1	Enhanced upward motion through the troposphere over the tropical
2	western Pacific and its implication <u>s</u> for the transport of trace gases
3	from the troposphere to the stratosphere
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15	The tropical western Pacific (TWP) is a preferential area of air uplifting from the
16	surface to the upper troposphere. A significantly intensified upward motion through
17	the troposphere over the TWP in the boreal wintertime (November to March of the
18	next year, NDJFM) has been detected using multiple reanalysis datasets. The upward
19	motion over the TWP is intensified at rates of 8.0±3.1% decade-1 and 3.6±3.3%
20	decade ⁻¹ in NDJFM at 150 hPa from 1958 to 2017 using JRA55 and ERA5 reanalysis
21	datasets, while the MERRA2 reanalysis data show a 7.5±7.1% decade ⁻¹ intensified
22	upward motion for the period 1980-2017. Model simulations using the Whole
23	Atmosphere Community Climate Model, version 4 (WACCM4) suggest that warming
24	global sea surface temperatures (SSTs), particularly SSTs over the eastern maritime
25	continent and tropical western Pacific, play a dominant role in the intensification of
26	the upward motion by strengthening the Pacific Walker circulation and enhancing the
27	deep convection over the TWP. Using CO as a tropospheric tracer, the WACCM4
28	simulations show that an increase of CO at a rate of 0.4 ppbv decade ⁻¹ at the layer
29	150-70 hPa in the tropics is mainly resulted from the global SST warming and the
30	subsequent enhanced upward motion over the TWP in the troposphere and
31	strengthened tropical upwelling of Brewer-Dobson (BD) circulation in the lower
32	stratosphere. This implies that more tropospheric trace gases and aerosols from both
33	natural maritime source and outflow from polluted air from South Asia may enter the
34	stratosphere through the TWP region and affect the stratospheric chemistry and
35	climate.

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

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39

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 <u>al., 2008; Pan et al., 2016).</u>

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 70 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) 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

74 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 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 78 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. 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

5

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,
<u>ERA5, and MERRA2</u> reanalysis datasets and <u>different WACCM4</u> simulations as
<u>described in Section 2</u>. The implication of the changes in the upward motion over the
TWP to the transport of trace gases from the surface to the <u>UTLS will be</u> discussed in
<u>Section 3</u>.

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 JRA55 from the Japan Meteorological Agency (JMA), ERA5 from the European 114 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. The JRA55 is produced by a four 117 dimensional variational (4D-Var) data assimilation system and provide 6-hourly data 118 (Harada et al. 2016). It has a vertical resolution of 60 levels from the surface to 0.1 119 hPa and a high horizontal resolution (T319). It covers the period from 1958 to present, 120 are interpolated to the standard pressure levels and 1.25°×1.25° horizontal mesh. 121 More details could be found in Kobayashi et al., 2015. The ERA5 reanalysis is the 122 newest generation product from the ECMWF (Hersbach et al., 2020). The ERA5 data 123 are based on the Integrated Forecasting System (IFS) Cy41r2, which includes the 124 improved model physics, core dynamics and data assimilation. The ERA5 has hourly 125 output, a horizontal resolution of 31 km, and a vertical resolution of 137 levels 126

127	extending into the middle atmosphere (0.01 hPa) and covers the period from 1979 to
128	present. The ERA5 data also extend back to 1958, which is coinciding with the time
129	that radiosonde observations in the Arctic became more systematic and regular. It
130	should be noted that the ERA5 data suffer from a bias during 2000-2006, and are
131	replaced by the ERA5.1 data in this period here. The MERRA2 data are also used,
132	which are only accessible after 1980 (Gelaro et al., 2017). The MERRA2 data are
133	produced by NASA/GMAO using Goddard Earth Observing System model (GEOS),
134	which have 3-hourly temporal resolution, 72 vertical levels up to 0.01 hPa, and a
135	horizontal resolution of 0.5°×0.625°. Although the horizontal and vertical resolution
136	of MERRA2 data are similar to MERRA data, the MERRA2 data represent UTLS
137	processes better (Gelaro et al., 2017). The monthly mean air temperature, horizontal
138	wind fields and vertical velocity at different pressure levels are extracted from the
139	three Reanalysis datasets. In the present study, we mainly focus on the upward motion
140	over TWP region in NDJFM, which is defined as 20°S-10°N, 100°E-180° due to the
141	strong upward motion (Fig. 1) and significantly increasing trends of the upward
142	motion (Fig. 2) over there.
143	A special caution is needed because of the limitations of reanalysis data. The
144	reanalysis datasets assimilate observational data based on the ground- and
145	space-based remote sensing platforms to provide more realistic data products.
146	However, previous studies suggested that there are still uncertainties in the reanalysis
147	data (e.g., Simmons et al., 2014; Long et al., 2017; Uma et al., 2021). The accuracy of

148 the vertical velocity in reanalysis data sets has been evaluated by the Reanalysis

149	Intercomparison Project (Fujiwara et al., 2017), which is initiated by the
150	Stratosphere-troposphere Processes And their Role in Climate (SPARC). Results of a
151	comparison between the radar observed data and the reanalysis data indicate that the
152	updrafts in the UTLS are captured well near the TWP even though there are still large
153	biases in the reanalysis datasets and the updrafts from the JRA55 data are stronger
154	than those from the ERA5 and MERRA2 data (Uma et al. 2021). Additionally,
155	discontinuities in the reanalysis data due to different observing systems (for example,
156	transition from TOVS to ATOVS) may still exist (e.g., Long et al., 2017), which could
157	lead to uncertainties in the long-term trend of a certain meteorological field.
158	Hitchcock (2019) suggested that the reanalysis uncertainty is larger in the radiosonde
159	era (after 1958) than in the satellite era (after 1979), but the radiosonde era is of
160	equivalent value to the satellite era because the dynamical uncertainty dominates in
161	the both eras. The data in the radiosonde era (1958-1978) used in the present study
162	may induce uncertainties in our results. Therefore, we discuss the trends for both the
163	periods of 1958-2017 and 1980-2017. In addition, we combine three most recent
164	reanalysis datasets (JRA55, ERA5, and MERRA2) to obtain relatively robust results.
165	Observed CO data. CO is used as a tropospheric tracer in this study to indicate
166	the vertical transport from the near-surface to the upper troposphere and the lower
167	stratosphere. The CO data used in the present study are from space-borne Microwave
168	Limb Sounder (MLS; Livesey et al., 2015) observation and Measurements Of
169	Pollution In The Troposphere instrument (MOPITT; Deeter et al., 2019). MLS is
170	carried by Aura, which has a sun-synchronous orbit at 705 km with a 16-day repeat

171	cycle. MLS observations are made from 82°S to 82°N and cover the period from 2005
172	to the present. MLS provides the CO data from the upper troposphere to the
173	mesosphere. MLS CO v4 level1 data used in the present study are processed using the
174	recommended procedures (Livesey et al., 2015) and interpolated into a $5^{\circ} \times 5^{\circ}$
175	horizontal mesh. MOPITT CO data are also used for comparison. MOPITT
176	instrument is aboard on the Terra satellite permitting retrievals of CO vertical profiles
177	using both thermal-infrared and near-infrared measurements and has a field of view of
178	22 km×22 km. The Terra satellite was launched in 1999 with a 705 km
179	sun-synchronous orbit. MOPITT provides the CO data from the surface to the upper
180	troposphere during the period of 2000/03 to the present. Here, we use the daytime
181	only MOPITT v8 level3 CO data. For comparison, we focus on the CO concentrations
182	in MLS and MOPITT data at similar level (215 hPa in MLS data and 200 hPa in
183	MOPITT data, respectively).
184	SST and outgoing longwave radiation (OLR) data. SST data are used in this
185	study to investigate the relationship between the upward motion and SSTs. The SST
186	data are from the HadISST dataset (1°×1° horizontal mesh) during 1958-2018
187	(Rayner et al., 2003). OLR is often utilized to reflect the deep convection in the
188	tropics. The OLR data are extracted from NOAA Interpolated OLR dataset on a
189	2.5°×2.5° horizontal mesh during 1974/11-2018/03 (Liebmann and Smith, 1996).

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

193	chemical-climate model with a horizontal resolution of 1.9°×2.5° (Marsh et al., 2013).
194	The WACCM4 has vertical 66 levels from the surface to 145 km with vertical
195	resolution of approximately 1 km in the UTLS, which is numerously used to
196	investigate the transport of the trace gases from the troposphere to the stratosphere
197	(e.g., Randel et al., 2010; Xie et al., 2014b; Minganti et al., 2020). A hindcast
198	simulation (Control simulation) is performed with observed greenhouse gases, solar
199	irradiances, and prescribed SSTs (HadISST dataset is used) during 1955-2018. A
200	single-factor controlling simulation (Fixsst simulation) is done for the same period
201	with the same forcings, except that the global SSTs are fixed to the climatological
202	mean values during 1955-2018 (long-term mean for each calendar month during
203	1955-2018).
204	To figure out the impact of the warming SST over the TWP region on the
204 205	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
205	intensification of the upward motion over the TWP region, a couple of time-slice
205 206	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
205 206 207	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
205 206 207 208	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
205 206 207 208 209	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
 205 206 207 208 209 210 	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
 205 206 207 208 209 210 211 	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,
 205 206 207 208 209 210 211 212 	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

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

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; θ , a, φ , and ρ 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 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 al., 2004): $\left(b-t_{1-\frac{\alpha}{2}}(n-2)\sigma_{b}, b+t_{1-\frac{\alpha}{2}}(n-2)\sigma\right)$

234 where b is the estimated slope, σ denotes the standard error of the slope, and

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

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

237 **3 Results**

238

3.1 Enhanced upward motion over the TWP

According to previous studies, the lapse-rate tropopause is a good proxy to 239 separate the tropospheric and the stratospheric dynamic behavior (vertical motion 240 dominated and horizontal mixing dominated, respectively) over the TWP (Pan et al., 241 2019). Since the lapse-rate tropopause over the TWP in the boreal winter is near 100 242 243 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 244 245 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 246 deep convection and the Pacific Walker circulation. It is noteworthy that there is 247 248 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 249 consistent with previous studies (Newell and Gould-Steward, 1981; Bergman et al., 250 2012). Therefore, we mainly focus on the changes in the upward motion in NDJFM, 251 which is more important to the transport of air over the TWP from the lower 252 troposphere to the TTL compared to the summer months (as shown in Fig. 1) and 253 subsequently to the lower stratosphere. As seen in Figs. 1a-c and 1k-l, the upward 254 motion (w) at 150 hPa is most evident over the region 20°S-10°N, 100°E-180°, which 255

256	is used to indicate the TWP in the following analysis. The climatological mean 150
257	hPa vertical velocity (w) in NDJFM in ERA5 during 1958-2017 and MERRA2 during
258	1980-2017 are also given in Supplementary Fig. 1. Comparing with the 150 hPa w in
259	NDJFM using JRA5, the 150 hPa w in ERA5 and MERRA2 data shows larger values
260	(maximum larger than 1.5 m s ⁻¹) over the land areas but smaller values (minimum less
261	than -0.4 m s ⁻¹) over the marine area. Notably, the 150 hPa w shows no subsidence
262	over the maritime continent, while there is descending motion over the maritime
263	continent at 100 hPa (Supplementary Fig. 2), which is referred to the "stratospheric
264	drain" (Gettleman et al., 2000; Sherwood, 2000).
265	Figure 2 displays the linear trends of w in the upper (150 hPa), middle (500 hPa)
265 266	Figure 2 displays the linear trends of <i>w</i> in the upper (150 hPa), middle (500 hPa) and lower (700 hPa) troposphere in NDJFM from 1958 to 2017 using <u>JRA55</u> , <u>ERA5</u> ,
266	and lower (700 hPa) troposphere in NDJFM from 1958 to 2017 using JRA55, ERA5,
266 267	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
266 267 268	and lower (700 hPa) troposphere in NDJFM from 1958 to 2017 using JRA55, ERA5, and MERRA2 reanalysis datasets. The 150 hPa <i>w</i> increased significantly over most areas of the TWP during 1958-2017 (Fig. 2). At the same time, the upward motion
266 267 268 269	and lower (700 hPa) troposphere in NDJFM from 1958 to 2017 using JRA55, ERA5, and MERRA2 reanalysis datasets. The 150 hPa <i>w</i> 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
266 267 268 269 270	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

274 reanalysis datasets. For example, the trends of the horizontal winds in the upper

275 troposphere in MERRA2 (Fig. 2c) are larger than those in JRA55 and ERA5 (Figs. 2a

276 and b). There are negative trends of vertical velocity in JRA55 and ERA5 while

277 positive trends of vertical velocity in MERRA2 over the northern Pacific. However,

278	these differences are mainly due to the different time periods which are used to
279	calculate the linear trends in JRA55 (1958-2017), ERA5 (1958-2017) and MERRA2
280	(1980-2017). Supplementary Fig. 3 gives the trends of w and horizontal winds in
281	NDJFM during 1980-2017 derived from JRA55, ERA5, and MERRA2 data, which
282	shows insignificant differences between these reanalysis datasets. The trend patterns
283	of the horizontal winds in JRA55, ERA5, and MERRA2 are consistent with each
284	other (Supplementary Fig. 3). For the trends of vertical velocity, significant positive
285	trends over the TWP region can be noted in the JRA55, ERA5, and MERRA2 datasets,
286	although the trends in ERA5 are slightly weaker than those in JRA55 and MERRA2
287	(Fig. 2 and Supplementary Fig. 3). Comparing to the negative trends of the vertical
200	velocity over the central Pacific in JRA55 and ERA5, the negative trends in MERRA2
288	verocity over the central radius in site is and Exercis, the negative dends in with (12)
288 289	extend more northward (Supplementary Fig. 3).
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289 290	extend more northward (Supplementary Fig. 3). The time series of the upward motion intensity over the TWP from different
289 290 291	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. <u>The intensity of the upward motion over the TWP used in</u>
289 290 291 292	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. <u>The intensity of the upward motion over the TWP used in</u> <u>Fig. 3 is simply defined as the area-averaged upward mass flux at a specific level, and</u>
289 290 291 292 293	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
289 290 291 292 293 294	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
289 290 291 292 293 294 295	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
289 290 291 292 293 294 295 296	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
289 290 291 292 293 294 295 296 297	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±1.2×10 ⁸ kg s ⁻¹

300	respectively. As shown in Figs. 3b and c, the intensity of the upward motion at 500
301	hPa and 700 hPa in JRA55 and the intensity of the upward motion at 500 hPa in
302	ERA5 over the TWP also increased significantly at 95% confidence level
303	$(4.6\pm2.6\times10^8 \text{ kg s}^{-1} \text{ decade}^{-1}, 2.9\pm1.7\times10^8 \text{ kg s}^{-1} \text{ decade}^{-1}, \text{ and } 2.5\pm2.5\times10^8 \text{ kg s}^{-1})$
304	decade ⁻¹ , respectively). The increasing trends of the intensity of the upward motion at
305	700 hPa in ERA5 and at 500 hPa and 700 hPa in MERRA2 are significant at the 90%
306	confidence level at rates of 1.9±1.6×10 ⁸ kg s ⁻¹ decade ⁻¹ , 5.4±5.3×10 ⁸ kg s ⁻¹ decade ⁻¹
307	and $3.9\pm3.8\times10^8$ kg s ⁻¹ decade ⁻¹ , respectively. This suggests a comprehensive
308	enhancement of vertical velocity through the whole troposphere, which is evident
309	from the surface to 100 hPa (Supplementary Fig. 4). It can also be inferred that the
310	upward motions over the TWP increased at different rates during the past decades due
311	to the difference between JRA55, ERA5, and MERRA2 data. Hence, caution is
312	suggested when investigating the trend of the upward motion over the TWP using the
313	reanalysis data. While the trace gases in the TTL are modulated by the upward motion
314	and subsequent vertical transport (e.g., Garfinkal et al., 2013; Xie et al., 2014b), such
315	a strengthening of the upward motion over the TWP may lead to more tropospheric
316	trace gases in the TTL.
317	The changes in the atmospheric circulation at the UTLS level in the tropics are

117 The changes in the atmospheric circulation at the OTLS level in the tropics are
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

322	analyzed, the changes in OLR could partly reflect the changes in the deep convection
323	during 1958-2017. The OLR shows significantly negative trends over the TWP which
324	indicates intensified deep convection over the TWP. The OLR trend pattern is very
325	similar to the trend pattern of the 150 hPa w (Figs. 2a-c), which indicates that the
326	increasing trends of 150 hPa w are closely related to the intensified deep convection
327	over the TWP. The intensified deep convection not only lead to the strengthened
328	upward motion in the UTLS (Highwood and Hoskins, 1998; Ryu and Lee, 2010), but
329	also result in the decreased temperature near the tropopause which plays a dominant
330	role in modulating the lower stratospheric water vapor concentration (e.g., Hu et al.,
331	2016; Wang et al., 2016). Corresponding to the enhanced deep convection over the
332	TWP, the CPTT derived from JRA55 data (see Fig. 4b) shows significantly decreasing
333	trends over the TWP in NDJFM during 1958-2017, which is consistent with Xie et al.,
334	(2014a). However, negative trends are also found in other regions in low and
335	mid-latitudes, except over the central and east Pacific. It should be noted that the
336	CPTT from different reanalysis datasets may show different trends even for the
337	satellite period (Tegtmeier et al., 2020). Additionally, the JRA55 data before 1978
338	may also lead to uncertainties in the CPTT trends. Caution is needed when discussing
339	the trends of CPTT from reanalysis datasets.

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).

The strengthened Pacific Walker circulation is closely related to the changes in 349 the SSTs (e.g., Meng et al., 2012; Ma and Zhou, 2016). The trends of the SSTs in 350 NDJFM during 1958-2017 are shown in Fig. 4c. The SST shows significantly 351 warming trends almost over the world except the central Pacific in NDJFM during 352 1958-2017. In addition, the intensity of the upward motion over the TWP is 353 significantly correlated with the SST (Fig. 4d), which suggests that the SST has 354 355 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 356 important impacts on the upward motion over the TWP (Levine et al., 2008). The 357 SSTs over the TWP are <u>mainly</u> positively correlated with the upward motion intensity 358 over the TWP with negative correlations shown over the western maritime continent, 359 360 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 361 Atlantic Ocean are poorly correlated with the upward motion intensity over the TWP 362 (not shown). This result suggests that the changes in global SSTs may be the primary 363 driver of the strengthened Pacific Walker circulation, which leads to enhanced deep 364 convection and intensified upward motion over the TWP. 365

366	It could be found that there are extreme minima (1982, 1991, and 1997) in Fig. 3,
367	which may be related to the El Niño events occurred in these years. To further figure
368	out the impact of ENSO events on the upward motion over the TWP, Supplementary
369	Fig. 5 displays the time series of the standardized intensity of the upward motion over
370	the TWP at 150 hPa, 500 hPa, and 700 hPa in NDJFM in JRA55, ERA5, and
371	MERRA2 with the ENSO signal removed using the linear regression method (Hu et
372	al., 2018; Qie et al., 2021). The extreme minima (1982, 1991, and 1997) become
373	much weaker in Supplementary Fig. 5 than those in Fig. 3, which indicates that the El
374	Niño events are responsible for the extreme minima. The upward motions over the
375	TWP at 150 hPa, 500 hPa, and 700 hPa in NDJFM in JRA55, ERA5, and MERRA2
376	still show statistically significant increasing trends after removing the ENSO signal in
377	Supplementary Fig. 5, which suggests that ENSO events exert limited impacts on the
378	trends of the upward motion over the TWP in NDJFM during 1958-2017.
379	3.2 Simulated trend of the upward motion over the TWP and its potential
380	mechanism
381	To verify the impact of SST on the trend of the upward motion over the TWP, a
382	couple of model simulations with WACCM4 are employed in the following analysis.
383	Consistent with the results shown using the reanalysis data (Figs. 2a-c), the simulated
384	150 hPa w (Control <u>simulation</u>) shows significantly increasing trends over the TWP
385	and decreasing trends over the tropical eastern Pacific in NDJFM during 1958-2017
386	(Fig. $5a$). Additionally, the 150 hPa w simulated in the Fixsst simulation shows weak

387 trends over the TWP (Fig. 5b). The difference between the Control and the Fixsst

388	simulations suggests that the trends of the 150 hPa w over the TWP region is
389	dominated by the changes in the global SSTs during 1958-2017. There are also
390	significantly positive trends of the vertical velocity over the TWP in the lower (700
391	hPa) and middle troposphere (500 hPa) in the Control simulation, while the zonal
392	winds are also enhanced over the tropical Pacific. The vertical velocity over the TWP
393	in the Fixsst simulation shows weak negative trends and the changes in zonal winds
394	over the tropical Pacific are very weak. This confirms the dominant role of the
395	changes in global SSTs on the enhancement of the Walker circulation.
396	Previous studies found that the changes in the intensity of the Pacific Walker
397	circulation and the stratospheric residual circulation are closely related to the changes
398	in tropical SST (Meng et al., 2012; Tokinaga et al., 2012; Lin et al., 2015). As
399	suggested by the correlation coefficients between the upward motion at 150 hPa over
400	the TWP and SSTs in Fig. 4d, warmer SSTs over the tropical central and eastern
401	Pacific, and Indian Ocean may lead to a weakened upward motion over the TWP
402	(negative correlation). The warming trends of SSTs over the eastern maritime
403	continent and tropical western Pacific may result in an intensification of the upward
404	motion over the TWP. To verify the impact of the changes in the SSTs over eastern
405	maritime continent and tropical western Pacific on the trends of the upward motion
406	over the TWP, a couple of <u>single-factor controlling</u> time-slice <u>simulations</u> (R1 and R2)
407	are performed with only SSTs over eastern maritime continent and tropical western
408	Pacific (20°S-20°N, 120°E-160°E) in NDJFM changed in these two simulations. In
409	R1, the SSTs over the eastern maritime continent and tropical western Pacific are

410	prescribed as the climatological mean SSTs during 1958-2017, while the SSTs over
411	the eastern maritime continent and tropical western Pacific in R2 are prescribed as the
412	climatological mean SSTs during 1958-1977 (more details are given in the section 2).
413	The differences of the wind fields between R1 and R2 are shown in Fig. 6 . The 150
414	hPa w shows significantly positive anomalies over the TWP and negative anomalies
415	over the tropical eastern Pacific, which is consistent with the trends of the 150 hPa w
416	in the Control <u>simulation</u> and the reanalysis datasets (Figs. 2 and <u>5</u>). <u>The upward mass</u>
417	flux over the TWP at 150 hPa increased approximately 27% in the R1 comparing with
418	R2 due to the warming SSTs over the eastern maritime continent and tropical western
419	Pacific (approximately 0.5 K). The upward motion in the lower and middle
420	troposphere over the TWP shows increasing trends due to the enhanced convergence
421	induced by the warmer SSTs over the TWP. This result is consistent with Hu et al.
422	(2016), which suggested that the increased zonal gradient of the SSTs over the
423	tropical Pacific could lead to a strengthened Pacific Walker circulation and an
424	enhanced upward motion over the TWP. Therefore, the warmer SSTs over the TWP
425	could contribute largely to the trend of the upward motion over the TWP in NDJFM
426	during 1958-2017.

The changes in the OLR <u>simulated in WACCM4</u> associated with the changes in the global SSTs are shown in Fig. <u>7</u>. There are significantly enhanced deep convection as indicated by OLR over the TWP due to the strengthened convergence in the Control <u>simulation</u>, while the deep convection shows weak and even decreasing trends over the TWP in the Fixsst <u>simulation (Figs. 7a and b)</u>. The enhanced deep

convection over the TWP could lead to the enhancing trends of the upward motion. 432 Hence, it can be inferred that the changes in the global SSTs are responsible for the 433 434 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 435 436 troposphere.

437

3.3 Implications for the concentrations of water vapor and CO in the TTL and lower stratosphere. 438

Previous studies showed that the enhanced deep convection and upward motion 439 440 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 441 the enhanced upward motion which could bring more wet air from low altitude to 442 443 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., 444 Highwood and Hoskins, 1998; Garfinkel et al., 2018; Pan et al., 2019). Hence, the 445 relationship between the intensity of upward motion and the water vapor 446 concentration in the UTLS is complex. Here, the relationship between the trends of 447 the upward motion over the TWP and the changes in CO and water vapor in the ULTS 448 simulated with WACCM4 are analyzed. 449 The trends of CPTT, the 100 hPa streamfunction, and the water vapor 450

- concentration are shown based on the Control and the Fixsst simulation as well as 451
- their difference in Figs. 7d-i. The changes in the deep convection could lead to the 452
- changes in the atmospheric circulation by releasing the latent heat. The changes in the 453

454	tropical deep convection lead to a Rossby-Kelvin wave response at the UTLS level
455	and then induce the changes in the air temperature near the tropopause (e.g., Gill,
456	1980; Highwood and Hoskins, 1998). The trends of the 100 hPa streamfunction show
457	a Rossby wave response over the TWP and a Kelvin wave response over the tropical
458	eastern Pacific in the Control simulation (Fig. 7d), which is caused by the changes in
459	the deep convection over the tropical Pacific. The Rossby-Kelvin wave response
460	further leads to the decrease the CPTT over the TWP and the increase of the CPTT
461	over the tropical eastern Pacific. Previous studies suggest that the lower stratospheric
462	water vapor is mainly influenced by the coldest temperature near the tropopause (e.g.,
463	Garfinkel et al., 2018; Zhou et al., 2021). Since the TWP has the coldest CPTT in the
464	boreal winter (e.g., Pan et al., 2016), the significantly decreased CPTT over the TWP
465	may result in significantly dried lower stratosphere (Fig. 7g). The intensity of the
466	upward motion over the TWP shows negative correlations with the concentration of
467	the tropical lower stratospheric water vapor (not shown). Hence, the enhanced upward
468	motion over the TWP may correspond to a dried lower stratosphere. The CPTT shows
469	weak trends over the TWP, and the tropical water vapor shows insignificant trends at
470	70 hPa in the Fixsst <u>simulation</u> . The comparison between the Control <u>simulation</u> and
471	the Fixsst simulation suggests that the trends of the deep convection, the CPTT, and
472	the lower stratospheric water vapor concentration in the tropics in NDJFM during
473	1958-2017 are dominated by the trends of the global SSTs, while other external
474	forcings may play minor roles.

475 Generally, the intensified <u>upward motion</u> may lead to more tropospheric trace

476	gases lifting to the upper troposphere and entering the lower stratosphere (e.g.,
477	Rosenlof, 2003; Lu et al., 2020). Here we use CO as a tropospheric tracer to detect the
478	possible influences of the enhanced upward motion over the TWP on the
479	transportation of the tropospheric trace gases to the upper troposphere and the lower
480	stratosphere. Due to the data limitation, it is not possible to show the corresponding
481	changes of trace gases by observations in NDJFM during 1958-2017. Here, the trends
482	of CO at around 200 hPa from MOPITT and MLS observations are shown in the Fig.
483	8. The CO increased significantly over the TWP in NDJFM in the upper troposphere
484	from the MOPITT (at 200 hPa during 2000-2017) and MLS data (at 215 hPa during
485	2005-2017). The concentration of MLS CO over the TWP is approximately 80 ppbv
486	at 215 hPa from MLS observations and 70 ppbv at 200 hPa from MOPITT
487	observations, which is consistent with previous study (e.g., Huang et al., 2016). The
488	MLS CO data show that the area-averaged CO increased approximately 2.0±3.7 ppbv
489	decade-1 over the TWP in NDJFM during 2005-2017. The area-averaged MOPITT CO
490	data show a stronger increase of approximately 5.0±3.1 ppbv decade-1 at 200 hPa
491	from 2000 to 2017 (significant at the 95% confidence level). It should be pointed out
492	that the linear trends of CO are calculated based on the satellite data which only cover
493	14 or 18 years due to the data limitation. Hence, the linear trends of CO may have
494	uncertainties particularly in the regions with large interannual variations. To partially
495	overcome this shortage, the trends of MLS CO at 215 hPa during time periods of
496	2005-2016, 2006-2016, 2006-2017, and 2007-2016 and the trends of MOPITT CO at
497	

are shown in Supplementary Fig. 6. It could be found that the CO in the upper 498 troposphere increased robustly over the TWP from both the MLS and MOPITT data. 499 Overall, though the observed CO only covers less than 20 years, the results from the 500 satellite data suggest a possible impact of the intensified upward motion over the 501 502 TWP on the trace gases in the upper troposphere. To further illustrate the impacts of the enhanced upward motion on the trace gas 503 in the upper troposphere and lower stratosphere, the Control and Fixsst simulations 504 with WACCM4 are used. The trends of the CO concentrations from the Control and 505 506 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 507 simulations at rates of 3.4 ppbv decade⁻¹ and 3.2 ppbv decade⁻¹, respectively, (Figs. 9a 508 509 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 510 trends at 150 hPa between the Control simulation and the Fixsst simulation are also 511 512 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 513 mainly caused by the changes in the atmospheric circulation induced by the changes 514 in the global SSTs. The difference of the CO concentration at 150 hPa between the 515 Control simulation and the Fixsst simulation shows a significantly increasing trend at 516 a rate of 0.2±0.1 ppbv decade⁻¹ over the TWP (significant at the 95% confidence 517 level). At the same time, decreasing trends over the central Africa exist, which 518 resembles to the trend patterns of the vertical velocity in the lower TTL and the deep 519

convection (Figs. 5i and 7c). This indicates that the enhanced deep convection in the 520 TWP lead to the strengthened upward motion over the TWP, which results in an extra 521 522 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). 523 524 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 525 then to the subtropics in the southern hemisphere through the southward flow during 526 boreal winter. The CO perturbation then spreads rapidly circling the globe following 527 528 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, 529 60°E-100°E) at a rate of approximately 0.2 m s⁻¹ decade⁻¹ and strengthened westerlies 530 531 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⁻¹ decade⁻¹ (Figs. 5c and f). 532

The trends of the zonal mean CO concentration from model simulations are 533 displayed in Figs. <u>10</u>a-c. The zonal mean CO shows significantly increasing trends at 534 535 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 536 significantly increasing trends in the TTL but negative trends in the middle 537 troposphere in the tropics and the Northern Hemisphere. At the same time, the 538 difference of CO concentration between the Control simulation and the Fixsst 539 simulation averaged in the western Pacific (100°E-180°E) shows significantly 540 increasing trends in the tropics (20° S- 10° N) from the surface to the TTL (Fig. <u>10</u>f). 541

542	The CO in the layer 150-70 hPa over the TWP increased 3.2 ppbv decade ⁻¹ and 2.8
543	ppbv decade ⁻¹ in the Control and Fixsst simulations in NDJFM during 1958-2017,
544	respectively. And the CO difference between the Control and Fixsst simulations
545	increased 0.4±0.2 ppbv decade ⁻¹ (significant at the 95% confidence level) in the layer
546	150-70 hPa over the TWP, which suggests that the intensifying upward motion over
547	the TWP and the tropical upwelling of BDC could lead to an extra 14% increasing
548	trend of CO. This indicates that the increased zonal mean CO in the TTL (Fig. <u>10</u> c) is
549	mainly transported through the western Pacific bands and highlights the importance of
550	the upward motion over the TWP in elevating trace gases from the surface to the
551	upper troposphere.

To understand the CO trends in the Control and Fixsst simulations and their 552 553 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 554 tropics while the w over the TWP intensified in the Control simulation in NDJFM 555 during 1958-2017. This is consistent with Fig. 5. While the SSTs fixed to 556 climatological values, the zonal mean w shows weak trends and the w over the TWP 557 shows significantly negative trends. The changes in the global SSTs therefore leads to 558 the increase of the w over the TWP region as indicated in the differences between the 559 two simulations in Fig. 11f. In summary, the CO shows increasing trends (3.5 ppby 560 decade-1) at 150 hPa over the TWP in NDJFM during 1958-2017 induced by the 561 changes in the surface emissions and the upward motion. The trends of CO at 150 hPa 562 over the TWP in NDJFM during 1958-2017 in the Fixsst simulation mainly include 563

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

586	over past decades as seen in the JRA55 data and the Control simulation (Figs. 12a and
587	12b). We found that the 70 hPa upward mass flux in NDJFM in the tropics
588	(15°S-15°N) increased 2.8±1.9% decade ⁻¹ (significant at the 95% confidence level) in
589	the JRA55 data from 1958 to 2017 (Fig. 12a) and 4.6±4.3% decade-1 (significant at
590	the 95% confidence level) in the MERRA2 data from 1980 to 2017 (Supplementary
591	Fig. 7b). From the ERA5 data, the 70 hPa upward mass flux in NDJFM increased in
592	the north hemisphere (0-15°N) at a rate of 5.0±2.8% decade ⁻¹ (significant at the 95%
593	confidence level), but decreased significantly in the south hemisphere (0-15°S) during
594	1958-2017 (Supplementary Fig. 7a). On average, the trend of the 70 hPa upward mass
595	flux in NDJFM in the tropics (15°S-15°N) is not significant in ERA5. In fact, many
596	previous studies have investigated the trends of BDC. For example, Abalos et al.
597	(2015) investigated the trends of BDC derived from JRA55, MERRA, and
598	ERA-Interim data during 1979-2012 and suggested that the BDC in JRA55 and
599	MERRA significantly strengthened throughout the layer 100-10 hPa with a rate of
600	2-5% decade ⁻¹ , while the BDC in ERA-Interim shows weakening trends. Diallo et al.
601	(2021) compared the trends of the BDC in the ERA5 and ERA-Interim during
602	1979-2018 and pointed out that the BDC in the ERA-Interim shows weakening trend
603	and the BDC in the ERA5 strengthened at a rate of 1.5% decade ⁻¹ which is more
604	consistent with other studies. In the present study, we only focus on the trend of the
605	BDC in the wintertime (NDJFM) in the tropics (15°S-15°N) during 1958-2017, which
606	may lead to some differences between our result and that in the previous studies.
607	Overall, the trends of the tropical upwelling of BDC derived from JRA55, MERRA2

608 data and the Control simulation are similar to that in previous studies using both

609 reanalysis datasets and model results (e.g., Butchart et al., 2010; Abalos et al., 2015;

610 Fu et al., 2019; Rao et al., 2019; Diallo et al., 2021). However, the tropical upwelling

- 611 of the BDC decreased in ERA5 data in the tropics (15°S-15°N), which are different
- 612 from the results in JRA55 and MERRA2.

In the Fixsst simulation, the trend of w^* is much weaker and not significant in 613 most areas. The changes in the global SSTs therefore play an important role in the 614 intensification of the shallow branch of the BDC as shown by the differences between 615 616 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 617 simulations, although there are some uncertainties since the ERA5 data show a 618 619 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. 620 9c and 10f may be due to a combined effect of the strengthened tropical upwelling of 621 622 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 623 the upper troposphere, works together with the strengthened BD circulation under 624 global warming may lead to an increase of tropospheric trace gases over the TWP in 625 the lower stratosphere. 626

627 **4 Summary and Discussion**

628 The recent trends of the upward motion from the lower to the upper troposphere 629 in boreal winter over the TWP is investigated for the first time based on the JRA55,

630	ERA5, MERRA2 datasets and four WACCM4 simulations (more details could be
631	found in Section 2). The upward motion at 150 hPa over the TWP in NDJFM
632	increased 8±3.1% decade-1 and 3.6±3.3% decade-1 in NDJFM from 1958 to 2017 in
633	JRA55 and ERA5 reanalysis datasets, respectively. Despite the possible
634	discontinuities between the radiosonde era (after 1958) and the satellite era (after
635	1979), the upward motion at 150 hPa over the TWP in NDJFM increased 7.5±7.1%
636	decade-1 during 1980-2017 in MERRA2 data. Such intensification of the upward
637	motion over the TWP also exist in the middle- and lower-troposphere in NDJFM in
638	JRA55, ERA5, and MERRA2, which can be confirmed by the WACCM4 model
639	simulations. Comparing the results between the Control and Fixsst simulations with
640	WACCM4, it is found that the trend of the upward motion over the TWP is closely
641	related to the changes in global SSTs, especially the SST warming over the eastern
642	maritime continent and tropical western Pacific (see the results from the experiments
643	<u>R1 and R2 in Fig. 7</u>). Warmer SSTs over the <u>eastern maritime continent and tropical</u>
644	western Pacific (approximately 0.5 K) lead to a strengthened Pacific Walker
645	circulation, enhanced deep convection and approximately 27% intensified upward
646	motion at 150 hPa over the TWP as shown by the results from the experiments R1 and
647	<u>R2</u> . The enhanced deep convection over the TWP could lead to a dryer lower
648	stratosphere over the TWP, as the strong upward motion and the Rossby-Kelvin wave
649	responses induce a colder tropopause over the TWP. It should be pointed out that the
650	results in the present study are mainly based on the reanalyses data, and some
651	uncertainties may exist. More observational data are expected to be used to obtain a

652 <u>more robust result in the future.</u>

653	Results from the Control simulation indicate that the CO concentrations
654	increased significantly from the surface to the stratosphere over the TWP. The CO at
655	150 hPa increased at a rate of approximately 3.4 ppbv decade ⁻¹ with increased surface
656	emissions and the enhanced upward motion over the TWP. Specifically, an
657	enhancement of tropospheric upward motion and subsequent upward transport of
658	trace gases over the TWP lead to an extra 6% increasing trend of CO concentrations
659	in the upper troposphere.
660	Furthermore, the upward mass fluxes at 70 hPa in the tropics (15°S-15°N) show
661	strengthening trends at rates of 2.8±1.9% decade-1 and 4.6±4.3% decade-1 in JRA55
662	data (during 1958-2017) and MERRA2 data (during 1980-2017) in NDJFM,
663	respectively, which is consistent with previous studies (e.g., Butchart et al., 2010; Fu
664	et al., 2019; Rao et al., 2019). However, such enhancement in tropical upward mass
665	flux at 70 hPa has large uncertainties since the ERA5 data show a negative and
666	insignificant trend (Supplementary Fig. 7a). The results from the Control and Fixsst
667	simulations indicate that the elevated CO in the upper troposphere is further uplifted
668	to the lower stratosphere by the intensified tropical upwelling of the BD circulation
669	due mainly to global SST warming and lead to an increase of CO in the lower
670	stratosphere. An extra 14% increasing trend of CO at the layer 150-70 hPa over the
671	TWP is derived from the Control and Fixsst simulations.
672	Tropospheric trace gases and aerosols have important impacts on the

673 stratospheric processes if they enter the stratosphere. For example, ozone-depleting

674	substances, CH ₄ and N ₂ O could influence on the stratospheric ozone significantly
675	(e.g., Shindell et al., 2013; Wang et al., 2014; WMO, 2018), which also modify the
676	temperature in the stratosphere significantly through their strong radiative effects.
677	Water vapor in the lower stratosphere, in particular, has a significant warming effect
678	on the surface climate (Solomon et al., 2010). Therefore, changes of trace gases in the
679	UTLS have important impacts on both tropospheric and stratospheric climate. Our
680	results suggest that the upward motion over the TWP and the vertical component of
681	the BDC at the lower stratosphere level have been intensified. These results suggest
682	that the emission from the maritime continent and surrounding areas may play a more
683	important role in the stratospheric processes and the global climate. In addition, more
684	very short lived substances emitted from the tropical ocean could be elevated to the
685	TTL by the enhanced convection and then transported into the stratosphere by the
686	large-scale uplifts and exert important effects on the stratospheric chemistry. However,
687	the quantitative impacts of the intensified upward motion over the TWP on
688	tropospheric and stratospheric trace gases and aerosols and their climate feedbacks
689	await further investigation using more observations and model simulations.
690	
691	Competing interests. The authors declare that they have no conflict of interest.
692	
693	Author contributions. WT designed the study. WW provided suggestions about the
694	statistical methods and model simulations. KQ ran the models and wrote the first draft.

695 RH, MX, and TW contributed to the manuscript writing. YP provided the data used in

696 the study. All authors contributed to the improvement of the results.

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- The SST data is obtained from HadISST:
- 707 https://www.metoffice.gov.uk/hadobs/hadisst/data/download.html.
- 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.
- The OLR data are from https://psl.noaa.gov/data/gridded/data.interp_OLR.html.
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1033 Figure captions:

- 1034Fig. 1. The climatological mean (averaged over 1958-2017) values of 150 hPa w (10-21035m s-1) in different months derived from the JRA55 data.
- 1036 Fig. 2. Trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, 700 hPa
- 1037 in NDJFM derived from JRA55, ERA5, and MERRA2 data. The trends of horizontal
- 1038 winds (arrows, units: 10^{-1} m s⁻¹ a⁻¹) and vertical velocity (shading, units: 10^{-4} m s⁻¹ a⁻¹)
- 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 <u>1980-2017 and the wind field data are from MERRA2. The vertical velocity trends</u>
- 1043 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 (<u>20°S-10°N, 100°E-180°E)</u>.

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.

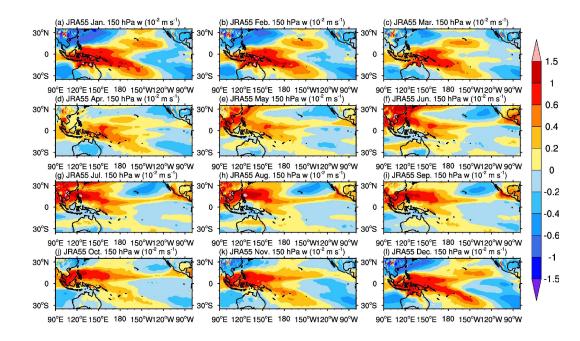
- 1049 The straight lines in each figure indicate the linear trends. The solid lines denote the
- 1050 linear trends are significant at the 95% confidence level, while the dashed lines denote
- 1051 the linear trends are significant at the 90% confidence level.
- 1052 Fig. 4. Trends of (a) observed outgoing longwave radiation (OLR, units: W m⁻² a⁻¹)
- 1053 provided by NOAA during 1974-2017; (b) cold-point tropopause temperature (CPTT,
- 1054 units: 10⁻¹ K a⁻¹) derived from JRA55 data and (c) SST (K a⁻¹) derived from HadISST
- 1055 during 1958-2017 in NDJFM. (d) The correlation coefficients between the intensity of
- 1056 the upward motion at 150 hPa over the TWP and SST in NDJFM during 1958-2017
- 1057 with the linear trends removed. The trends and correlation coefficients over the dotted
- 1058 regions are statistically significant at the 95% confidence level. The black rectangles
- 1059 denote the TWP region $(20^{\circ}\text{S}-10^{\circ}\text{N}, 100^{\circ}\text{E}-180^{\circ}\text{E})$.
- 1060 Fig. 5. The trends of vertical velocity and horizontal winds at 150 hPa, 500 hPa, and
- 1061 <u>700 hPa in NDJFM during 1958-2017 in the Control and Fixsst simulations as well as</u>
- 1062 their difference. The trends of 150 hPa w (shading, units: 10⁻⁴ m s⁻¹ a⁻¹) and horizontal
- 1063 winds (arrows; 10⁻¹ m s⁻¹ a⁻¹) from (a) Control simulation; (b) Fixsst simulation; and
- 1064 (c) difference between the Control simulation and the Fixsst simulation in NDJFM
- 1065 during 1958-2017. (d)-(f) are similar to (a)-(c) but for the results at 500 hPa. (g)-(i)
- 1066 are similar to (d)-(f) but for the results at 700 hPa. The vertical velocity trends over
- 1067 the dotted regions are statistically significant at the 95% confidence level. The black
- 1068 rectangles denote the TWP region (20°S-10°N, 100°E-180°E).
- 1069 Fig. 6. The difference of vertical velocity (shading, units: 10⁻² m s⁻¹) and horizontal

1070	winds (arrows, units: m s ⁻¹) at (a) 150 hPa; (b) 500 hPa; and (c) 700 hPa in NDJFM
1071	between experiments R1 and R2. The differences between vertical velocity over the
1072	dotted regions are statistically significant at the 95% confidence level. The black
1073	rectangles denote the TWP region (20°S-10°N, 100°E-180°E).
1074	Fig. 7. Trends of OLR (W m ⁻² a ⁻¹) (a)-(c), CPTT (shading, units: 10 ⁻¹ K a ⁻¹) and 100
1075	hPa streamfunction (contour, units: 10 ⁶ m ² s ⁻¹ a ⁻¹) (d)-(f), and 70 hPa water vapor
1076	concentration (units: 10 ⁻² ppmv a ⁻¹) (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
1078	results in the Control simulation; (b), (e), and (h) are the results in the Fixsst
1079	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
1081	statistically significant at the 95% confidence level. The CPTT trends in (d)-(f) over
1082	the dotted regions are statistically significant at the 95% confidence level. The black
1083	rectangles denote the TWP region (20°S-10°N, 100°E-180°E).
1084	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
1086	trends of CO (10 ⁻¹ ppbv a ⁻¹) 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 ⁻⁴ ppmv a ⁻¹) from (a) Control
1090	simulation; (b) Fixsst simulation; and (c) difference between the Control simulation
1091	and the Fixsst simulation in NDJFM during 1958-2017. The trends in (a)-(c) over the

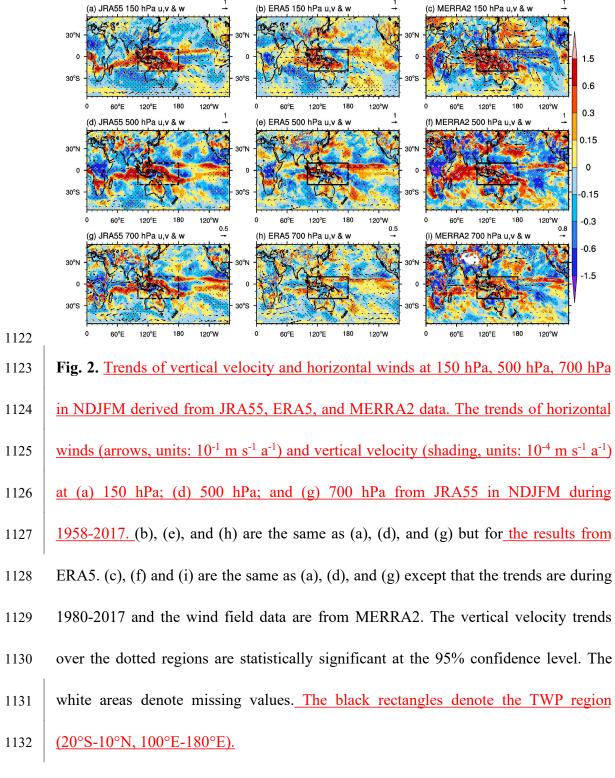
- 1092 dotted regions are statistically significant at the 95% confidence level. The black
 1093 rectangles denote the TWP region (20°S-10°N, 100°E-180°E).
- 1094 Fig. 10. Latitude-pressure cross sections of the trends of (a)-(c) zonal mean CO
- 1095 concentration (10⁻⁴ ppmv a⁻¹) and (d)-(f) CO concentration (10⁻⁴ ppmv a⁻¹) over the
- 1096 TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and
- 1097 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
- 1099 the results derived from the difference between the Control and Fixsst simulations.
- 1100 The trends over the dotted regions are statistically significant at the 95% confidence
- 1101 <u>level.</u>
- 1102 Fig. 11. Latitude-pressure cross sections of the trends of (a)-(c) the zonal mean w (10⁻⁴)
- 1103 <u>m s⁻¹ a⁻¹</u>) and $v (10^{-1} \text{ m s}^{-1} \text{ a}^{-1})$ and (d)-(f) $w (10^{-4} \text{ m s}^{-1} \text{ a}^{-1})$ and $v (10^{-1} \text{ m s}^{-1} \text{ a}^{-1})$ over
- 1104 the TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and
- 1105 Fixsst simulation as well as their difference. (a) and (d) are the trends of w and v in
- 1106 the Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f)
- 1107 are the results derived from the difference between the Control and Fixsst simulations.
- 1108 The shadings denote the trends of the w (10⁻⁴ m s⁻¹ a⁻¹). The trends over the dotted
- 1109 regions are statistically significant at the 90% confidence level.
- 1110 Fig. 12. Trends of the BDC (vectors, units in the horizontal and vertical components
- 1111 are 10⁻² and 10⁻⁵ m s⁻¹ a⁻¹, respectively) calculated using the TEM formula from (a)
- 1112 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

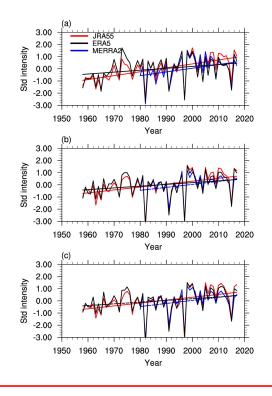
- 1114 shadings are the trends of the w^* (10⁻⁵ m s⁻¹ a⁻¹). The trends of the vertical velocity
- 1115 over the dotted regions are statistically significant at the 90% confidence level.

1117 Figures



- **Fig. 1.** The climatological mean (averaged over 1958-2017) values of 150 hPa w (10⁻²
- 1120 m s⁻¹) in different months <u>derived from</u> the JRA55 reanalysis data.









1136 Fig. 3. The time series of the standardized intensity of the upward motion over the

1137 tropical western Pacific (20°S-10°N, 100°E-180°E) at (a) 150 hPa; (b) 500 hPa; and

1138 (c) 700 hPa extracted from JRA55 (red), ERA5 (black) and MERRA2 (blue) datasets.

1139 The straight lines in each figure indicate the linear trends. The solid lines denote the

- 1140 linear trends are significant at the 95% confidence level, while the dashed lines denote
- 1141 the linear trends are significant at the 90% confidence level.
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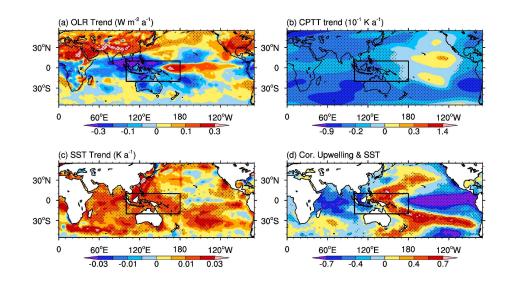
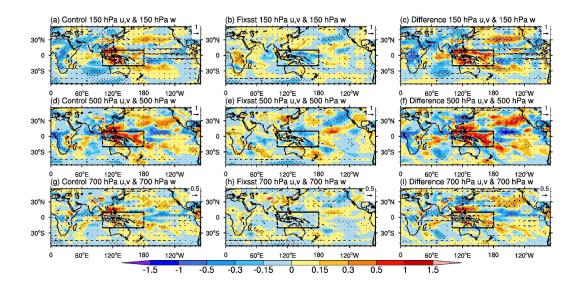
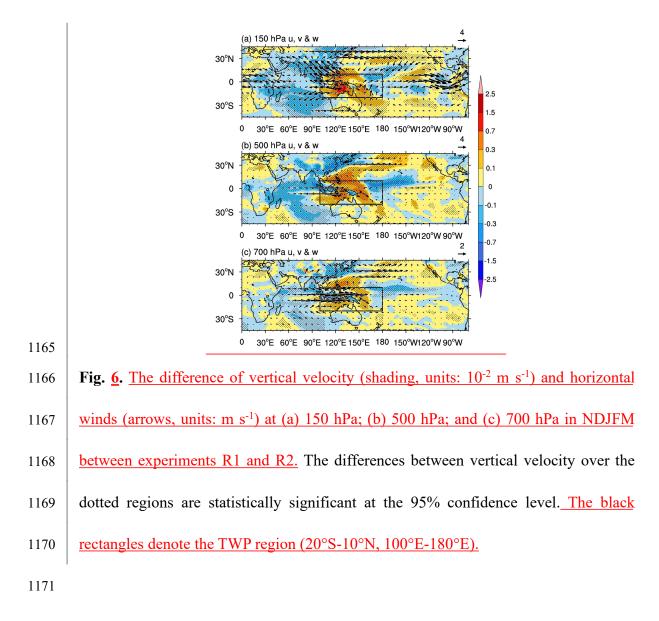
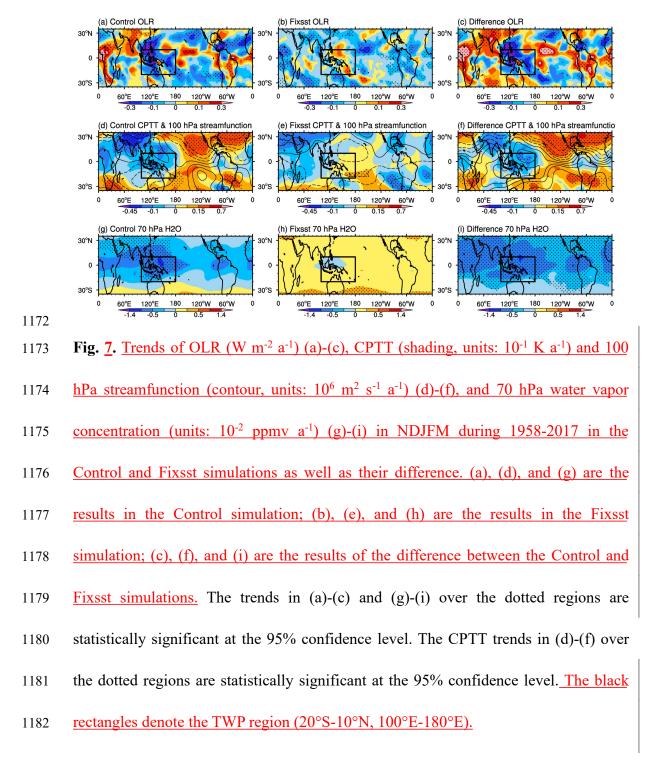


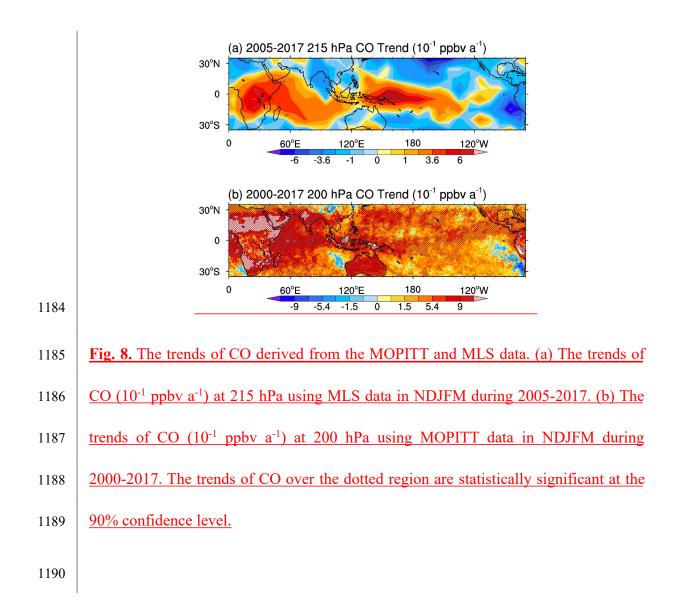
Fig. 4. Trends of (a) observed outgoing longwave radiation (OLR, units: W m⁻² a⁻¹) 1144 provided by NOAA during 1974-2017; (b) cold-point tropopause temperature (CPTT, 1145 1146 units: 10⁻¹ K a⁻¹) derived from JRA55 data and (c) SST (K a⁻¹) derived from HadISST during 1958-2017 in NDJFM. (d) The correlation coefficients between the intensity of 1147 the upward motion at 150 hPa over the TWP and SST in NDJFM during 1958-2017 1148 1149 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 1150 denote the TWP region (20°S-10°N, 100°E-180°E). 1151 1152



1154 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 1155 their difference. The trends of 150 hPa w (shading, units: 10⁻⁴ m s⁻¹ a⁻¹) and horizontal 1156 winds (arrows; 10⁻¹ m s⁻¹ a⁻¹) from (a) Control simulation; (b) Fixsst simulation; and 1157 (c) difference between the Control simulation and the Fixsst simulation in NDJFM 1158 1159 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 1160 1161 the dotted regions are statistically significant at the 95% confidence level. The black 1162 rectangles denote the TWP region (20°S-10°N, 100°E-180°E).







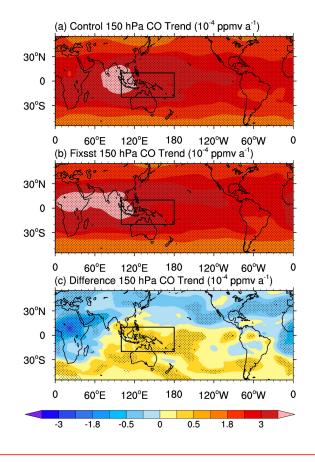
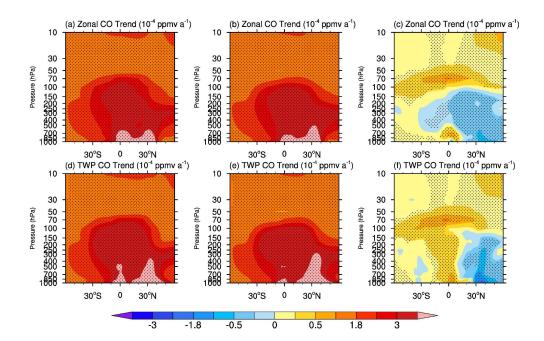


Fig. 9. The trends of 150 hPa CO concentration (10⁻⁴ ppmv a⁻¹) from (a) Control simulation; (b) Fixsst simulation; and (c) difference between the Control simulation and the Fixsst simulation in NDJFM during 1958-2017. The trends in (a)-(c) over the dotted regions are statistically significant at the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

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1202 Fig. 10. Latitude-pressure cross sections of the trends of (a)-(c) zonal mean CO concentration (10⁻⁴ ppmv a⁻¹) and (d)-(f) CO concentration (10⁻⁴ ppmv a⁻¹) over the 1203 TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and 1204 Fixsst simulation as well as their difference. (a) and (d) are the CO trends in the 1205 Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f) are 1206 the results derived from the difference between the Control and Fixsst simulations. 1207 The trends over the dotted regions are statistically significant at the 95% confidence 1208 level. 1209

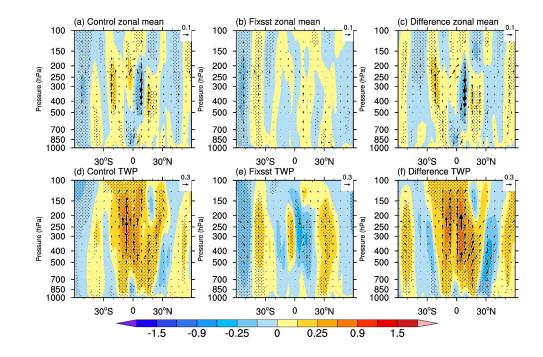


Fig. <u>11</u>. Latitude-pressure cross sections of the trends of (a)-(c) the zonal mean w (10⁻⁴ 1212 <u>m s⁻¹ a⁻¹</u>) and v (10⁻¹ m s⁻¹ a⁻¹) and (d)-(f) w (10⁻⁴ m s⁻¹ a⁻¹) and v (10⁻¹ m s⁻¹ a⁻¹) over 1213 the TWP (100°E-180°E) in NDJFM during 1958-2017 in the Control simulation and 1214 Fixsst simulation as well as their difference. (a) and (d) are the trends of w and v in 1215 the Control simulation. (b) and (e) are the results in the Fixsst simulation. (c) and (f) 1216 1217 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⁻¹ a⁻¹). The trends over the dotted 1218 regions are statistically significant at the 90% confidence level. 1219

