

346

347

348

349

350

351

352

353

354

355

356

357

358

359

360

361

362

363

364

365

The strengthened Pacific Walker circulation is closely related to the changes in the SSTs (e.g., Meng et al., 2012; Ma and Zhou, 2016). The trends of the SSTs in NDJFM during 1958-2017 are shown in Fig. 4c. The SST shows significantly warming trends almost over the world except the central Pacific in NDJFM during 1958-2017. In addition, the intensity of the upward motion over the TWP is significantly correlated with the SST (Fig. 4d), which suggests that the SST has important effects on the upward motion over the TWP. The correlation coefficient in Fig. 4d shows a La Niña-like pattern and indicates that the ENSO events exert important impacts on the upward motion over the TWP (Levine et al., 2008). The SSTs over the TWP are mainly positively correlated with the upward motion intensity over the TWP with negative correlations shown over the western maritime continent, while the SSTs over tropical central, eastern Pacific, and Indian Ocean show negative correlations with the intensity of the upward motion over the TWP. The SSTs over the Atlantic Ocean are poorly correlated with the upward motion intensity over the TWP (not shown). This result suggests that the changes in global SSTs may be the primary driver of the strengthened Pacific Walker circulation, which leads to enhanced deep convection and intensified upward motion over the TWP.

It could be found that there are extreme minima (1982, 1991, and 1997) in Fig. 3, which may be related to the El Niño events occurred in these years. To further figure out the impact of ENSO events on the upward motion over the TWP, Supplementary Fig. 5 displays the time series of the standardized intensity of the upward motion over the TWP at 150 hPa, 500 hPa, and 700 hPa in NDJFM in JRA55, ERA5, and MERRA2 with the ENSO signal removed using the linear regression method (Hu et al., 2018; Qie et al., 2021). The extreme minima (1982, 1991, and 1997) become much weaker in Supplementary Fig. 5 than those in Fig. 3, which indicates that the El Niño events are responsible for the extreme minima. The upward motions over the TWP at 150 hPa, 500 hPa, and 700 hPa in NDJFM in JRA55, ERA5, and MERRA2 still show statistically significant increasing trends after removing the ENSO signal in Supplementary Fig. 5, which suggests that ENSO events exert limited impacts on the trends of the upward motion over the TWP in NDJFM during 1958-2017.

## 3.2 Simulated trend of the upward motion over the TWP and its potential mechanism

To verify the impact of SST on the trend of the upward motion over the TWP, a couple of model simulations with WACCM4 are employed in the following analysis. Consistent with the results shown using the reanalysis data (Figs. 2a-c), the simulated 150 hPa w (Control simulation) shows significantly increasing trends over the TWP and decreasing trends over the tropical eastern Pacific in NDJFM during 1958-2017 (Fig. 5a). Additionally, the 150 hPa w simulated in the Fixsst simulation shows weak trends over the TWP (Fig. 5b). The difference between the Control and the Fixsst

simulations suggests that the trends of the 150 hPa w over the TWP region is dominated by the changes in the global SSTs during 1958-2017. There are also significantly positive trends of the vertical velocity over the TWP in the lower (700 hPa) and middle troposphere (500 hPa) in the Control simulation, while the zonal winds are also enhanced over the tropical Pacific. The vertical velocity over the TWP in the Fixsst simulation shows weak negative trends and the changes in zonal winds over the tropical Pacific are very weak. This confirms the dominant role of the changes in global SSTs on the enhancement of the Walker circulation.

Previous studies found that the changes in the intensity of the Pacific Walker circulation and the stratospheric residual circulation are closely related to the changes in tropical SST (Meng et al., 2012; Tokinaga et al., 2012; Lin et al., 2015). As suggested by the correlation coefficients between the upward motion at 150 hPa over the TWP and SSTs in Fig. 4d, warmer SSTs over the tropical central and eastern Pacific, and Indian Ocean may lead to a weakened upward motion over the TWP (negative correlation). The warming trends of SSTs over the eastern maritime continent and tropical western Pacific may result in an intensification of the upward motion over the TWP. To verify the impact of the changes in the SSTs over eastern maritime continent and tropical western Pacific on the trends of the upward motion over the TWP, a couple of single-factor controlling time-slice simulations (R1 and R2) are performed with only SSTs over eastern maritime continent and tropical western Pacific (20°S-20°N, 120°E-160°E) in NDJFM changed in these two simulations. In R1, the SSTs over the eastern maritime continent and tropical western Pacific are

prescribed as the climatological mean SSTs during 1958-2017, while the SSTs over the eastern maritime continent and tropical western Pacific in R2 are prescribed as the climatological mean SSTs during 1958-1977 (more details are given in the section 2). The differences of the wind fields between R1 and R2 are shown in Fig. 6. The 150 hPa w shows significantly positive anomalies over the TWP and negative anomalies over the tropical eastern Pacific, which is consistent with the trends of the 150 hPa w in the Control simulation and the reanalysis datasets (Figs. 2 and 5). The upward mass flux over the TWP at 150 hPa increased approximately 27% in the R1 comparing with R2 due to the warming SSTs over the eastern maritime continent and tropical western Pacific (approximately 0.5 K). The upward motion in the lower and middle troposphere over the TWP shows increasing trends due to the enhanced convergence induced by the warmer SSTs over the TWP. This result is consistent with Hu et al. (2016), which suggested that the increased zonal gradient of the SSTs over the tropical Pacific could lead to a strengthened Pacific Walker circulation and an enhanced upward motion over the TWP. Therefore, the warmer SSTs over the TWP could contribute largely to the trend of the upward motion over the TWP in NDJFM during 1958-2017. The changes in the OLR simulated in WACCM4 associated with the changes in the global SSTs are shown in Fig. 7. There are significantly enhanced deep convection as indicated by OLR over the TWP due to the strengthened convergence in the Control simulation, while the deep convection shows weak and even decreasing

410

411

412

413

414

415

416

417

418

419

420

421

422

423

424

425

426

427

428

429

430

431

trends over the TWP in the Fixsst simulation (Figs. 7a and b). The enhanced deep

convection over the TWP could lead to the enhancing trends of the upward motion. Hence, it can be inferred that the changes in the global SSTs are responsible for the intensification of the Pacific Walker circulation, and the enhanced deep convection and a stronger upward motion over the TWP which could extend to the upper troposphere.

# 3.3 Implications for the concentrations of water vapor and CO in the TTL and lower stratosphere.

Previous studies showed that the enhanced deep convection and upward motion could lead to increased CO in the UTLS (e.g., Duncan et al., 2007; Livesey et al., 2013). At the same time, water vapor mixing ratios in the UTLS may increase due to the enhanced upward motion which could bring more wet air from low altitude to high altitude (e.g., Rosenlof, 2003; Lu et al., 2020). However, the water vapor mixing ratios in the lower stratosphere also depend on the tropopause temperature (e.g., Highwood and Hoskins, 1998; Garfinkel et al., 2018; Pan et al., 2019). Hence, the relationship between the intensity of upward motion and the water vapor concentration in the UTLS is complex. Here, the relationship between the trends of the upward motion over the TWP and the changes in CO and water vapor in the ULTS simulated with WACCM4 are analyzed.

The trends of CPTT, the 100 hPa streamfunction, and the water vapor concentration are shown based on the Control and the Fixsst simulation as well as their difference in Figs. 7d-i. The changes in the deep convection could lead to the changes in the atmospheric circulation by releasing the latent heat. The changes in the

tropical deep convection lead to a Rossby-Kelvin wave response at the UTLS level and then induce the changes in the air temperature near the tropopause (e.g., Gill, 1980; Highwood and Hoskins, 1998). The trends of the 100 hPa streamfunction show a Rossby wave response over the TWP and a Kelvin wave response over the tropical eastern Pacific in the Control simulation (Fig. 7d), which is caused by the changes in the deep convection over the tropical Pacific. The Rossby-Kelvin wave response further leads to the decrease the CPTT over the TWP and the increase of the CPTT over the tropical eastern Pacific. Previous studies suggest that the lower stratospheric water vapor is mainly influenced by the coldest temperature near the tropopause (e.g., Garfinkel et al., 2018; Zhou et al., 2021). Since the TWP has the coldest CPTT in the boreal winter (e.g., Pan et al., 2016), the significantly decreased CPTT over the TWP may result in significantly dried lower stratosphere (Fig. 7g). The intensity of the upward motion over the TWP shows negative correlations with the concentration of the tropical lower stratospheric water vapor (not shown). Hence, the enhanced upward motion over the TWP may correspond to a dried lower stratosphere. The CPTT shows weak trends over the TWP, and the tropical water vapor shows insignificant trends at 70 hPa in the Fixsst simulation. The comparison between the Control simulation and the Fixsst simulation suggests that the trends of the deep convection, the CPTT, and the lower stratospheric water vapor concentration in the tropics in NDJFM during 1958-2017 are dominated by the trends of the global SSTs, while other external forcings may play minor roles.

454

455

456

457

458

459

460

461

462

463

464

465

466

467

468

469

470

471

472

473

474

475

Generally, the intensified upward motion may lead to more tropospheric trace

gases lifting to the upper troposphere and entering the lower stratosphere (e.g., Rosenlof, 2003; Lu et al., 2020). Here we use CO as a tropospheric tracer to detect the possible influences of the enhanced upward motion over the TWP on the transportation of the tropospheric trace gases to the upper troposphere and the lower stratosphere. Due to the data limitation, it is not possible to show the corresponding changes of trace gases by observations in NDJFM during 1958-2017. Here, the trends of CO at around 200 hPa from MOPITT and MLS observations are shown in the Fig. 8. The CO increased significantly over the TWP in NDJFM in the upper troposphere from the MOPITT (at 200 hPa during 2000-2017) and MLS data (at 215 hPa during 2005-2017). The concentration of MLS CO over the TWP is approximately 80 ppbv at 215 hPa from MLS observations and 70 ppbv at 200 hPa from MOPITT observations, which is consistent with previous study (e.g., Huang et al., 2016). The MLS CO data show that the area-averaged CO increased approximately 2.0±3.7 ppbv decade<sup>-1</sup> over the TWP in NDJFM during 2005-2017. The area-averaged MOPITT CO data show a stronger increase of approximately 5.0±3.1 ppbv decade-1 at 200 hPa from 2000 to 2017 (significant at the 95% confidence level). It should be pointed out that the linear trends of CO are calculated based on the satellite data which only cover 14 or 18 years due to the data limitation. Hence, the linear trends of CO may have uncertainties particularly in the regions with large interannual variations. To partially overcome this shortage, the trends of MLS CO at 215 hPa during time periods of 2005-2016, 2006-2016, 2006-2017, and 2007-2016 and the trends of MOPITT CO at 200 hPa during time periods of 2000-2016, 2001-2016, 2001-2017, and 2002-2016

476

477

478

479

480

481

482

483

484

485

486

487

488

489

490

491

492

493

494

495

496

497

are shown in Supplementary Fig. 6. It could be found that the CO in the upper troposphere increased robustly over the TWP from both the MLS and MOPITT data. Overall, though the observed CO only covers less than 20 years, the results from the satellite data suggest a possible impact of the intensified upward motion over the TWP on the trace gases in the upper troposphere.

498

499

500

501

502

503

504

505

506

507

508

509

510

511

512

513

514

515

516

517

518

519

To further illustrate the impacts of the enhanced upward motion on the trace gas in the upper troposphere and lower stratosphere, the Control and Fixsst simulations with WACCM4 are used. The trends of the CO concentrations from the Control and Fixsst simulations as well as their differences are shown in Fig. 9. The tropical CO at 150 hPa shows significantly increasing trends both in the Control and the Fixsst simulations at rates of 3.4 ppbv decade<sup>-1</sup> and 3.2 ppbv decade<sup>-1</sup>, respectively, (Figs. 9a and b). This suggests that the surface emission of the CO exerts the most important effect on the increase of the tropical CO concentration. The differences of the CO trends at 150 hPa between the Control simulation and the Fixsst simulation are also displayed in Fig. 9c. Since the surface emission inventories of the two simulations are the same, it can be inferred that the trends of the CO concentration in Fig. 9c are mainly caused by the changes in the atmospheric circulation induced by the changes in the global SSTs. The difference of the CO concentration at 150 hPa between the Control simulation and the Fixsst simulation shows a significantly increasing trend at a rate of 0.2±0.1 ppbv decade-1 over the TWP (significant at the 95% confidence level). At the same time, decreasing trends over the central Africa exist, which resembles to the trend patterns of the vertical velocity in the lower TTL and the deep

convection (Figs. 5i and 7c). This indicates that the enhanced deep convection in the TWP lead to the strengthened upward motion over the TWP, which results in an extra 6% increasing trend of CO in the upper troposphere over the TWP. It could also be found that CO also increased in the mid latitudes of the southern hemisphere (Fig. 9c). According to previous studies, the CO perturbation from the Indonesian fires at upper troposphere could be transported to the tropical Indian Ocean by easterly winds and then to the subtropics in the southern hemisphere through the southward flow during boreal winter. The CO perturbation then spreads rapidly circling the globe following the subtropical jet (Duncan et al., 2007). This is consistent with our results which show intensified northerlies over the subtropical Indian Ocean (15°S-25°S, 60°E-100°E) at a rate of approximately 0.2 m s<sup>-1</sup> decade<sup>-1</sup> and strengthened westerlies over the subtropical Indian Ocean and western Pacific (20°N-35°N, 60°E-160°E) at a rate of approximately 0.3 m s<sup>-1</sup> decade<sup>-1</sup> (Figs. 5c and f).

The trends of the zonal mean CO concentration from model simulations are displayed in Figs. 10a-c. The zonal mean CO shows significantly increasing trends at all levels in the Control simulation and the Fixsst simulation, while the difference of the zonal mean CO between the Control simulation and the Fixsst simulation shows significantly increasing trends in the TTL but negative trends in the middle troposphere in the tropics and the Northern Hemisphere. At the same time, the difference of CO concentration between the Control simulation and the Fixsst simulation averaged in the western Pacific (100°E-180°E) shows significantly increasing trends in the tropics (20°S-10°N) from the surface to the TTL (Fig. 10f).

The CO in the layer 150-70 hPa over the TWP increased 3.2 ppbv decade<sup>-1</sup> and 2.8 ppbv decade<sup>-1</sup> in the Control and Fixsst simulations in NDJFM during 1958-2017, respectively. And the CO difference between the Control and Fixsst simulations increased 0.4±0.2 ppbv decade<sup>-1</sup> (significant at the 95% confidence level) in the layer 150-70 hPa over the TWP, which suggests that the intensifying upward motion over the TWP and the tropical upwelling of BDC could lead to an extra 14% increasing trend of CO. This indicates that the increased zonal mean CO in the TTL (Fig. 10c) is mainly transported through the western Pacific bands and highlights the importance of the upward motion over the TWP in elevating trace gases from the surface to the upper troposphere.

To understand the CO trends in the Control and Fixsst simulations and their differences, the trends of vertical velocity averaged over the globe and the TWP band are given in Fig. 11. The zonal mean w shows weak and even decreasing trends in the tropics while the w over the TWP intensified in the Control simulation in NDJFM during 1958-2017. This is consistent with Fig. 5. While the SSTs fixed to climatological values, the zonal mean w shows weak trends and the w over the TWP shows significantly negative trends. The changes in the global SSTs therefore leads to the increase of the w over the TWP region as indicated in the differences between the two simulations in Fig. 11f. In summary, the CO shows increasing trends (3.5 ppbv decade<sup>-1</sup>) at 150 hPa over the TWP in NDJFM during 1958-2017 induced by the changes in the surface emissions and the upward motion. The trends of CO at 150 hPa over the TWP in NDJFM during 1958-2017 in the Fixsst simulation mainly include

the impact induced by the increased surface emissions since the upward motion over the TWP in the Fixsst simulation shows weak trends. The difference between the Control and Fixsst simulations indicates that the enhanced tropospheric upward motion over the TWP forced by the changes in the global SSTs leads to some extra increase of CO concentrations in the upper troposphere. It should be mentioned that the increasing trends of CO in the lower troposphere in Fig. 10f may be mainly caused by the changes in the horizontal winds. Girach and Nair (2014) suggested that enhanced deep convection and the subsequent intensified upward motion may lead to a decreased CO concentration in the lower troposphere and an increased CO concentration in the upper troposphere. The trends of horizontal winds at 925 hPa are shown in Supplementary Fig. 8c. There are northerly trends over east Asia and northeasterly trends near the south Asia (Supplementary Fig. 8c), which suggests that more CO-rich air from east Asia and south Asia could be transported to the TWP in the Control simulation comparing to the Fixsst simulation. Since the CO concentration in the lower troposphere over the northern Pacific is higher than that over southern Pacific, the northerly trends over the western and central Pacific may also contribute to the increased CO in the lower troposphere over the TWP in Fig. 10f. As discussed in the Introduction, the tropospheric trace gases enter the stratosphere mainly through the large-scale tropical upwelling associated with the BD circulation. The trends of the BD circulation in different model simulations as well as their differences are displayed in Fig. 12. The tropical upwelling of BDC  $(w^*)$ calculated using the TEM formula increased significantly in the lower stratosphere

564

565

566

567

568

569

570

571

572

573

574

575

576

577

578

579

580

581

582

583

584

585

over past decades as seen in the JRA55 data and the Control simulation (Figs. 12a and 12b). We found that the 70 hPa upward mass flux in NDJFM in the tropics (15°S-15°N) increased 2.8±1.9% decade<sup>-1</sup> ( significant at the 95% confidence level) in the JRA55 data from 1958 to 2017 (Fig. 12a) and 4.6±4.3% decade<sup>-1</sup> ( significant at the 95% confidence level) in the MERRA2 data from 1980 to 2017 (Supplementary Fig. 7b). From the ERA5 data, the 70 hPa upward mass flux in NDJFM increased in the north hemisphere (0-15°N) at a rate of 5.0±2.8% decade-1 ( significant at the 95% confidence level), but decreased significantly in the south hemisphere (0-15°S) during 1958-2017 (Supplementary Fig. 7a). On average, the trend of the 70 hPa upward mass flux in NDJFM in the tropics (15°S-15°N) is not significant in ERA5. In fact, many previous studies have investigated the trends of BDC. For example, Abalos et al. (2015) investigated the trends of BDC derived from JRA55, MERRA, and ERA-Interim data during 1979-2012 and suggested that the BDC in JRA55 and MERRA significantly strengthened throughout the layer 100-10 hPa with a rate of 2-5% decade<sup>-1</sup>, while the BDC in ERA-Interim shows weakening trends. Diallo et al. (2021) compared the trends of the BDC in the ERA5 and ERA-Interim during 1979-2018 and pointed out that the BDC in the ERA-Interim shows weakening trend and the BDC in the ERA5 strengthened at a rate of 1.5% decade-1 which is more consistent with other studies. In the present study, we only focus on the trend of the BDC in the wintertime (NDJFM) in the tropics (15°S-15°N) during 1958-2017, which may lead to some differences between our result and that in the previous studies. Overall, the trends of the tropical upwelling of BDC derived from JRA55, MERRA2

586

587

588

589

590

591

592

593

594

595

596

597

598

599

600

601

602

603

604

605

606

607

data and the Control simulation are similar to that in previous studies using both reanalysis datasets and model results (e.g., Butchart et al., 2010; Abalos et al., 2015; Fu et al., 2019; Rao et al., 2019; Diallo et al., 2021). However, the tropical upwelling of the BDC decreased in ERA5 data in the tropics (15°S-15°N), which are different from the results in JRA55 and MERRA2.

In the Fixsst simulation, the trend of w\* is much weaker and not significant in most areas. The changes in the global SSTs therefore play an important role in the intensification of the shallow branch of the BDC as shown by the differences between the two simulations in Fig. 12d. In summary, the tropical upwelling of the BDC is likely strengthened as shown in JRA55 and MERRA2 reanalyses as well as model simulations, although there are some uncertainties since the ERA5 data show a negative trend. This may impact on the transport of the tropospheric trace gases from the TTL to a higher altitude. The increased concentration of CO in the UTLS in Fig. 9c and 10f may be due to a combined effect of the strengthened tropical upwelling of the BD circulation and the enhanced upward motion over the TWP. The enhancement of upward motion over the TWP, which transported more tropospheric trace gases to the upper troposphere, works together with the strengthened BD circulation under global warming may lead to an increase of tropospheric trace gases over the TWP in the lower stratosphere.

#### 4 Summary and Discussion

The recent trends of the upward motion from the lower to the upper troposphere in boreal winter over the TWP is investigated for the first time based on the JRA55,

ERA5, MERRA2 datasets and four WACCM4 simulations (more details could be found in Section 2). The upward motion at 150 hPa over the TWP in NDJFM increased 8±3.1% decade<sup>-1</sup> and 3.6±3.3% decade<sup>-1</sup> in NDJFM from 1958 to 2017 in and ERA5 reanalysis datasets, respectively. Despite the possible discontinuities between the radiosonde era (after 1958) and the satellite era (after 1979), the upward motion at 150 hPa over the TWP in NDJFM increased 7.5±7.1% decade-1 during 1980-2017 in MERRA2 data. Such intensification of the upward motion over the TWP also exist in the middle- and lower-troposphere in NDJFM in JRA55, ERA5, and MERRA2, which can be confirmed by the WACCM4 model simulations. Comparing the results between the Control and Fixsst simulations with WACCM4, it is found that the trend of the upward motion over the TWP is closely related to the changes in global SSTs, especially the SST warming over the eastern maritime continent and tropical western Pacific (see the results from the experiments R1 and R2 in Fig. 7). Warmer SSTs over the eastern maritime continent and tropical western Pacific (approximately 0.5 K) lead to a strengthened Pacific Walker circulation, enhanced deep convection and approximately 27% intensified upward motion at 150 hPa over the TWP as shown by the results from the experiments R1 and R2. The enhanced deep convection over the TWP could lead to a dryer lower stratosphere over the TWP, as the strong upward motion and the Rossby-Kelvin wave responses induce a colder tropopause over the TWP. It should be pointed out that the results in the present study are mainly based on the reanalyses data, and some uncertainties may exist. More observational data are expected to be used to obtain a

630

631

632

633

634

635

636

637

638

639

640

641

642

643

644

645

646

647

648

649

650

651

more robust result in the future.

Results from the Control simulation indicate that the CO concentrations increased significantly from the surface to the stratosphere over the TWP. The CO at 150 hPa increased at a rate of approximately 3.4 ppbv decade-1 with increased surface emissions and the enhanced upward motion over the TWP. Specifically, an enhancement of tropospheric upward motion and subsequent upward transport of trace gases over the TWP lead to an extra 6% increasing trend of CO concentrations in the upper troposphere.

Furthermore, the upward mass fluxes at 70 hPa in the tropics (15°S-15°N) show strengthening trends at rates of 2.8±1.9% decade<sup>-1</sup> and 4.6±4.3% decade<sup>-1</sup> in JRA55 data (during 1958-2017) and MERRA2 data (during 1980-2017) in NDJFM, respectively, which is consistent with previous studies (e.g., Butchart et al., 2010; Fu et al., 2019; Rao et al., 2019). However, such enhancement in tropical upward mass flux at 70 hPa has large uncertainties since the ERA5 data show a negative and insignificant trend (Supplementary Fig. 7a). The results from the Control and Fixsst simulations indicate that the elevated CO in the upper troposphere is further uplifted to the lower stratosphere by the intensified tropical upwelling of the BD circulation due mainly to global SST warming and lead to an increase of CO in the lower stratosphere. An extra 14% increasing trend of CO at the layer 150-70 hPa over the TWP is derived from the Control and Fixsst simulations.

Tropospheric trace gases and aerosols have important impacts on the stratospheric processes if they enter the stratosphere. For example, ozone-depleting

substances, CH<sub>4</sub> and N<sub>2</sub>O could influence on the stratospheric ozone significantly (e.g., Shindell et al., 2013; Wang et al., 2014; WMO, 2018), which also modify the temperature in the stratosphere significantly through their strong radiative effects. Water vapor in the lower stratosphere, in particular, has a significant warming effect on the surface climate (Solomon et al., 2010). Therefore, changes of trace gases in the UTLS have important impacts on both tropospheric and stratospheric climate. Our results suggest that the upward motion over the TWP and the vertical component of the BDC at the lower stratosphere level have been intensified. These results suggest that the emission from the maritime continent and surrounding areas may play a more important role in the stratospheric processes and the global climate. In addition, more very short lived substances emitted from the tropical ocean could be elevated to the TTL by the enhanced convection and then transported into the stratosphere by the large-scale uplifts and exert important effects on the stratospheric chemistry. However, the quantitative impacts of the intensified upward motion over the TWP on tropospheric and stratospheric trace gases and aerosols and their climate feedbacks await further investigation using more observations and model simulations.

690

674

675

676

677

678

679

680

681

682

683

684

685

686

687

688

689

**Competing interests.** The authors declare that they have no conflict of interest.

692

693

694

695

691

**Author contributions.** WT designed the study. WW provided suggestions about the statistical methods and model simulations. KQ ran the models and wrote the first draft. RH, MX, and TW contributed to the manuscript writing. YP provided the data used in

- the study. All authors contributed to the improvement of the results.
- 697
- 698
- 699 Acknowledgements. This research is supported by Strategic Priority Research
- 700 Program of Chinese Academy of Sciences (XDA17010106), the National Natural
- Science Foundation of China (42075055) and the Supercomputing Center of Lanzhou
- 702 University.
- The authors gratefully acknowledge the data used in the present study provided by the
- 704 corresponding scientific groups. The JRA55 data are from:
- 705 http://rda.ucar.edu/datasets/ds628.0/.
- 706 The SST data is obtained from HadISST:
- 707 https://www.metoffice.gov.uk/hadobs/hadisst/data/download.html.
- 708 The ERA5 data and ERA5.1 data are extracted from:
- 709 https://cds.climate.copernicus.eu/#!/search?text=ERA5&type=dataset.
- 710 The MERRA2 data are downloaded from:
- 711 https://search.earthdata.nasa.gov/search?q=MERRA2&fst0=Atmosphere.
- 712 The OLR data are from https://psl.noaa.gov/data/gridded/data.interp OLR.html.
- 713
- 714 **References:**
- Abalos, M., Legras, B., Ploeger, F., and Randel, W. J.: Evaluating the advective
- Brewer-Dobson circulation in three reanalyses for the period 1979-2012, J.
- 717 Geophys. Res., 120,7534-7554, doi:10.1002/2015JD023182, 2015.
- Andrews, D. G., and Mcintyre, M. E.: Planetary waves in horizontal and vertical shear:
- 719 the generalized eliassen-palm relation and the mean zonal acceleration, J. Atmos.
- 720 Sci., 33, 2031 2048, 1976
- Ashfold, M. J., Pyle, J. A., Robinson, A. D., Meneguz, E., Nadzir, M. S. M., Phang, S.
- M., Samah, A. A., Ong, S., Ung, H. E., Peng, L. K., Yong, S. E., and Harris, N. R.

- P.: Rapid transport of East Asian pollution to the deep tropics, Atmos. Chem.
- 724 Phys., 15, 3565-3573, doi:10.5194/acp-15-3565-2015, 2015.
- Bergman, J. W., Jensen, E. J., Pfister, L., and Yang, Q.: Seasonal differences of
- vertical-transport efficiency in the tropical tropopause layer: On the interplay
- between tropical deep convection, large-scale vertical ascent, and horizontal
- 728 circulations, J. Geophys. Res., 117, D05302, doi:10.1029/2011JD016992, 2012.
- Brewer, A. M.: Evidence for a world circulation provides by the measurements of
- helium and water vapor distribution in the stratosphere, Q. J. R. Meteorol. Soc.,
- 731 75, 351–363, 1949.
- Butchart, N., Cionni, I., Eyring, V., Shepherd, T. G., Waugh, D. W., Akiyoshi, H.,
- Austin, J., Brühl, C., Chipperfield, M. P., Cordero, E., Dameris, M., Deckert, R.,
- Dhomse, S., Frith, S. M., Garcia, R. R., Gettelman, A., Giorgetta, M. A.,
- Kinnison, D. E., Li, F., Mancini, E., McLandress, C., Pawson, S., Pitari, G.,
- Plummer, D. A., Rozanov, E., Sassi, F., Scinocca, J. F., Shibata, K., Steil, B., and
- 737 Tian, W.: Chemistry-Climate Model simulations of twenty-first century
- stratospheric climate and circulation changes, J. Climate, 23, 5349–5374,
- 739 https://doi.org/10.1175/2010JCLI3404.1, 2010.
- Cane, M. A., Clement, A. C., Kaplan, A., Kushnir, Y., Pozdnyakov, D., and Seager, R.,
- et al.: Twentieth-century sea surface temperature trends, Science, 275(5302),
- 742 957-960, DOI: 10.1126/science.275.5302.957, 1997.
- Deeter, M. N., Edwards, D. P., Francis, G. L., Gille, J. C., Mao, D., Martinez-Alonso,
- S., Worden, H. M., Ziskin, D., and Andreae, M. O.: Radiance-based retrieval bias
- mitigation for the MOPITT instrument: the version 8 product, Atmos. Meas.
- 746 Tech., 12, 4561–4580, https://doi.org/10.5194/amt-12-4561-2019, 2019.
- Deser, C., Phillips, A. S., and Alexander, M. A.: Twentieth century tropical sea surface
- temperature trends revisited, Geophys. Res. Lett., 37(10), L10701,
- 749 doi:10.1029/2010GL043321, 2010.
- Diallo, M., Ern, M., and Ploeger, F.: The advective Brewer Dobson circulation in
- 751 the ERA5 reanalysis: climatology, variability, and trends, Atmos. Chem. Phys.,

- 752 21, 7515 7544, https://doi.org/10.5194/acp-21-7515-2021, 2021.
- 753 Dobson G. M. B.: Origin and distribution of the polyatomic molecules in the
- 754 atmosphere, Proc. R. Soc. Lond. Ser. A. 236: 187-193, 1956.
- Duncan, B. N., Logan J. A., Bey, I., Megretskaia, I. A., Yantosca, R. M., Novelli, P. C.,
- Jones, N. B. and Rinsland, C. P.: Global budget of CO, 1988-1997: Source
- estimates and validation with a global model, J. Geophys. Res., 112, D22301,
- 758 doi:10.1029/2007JD008459, 2007.
- Feng, W., Chipperfifield, M. P., Dorf, M., Pfeilsticker, K., and Ricaud, P.: Mid-latitude
- ozone changes: studies with a 3-D CTM forced by ERA-40 analyses, Atmos.
- 761 Chem. Phys., 7, 2357 2369, doi:10.5194/acp-7-2357-2007, 2007.
- 762 Fu, Q., Solomon, S., Pahlavan, H. A., and Lin, P.: Observed changes in
- Brewer–Dobson circulation for 1980–2018, Environ. Res. Lett., 14, 114 026,
- 764 https://doi.org/10.1088/1748-9326/ab4de7, 2019.
- Fueglistaler, S., Desller, A. E., Dunkerton, T. J., Folkins, I., Fu, Q., and Mote P.:
- 766 Tropical tropopause layer, Rev. Geophys., 47(1), RG1004,
- 767 doi:10.1029/2008RG000267, 2009.
- Fueglistaler, S., Wernli, H., and Peter, T.: Tropical troposphere-to-stratosphere
- transport inferred from trajectory calculations, J. Geophys. Res., 109, D03108,
- 770 doi:10.1029/2003JD004069, 2004.
- Fujiwara, M., Wright, J. S., Manney, G. L., Gray, L. J., Anstey, J., Birner, T., Davis, S.,
- Gerber, E. P., Harvey, V. L., Hegglin, M. I., Homeyer, C. R., Knox, J. A., Krüger,
- K., Lambert, A., Long, C. S., Martineau, P., Molod, A., Monge-Sanz, B. M.,
- Santee, M. L., Tegtmeier, S., Chabrillat, S., Tan, D. G. H., Jackson, D. R.,
- Polavarapu, S., Compo, G. P., Dragani, R., Ebisuzaki, W., Harada, Y., Kobayashi,
- C., McCarty, W., Onogi, K., Pawson, S., Simmons, A., Wargan, K., Whitaker, J.
- 777 S., and Zou, C.-Z.: Introduction to the SPARC Reanalysis Intercomparison
- Project (S-RIP) and overview of the reanalysis systems, Atmos. Chem. Phys., 17,
- 779 1417-1452, https://doi.org/10.5194/acp-17-1417-2017, 2017
- Garfinkel, C. I., Waugh, D. W., and Polvani, L. M.: Recent Hadley cell expansion:
- The role of internal atmospheric variability in reconciling modeled and observed

- 782 trends, Geophys. Res. Lett., 42(24), 10824-10831,
- 783 https://doi.org/10.1002/2015GL066942, 2015
- Garfinkel, C. I., Waugh, D. W., Oman, L. D., Wang, L., and Hurwitz, M. M.:
- 785 Temperature trends in the tropical upper troposphere and lower stratosphere:
- Connections with sea surface temperatures and implications for water vapor and
- 787 ozone, J. Geophys. Res., 118, 9658-9672, doi:10.1002/jgrd.50772, 2013.
- Garfinkel, C. O., Gordon, A., Oman, L. D., Li, F., Davis, S., and Pawson, S.:
- Nonlinear response of tropical lower-stratospheric temperature and water vapor
- 790 to ENSO, Atmos. Chem. Phys., 18, 4597-4615,
- 791 https://doi.org/10.5194/acp-18-4597-2018, 2018.
- 792 Gelaro, R., Mccarty, W., and Suárez, M. J.: The modern-era retrospective analysis for
- research and applications, version 2 (merra-2), J. Climate, 30(14), 5419-5454,
- 794 https://doi.org/10.1175/JCLI-D-16-0758.1, 2017
- 795 Gettelman, A., Holton, J. R., and Douglass, A. R.: Simulations of water vapor in the
- lower stratosphere and upper troposphere, J. Geophys. Res., 105(D7), 9003-9023,
- 797 https://doi.org/10.1029/1999JD901133, 2000.
- 798 Gill, A. E.: Some simple solutions for heat-induced tropical circulation, Q. J. R.
- 799 Meteorolog. Soc., 106, 447-462, doi:10.1002/qj.49710644905, 1980.
- 800 Girach, I. A., and Nair, P. R.: Carbon monoxide over Indian region as observed by
- 801 MOPITT, Atmos. Environ., 99, 599-609,
- http://dx.doi.org/10.1016/j.atmosenv.2014.10.019, 2014.
- Haines, P. E., and Esler, J. G.: Determination of the source regions for surface to
- stratosphere transport: An Eulerian backtracking approach, Geophys. Res. Lett.,
- 41, 1343-1349, doi:10.1002/2013GL058757, 2014.
- Harada, Y., Kamahori, H., Kobayashi, C., Endo, H., Kobayashi, S., Ota, Y., Onoda, H.,
- Onogi, K., Miyaoka, K., and Takahashi, K.: The JRA-55 Reanalysis:
- Representation of atmospheric circulation and climate variability, J. Meteorol.
- 809 Soc. Jpn., 94(3), 269-302, https://doi.org/10.2151/jmsj.2016-015, 2016.
- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J.,
- and Nicolas, J., et al.: The ERA5 global reanalysis, Q. J. R. Meteorol. Soc., 146,

- 812 1999-2049, DOI: 10.1002/qj.3803, 2020.
- Highwood, E. J., and Hoskins, B. J.: The tropical tropopause, Q. J. R. Meteorol. Soc.,
- 814 124(549), 1579-1604, DOI: 10.1002/qj.49712454911, 1998.
- Hitchcock, P.: On the value of reanalysis prior to 1979 for dynamical studies of
- stratosphere-troposphere coupling, Atmos. Chem. Phys., 19, 2749-2764,
- https://doi.org/10.5194/acp-19-2749-2019, 2019
- Holton, J. R., Haynes, P. H., McIntyre, M. E., Douglass, A. R., Rood, R. B., and
- Pfister, L.: Stratosphere-troposphere exchange, Rev. Geophys, 33,
- 403-439, https://doi.org/10.1029/95RG02097, 1995.
- Hosking, J. S., Russo, M. R., Braesicke, P., and Pyle, J. A.: Tropical convective
- transport and the Walker circulation, Atmos. Chem. Phys., 12, 9791-9797,
- 823 doi:10.5194/acp-12-9791-2012, 2012
- Hu, D., Guan, Z., Tian, W., and Ren, R.: Recent strengthening of the stratospheric
- Arctic vortex response to warming in the central North Pacific, 9, 1697,
- https://doi.org/10.1038/s41467-018-04138-3, 2018.
- 827 Hu, D., Guo, Y., Wang, F., Xu, Q., Li, Y., Sang, W., Wang, X., and Liu, M.:
- Brewer-Dobson circulation: Recent-Past and near-future trends simulated by
- chemistry-climate models, Adv. Meteorol., 2017, 1-13,
- https://doi.org/10.1155/2017/2913895, 2017.
- Hu, D., Tian, W., Guan, Z., Guo, Y., and Dhomse, S.: Longitudinal asymmetric trends
- of tropical cold-point tropopause temperature and their link to strengthened
- 833 Walker circulation, J. Climate, 29(21), 7755–7771,
- https://doi.org/10.1175/JCLI-D-15-0851.1, 2016.
- Hu, P., Huangfu, J., Chen, W., and Huang, R.: Impacts of early/late South China Sea
- summer monsoon withdrawal on tropical cyclone genesis over the western North
- 837 Pacific, Clim. Dynam., 55(19), 1507-1520,
- https://doi.org/10.1007/s00382-020-05339-7, 2020.
- Huang, L., Jiang, J. H., Murray, L. T., Damon, M. R., Su, H., and Livesey, N.:
- 840 Evaluation of UTLS carbon monoxide simulations in GMI and GEOS-Chem
- chemical transport models using Aura MLS observations, Atmos. Chem. Phys.,

- 842 16, 5641-5663, doi:10.5194/acp-16-5641-2016, 2016.
- Kobayashi, S., Ota, Y., Harada, Y., Ebita, A., Moriya, M., and Onoda, H., et al.: The
- JRA-55 Reanalysis: General specifications and basic characteristics, J. Meteorol.
- 845 Soc. Jpn., 93, 5-48, DOI:10.2151/jmsj.2015-001, 2015.
- Krüger, K., Tegtmeier, S., and Rex, M.: Long-term climatology of air mass transport
- through the Tropical Tropopause Layer (TTL) during NH winter, Atmos. Chem.
- Phys., 8, 813–823, doi:10.5194/acpd-7-13989-2007, 2008.
- L'Heureux, M. L., Lee, S., and Lyon, B.: Recent multidecadal strengthening of the
- Walker circulation across the tropical Pacific, Nat. Clim. Change, 3, 571–576,
- doi: 10.1038/nclimate1840, 2013.
- Levine, J. G., Braesicke, P., Harris, N. R. P., Pyle, J. A.: Seasonal and inter-annual
- variations in troposphere-to-stratosphere transport from the tropical tropopause
- layer, Atmos. Chem. Phys., 8, 3689-3703, DOI:10.5194/acpd-8-489-2008, 2008.
- Levine, J. G., Braesicke, P., Harris, N. R. P., Savage, N. H., and Pyle, J. A.: Pathways
- and timescales for troposphere-to-stratosphere transport via the tropical
- tropopause layer and their relevance for very short lived substances, J. Geophys.
- 858 Res., 112, D04308, doi:10.1029/2005JD006940, 2007.
- 859 Liebmann, B., and Smith, C. A.: Description of a complete (interpolated) outgoing
- longwave radiation dataset, B. Am. Meteorol. Soc., 77, 1275-1277, doi:
- 861 10.1175/1520-0477(1996)077<1255:EA>2.0.CO;2, 1996.
- Livesey, N. J., Logan, J. A., Santee, M. L., Waters, J. W., Doherty, R. M., Read, W. G.,
- Froidevaux, L., and Jiang, J. H.: Interrelated variatinos of O3, CO and deep
- convection in the tropical/subtropical upper troposphere observed by the Aura
- Microwave Lim Sounder (MLS) during 2004-2011, Atmos. Chem. Phys, 13,
- 866 579-598, doi:10.5194/acp-13-579-2013, 2013.
- Livesey, N. J., Read, W. G., Wagner, P. A., Froidevaux, L., Lambert, A., Manney, G. L.,
- Millán, L., Pumphrey, H. C., Santee, M. L., Schwartz, M. J., Wang, S., Fuller, R.
- A., Jarnot, R. F., Knosp, B. W., and Martinez, E.: EOS MLS Version 4.2x Level
- 2 data quality and description document, Jet Propulsion Laboratory, California
- Institute of Technology, Pasadena, CA, 2015.

- Lin, P., Ming, Y., and Ramaswamy, V.: Tropical climate change control of the lower
- stratospheric circulation, Geophys. Res. Lett., 42(3), 941-948, doi:
- 874 10.1002/2014GL062823, 2015.
- Logan, J. A., Megretskaia, I. A., Nassar, R., Murray, L. T., Zhang, L., Bowman, K. W.,
- Worden, H. M., and Luo, M.: Effects of the 2006 El Niño on tropospheric
- composition as revealed by data from the Tropospheric Emission Spectrometer
- 878 (TES), Geophys. Res. Lett., 35, L03816,
- https://doi.org/10.1029/2007GL031698, 2008.
- Long, C. S., Fujiwara, M., Davis, S., Mitchell, D. M., and Wright, C. J.: Climatology
- and interannual variability of dynamic variables in multiply reanalyses evaluated
- by the SPARC Reanalysis Intercomparison Project (S-RIP), Atmos. Chem. Phys.,
- 17, 14593-14629, https://doi.org/10.5194/acp-17-14593-2017, 2017.
- Lu, J., Vecchi, G. A., and Reichler, T.: Expansion of the Hadley cell under global
- warming, Geophys. Res. Lett., 34(6), L06805, doi:10.1029/2006GL028443,
- 886 2007
- Lu, J., Xie, F., Sun, C., Luo, J., Cai, Q., Zhang, J., Li, J., and Tian, H.: Analysis of
- factors influencing tropical lower stratospheric water vapor during 1980–2017,
- npj Clim. Atmos. Sci., 3(1), 35, https://doi.org/10.1038/s41612-020-00138-7,
- 890 2020.
- 891 Ma, S., and Zhou, T.: Robust Strengthening and Westward Shift of the Tropical
- Pacific Walker Circulation during 1979–2012: A Comparison of 7 Sets of
- Reanalysis Data and 26 CMIP5 Models, J. Climate, 29, 3097–3118, doi:
- 894 10.1175/JCLI-D-15-0398.1, 2016.
- Marsh, D. R., Mills, M. J., Kinnison, D. E., Lamarque, J., Calvo, N., and Polvani, L.
- M.: Climate change from 1850 to 2005 simulated in CESM1 (WACCM), J.
- 897 Climate, 26, 7372-7391, https://doi.org/10.1175/JCLI-D-12-00558.1, 2013.
- McGregor, S., Timmermann, A., Stuecker, M. F., England, M. H., Merrifield, M., Jin,
- F., and Chikamoto, Y.: Recent Walker circulation strengthening and Pacifific
- cooling amplifified by Atlantic warming, Nat. Clim. Change, 4, 888–882,
- 901 https://doi.org/10.1038/nclimate2330, 2014.

- 902 Meng, Q., Latif, M., Park, W., Keenlyside, N. S., Semenov, V. A., and Martin, T.:
- Twentieth century Walker circulation change: Data analysis and model
- 904 experiments, Clim. Dynam., 38, 1757–1773,
- 905 https://doi.org/10.1007/s00382-011-1047-8, 2012.
- 906 Minganti, D., Chabrillat, S., Christophe, Y., Errera, Q., Abalos, M., Prignon, M.,
- Kinnison, D. E., and Mahieu, E.: Climatological impact of the Brewer–Dobson
- or circulation on the N2O budget in WACCM, a chemical reanalysis and a CTM
- driven by four dynamical reanalyses, Atmos. Chem. Phys., 20, 12609–12631,
- 910 https://doi.org/10.5194/acp-20-12609-2020, 2020.
- Navarro, M. A., Atlas, E. L., Saiz-Lopez, A., Rodriguez-Lloveras, X., Kinnison, D. E.,
- Lamarque, J., Tilmes, S., Filus, M., and Harris, N. R. P., et al.: Airborne
- measurements of organic bromine compounds in the Pacific tropical tropopause
- layer, P. Natl. Acad. Sci. USA, 112, 13789-13793, doi:10.1073/pnas.1511463112,
- 915 2015.
- Newell, R. E., and Gould-Stewart, S.: A stratospheric fountain? J. Atmos. Sci., 38,
- 917 2789–2796, DOI: 10.1175/1520-0469(1981)038<2789:ASF>2.0.CO;2, 1981.
- Newton, R., Vaughan, G., Hintsa, E., Filus, M. T., Pan, L. L., and Honomichl, S., et al.:
- Observations of ozone-poor air in the tropical tropopause layer, Atmos. Chem.
- 920 Phys., 18, 5157–5171, https://doi.org/10.5194/acp-18-5157-2018, 2018.
- Pan, L. L., Atlas, E. L., Salawitch, R. J., Honomichl, S. B., Bresch, J. F., and Randel,
- W. J., et al.: The Convective Transport of Active Species in the Tropics
- 923 (CONTRAST) Experiment, B. Am. Meteorol. Soc., 98(1), 106-128,
- 924 DOI: 10.1175/BAMS-D-14-00272.1, 2016.
- Pan, L. L., Honomichl, S. B., Thornberry, T., Rollins, A., Bui, T. P., Pfister, L., and
- Jensen E. E.: Observational Evidence of Horizontal Transport-Driven
- Dehydration in the TTL, Geophys. Res. Lett., 46(13), 7848-7856,
- 928 DOI: 10.1029/2019GL083647, 2019.
- 929 Qie, K., Qie, X., and Tian, W.: Increasing trend of lightning activity in the South Asia
- 930 region, Sci. Bull., 66, 78-84, https://doi.org/10.1016/j.scib.2020.08.033, 2021.
- Randel, W. J., and Jensen, E. J.: Physical processes in the tropical tropopause layer

- and their roles in a changing climate, Nat. Geosci., 6, 169–176,
- 933 DOI:10.1038/ngeo1733, 2013.
- Randel, W. J., Park, M., Emmons, L., Kinnison, D., Bernath, P., Walker, K. A., Boone,
- 935 C., and Pumphrey. H., Asian monsoon transport of pollution to the Stratosphere,
- 936 Science, 328, 611-613, DOI: 10.1126/science.1182274, 2010.
- Rao, J., Yu, Y., Guo, D., Shi, C., Chen, D., and Hu, D.: Evaluating the Brewer-Dobson
- circulation and its responses to ENSO, QBO, and the solar cycle in different
- 939 reanalyses, Earth Planet. Phys., 3(2), 1-16, http://doi.org/10.26464/epp2019012,
- 940 2019.
- Rayner, N., Parker, D., Horton, E., Folland, C. K., Alexander, L., and Rowell, D. P., et
- al.: Global analyses of sea surface temperature, sea ice, and night marine air
- temperature since the late nineteenth century, J. Geophys. Res., 108(D14), 4407,
- 944 doi:10.1029/2002JD002670, 2003.
- Rosenlof, K. H. How water enters the stratosphere, Science, 302, 1691-1692,
- 946 doi:10.1126/science.1092703, 2003.
- 947 Ryu, J., and Lee, S.: Effect of tropical waves on the tropical tropopause transition
- 948 layer upwelling, J. Atmos. Sci., 67(10), 3130-3148,
- 949 DOI: 10.1175/2010JAS3434.1, 2010.
- 950 Saiz-Lopez, A., and von Glasow, R.: Reactive halogen chemistry in the troposphere,
- 951 Chem. Soc. Rev., 41, 6448–6472, DOI:10.1039/c2cs35208g, 2012.
- Schoeberl, M. R., Jensen, E. J., Pfister, L., Ueyama, R., Avery, M., and Dessler, A. E.:
- Convective hydration of the upper troposphere and lower stratosphere, J.
- 954 Geophys. Res., 123(9), 4583-4593, https://doi.org/10.1029/2018JD028286, 2018.
- Sherwood, S. C.: A stratospheric "drain" over the maritime continent, Geophys. Res.
- 956 Lett., 27(5), 677-680, https://doi.org/10.1029/1999GL010868, 2000.
- 957 Shindell, D. T., Pechony, O., Voulgarakis, A., Faluvegi, G., Nazarenko, L., Lamarque,
- J.-F., Bowman, K., Milly, G., Kovari, B., Ruedy, R., and Schmidt, G. A.:
- Interactive ozone and methane chemistry in GISS-E2 historical future climate
- simulations, Atmos. Chem. Phys., 13, 2653-2689,
- 961 doi:10.5194/acp-13-2653-2013, 2013.

- Shirley, D., Stanley, W., & Daniel, C.: Statistics for Research (Third Edition), (p. 627),
- Hoboken, New Jersey: John Wiley & Sons Inc., 2004.
- Simmons, A. J., Poli, P., Dee, D. P., Berrisford, P., Hersbach, H., Kobayashi, S., and
- Peubey, C.: Estimating lowfrequency variability and trends in atmospheric
- 966 temperature using ERA-Interim, Q. J. Roy. Meteorol. Soc., 140, 329 353,
- 967 https://doi.org/10.1002/qj.2317, 2014.
- Sinnhuber, B.-M., Sheode, N., Sinnhuber, M., Chipperfield, M. P., and Feng, W.: The
- ontribution of anthropogenic bromine emis sions to past stratospheric ozone
- 970 trends: a modelling study, Atmos. Chem. Phys., 9, 2863 2871,
- 971 doi:10.5194/acp-9-2863-2009, 2009.
- 972 Solomon, S., Rosenlof, K. H., Portmann, R. W., Daniel, J. S., Davis, S. M., Sanford, T.
- J., and Plattner, G.-K.: Contributions of stratospheric water vapor to decadal
- changes in the rate of global warming, Science, 327, 1219-1223,
- 975 DOI:10.1126/science.1182488, 2010.
- 976 Tegtmeier, S., Anstey, J., Davis, S., Dragani, R., Harada, Y., Ivanciu, I., Kedzierski, R.
- P., Krüger, K., Legras, B., Long, C., Wang, J. S., Wargan, K., and Wright, J. S.:
- Temperature and tropopause characteristics from reanalyses data in the tropical
- 979 tropopause layer, Atmos. Chem. Phys., 20, 753-770,
- 980 https://doi.org/10.5194/acp-20-753-2020, 2020.
- Tokinaga, H., Xie, S. P., Deser, C., Kosaka, Y., and Okumura, Y. M.: Slowdown of the
- Walker circulation driven by tropical Indo-Pacifific warming. Nature, 491,
- 983 439–443, DOI: 10.1038/nature11576, 2012.
- Uma, K. N., Das, S. S., Ratnam, M. V., and Suneeth, K. V.: Assessment of vertical air
- 985 motion among reanalyses and qualitative comparison with very-high-frequency
- radar measurements over two tropical stations, Atmos. Chem. Phys., 21,
- 987 2083-2103, https://doi.org/10.5194/acp-21-2083-2021, 2021.
- Wales, P. A., Salawitch, R. J., Nicely, J. M., Anderson, D. C., Canty, T. P. and Baidar,
- S., et al.: Stratospheric injection of Brominated very short-lived substances:
- Aircraft observations in the western Pacific and representation in global models,
- 991 J. Geophys. Res., 123(10), 5690-5719, https://doi.org/10.1029/2017JD027978,

- 992 2017.
- Wang, S., Schmidt, J. A., Baidar, S., Coburn, S., Dix, B., and Koenig, T. K., et al.:
- Active and wide-spread halogen chemistry in the tropical and subtropical free
- 995 troposphere, P. Natl. Acad. Sci. USA, 112, 9281–9286,
- 996 DOI: 10.1073/pnas.1505142112, 2015.
- 997 Wang, W., Matthes, K., and Schmidt, T.: Quantifying contributions to the recent
- temperature variability in the tropical tropopause layer, Atmos. Chem. Phys., 15,
- 999 5815–5826, doi:10.5194/acp-15-5815-2015, 2015.
- 1000 Wang, W., Matthes, K., Omarani, N.-E., and Latif, M.: Decadal variability of tropical
- tropopause temperature and its relationship to the Pacific Decadal Oscillation,
- Sci. Rep., 6, 29537, https://doi.org/10.1038/srep29537, 2016.
- Wang, W., Tian, W., Dhomse, S., Xie, F., Shu, J., and Austin, J.: Stratospheric ozone
- depletion from future nitrous oxide increases, Atmos. Chem. Phys., 14,
- 1005 12967-12982, doi:10.5194/acp-14-12967-2014, 2014.
- 1006 Webster, P. J., Lukas, R.: TOGA COARE: the coupled ocean-atmosphere response
- 1007 experiment, B. Am. Meteorol. Soc., 73(9), 1377-1416,
- 1008 DOI: 10.1175/1520-0477(1992)073<1377:TCTCOR>2.0.CO;2, 1992.
- 1009 Webster, P. J., Clayson, C. A., Curry, J. A.: Clouds, radiation, and the diurnal cycle of
- sea surface temperature in the tropical western Pacific, J. Climate, 8(8),
- 1011 1712-1730, DOI: 10.1175/1520-0442(1996)009<1712:CRATDC>2.0.CO;2,
- 1012 1996.
- 1013 WMO.: Low-cost Sensors for the Measurement of Atmospheric Composition:
- Overview of Topic and Future Applications, 2018.
- 1015 Xie, F., Li, J., Tian, W., Feng, J., and Huo, Y.: Signals of El Niño Modoki in the
- tropical tropopause layer and stratosphere, Atmos. Chem. Phys., 12, 5259-5273,
- doi:10.5194/acp-12-5259-2012, 2012.
- 1018 Xie, F., Li, J., Tian, W., Li, Y., and Feng, J.: Indo-Pacific Warm Pool Area Expansion,
- Modoki Activity, and Tropical Cold-Point Tropopause Temperature Variations,
- 1020 Sci. Rep., 4, 4552, https://doi.org/10.1038/srep04552, 2014a.
- 1021 Xie, F., Li, J., Tian, W., Zhang, J., and Shu, J.: The impacts of two types of El Niño on

- global ozone variations in the last three decades, Adv. Atmos. Sci., 31(5),
- 1023 1113-1126, doi: 10.1007/s00376-013-3166-0, 2014b.
- 1024 Xie, F., Tian, W., Zhou, X., Zhang, J., Xia, Y., and Lu, J.: Increase in lower
- stratospheric water vapor in the past 100 years related to tropical Atlantic
- warming, Geophys. Res. Lett., 47, e2020GL090539,
- DOI: 10.1029/2020GL090539, 2020.
- Zhou, X., Chen, Q., Li, Y., Zhao, Y., Lin, Y., and Jiang, Y.: Impacts of the Indo-Pacific
- warm pool on lower stratospheric water vapor: Seasonality and hemispheric
- 1030 contrasts, J. Geophys. Res., 126, e2020JD034363, DOI: 10.1029/2020JD034363,
- 1031 2021.

## 1033 Figure captions:

- Fig. 1. The climatological mean (averaged over 1958-2017) values of 150 hPa w (10<sup>-2</sup>)
- 1035 m s<sup>-1</sup>) in different months derived from the JRA55 data.
- Fig. 2. Trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, 700 hPa
- in NDJFM derived from JRA55, ERA5, and MERRA2 data. The trends of horizontal
- winds (arrows, units: 10<sup>-1</sup> m s<sup>-1</sup> a<sup>-1</sup>) and vertical velocity (shading, units: 10<sup>-4</sup> m s<sup>-1</sup> a<sup>-1</sup>)
- 1039 at (a) 150 hPa; (d) 500 hPa; and (g) 700 hPa from JRA55 in NDJFM during
- 1040 1958-2017. (b), (e), and (h) are the same as (a), (d), and (g) but for the results from
- 1041 ERA5. (c), (f) and (i) are the same as (a), (d), and (g) except that the trends are during
- 1042 1980-2017 and the wind field data are from MERRA2. The vertical velocity trends
- over the dotted regions are statistically significant at the 95% confidence level. The
- 1044 white areas denote missing values. The black rectangles denote the TWP region
- 1045 (20°S-10°N, 100°E-180°E).
- 1046 Fig. 3. The time series of the standardized intensity of the upward motion over the
- tropical western Pacific (20°S-10°N, 100°E-180°E) at (a) 150 hPa; (b) 500 hPa; and

1048 (c) 700 hPa extracted from JRA55 (red), ERA5 (black) and MERRA2 (blue) datasets. The straight lines in each figure indicate the linear trends. The solid lines denote the 1049 linear trends are significant at the 95% confidence level, while the dashed lines denote 1050 the linear trends are significant at the 90% confidence level. 1051 Fig. 4. Trends of (a) observed outgoing longwave radiation (OLR, units: W m<sup>-2</sup> a<sup>-1</sup>) 1052 1053 provided by NOAA during 1974-2017; (b) cold-point tropopause temperature (CPTT, units: 10<sup>-1</sup> K a<sup>-1</sup>) derived from JRA55 data and (c) SST (K a<sup>-1</sup>) derived from HadISST 1054 during 1958-2017 in NDJFM. (d) The correlation coefficients between the intensity of 1055 the upward motion at 150 hPa over the TWP and SST in NDJFM during 1958-2017 1056 with the linear trends removed. The trends and correlation coefficients over the dotted 1057 regions are statistically significant at the 95% confidence level. The black rectangles 1058 1059 denote the TWP region (20°S-10°N, 100°E-180°E). Fig. 5. The trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, and 1060 700 hPa in NDJFM during 1958-2017 in the Control and Fixsst simulations as well as 1061 their difference. The trends of 150 hPa w (shading, units:  $10^{-4}$  m s<sup>-1</sup> a<sup>-1</sup>) and horizontal 1062 winds (arrows; 10<sup>-1</sup> m s<sup>-1</sup> a<sup>-1</sup>) from (a) Control simulation; (b) Fixsst simulation; and 1063 (c) difference between the Control simulation and the Fixsst simulation in NDJFM 1064 1065 during 1958-2017. (d)-(f) are similar to (a)-(c) but for the results at 500 hPa. (g)-(i) are similar to (d)-(f) but for the results at 700 hPa. The vertical velocity trends over 1066 the dotted regions are statistically significant at the 95% confidence level. The black 1067 rectangles denote the TWP region (20°S-10°N, 100°E-180°E). 1068

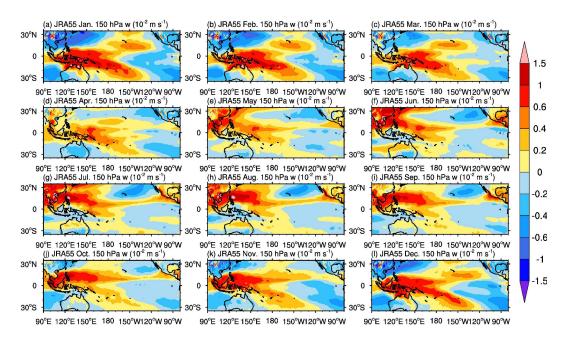
Fig. 6. The difference of vertical velocity (shading, units: 10<sup>-2</sup> m s<sup>-1</sup>) and horizontal

- winds (arrows, units: m s<sup>-1</sup>) at (a) 150 hPa; (b) 500 hPa; and (c) 700 hPa in NDJFM
- between experiments R1 and R2. The differences between vertical velocity over the
- dotted regions are statistically significant at the 95% confidence level. The black
- rectangles denote the TWP region (20°S-10°N, 100°E-180°E).
- 1074 **Fig. 7.** Trends of OLR (W m<sup>-2</sup> a<sup>-1</sup>) (a)-(c), CPTT (shading, units: 10<sup>-1</sup> K a<sup>-1</sup>) and 100
- 1075 hPa streamfunction (contour, units: 106 m<sup>2</sup> s<sup>-1</sup> a<sup>-1</sup>) (d)-(f), and 70 hPa water vapor
- 1076 concentration (units: 10<sup>-2</sup> ppmv a<sup>-1</sup>) (g)-(i) in NDJFM during 1958-2017 in the
- 1077 Control and Fixsst simulations as well as their difference. (a), (d), and (g) are the
- results in the Control simulation; (b), (e), and (h) are the results in the Fixsst
- simulation; (c), (f), and (i) are the results of the difference between the Control and
- 1080 Fixsst simulations. The trends in (a)-(c) and (g)-(i) over the dotted regions are
- statistically significant at the 95% confidence level. The CPTT trends in (d)-(f) over
- the dotted regions are statistically significant at the 95% confidence level. The black
- rectangles denote the TWP region (20°S-10°N, 100°E-180°E).
- Fig. 8. The trends of CO derived from the MOPITT and MLS data. (a) The trends of
- 1085 CO ( $10^{-1}$  ppbv  $a^{-1}$ ) at 215 hPa using MLS data in NDJFM during 2005-2017. (b) The
- trends of CO (10<sup>-1</sup> ppbv a<sup>-1</sup>) at 200 hPa using MOPITT data in NDJFM during
- 1087 2000-2017. The trends of CO over the dotted region are statistically significant at the
- 1088 90% confidence level.
- 1089 Fig. 9. The trends of 150 hPa CO concentration (10<sup>-4</sup> ppmv a<sup>-1</sup>) from (a) Control
- simulation; (b) Fixsst simulation; and (c) difference between the Control simulation
- and the Fixsst simulation in NDJFM during 1958-2017. The trends in (a)-(c) over the

- dotted regions are statistically significant at the 95% confidence level. The black rectangles denote the TWP region (20°S-10°N, 100°E-180°E).
- 1094 Fig. 10. Latitude-pressure cross sections of the trends of (a)-(c) zonal mean CO
- 1095 concentration (10<sup>-4</sup> ppmv a<sup>-1</sup>) and (d)-(f) CO concentration (10<sup>-4</sup> ppmv a<sup>-1</sup>) over the
- 1096 TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and
- Fixsst simulation as well as their difference. (a) and (d) are the CO trends in the
- 1098 Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f) are
- the results derived from the difference between the Control and Fixsst simulations.
- The trends over the dotted regions are statistically significant at the 95% confidence
- level.
- Fig. 11. Latitude-pressure cross sections of the trends of (a)-(c) the zonal mean w (10<sup>-4</sup>)
- 1103 m s<sup>-1</sup> a<sup>-1</sup>) and v (10<sup>-1</sup> m s<sup>-1</sup> a<sup>-1</sup>) and (d)-(f) w (10<sup>-4</sup> m s<sup>-1</sup> a<sup>-1</sup>) and v (10<sup>-1</sup> m s<sup>-1</sup> a<sup>-1</sup>) over
- the TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and
- Fixsst simulation as well as their difference. (a) and (d) are the trends of w and v in
- the Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f)
- are the results derived from the difference between the Control and Fixsst simulations.
- The shadings denote the trends of the w (10<sup>-4</sup> m s<sup>-1</sup> a<sup>-1</sup>). The trends over the dotted
- regions are statistically significant at the 90% confidence level.
- 1110 Fig. 12. Trends of the BDC (vectors, units in the horizontal and vertical components
- are 10<sup>-2</sup> and 10<sup>-5</sup> m s<sup>-1</sup> a<sup>-1</sup>, respectively) calculated using the TEM formula from (a)
- JRA55; (b) Control simulation; (c) Fixsst simulation; and (d) difference between the
- 1113 Control simulation and the Fixsst simulation in NDJFM during 1958-2017. The

- shadings are the trends of the  $w^*$  (10<sup>-5</sup> m s<sup>-1</sup> a<sup>-1</sup>). The trends of the vertical velocity
- over the dotted regions are statistically significant at the 90% confidence level.

## 1117 Figures



**Fig. 1.** The climatological mean (averaged over 1958-2017) values of 150 hPa w (10<sup>-2</sup> m s<sup>-1</sup>) in different months derived from the JRA55 reanalysis data.

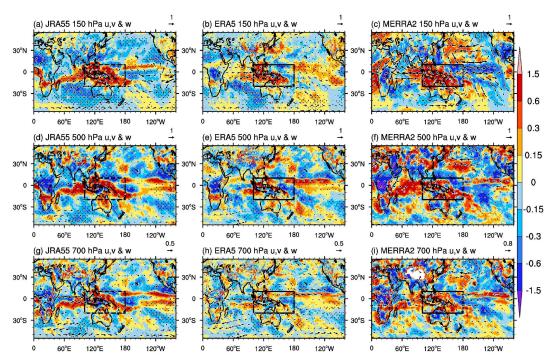
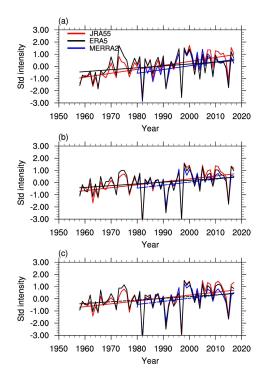
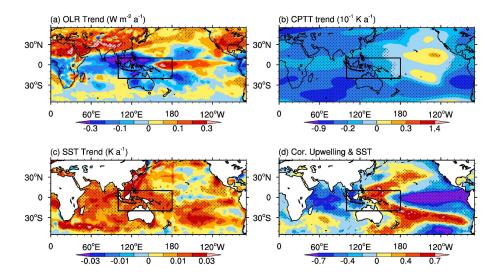


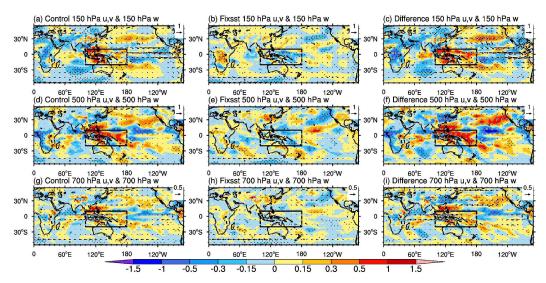
Fig. 2. Trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, 700 hPa in NDJFM derived from JRA55, ERA5, and MERRA2 data. The trends of horizontal winds (arrows, units: 10<sup>-1</sup> m s<sup>-1</sup> a<sup>-1</sup>) and vertical velocity (shading, units: 10<sup>-4</sup> m s<sup>-1</sup> a<sup>-1</sup>) at (a) 150 hPa; (d) 500 hPa; and (g) 700 hPa from JRA55 in NDJFM during 1958-2017. (b), (e), and (h) are the same as (a), (d), and (g) but for the results from ERA5. (c), (f) and (i) are the same as (a), (d), and (g) except that the trends are during 1980-2017 and the wind field data are from MERRA2. The vertical velocity trends over the dotted regions are statistically significant at the 95% confidence level. The white areas denote missing values. The black rectangles denote the TWP region (20°S-10°N, 100°E-180°E).



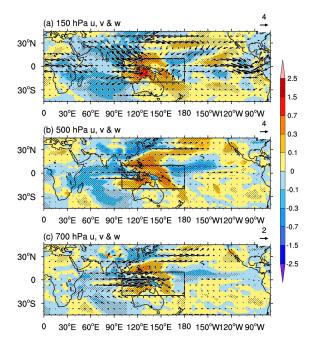
**Fig. 3.** The time series of the standardized intensity of the upward motion over the tropical western Pacific (20°S-10°N, 100°E-180°E) at (a) 150 hPa; (b) 500 hPa; and (c) 700 hPa extracted from JRA55 (red), ERA5 (black) and MERRA2 (blue) datasets. The straight lines in each figure indicate the linear trends. The solid lines denote the linear trends are significant at the 95% confidence level, while the dashed lines denote the linear trends are significant at the 90% confidence level.



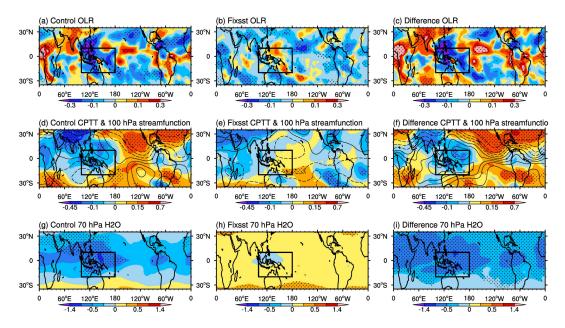
**Fig. 4.** Trends of (a) observed outgoing longwave radiation (OLR, units: W m<sup>-2</sup> a<sup>-1</sup>) provided by NOAA during 1974-2017; (b) cold-point tropopause temperature (CPTT, units: 10<sup>-1</sup> K a<sup>-1</sup>) derived from JRA55 data and (c) SST (K a<sup>-1</sup>) derived from HadISST during 1958-2017 in NDJFM. (d) The correlation coefficients between the intensity of the upward motion at 150 hPa over the TWP and SST in NDJFM during 1958-2017 with the linear trends removed. The trends and correlation coefficients over the dotted regions are statistically significant at the 95% confidence level. The black rectangles denote the TWP region (20°S-10°N, 100°E-180°E).



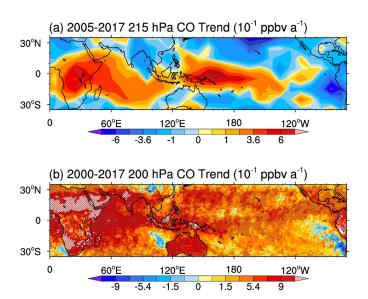
**Fig. 5.** The trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, and 700 hPa in NDJFM during 1958-2017 in the Control and Fixsst simulations as well as their difference. The trends of 150 hPa *w* (shading, units:  $10^{-4}$  m s<sup>-1</sup> a<sup>-1</sup>) and horizontal winds (arrows;  $10^{-1}$  m s<sup>-1</sup> a<sup>-1</sup>) from (a) Control simulation; (b) Fixsst simulation; and (c) difference between the Control simulation and the Fixsst simulation in NDJFM during 1958-2017. (d)-(f) are similar to (a)-(c) but for the results at 500 hPa. (g)-(i) are similar to (d)-(f) but for the results at 700 hPa. The vertical velocity trends over the dotted regions are statistically significant at the 95% confidence level. The black rectangles denote the TWP region (20°S-10°N, 100°E-180°E).



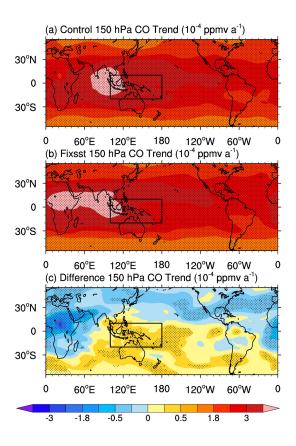
**Fig. 6.** The difference of vertical velocity (shading, units:  $10^{-2}$  m s<sup>-1</sup>) and horizontal winds (arrows, units: m s<sup>-1</sup>) at (a) 150 hPa; (b) 500 hPa; and (c) 700 hPa in NDJFM between experiments R1 and R2. The differences between vertical velocity over the dotted regions are statistically significant at the 95% confidence level. The black rectangles denote the TWP region ( $20^{\circ}$ S- $10^{\circ}$ N,  $100^{\circ}$ E- $180^{\circ}$ E).



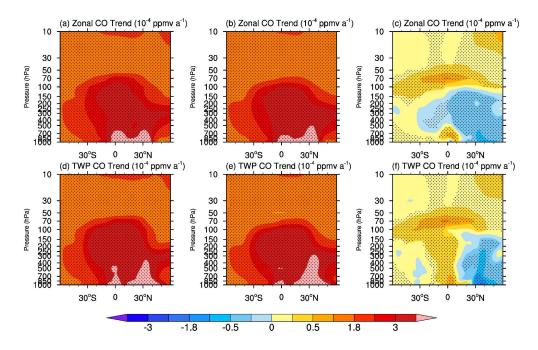
**Fig. 7.** Trends of OLR (W m<sup>-2</sup> a<sup>-1</sup>) (a)-(c), CPTT (shading, units: 10<sup>-1</sup> K a<sup>-1</sup>) and 100 hPa streamfunction (contour, units: 10<sup>6</sup> m<sup>2</sup> s<sup>-1</sup> a<sup>-1</sup>) (d)-(f), and 70 hPa water vapor concentration (units: 10<sup>-2</sup> ppmv a<sup>-1</sup>) (g)-(i) in NDJFM during 1958-2017 in the Control and Fixsst simulations as well as their difference. (a), (d), and (g) are the results in the Control simulation; (b), (e), and (h) are the results in the Fixsst simulation; (c), (f), and (i) are the results of the difference between the Control and Fixsst simulations. The trends in (a)-(c) and (g)-(i) over the dotted regions are statistically significant at the 95% confidence level. The CPTT trends in (d)-(f) over the dotted regions are statistically significant at the 95% confidence level. The black rectangles denote the TWP region (20°S-10°N, 100°E-180°E).



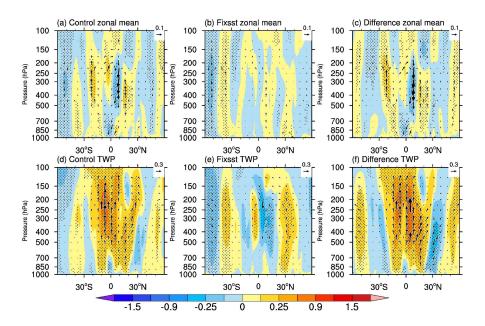
**Fig. 8.** The trends of CO derived from the MOPITT and MLS data. (a) The trends of CO (10<sup>-1</sup> ppbv a<sup>-1</sup>) at 215 hPa using MLS data in NDJFM during 2005-2017. (b) The trends of CO (10<sup>-1</sup> ppbv a<sup>-1</sup>) at 200 hPa using MOPITT data in NDJFM during 2000-2017. The trends of CO over the dotted region are statistically significant at the 90% confidence level.



**Fig. 9.** The trends of 150 hPa CO concentration (10<sup>-4</sup> ppmv a<sup>-1</sup>) from (a) Control simulation; (b) Fixsst simulation; and (c) difference between the Control simulation and the Fixsst simulation in NDJFM during 1958-2017. The trends in (a)-(c) over the dotted regions are statistically significant at the 95% confidence level. The black rectangles denote the TWP region (20°S-10°N, 100°E-180°E).



**Fig. 10.** Latitude-pressure cross sections of the trends of (a)-(c) zonal mean CO concentration (10<sup>-4</sup> ppmv a<sup>-1</sup>) and (d)-(f) CO concentration (10<sup>-4</sup> ppmv a<sup>-1</sup>) over the TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and Fixsst simulation as well as their difference. (a) and (d) are the CO trends in the Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f) are the results derived from the difference between the Control and Fixsst simulations. The trends over the dotted regions are statistically significant at the 95% confidence level.



**Fig. 11.** Latitude-pressure cross sections of the trends of (a)-(c) the zonal mean w ( $10^{-4}$  m s<sup>-1</sup> a<sup>-1</sup>) and v ( $10^{-1}$  m s<sup>-1</sup> a<sup>-1</sup>) and v ( $10^{-1}$  m s<sup>-1</sup> a<sup>-1</sup>) and v ( $10^{-1}$  m s<sup>-1</sup> a<sup>-1</sup>) over the TWP ( $100^{\circ}$ E- $180^{\circ}$ E) in NDJFM during 1958-2017 in the Control simulation and Fixsst simulation as well as their difference. (a) and (d) are the trends of w and v in the Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f) are the results derived from the difference between the Control and Fixsst simulations. The shadings denote the trends of the w ( $10^{-4}$  m s<sup>-1</sup> a<sup>-1</sup>). The trends over the dotted regions are statistically significant at the 90% confidence level.

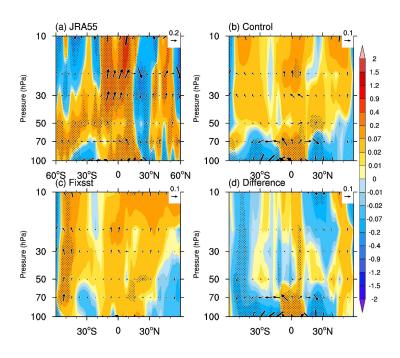


Fig. 12. Trends of the BDC (vectors, units in the horizontal and vertical components are 10<sup>-2</sup> and 10<sup>-5</sup> m s<sup>-1</sup> a<sup>-1</sup>, respectively) calculated using the TEM formula from (a) JRA55; (b) Control simulation; (c) Fixsst simulation; and (d) difference between the Control simulation and the Fixsst simulation in NDJFM during 1958-2017. The shadings are the trends of the  $w^*$  (10<sup>-5</sup> m s<sup>-1</sup> a<sup>-1</sup>). The trends of the vertical velocity

over the dotted regions are statistically significant at the 90% confidence level.

1221

1222

1223

1224

1225

1226

1227