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
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#### 14 Abstract

The tropical western Pacific (TWP) is a preferential area of air uplifting from the 15 16 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 17 next year, NDJFM) has been detected using multiple reanalysis datasets. The upward 18 19 motion over the TWP is intensified at rates of  $8.0\pm3.1\%$  decade<sup>-1</sup> and  $3.6\pm3.3\%$ decade<sup>-1</sup> in NDJFM at 150 hPa from 1958 to 2017 using JRA55 and ERA5 reanalysis 20 datasets, while the MERRA2 reanalysis data show a  $7.5\pm7.1\%$  decade<sup>-1</sup> intensified 21 upward motion for the period 1980-2017. Model simulations using the Whole 22 23 Atmosphere Community Climate Model, version 4 (WACCM4) suggest that warming global sea surface temperatures (SSTs), particularly SSTs over the eastern maritime 24 25 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 26 deep convection over the TWP. Using CO as a tropospheric tracer, the WACCM4 27 simulations show that an increase of CO at a rate of 0.4 ppbv decade<sup>-1</sup> at the layer 28 150-70 hPa in the tropics is mainly resulted from the global SST warming and the 29 subsequent enhanced upward motion over the TWP in the troposphere and 30 strengthened tropical upwelling of Brewer-Dobson (BD) circulation in the lower 31 stratosphere. This implies that more tropospheric trace gases and aerosols from both 32 natural maritime source and outflow from polluted air from South Asia may enter the 33 stratosphere through the TWP region and affect the stratospheric chemistry and 34 climate. 35

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

#### 40 **1 Introduction**

41 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 42 surface temperature (exceeding 28 °C) which fuels intense and massive deep 43 44 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 45 source of tropospheric air entering the stratosphere due to the strong upward motion 46 and deep convection over this region (e.g., Fueglistaler et al., 2004; Pan et al., 2016). 47 Through the TWP region, tropospheric trace gases, e.g., the natural maritime 48 bromine-containing substances and outflow from anthropogenic emissions from South 49 50 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 51 large-scale upwelling (e.g., Levine et al., 2007, 2008; Navarro et al., 2015), which 52 53 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 54 lowest cold-point tropopause temperature (CPTT) over the globe and plays an 55 important role in controlling the water vapor concentration in the stratosphere (e.g., 56 Fueglistaler et al., 2009; Newell and Gould-Steward, 1981; Pan et al., 2016; Randel 57 and Jensen, 2013). The TWP is an important region for tropospheric trace gases being 58 transported from the troposphere to the stratosphere, and therefore influencing the 59 stratospheric chemistry (e.g., Fueglistaler et al., 2004; Levine et al., 2007; Krüger et 60

61 al., 2008; Pan et al., 2016).

The TWP was thought to be the main pathway of the troposphere-to-stratosphere 62 transport. A concept of "stratospheric fountain" was proposed by Newell and 63 Gould-Steward (1981), which suggested that the poor-water vapor air in the 64 stratosphere stems mainly from the TWP region. However, following studies using the 65 66 observational and reanalysis data showed that there is subsidence at the near-tropopause level over the maritime continent, which is named as the 67 "stratospheric drain" (Gettelman et al., 2000; Sherwood, 2000; Fueglistaler et al., 68 2004). Further studies verified that the large-scale transport from the tropical 69 tropopause layer (TTL) to the stratosphere is dominated by the upward branch of the 70 Brewer-Dobson (BD) circulation (Brewer, 1949; Dobson, 1956; Holton et al., 1995) 71 72 while the local upwelling may play a minor role (e.g., Levine et al., 2007; Fueglistaler et al., 2009; Schoeberl et al., 2018). 73

Though the vertical transport from TTL to the lower stratosphere is dominated by 74 the BD circulation, numerous studies confirmed that the TWP region is an important 75 pathway of the surface air entering the TTL (Fueglistaler et al., 2004; Levine et al., 76 2007; Krüger et al., 2008; Haines and Esler, 2014). Based on a trajectory model, 77 Fueglistaler et al. (2004) pointed out that approximately 80% of the trajectories 78 ascending into the stratosphere from the TTL are originated from the TWP region. 79 Bergman et al. (2012) suggested that the tropospheric air over the TWP enters the 80 stratosphere mainly in boreal winter, while less air over the TWP could be transported 81 into the stratosphere during boreal summer. Other studies also found that the TWP 82

region is an important source of the tropospheric trace gases in the TTL (e.g., Newton 83 et al., 2018; Pan et al., 2016; Wales et al., 2018), even the polluted air from East Asia 84 85 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 86 deep convection (Ashfold et al., 2015). Hence, the strength of the upward motion over 87 the TWP region during boreal winter is a key feature for understanding the variations 88 of trace gases in the TTL and therefore important for stratospheric chemistry and 89 climate. 90

The strength of the TWP upward motion is closely related to atmospheric 91 circulation and deep convection. The ascending branch of the Pacific Walker 92 circulation and the strong deep convection over the TWP allow rapid transport from 93 94 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 95 (e.g., sea surface temperature; SST) have been changed. For example, the Hadley cell 96 97 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., 98 2015; Ma and Zhou, 2016). At the same time, SSTs over most of areas are getting 99 100 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 101 atmospheric circulations from the troposphere and the stratosphere (e.g., Garfinkel et 102 al., 2013; Xie et al., 2012, 2014a; Wang et al., 2015; Hu et al., 2016; Lu et al., 2020). 103 However, how the strength of the upward motion in the lower TTL over the TWP 104

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.

111 **2 Data and method** 

Reanalysis data. To investigate the long-term trend of the upward motion over 112 113 the TWP through the troposphere, three most recent reanalysis datasets, including 114 JRA55 from the Japan Meteorological Agency (JMA), ERA5 from the European Centre for Medium Range Weather Forecasting (ECMWF) and MERRA2 from the 115 116 National Aeronautics and Space Administration/Global Modeling and Assimilation Office (NASA/GMAO) are used in this study (Table 1). The JRA55 data, covering the 117 period from 1958 to present, are interpolated to the standard pressure levels and 118 1.25°×1.25° horizontal mesh (Harada et al. 2016). The ERA5 reanalysis is the newest 119 generation product from the ECMWF (Hersbach et al., 2020). The ERA5 data are 120 based on the Integrated Forecasting System (IFS) Cy41r2, which includes the 121 improved model physics, core dynamics and data assimilation. The ERA5 data also 122 extend back to 1958, which is coinciding with the time that radiosonde observations 123 in the Arctic became more systematic and regular. It should be noted that the ERA5 124 data suffer from a bias during 2000-2006, and are replaced by the ERA5.1 data in this 125 period here. The MERRA2 data are also used (Gelaro et al., 2017), which are 126

produced by NASA/GMAO using Goddard Earth Observing System model (GEOS). 127 Although the horizontal and vertical resolution of MERRA2 data are similar to 128 129 MERRA data, the MERRA2 data represent UTLS processes better (Gelaro et al., 2017). The monthly mean air temperature, horizontal wind fields and vertical velocity 130 131 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, 132 which is defined as 20°S-10°N, 100°E-180° due to the strong upward motion (Fig. 1) 133 and significantly increasing trends of the upward motion (Fig. 2) over there. 134

135 A special caution is needed because of the limitations of reanalysis data. The reanalysis datasets assimilate observational data based on the ground- and 136 space-based remote sensing platforms to provide more realistic data products. 137 138 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 139 the vertical velocity in reanalysis data sets has been evaluated by the Reanalysis 140 Intercomparison Project (Fujiwara et al., 2017), which is initiated by the 141 Stratosphere-troposphere Processes And their Role in Climate (SPARC). Results of a 142 comparison between the radar observed data and the reanalysis data indicate that the 143 updrafts in the UTLS are captured well near the TWP even though there are still large 144 biases in the reanalysis datasets and the updrafts from the JRA55 data are stronger 145 than those from the ERA5 and MERRA2 data (Uma et al. 2021). Additionally, 146 discontinuities in the reanalysis data due to different observing systems (for example, 147 transition from TOVS to ATOVS) may still exist (e.g., Long et al., 2017), which could 148

149	lead to uncertainties in the long-term trend of a certain meteorological field.
150	Hitchcock (2019) suggested that the reanalysis uncertainty is larger in the radiosonde
151	era (after 1958) than in the satellite era (after 1979), but the radiosonde era is of
152	equivalent value to the satellite era because the dynamical uncertainty dominates in
153	the both eras. The data in the radiosonde era (1958-1978) used in the present study
154	may induce uncertainties in our results. Therefore, we discuss the trends for both the
155	periods of 1958-2017 and 1980-2017. In addition, we combine three most recent
156	reanalysis datasets (JRA55, ERA5, and MERRA2) to obtain relatively robust results.
157	Observed CO data. Since CO has a photochemical lifetime in the range of 2-3
158	months (Xiao et al., 2007), it could be utilized as a tracer of cross-region transport in
159	the troposphere and the lower stratosphere (Park et al., 2009). Here, CO is used as a
160	tropospheric tracer to indicate the vertical transport from the near-surface to the upper
161	troposphere and the lower stratosphere. The CO data used in the present study are
162	from space-borne Microwave Limb Sounder (MLS; Livesey et al., 2015) observation
163	and Measurements Of Pollution In The Troposphere instrument (MOPITT; Deeter et
164	al., 2019). MLS is carried by Aura, which has a sun-synchronous orbit at 705 km with
165	a 16-day repeat cycle. MLS observations are made from 82°S to 82°N and cover the
166	period from 2005 to the present. MLS provides the CO data from the upper
167	troposphere to the mesosphere. MLS CO v4 level1 data used in the present study are
168	processed using the recommended procedures (Livesey et al., 2015) and interpolated
169	into a $5^{\circ} \times 5^{\circ}$ horizontal mesh. MOPITT CO data are also used for comparison.
170	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).

178 SST and outgoing longwave radiation (OLR) data. SST data are used in this 179 study to investigate the relationship between the upward motion and SSTs. The SST 180 data are from the HadISST dataset  $(1^{\circ}\times1^{\circ}$  horizontal mesh) during 1958-2018 181 (Rayner et al., 2003). OLR is often utilized to reflect the deep convection in the 182 tropics. The OLR data are extracted from NOAA Interpolated OLR dataset on a 183  $2.5^{\circ}\times2.5^{\circ}$  horizontal mesh during 1974/11-2018/03 (Liebmann and Smith, 1996).

Model simulations. A series of model simulations with the Whole Atmosphere 184 185 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 (Table 2). The 186 WACCM4 is a chemical-climate model with a horizontal resolution of 1.9°×2.5° 187 (Marsh et al., 2013). The WACCM4 has vertical 66 levels from the surface to 145 km 188 with vertical resolution of approximately 1 km in the UTLS, which is numerously 189 used to investigate the transport of the trace gases from the troposphere to the 190 stratosphere (e.g., Randel et al., 2010; Xie et al., 2014b; Minganti et al., 2020). A 191 hindcast simulation (Control simulation) is performed with observed greenhouse 192

193 gases, solar irradiances, and prescribed SSTs (HadISST dataset is used) during 194 1955-2018. A single-factor controlling simulation (Fixsst simulation) is done for the 195 same period with the same forcings, except that the global SSTs are fixed to the 196 climatological mean values during 1955-2018 (long-term mean for each calendar 197 month during 1955-2018).

To figure out the impact of the warming SST over the TWP region on the 198 intensification of the upward motion over the TWP region, a couple of time-slice 199 simulations (R1 and R2) are also integrated for 33 years. The SSTs over the eastern 200 201 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 202 prescribed as the climatological mean SSTs during 1998-2017, while the SSTs over 203 204 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, 205 120°E-160°E) in NDJFM are prescribed as the climatological mean SSTs during 206 1958-1977. Since the SSTs over the eastern maritime continent and tropical western 207 Pacific (20°S-20°N, 120°E-160°E) show significantly warming trends, the SSTs 208 during 1998-2017 are higher than the SSTs during 1958-1977 (approximately 0.5 K). 209 Hence, the difference between R1 and R2 reflects the impact of the warmed SSTs 210 over the eastern maritime continent and tropical western Pacific (20°S-20°N, 211 120°E-160°E) on the atmospheric circulation. The first 3 years of the numeric 212 simulations are not used in the present study to provide a spin-up. 213

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Transformed Eulerian Mean (TEM) calculation. To diagnose the changes in

the BD circulation, the meridional and vertical velocities of the BD circulation are calculated by the TEM equations (Andrews and Mcintyre, 1976):

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

218 
$$w^* = \overline{w} + \frac{1}{a\cos\varphi} \left(\cos\varphi \frac{\overline{v\theta'}}{\overline{\theta_z}}\right)_{\varphi}$$

219 Where  $v^*$  and  $w^*$  denote the meridional and vertical velocities of the BD circulation; 220 the overbar represents the zonal mean; the prime denotes the deviation from the zonal 221 mean;  $\theta$ , a,  $\varphi$ , and  $\rho$  indicate the potential temperature, the radius of the earth, 222 the latitude, and the standard density.

Linear trends and the significance test. The linear trends are estimated using a 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

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

228 where b is the estimated slope,  $\sigma$  denotes the standard error of the slope, and 229  $t_{1-\frac{\alpha}{2}}(n-2)$  represents the value of t-distribution with the degree of freedom equal to

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

**3 Results** 

#### **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., 235 2019). Since the lapse-rate tropopause over the TWP in the boreal winter is near 100 236 237 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 238 velocity at 150 hPa for each month averaged over 60 years from 1958 to 2017. The 239 TWP region at the UTLS level has strong upward motion due to the frequent intense 240 deep convection and the Pacific Walker circulation. It is noteworthy that there is 241 strong upward motion at 150 hPa in NDJFM over the TWP, while the upward motion 242 243 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., 244 2012). Therefore, we mainly focus on the changes in the upward motion in NDJFM, 245 246 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 247 subsequently to the lower stratosphere. As seen in Figs. 1a-c and 1k-l, the upward 248 motion (w) at 150 hPa is most evident over the region  $20^{\circ}$ S- $10^{\circ}$ N,  $100^{\circ}$ E- $180^{\circ}$ , which 249 is used to indicate the TWP in the following analysis. The climatological mean 150 250 hPa vertical velocity (w) in NDJFM in ERA5 during 1958-2017 and MERRA2 during 251 1980-2017 are also given in Supplementary Fig. 1. Comparing with the 150 hPa w in 252 NDJFM using JRA55, the 150 hPa w in ERA5 and MERRA2 data shows larger 253 values (maximum larger than 1.5 cm s<sup>-1</sup>) over the land areas but smaller values 254 (minimum less than -0.4 cm s<sup>-1</sup>) over the marine area. Notably, the 150 hPa w shows 255 no subsidence over the maritime continent, while there is descending motion over the 256

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maritime continent at 100 hPa (Supplementary Fig. 2), which is referred to the "stratospheric drain" (Gettleman et al., 2000; Sherwood, 2000).

259 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, 260 and MERRA2 reanalysis datasets. The 150 hPa w increased significantly over most 261 areas of the TWP during 1958-2017 (Fig. 2). At the same time, the upward motion 262 over the TWP in the lower and middle troposphere also mainly shows positive trends 263 (Figs. 2d and g). This indicates that the upward motion over the TWP is increasing 264 265 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, 266 ERA5, and MERRA2), although there are also some differences between the three 267 268 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 269 and b). There are negative trends of vertical velocity in JRA55 and ERA5 while 270 positive trends of vertical velocity in MERRA2 over the northern Pacific. However, 271 these differences are mainly due to the different time periods which are used to 272 calculate the linear trends in JRA55 (1958-2017), ERA5 (1958-2017) and MERRA2 273 (1980-2017). Supplementary Fig. 3 gives the trends of w and horizontal winds in 274 NDJFM during 1980-2017 derived from JRA55, ERA5, and MERRA2 data, which 275 shows insignificant differences between these reanalysis datasets. The trend patterns 276 of the horizontal winds in JRA55, ERA5, and MERRA2 are consistent with each 277 other (Supplementary Fig. 3). For the trends of vertical velocity, significantly positive 278

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 284 datasets are given in Fig. 3. The intensity of the upward motion over the TWP used in 285 Fig. 3 is simply defined as the area-averaged upward mass flux at a specific level, and 286 287 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 288 motion over the TWP at 150 hPa increased significantly in NDJFM during last 289 290 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  $3.0\pm1.2\times10^9$  kg s<sup>-1</sup> 291 decade<sup>-1</sup> (8.0±3.1% decade<sup>-1</sup>), 1.3±1.2×10<sup>9</sup> kg s<sup>-1</sup> decade<sup>-1</sup> (3.6±3.3% decade<sup>-1</sup>), and 292 3.0±2.8×10<sup>9</sup> kg s<sup>-1</sup> decade<sup>-1</sup> (7.5±7.1% decade<sup>-1</sup>) in JRA55, ERA5, and MERRA2 data, 293 respectively (Table 3). As shown in Figs. 3b and c, the intensity of the upward motion 294 at 500 hPa and 700 hPa in JRA55 and the intensity of the upward motion at 500 hPa 295 in ERA5 over the TWP also increased significantly at 95% confidence level 296 (4.6±2.6×10<sup>9</sup> kg s<sup>-1</sup> decade<sup>-1</sup>, 2.9±1.7×10<sup>9</sup> kg s<sup>-1</sup> decade<sup>-1</sup>, and 2.5±2.5×10<sup>9</sup> kg s<sup>-1</sup> 297 decade<sup>-1</sup>, respectively). The increasing trends of the intensity of the upward motion at 298 700 hPa in ERA5 and at 500 hPa and 700 hPa in MERRA2 are significant at the 90% 299 confidence level at rates of  $1.9\pm1.6\times10^9$  kg s<sup>-1</sup> decade<sup>-1</sup>,  $5.4\pm5.3\times10^9$  kg s<sup>-1</sup> decade<sup>-1</sup> 300

and  $3.9\pm3.8\times10^9$  kg s<sup>-1</sup> decade<sup>-1</sup>, respectively. This suggests a comprehensive 301 enhancement of vertical velocity through the whole troposphere, which is evident 302 303 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 304 to the difference between JRA55, ERA5, and MERRA2 data. Hence, caution is 305 suggested when investigating the trend of the upward motion over the TWP using the 306 reanalysis data. While the trace gases in the TTL are modulated by the upward motion 307 and subsequent vertical transport (e.g., Garfinkal et al., 2013; Xie et al., 2014b), such 308 a strengthening of the upward motion over the TWP may lead to more tropospheric 309 310 trace gases in the TTL.

The changes in the atmospheric circulation at the UTLS level in the tropics are 311 312 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 313 provided by NOAA (see Section 2) in NDJFM during 1974-2017 are shown in Fig. 4a. 314 Though the time period of the observed OLR data is shorter than the time period we 315 analyzed, the changes in OLR could partly reflect the changes in the deep convection 316 during 1958-2017. The OLR shows significantly negative trends over the TWP which 317 indicates intensified deep convection over the TWP. The OLR trend pattern is very 318 similar to the trend pattern of the 150 hPa w (Figs. 2a-c), which indicates that the 319 increasing trends of 150 hPa w are closely related to the intensified deep convection 320 over the TWP. The intensified deep convection not only lead to the strengthened 321 upward motion in the UTLS (Highwood and Hoskins, 1998; Ryu and Lee, 2010), but 322

also result in the decreased temperature near the tropopause which plays a dominant 323 role in modulating the lower stratospheric water vapor concentration (e.g., Hu et al., 324 325 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 326 327 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 328 mid-latitudes, except over the central and east Pacific. It should be noted that the 329 CPTT from different reanalysis datasets may show different trends even for the 330 331 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 332 the trends of CPTT from reanalysis datasets. 333

334 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 335 significant intensification over the past decades (e.g., Meng et al., 2012; L'Heureux et 336 al., 2013; McGregor et al., 2014). The vertical velocity at 500 hPa and 150 hPa shows 337 significantly positive trends over the TWP in NDJFM during 1958-2017 (Fig. 2). 338 339 Meanwhile, the lower tropospheric zonal wind shows easterly trends over the tropical Pacific, while the upper tropospheric zonal wind shows westerly trends over the 340 tropical Pacific, which suggests a strengthened Pacific Walker circulation and is 341 consistent with previous studies (Hu et al., 2016; Ma and Zhou, 2016). 342

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

345 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 346 1958-2017. In addition, the intensity of the upward motion over the TWP is 347 significantly correlated with the SST (Fig. 4d), which suggests that the SST has 348 important effects on the upward motion over the TWP. The correlation coefficient in 349 Fig. 4d shows a La Niña-like pattern and indicates that the ENSO events exert 350 important impacts on the upward motion over the TWP (Levine et al., 2008). The 351 SSTs over the TWP are mainly positively correlated with the upward motion intensity 352 353 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 354 correlations with the intensity of the upward motion over the TWP. The SSTs over the 355 356 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 357 driver of the strengthened Pacific Walker circulation, which leads to enhanced deep 358 convection and intensified upward motion over the TWP. 359

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.

# 373 3.2 Simulated trend of the upward motion over the TWP and its potential 374 mechanism

375 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. 376 Consistent with the results shown using the reanalysis data (Figs. 2a-c), the simulated 377 378 150 hPa w (Control simulation) shows significantly increasing trends over the TWP and decreasing trends over the tropical eastern Pacific in NDJFM during 1958-2017 379 (Fig. 5a). Additionally, the 150 hPa w simulated in the Fixsst simulation shows weak 380 trends over the TWP (Fig. 5b). The difference between the Control and the Fixsst 381 simulations suggests that the trends of the 150 hPa w over the TWP region is 382 dominated by the changes in the global SSTs during 1958-2017. There are also 383 significantly positive trends of the vertical velocity over the TWP in the lower (700 384 hPa) and middle troposphere (500 hPa) in the Control simulation, while the zonal 385 winds are also enhanced over the tropical Pacific. The vertical velocity over the TWP 386 in the Fixsst simulation shows weak negative trends and the changes in zonal winds 387 over the tropical Pacific are very weak. This confirms the dominant role of the 388

389 changes in global SSTs on the enhancement of the Walker circulation.

Previous studies found that the changes in the intensity of the Pacific Walker 390 391 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 392 suggested by the correlation coefficients between the upward motion at 150 hPa over 393 the TWP and SSTs in Fig. 4d, warmer SSTs over the tropical central and eastern 394 Pacific, and Indian Ocean may lead to a weakened upward motion over the TWP 395 (negative correlation). The warming trends of SSTs over the eastern maritime 396 397 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 398 maritime continent and tropical western Pacific on the trends of the upward motion 399 400 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 401 Pacific (20°S-20°N, 120°E-160°E) in NDJFM changed in these two simulations. In 402 403 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 404 the eastern maritime continent and tropical western Pacific in R2 are prescribed as the 405 climatological mean SSTs during 1958-1977 (more details are given in the section 2). 406 The differences of the wind fields between R1 and R2 are shown in Fig. 6. The 150 407 hPa w shows significantly positive anomalies over the TWP and negative anomalies 408 over the tropical eastern Pacific, which is consistent with the trends of the 150 hPa w 409 in the Control simulation and the reanalysis datasets (Figs. 2 and 5). The upward mass 410

flux over the TWP at 150 hPa increased approximately 27% in the R1 comparing with 411 R2 due to the warming SSTs over the eastern maritime continent and tropical western 412 413 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 414 induced by the warmer SSTs over the TWP. This result is consistent with Hu et al. 415 (2016), which suggested that the increased zonal gradient of the SSTs over the 416 tropical Pacific could lead to a strengthened Pacific Walker circulation and an 417 enhanced upward motion over the TWP. Therefore, the warmer SSTs over the TWP 418 419 could contribute largely to the trend of the upward motion over the TWP in NDJFM during 1958-2017. 420

The changes in the OLR simulated in WACCM4 associated with the changes in 421 422 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 423 Control simulation, while the deep convection shows weak and even decreasing 424 trends over the TWP in the Fixsst simulation (Figs. 7a and b). The enhanced deep 425 convection over the TWP could lead to the enhancing trends of the upward motion. 426 427 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 428 and a stronger upward motion over the TWP which could extend to the upper 429 troposphere. 430

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 433 could lead to increased CO in the UTLS (e.g., Duncan et al., 2007; Livesey et al., 434 435 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 436 437 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., 438 Highwood and Hoskins, 1998; Garfinkel et al., 2018; Pan et al., 2019). Hence, the 439 relationship between the intensity of upward motion and the water vapor 440 441 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 442 simulated with WACCM4 are analyzed. 443

444 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 445 their difference in Figs. 7d-i. The changes in the deep convection could lead to the 446 447 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 448 449 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 450 a Rossby wave response over the TWP and a Kelvin wave response over the tropical 451 eastern Pacific in the Control simulation (Fig. 7d), which is caused by the changes in 452 the deep convection over the tropical Pacific. The Rossby-Kelvin wave response 453 further leads to the decrease the CPTT over the TWP and the increase of the CPTT 454

over the tropical eastern Pacific. Previous studies suggest that the lower stratospheric 455 water vapor is mainly influenced by the coldest temperature near the tropopause (e.g., 456 Garfinkel et al., 2018; Zhou et al., 2021). Since the TWP has the coldest CPTT in the 457 boreal winter (e.g., Pan et al., 2016), the significantly decreased CPTT over the TWP 458 459 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 460 the tropical lower stratospheric water vapor (not shown). Hence, the enhanced upward 461 motion over the TWP may correspond to a dried lower stratosphere. The CPTT shows 462 463 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 464 the Fixsst simulation suggests that the trends of the deep convection, the CPTT, and 465 466 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 467 forcings may play minor roles. 468

469 Generally, the intensified upward motion may lead to more tropospheric trace gases lifting to the upper troposphere and entering the lower stratosphere (e.g., 470 Rosenlof, 2003; Lu et al., 2020). Here we use CO as a tropospheric tracer to detect the 471 possible influences of the enhanced upward motion over the TWP on the 472 transportation of the tropospheric trace gases to the upper troposphere and the lower 473 stratosphere. Due to the data limitation, it is not possible to show the corresponding 474 changes of trace gases by observations in NDJFM during 1958-2017. Here, the trends 475 of CO at around 200 hPa from MOPITT and MLS observations are shown in the Fig. 476

477	8. The CO increased significantly over the TWP in NDJFM in the upper troposphere
478	from the MOPITT (at 200 hPa during 2000-2017) and MLS data (at 215 hPa during
479	2005-2017). The concentration of MLS CO over the TWP is approximately 80 ppbv
480	at 215 hPa from MLS observations and 70 ppbv at 200 hPa from MOPITT
481	observations, which is consistent with previous study (e.g., Huang et al., 2016). The
482	MLS CO data show that the area-averaged CO increased approximately 2.0±3.7 ppbv
483	decade <sup>-1</sup> over the TWP in NDJFM during 2005-2017. The area-averaged MOPITT CO
484	data show a stronger increase of approximately 5.0±3.1 ppbv decade-1 at 200 hPa
485	from 2000 to 2017 (significant at the 95% confidence level). It should be pointed out
486	that the linear trends of CO are calculated based on the satellite data which only cover
487	14 or 18 years due to the data limitation. Hence, the linear trends of CO may have
488	uncertainties particularly in the regions with large interannual variations. To partially
489	overcome this shortage, the trends of MLS CO at 215 hPa during time periods of
490	2005-2016, 2006-2016, 2006-2017, and 2007-2016 and the trends of MOPITT CO at
491	200 hPa during time periods of 2000-2016, 2001-2016, 2001-2017, and 2002-2016
492	are shown in Supplementary Fig. 6. It could be found that the CO in the upper
493	troposphere increased robustly over the TWP from both the MLS and MOPITT data.
494	Overall, though the observed CO only covers less than 20 years, the results from the
495	satellite data suggest a possible impact of the intensified upward motion over the
496	TWP on the trace gases in the upper troposphere.

497 To further illustrate the impacts of the enhanced upward motion on the trace gas498 in the upper troposphere and lower stratosphere, the Control and Fixsst simulations

499	with WACCM4 are used. The trends of the CO concentrations from the Control and
500	Fixsst simulations as well as their differences are shown in Fig. 9. The tropical CO at
501	150 hPa shows significantly increasing trends both in the Control and the Fixsst
502	simulations at rates of 3.4 ppbv decade <sup>-1</sup> and 3.2 ppbv decade <sup>-1</sup> , respectively, (Figs. 9a
503	and b). This suggests that the surface emission of the CO exerts the most important
504	effect on the increase of the tropical CO concentration. The differences of the CO
505	trends at 150 hPa between the Control simulation and the Fixsst simulation are also
506	displayed in Fig. 9c. Since the surface emission inventories of the two simulations are
507	the same, it can be inferred that the trends of the CO concentration in Fig. 9c are
508	mainly caused by the changes in the atmospheric circulation induced by the changes
509	in the global SSTs. The difference of the CO concentration at 150 hPa between the
510	Control simulation and the Fixsst simulation shows a significantly increasing trend at
511	a rate of $0.2\pm0.1$ ppbv decade <sup>-1</sup> over the TWP (significant at the 95% confidence
512	level). At the same time, decreasing trends over the central Africa exist, which
513	resembles to the trend patterns of the vertical velocity in the lower TTL and the deep
514	convection (Figs. 5i and 7c). This indicates that the enhanced deep convection in the
515	TWP lead to the strengthened upward motion over the TWP, which results in an extra
516	6% increasing trend of CO in the upper troposphere over the TWP. It could also be
517	found that CO also increased in the mid latitudes of the southern hemisphere (Fig. 9c).
518	According to previous studies, the CO perturbation from the Indonesian fires at upper
519	troposphere could be transported to the tropical Indian Ocean by easterly winds and
520	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 ( $15^{\circ}S-25^{\circ}S$ ,  $60^{\circ}E-100^{\circ}E$ ) 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 ( $20^{\circ}N-35^{\circ}N$ ,  $60^{\circ}E-160^{\circ}E$ ) 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 527 displayed in Figs. 10a-c. The zonal mean CO shows significantly increasing trends at 528 529 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 530 significantly increasing trends in the TTL but negative trends in the middle 531 532 troposphere in the tropics and the Northern Hemisphere. At the same time, the difference of CO concentration between the Control simulation and the Fixsst 533 simulation averaged in the western Pacific (100°E-180°E) shows significantly 534 increasing trends in the tropics (20°S-10°N) from the surface to the TTL (Fig. 10f). 535 The CO in the layer 150-70 hPa over the TWP increased 3.2 ppbv decade<sup>-1</sup> and 2.8 536 ppbv decade<sup>-1</sup> in the Control and Fixsst simulations in NDJFM during 1958-2017, 537 respectively. And the CO difference between the Control and Fixsst simulations 538 increased  $0.4\pm0.2$  ppbv decade<sup>-1</sup> (significant at the 95% confidence level) in the layer 539 150-70 hPa over the TWP, which suggests that the intensifying upward motion over 540 the TWP and the tropical upwelling of BDC could lead to an extra 14% increasing 541 trend of CO. This indicates that the increased zonal mean CO in the TTL (Fig. 10c) is 542

543 mainly transported through the western Pacific bands and highlights the importance of 544 the upward motion over the TWP in elevating trace gases from the surface to the 545 upper troposphere.

To understand the CO trends in the Control and Fixsst simulations and their 546 547 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 548 tropics while the w over the TWP intensified in the Control simulation in NDJFM 549 during 1958-2017. This is consistent with Fig. 5. While the SSTs fixed to 550 551 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 552 the increase of the *w* over the TWP region as indicated in the differences between the 553 554 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 555 changes in the surface emissions and the upward motion. The trends of CO at 150 hPa 556 over the TWP in NDJFM during 1958-2017 in the Fixsst simulation mainly include 557 558 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 559 Control and Fixsst simulations indicates that the enhanced tropospheric upward 560 motion over the TWP forced by the changes in the global SSTs leads to some extra 561 increase of CO concentrations in the upper troposphere. It should be mentioned that 562 the increasing trends of CO in the lower troposphere in Fig. 10f may be mainly caused 563 by the changes in the horizontal winds. Girach and Nair (2014) suggested that 564

enhanced deep convection and the subsequent intensified upward motion may lead to 565 a decreased CO concentration in the lower troposphere and an increased CO 566 567 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 568 northeasterly trends near the south Asia (Supplementary Fig. 8c), which suggests that 569 more CO-rich air from east Asia and south Asia could be transported to the TWP in 570 the Control simulation comparing to the Fixsst simulation. Since the CO 571 concentration in the lower troposphere over the northern Pacific is higher than that 572 over southern Pacific, the northerly trends over the western and central Pacific may 573 also contribute to the increased CO in the lower troposphere over the TWP in Fig. 10f. 574 As discussed in the Introduction, the tropospheric trace gases enter the 575 576 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 577 their differences are displayed in Fig. 12. The tropical upwelling of BDC  $(w^*)$ 578 calculated using the TEM formula increased significantly in the lower stratosphere 579 over past decades as seen in the JRA55 data and the Control simulation (Figs. 12a and 580 12b). We found that the 70 hPa upward mass flux in NDJFM in the tropics 581 (15°S-15°N) increased 2.8±1.9% decade<sup>-1</sup> (significant at the 95% confidence level) in 582 the JRA55 data from 1958 to 2017 (Fig. 12a) and 4.6±4.3% decade<sup>-1</sup> (significant at 583 the 95% confidence level) in the MERRA2 data from 1980 to 2017 (Supplementary 584 Fig. 7b). From the ERA5 data, the 70 hPa upward mass flux in NDJFM increased in 585 the north hemisphere (0-15°N) at a rate of  $5.0\pm2.8\%$  decade<sup>-1</sup> (significant at the 95%) 586

confidence level), but decreased significantly in the south hemisphere (0-15°S) during 587 1958-2017 (Supplementary Fig. 7a). On average, the trend of the 70 hPa upward mass 588 flux in NDJFM in the tropics (15°S-15°N) is not significant in ERA5. In fact, many 589 previous studies have investigated the trends of BDC. For example, Abalos et al. 590 (2015) investigated the trends of BDC derived from JRA55, MERRA, and 591 ERA-Interim data during 1979-2012 and suggested that the BDC in JRA55 and 592 MERRA significantly strengthened throughout the layer 100-10 hPa with a rate of 593 2-5% decade<sup>-1</sup>, while the BDC in ERA-Interim shows weakening trends. Diallo et al. 594 (2021) compared the trends of the BDC in the ERA5 and ERA-Interim during 595 1979-2018 and pointed out that the BDC in the ERA-Interim shows weakening trend 596 and the BDC in the ERA5 strengthened at a rate of 1.5% decade<sup>-1</sup> which is more 597 598 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 599 may lead to some differences between our result and that in the previous studies. 600 Overall, the trends of the tropical upwelling of BDC derived from JRA55, MERRA2 601 data and the Control simulation are similar to that in previous studies using both 602 603 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 604 of the BDC decreased in ERA5 data in the tropics (15°S-15°N), which are different 605 from the results in JRA55 and MERRA2. 606

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 609 the two simulations in Fig. 12d. In summary, the tropical upwelling of the BDC is 610 611 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 612 613 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. 614 9c and 10f may be due to a combined effect of the strengthened tropical upwelling of 615 the BD circulation and the enhanced upward motion over the TWP. The enhancement 616 617 of upward motion over the TWP, which transported more tropospheric trace gases to the upper troposphere, works together with the strengthened BD circulation under 618 global warming may lead to an increase of tropospheric trace gases over the TWP in 619 620 the lower stratosphere.

621

#### 4 Summary and Discussion

The recent trends of the upward motion from the lower to the upper troposphere 622 in boreal winter over the TWP is investigated for the first time based on the JRA55, 623 ERA5, MERRA2 datasets and four WACCM4 simulations (Table 2). The upward 624 motion at 150 hPa over the TWP in NDJFM increased  $8\pm3.1\%$  decade<sup>-1</sup> and  $3.6\pm3.3\%$ 625 decade<sup>-1</sup> in NDJFM from 1958 to 2017 in JRA55 and ERA5 reanalysis datasets, 626 respectively (Table 3). Despite the possible discontinuities between the radiosonde era 627 (after 1958) and the satellite era (after 1979), the upward motion at 150 hPa over the 628 TWP in NDJFM increased 7.5±7.1% decade<sup>-1</sup> during 1980-2017 in MERRA2 data. 629 Such intensification of the upward motion over the TWP also exist in the middle and 630

lower troposphere in NDJFM in JRA55, ERA5, and MERRA2, which can be 631 confirmed by the WACCM4 model simulations. Comparing the results between the 632 Control and Fixsst simulations with WACCM4, it is found that the trend of the 633 upward motion over the TWP is closely related to the changes in global SSTs, 634 especially the SST warming over the eastern maritime continent and tropical western 635 Pacific (see the results from the experiments R1 and R2 in Fig. 7). Warmer SSTs over 636 the eastern maritime continent and tropical western Pacific (approximately 0.5 K) lead 637 to a strengthened Pacific Walker circulation, enhanced deep convection and 638 639 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 640 TWP could lead to a dryer lower stratosphere over the TWP, as the strong upward 641 642 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 643 the reanalyses data, and some uncertainties may exist. The availability of more high 644 resolution observations in the future may enhance the quality of the reanalysis data. 645 Results from the Control simulation indicate that the CO concentrations 646 increased significantly from the surface to the stratosphere over the TWP. The CO at 647

648 150 hPa increased at a rate of approximately 3.4 ppbv decade<sup>-1</sup> with increased surface 649 emissions and the enhanced upward motion over the TWP. Specifically, an 650 enhancement of tropospheric upward motion and subsequent upward transport of 651 trace gases over the TWP lead to an extra 6% increasing trend of CO concentrations 652 in the upper troposphere.

653	Furthermore, the upward mass fluxes at 70 hPa in the tropics (15°S-15°N) show
654	strengthening trends at rates of $2.8\pm1.9\%$ decade <sup>-1</sup> and $4.6\pm4.3\%$ decade <sup>-1</sup> in JRA55
655	data (during 1958-2017) and MERRA2 data (during 1980-2017) in NDJFM,
656	respectively, which is consistent with previous studies (e.g., Butchart et al., 2010; Fu
657	et al., 2019; Rao et al., 2019). However, such enhancement in tropical upward mass
658	flux at 70 hPa has large uncertainties since the ERA5 data show a negative and
659	insignificant trend (Supplementary Fig. 7a). The results from the Control and Fixsst
660	simulations indicate that the elevated CO in the upper troposphere is further uplifted
661	to the lower stratosphere by the intensified tropical upwelling of the BD circulation
662	due mainly to global SST warming and lead to an increase of CO in the lower
663	stratosphere. An extra 14% increasing trend of CO at the layer 150-70 hPa over the
664	TWP is derived from the Control and Fixsst simulations.

Tropospheric trace gases and aerosols have important impacts on the 665 stratospheric processes if they enter the stratosphere. For example, ozone-depleting 666 substances, CH<sub>4</sub> and N<sub>2</sub>O could influence on the stratospheric ozone significantly 667 (e.g., Shindell et al., 2013; Wang et al., 2014; WMO, 2018), which also modify the 668 temperature in the stratosphere significantly through their strong radiative effects. 669 Water vapor in the lower stratosphere, in particular, has a significant warming effect 670 on the surface climate (Solomon et al., 2010). Therefore, changes of trace gases in the 671 UTLS have important impacts on both tropospheric and stratospheric climate. Our 672 results suggest that the upward motion over the TWP and the vertical component of 673 674 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 675 important role in the stratospheric processes and the global climate. In addition, more 676 677 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 678 679 large-scale uplifts and exert important effects on the stratospheric chemistry. However, the quantitative impacts of the intensified upward motion over the TWP on 680 tropospheric and stratospheric trace gases and aerosols and their climate feedbacks 681 682 await further investigation using more observations and model simulations. 683 **Competing interests.** The authors declare that they have no conflict of interest. 684 685 686 Author contributions. KQ ran the models and wrote the first draft. WW provided suggestions about the statistical methods and model simulations. WT designed the 687 study. RH, MX, and TW contributed to the manuscript writing. YP provided the data 688 689 used in the study. All authors contributed to the improvement of the results. 690 691 692 Acknowledgements. This research is supported by Strategic Priority Research Program of Chinese Academy of Sciences (XDA17010106), the National Natural 693 694 Science Foundation of China (42075055) and the Supercomputing Center of Lanzhou University. 695 The authors gratefully acknowledge the data used in the present study provided by the 696

697 corresponding scientific groups. The JRA55 data are from:

698 http://rda.ucar.edu/datasets/ds628.0/.

- 699 The SST data is obtained from HadISST:
- 700 https://www.metoffice.gov.uk/hadobs/hadisst/data/download.html.
- 701 The ERA5 data and ERA5.1 data are extracted from:
- 702 https://cds.climate.copernicus.eu/#!/search?text=ERA5&type=dataset.
- 703 The MERRA2 data are downloaded from:
- 704 https://search.earthdata.nasa.gov/search?q=MERRA2&fst0=Atmosphere.
- 705 The OLR data are from https://psl.noaa.gov/data/gridded/data.interp\_OLR.html.

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#### 1030 **Figure captions:**

1031Fig. 1. The climatological mean (averaged over 1958-2017) values of 150 hPa w (10-21032m s<sup>-1</sup>) in Jan.-Dec. (a-1) derived from the JRA55 data.

Fig. 2. Trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, 700 hPa 1033 1034 in NDJFM derived from JRA55, ERA5, and MERRA2 data. The trends of horizontal winds (arrows, units:  $10^{-1}$  m s<sup>-1</sup> a<sup>-1</sup>) and vertical velocity (shading, units:  $10^{-4}$  m s<sup>-1</sup> a<sup>-1</sup>) 1035 1036 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 1037 ERA5. (c), (f) and (i) are the same as (a), (d), and (g) except that the trends are during 1038 1980-2017 and the wind field data are from MERRA2. The vertical velocity trends 1039 1040 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 1041 (20°S-10°N, 100°E-180°E). 1042

1043 Fig. 3. The time series of the standardized intensity of the upward motion over the

1044 tropical western Pacific (20°S-10°N, 100°E-180°E) at (a) 150 hPa; (b) 500 hPa; and

- 1045 (c) 700 hPa extracted from JRA55 (red), ERA5 (black) and MERRA2 (blue) datasets.
- 1046 The straight lines in each figure indicate the linear trends. The solid lines denote the
- 1047 linear trends are significant at the 95% confidence level, while the dashed lines denote
- 1048 the linear trends are significant at the 90% confidence level.
- 1049 Fig. 4. Trends of (a) observed outgoing longwave radiation (OLR, units: W m<sup>-2</sup> a<sup>-1</sup>)
- 1050 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).

1057 Fig. 5. Same as Fig. 2 but for the Control simulation (a, d, and g), Fixsst simulation (b,

1058 <u>e, and h), and the difference between the simulations (c, f, and i).</u> The vertical velocity

1059 trends over the dotted regions are statistically significant at the 95% confidence level.

1060 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°S-10°N, 100°E-180°E).

1066 Fig. 7. Same as Fig. 5 but for the trends of (a)-(c) OLR (W m<sup>-2</sup> a<sup>-1</sup>), (d-f) CPTT

1067 (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>),

1068 and (g-i) 70 hPa water vapor concentration (units: 10<sup>-2</sup> ppmv a<sup>-1</sup>). The trends in (a)-(c)

1069 and (g)-(i) over the dotted regions are statistically significant at the 95% confidence

1070 level. The CPTT trends in (d)-(f) over the dotted regions are statistically significant at

1071 the 95% confidence level. The black rectangles denote the TWP region (20°S-10°N,

1072 <u>100°E-180°E).</u>

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 1083 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 1084 TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and 1085 Fixsst simulation as well as their difference. (a) and (d) are the CO trends in the 1086 Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f) are 1087 the results derived from the difference between the Control and Fixsst simulations. 1088 The trends over the dotted regions are statistically significant at the 95% confidence 1089 level. 1090

# 1091Fig. 11. Same as Fig. 10 but for the trends of tropospheric w (10<sup>-4</sup> m s<sup>-1</sup> a<sup>-1</sup>) and v (10<sup>-1</sup>1092m s<sup>-1</sup> a<sup>-1</sup>). 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

1093 dotted regions are statistically significant at the 90% confidence level.

1094 Fig. 12. Trends of the BDC (vectors, units in the horizontal and vertical components

1095	are $10^{-2}$ and $10^{-5}$ m s <sup>-1</sup> a <sup>-1</sup> , respectively) calculated using the TEM formula from (a)
1096	JRA55; (b) Control simulation; (c) Fixsst simulation; and (d) difference between the

- 1097 Control simulation and the Fixsst simulation in NDJFM during 1958-2017. The
- 1098 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
- 1099 over the dotted regions are statistically significant at the 90% confidence level.

## **<u>Tables:</u>**

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Table 1. Basic specifications of JRA55, ERA5, and MERRA2 used in this study.

Name	Organization	Time period	Spatial resolution	Temporal resolution	<u>Data</u> assimilation
<u>JRA55</u>	<u>JMA</u>	1958-present	<u>55 km; L60</u>	<u>6-hourly</u>	4D-Var
ERA5	ECMWF	1950-present	<u>31 km; L137</u>	hourly	<u>4D-Var</u>
MERRA2	<u>NASA</u>	1980-present	<u>0.5°×0.625°;</u>	<u>3-hourly</u>	<u>3D-Var</u>
	<u>GMAO</u>		<u>L72</u>		

#### **Table 2.** Description of simulations with WACCM4.

Experiment	Description			
Control	Transient simulation. Observed greenhouse gases and solar irradiances.			
	Prescribed SST forcing using observed SST.			
<u>Fixsst</u>	Transient simulation. Observed greenhouse gases and solar irradiances.			
	Prescribed SST forcing using monthly mean climatology from 1958 to			
	<u>2017.</u>			
<u>R1</u>	Time-slice simulation. SSTs prescribed as the climatological mean of			
	1998-2017 over the region 20°S-20°N, 120°E-160°E in NDJFM, but			
	fixed as climatological mean of 1958-2017 over other regions.			
<u>R2</u>	Same as R1, but the SSTs over the region 20°S-20°N, 120°E-160°E are			
	prescribed as the climatological mean SSTs during 1958-1977.			

**Table 3.** The trends of the upward motion over the TWP at 150 hPa, 500 hPa, and 700

hPa in NDJFM during 1958-2017 from JRA55, ERA5, MERRA2, Control simulation and Fixsst simulation. And the trends of 150 hPa CO from the Control and Fixsst simulations.

	<u>JRA55</u>	ERA5	MERRA2	<u>Control</u>	<u>Fixsst</u>
<u>150 hPa</u>	3.0±1.2×10 <sup>9</sup>	1.3±1.2×10 <sup>9</sup>	3.0±2.8×10 <sup>9</sup>	2.0±1.2×10 <sup>9</sup>	<u>-4.8±6.4×10<sup>8</sup></u>
<u>Upward</u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>
motion	decade-1	decade-1	decade-1	decade-1	decade-1
<u>500 hPa</u>	4.6±2.6×109	2.5±2.5×10 <sup>9</sup>	<u>5.4±5.3×10<sup>9</sup></u>	<u>3.5±2.4×10<sup>9</sup></u>	<u>-1.0±1.3×10<sup>9</sup></u>
<u>Upward</u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>
motion	decade-1	decade-1	decade-1	decade-1	decade-1
<u>700 hPa</u>	2.9±1.7×10 <sup>9</sup>	<u>1.9±1.6×10<sup>9</sup></u>	<u>3.9±3.8×10<sup>9</sup></u>	$1.8 \pm 1.4 \times 10^{9}$	<u>-6.3±8.1×10<sup>8</sup></u>
<u>Upward</u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>	<u>kg s<sup>-1</sup></u>
motion	decade-1	decade-1	decade-1	decade-1	decade-1
<u>150 hPa</u>				<u>3.4 ppbv</u>	<u>3.2 ppbv</u>
<u>CO</u>				decade-1	decade-1

#### 1112 Figures



1114 Fig. 1. The climatological mean (averaged over 1958-2017) values of 150 hPa w (10<sup>-2</sup>

- 1115 m s<sup>-1</sup>) in <u>Jan.-Dec. (a-1)</u> derived from the JRA55 reanalysis data.
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Fig. 2. Trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, 700 hPa 1118 in NDJFM derived from JRA55, ERA5, and MERRA2 data. The trends of horizontal 1119 winds (arrows, units:  $10^{-1}$  m s<sup>-1</sup> a<sup>-1</sup>) and vertical velocity (shading, units:  $10^{-4}$  m s<sup>-1</sup> a<sup>-1</sup>) 1120 1121 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 1122 1123 ERA5. (c), (f) and (i) are the same as (a), (d), and (g) except that the trends are during 1124 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 1125 white areas denote missing values. The black rectangles denote the TWP region 1126 (20°S-10°N, 100°E-180°E). 1127



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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>) 1139 provided by NOAA during 1974-2017; (b) cold-point tropopause temperature (CPTT, 1140 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 1141 during 1958-2017 in NDJFM. (d) The correlation coefficients between the intensity of 1142 the upward motion at 150 hPa over the TWP and SST in NDJFM during 1958-2017 1143 with the linear trends removed. The trends and correlation coefficients over the dotted 1144 regions are statistically significant at the 95% confidence level. The black rectangles 1145 denote the TWP region (20°S-10°N, 100°E-180°E). 1146



- 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).



1155

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).





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).

1185



Fig. 10. Latitude-pressure cross sections of the trends of (a)-(c) zonal mean CO 1189 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 1190 TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and 1191 Fixsst simulation as well as their difference. (a) and (d) are the CO trends in the 1192 Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f) are 1193 the results derived from the difference between the Control and Fixsst simulations. 1194 The trends over the dotted regions are statistically significant at the 95% confidence 1195 level. 1196



![](_page_62_Figure_0.jpeg)

![](_page_62_Figure_1.jpeg)

Fig. 12. Trends of the BDC (vectors, units in the horizontal and vertical components are  $10^{-2}$  and  $10^{-5}$  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  $w^*$  ( $10^{-5}$  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.