



## Characteristics of intercontinental transport of tropospheric ozone from Africa to Asia

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**Abstract.** Based on 20-year simulations using a global chemical transport model, GEOS-Chem, and a trajectory model, HYSPLIT, the transport of ozone produced in the African troposphere to Asia is investigated. The study shows that the influence of African ozone on Asia varies largely in time and space. In the middle and upper troposphere, the inflow of African ozone to Asia peaks around 25°N, being the largest in boreal winter and early spring (>10 ppbv) and the lowest in boreal summer (<6 ppbv). In the lower troposphere, imported African ozone ranges 2-6 ppbv over Asia, being highest in boreal winter (~4 ppbv) and lowest in boreal fall (~2 ppbv). The seasonality of the transport mainly results from the seasonal swing of the Hadley circulation and the Intertropical Convergence Zone (ITCZ) in Africa and subtropical westerlies along the transport pathways. These meteorological conditions combined with the seasonality of ozone precursor emissions from various sources in Africa can modulate the influence of African ozone on Asia. Ozone from the northern hemispheric Africa accounts for over 70% of the total African ozone over Asia in most altitudes and seasons. The interhemispheric

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transport of ozone from the southern hemispheric Africa is observed most evidently in boreal winter  
25 over the Asian upper troposphere and in boreal summer over the Asian lower troposphere. The former is  
facilitated by the convective divergence in the upper troposphere over the ITCZ in Africa, while the  
latter is driven by the Somali jet. In boreal winter, the intensity of the ITCZ in Africa is positively  
correlated with the interannual variation of imported African ozone over Asia, because a stronger ITCZ  
can uplift more surface ozone and ozone precursors and enhance lightning NO<sub>x</sub> generation, resulting in  
30 more African ozone transported to Asia.

## 1 Introduction

Tropospheric ozone is a major air pollutant, harmful to human health (Anenberg et al., 2010),  
agricultural crops, and natural ecosystems (Hollaway et al., 2012; Lefohn et al., 2017). It also acts as a  
35 greenhouse gas, whose global mean radiative forcing is about  $0.4 \pm 0.2 \text{ W/m}^2$  (Myhre et al., 2013). The  
dominant source of tropospheric ozone is from the photochemical reactions in which volatile organic  
compounds (VOCs) and carbon monoxide (CO) are oxidized in the presence of nitrogen oxides (NO<sub>x</sub>)  
(Lelieveld and Dentener, 2000). Transport of ozone from the stratosphere downward is another  
important source of tropospheric ozone (Neu et al., 2014; Akritidis et al., 2016). Due to its relatively  
40 long lifetime (days to weeks in the free troposphere), ozone can be transported over long distances  
across continents, as seen in a wealth of observation evidence (Huntrieser et al., 2005; Lewis et al.,  
2007; Rastigejev et al., 2010; Verstraeten et al., 2015). Consequently, tropospheric ozone in a region is  
greatly influenced by ozone transport from upwind regions (Doherty et al., 2013; Derwent et al., 2015).  
Therefore, intercontinental transport of ozone has been a significant issue to scientists, public, and  
45 policymakers concerned with air quality and climate change (HTAP, 2010; Cooper et al., 2015; Doherty,  
2015).

There have been numerous studies on the three major source-receptor relationships in the Northern



Hemisphere for ozone transport among Asia, North America, and Europe, which are trans-Pacific (Jaffe et al., 2003; Zhao et al., 2003; Cooper et al., 2010), trans-Atlantic (Wild et al., 1996; Cooper et al., 2002; 50 Guerova et al., 2006), and trans-Eurasian (Wild et al., 2004; Fiore et al., 2009; Li et al., 2014). The trans-Pacific transport from Asia to North America has been studied most because of high pollutant emissions in Asia and their efficient eastward export (Cooper et al., 2010, 2011, 2015; Verstraeten et al., 2015; Huang et al., 2017; Lin et al., 2017).

Transport of ozone to Asia from various source regions in the world has received some attention 55 recently (Holloway et al., 2008; Nagashima et al., 2010; Wang et al., 2011; Li et al., 2014; Zhu et al., 2016; Zhu et al., 2017b). Imported ozone, its seasonal variation, and spatial distribution depend on numerous processes, involving chemistry (Wild et al., 2004; Li et al., 2014), and meteorology in Asia, the source regions, and along the transport pathways. Previous studies illustrated the important role that meteorology plays in these processes (Sudo and Akimoto, 2007; Sekiya and Sudo, 2012, 2014; Zhu et 60 al., 2017b). Governed by the westerlies in the Northern Hemisphere, the influence of European and North American ozone is larger in higher latitudes than in lower altitudes of Asia (Wild et al., 2004; Holloway et al., 2008; Zhu et al., 2017b). The seasonal switch in Asian monsoonal winds significantly affects the seasonal variation of ozone transported to Asia (Nagashima et al., 2010; Wang et al., 2011; Zhu et al., 2017b). The Asian monsoon anticyclone (South Asian High, SAH) has remarkable effects on 65 the upper tropospheric ozone transport over Eurasia (Vogel et al., 2014; Garny and Randel, 2016).

Africa covers areas from the Northern Hemisphere to the Southern Hemisphere; around three quarters of the continent are located in the tropics. Comparing with other continents, Africa has the most frequent lightning (Albrecht et al., 2016) and the largest burned areas (Giglio et al., 2013). Over 70% of tropospheric ozone produced in Africa from natural and anthropogenic sources is exported out of Africa 70 (Aghedo et al., 2007), making African ozone an obvious influence on the global tropospheric ozone budget (Piketh and Walton, 2004; Thompson, 2004; Williams et al., 2009; Bouarar et al., 2011; Zare et



al., 2014). There have been a few studies on the transport of African ozone to some locations in Asia (Liu et al., 2002; Sudo and Akimoto, 2007; Lal et al., 2014). Liu et al. (2002) found that African outflow makes a contribution of about 10-20 ppbv to ozone in the middle and upper troposphere over Hong Kong, southeast of China, during the November-April period. Sudo and Akimoto (2007) showed that there is a significant interhemispheric ozone transport from South America to Japan in the upper troposphere in conjunction with ozone export from northern Africa. Lal et al. (2014) reported that the ozone distribution over western India in the lower troposphere during the summer monsoon seasons is affected by long-range transport from the east coast of Africa. Nevertheless, questions still remain on how and how much African ozone is transported to Asia, the transport pathways and their seasonality, as well as underlying mechanisms for the transport.

The Intertropical Convergence Zone (ITCZ), defined as the convergence of the trade winds, is one of the most prominent meteorological phenomena in the tropics (Waliser and Gautier, 1993; Žagar et al., 2011; Suzuki, 2011). ITCZ is a heat engine driving the ascending branch of the Hadley circulation (Nicholson, 2009; Suzuki, 2011). ITCZ and its seasonality can significantly impact the meteorological conditions over Africa (Nicholson, 2009; Collier and Hughes, 2011; Suzuki, 2011). Characterized with deep and strong convection, the equatorial region between the ITCZ branches is a region with effective interhemispheric mixing (Avery et al., 2001). Meanwhile, the ITCZ can be a barrier to interhemispheric exchange in the lower troposphere (Raper et al., 2001). The convective divergence in the upper troposphere over the ITCZ was proposed to be one of the primary mechanisms for the interhemispheric transport (Hartley and Black, 1995; Avery et al., 2001). Based on aircraft and satellite observations, an anti-correlation was found between ozone abundances and convective outflow over central American ITCZ region by Avery et al. (2010). Together with the Southeast Asian monsoon, the movement of the ITCZ can modulate pollution transport to Southeast Asia (Pochanart et al., 2003, 2004). In addition, the ITCZ over western Africa is found to play a significant role in controlling the transport of Saharan



mineral dust to northern South America, modifying its air quality and climate (Piketh and Walton, 2004; Doherty et al., 2012, 2014; Rodríguez et al., 2015). However, the influences of the ITCZ on ozone transport from Africa to Asia have not been well understood.

In this study, a comprehensive investigation is made on the transport of African ozone to Asia through integrated numerical analysis of tagged tracer and trajectory simulations. African ozone here refers to ozone generated in the African troposphere below the tropopause from all natural and anthropogenic sources, as Sudo and Akimoto (2007) suggested that investigation of ozone production from both sources is necessary. Our specific objectives are (1) to characterize the seasonal variation of ozone transport from Africa to Asia, (2) to investigate the underlying mechanisms responsible for such seasonal variation, and (3) to explore a possible connection between the ITCZ and the interannual variation of imported African ozone over Asia. Our analysis is based on the simulations from a 3-dimensional global chemical transport model, GEOS-Chem (Bey et al., 2001a), for 20 years from 1987 to 2006 and simulations from a trajectory climatology for the same period from the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Draxler and Hess, 1998; Stein et al., 2015). In section 2, we describe the two models and the meteorological data used in this study. The validation of GEOS-Chem simulations is also presented in section 2. We address the first objective in section 3 and the second and third in section 4. Section 5 offers an overview of the processes involved in the transport of African ozone to Asia. Finally, the conclusions are drawn in section 6.

## 2 Methodology

### 2.1 The GEOS-Chem model and its validation

A global 3-dimensional chemistry transport model, GEOS-Chem (version v9-02) (Bey et al., 2001a, <http://geos-chem.org>), is employed to simulate the global ozone distributions and the source-receptor relationships between Africa and Asia. GEOS-Chem includes a detailed simulation of tropospheric O<sub>3</sub>-



120 NO<sub>x</sub>-hydrocarbon-aerosol. It is driven by assimilated meteorological data from the GEOS at NASA  
Global Modeling and Assimilation Office (GMAO). The model has been used extensively in studying  
pollution transport (Bey et al., 2001b; Koumoutsaris et al., 2008; Zhang et al., 2008; Liu et al., 2011;  
Wang et al., 2011; Ridder et al., 2012; Long et al., 2015; Jiang et al., 2016; Huang et al., 2017; Ikeda et  
al., 2017; Zhu et al., 2017a; Zhu et al., 2017b).

125 In this version of GEOS-Chem, the global anthropogenic emission inventories are from EDGAR 3.2  
for NO<sub>x</sub>, CO, SO<sub>x</sub> (Olivier and Berdowski, 2001) and RETRO for VOC emissions (Pulles et al., 2007),  
with the following regional inventories: the US Environmental Protection Agency National Emission  
Inventory 2005 (NEI05) for North America, the Cooperative Programme for Monitoring and Evaluation  
of the Long-range Transmission of Air Pollutants in Europe (EMEP) inventory for Europe (Vestreng  
130 and Klein, 2002), Big Bend Regional Aerosol and Visibility Observational (BRAVO) Study Emissions  
Inventory for 1999 in Mexico (Kuhns et al., 2003), and the Criteria Air Contaminants (CAC) inventory  
for Canada. The emission inventories for China and SE Asia are based on Streets et al. (2003, 2006).  
Biofuel emission inventory is from Yevich and Logan (2003). Biomass burning and Biogenic emissions  
are from the GFED3 inventory (van der Werf et al., 2010) and MEGAN 2.1 (Guenther et al., 2012)  
135 respectively. Lightning NO<sub>x</sub> emissions are calculated with the scheme of Allen et al. (2010) and the  
vertical distribution suggested by Ott et al. (2010).

To investigate the influence of African ozone on Asia, we used the standard mode for tagged ozone  
simulation in GEOS-Chem (Wang et al., 1998; Fiore et al., 2002; Zhang et al., 2008). The simulation is  
driven by GEOS-4 meteorology, which uses the deep convection scheme of Zhang and McFarlane  
140 (1995) and the shallow convection scheme of Hack (1994). The simulation covers a period from  
January 1986 to December 2006 (using 1986 for model spin-up) at 4° latitude by 5° longitude horizontal  
resolution with 30 vertical layers, 17 levels in the troposphere. Transport and chemical timesteps used in  
the study are 10 minutes and 20 minutes, respectively, as suggested by Philip et al. (2016). The tagged



ozone simulation calculates the contributions of the source regions to the receptor region using ozone  
145 production rates and loss frequencies of odd oxygen defined in GEOS-Chem ( $O_x = O_3 + NO_2 + 2NO_3 + 3N_2O_5 + HNO_3 + HNO_4 + PAN + PMN + PPN$ , Fiore et al., 2002; Zhang et al., 2008). Since  
ozone accounts for most of  $O_x$ , we refer to ozone instead of  $O_x$  for clarity. The ozone production rates  
and loss frequencies were generated and archived from a full chemistry simulation before the tagged  
simulation (Zhang et al., 2008, Liu et al., 2011). The source region, Africa, is defined as the region of  
150 35°S-15°N, 20°W-55°E and 15°N-35°N, 20°W-30°E. Therefore, ozone produced in Africa below the  
tropopause is tagged as a tracer. We further divided Africa into northern hemispheric Africa (NHAF)  
and southern hemispheric Africa (SHAF) separated by the equator and thus added two more tracers. The  
receptor region, Asia, is defined as the region of 5°N-40°N, 60°E-145°E. In the simulation, seasonal  
variation of chemistry and meteorology are both considered. For the interannual variation, our purpose  
155 is focused on the impact of meteorology on the transport of African ozone to Asia. Therefore, the  
meteorology was set to vary from year to year, while a fixed chemistry was used for the 20-year  
simulation, i. e., with the archived daily ozone production and loss data in 2005, similar to the  
treatments in previous studies that examined the influences of meteorology on pollution transport (Liu  
et al., 2011; Zhao et al., 2012; Sekiya and Sudo, 2014; Zhu et al., 2017b).

160 To quantify accurately the contribution of different sources to ozone abundances in a receptor region  
using a numerical model, a good agreement between the model simulations and observations is required  
(Cooper et al., 2015; Derwent et al., 2016). Although the GEOS-Chem simulations have been  
extensively validated (Zhang et al., 2008; Liu et al., 2009; Wang et al., 2011; Kim et al., 2015; Zhu et al.,  
2017a; Zhu et al., 2017b), we specifically validated the model in Africa for an enhanced confidence on  
165 the model performance, using the ozonesonde measurements acquired from the World Ozone and  
Ultraviolet Radiation Data Centre (WOUDC) (<http://www.woudc.org/home.php>). We selected three  
ozonesonde stations at Santa Cruz in North Africa (28.42°N, 16.26°W, 36 m), Nairobi in East Africa



(1.27°S, 36,8°E, 1745 m) and Irene in South Africa (25.91°S, 28.21°E, 1524 m). The ozonesonde observations have been used in studies on African tropospheric ozone widely (Clain et al., 2009; Thompson et al., 2012, 2014). Fig. 1 shows the vertical ozone profiles averaged from 1990 to 2003 for Santa Cruz, from 1996 to 2006 for Nairobi, and from 1990 to 2006 for Irene by season. The simulations and the observations exhibit good agreements. However, the model underestimates upper tropospheric ozone in Nairobi in the four seasons and somewhat overestimates ozone near the surface in Irene.

## 2.2 The HYSPLIT trajectory model

We use HYSPLIT\_4 (Hybrid Single-Particle Lagrangian Integrated Trajectory, version 4) (Draxler and Hess, 1998; Stein et al., 2015) forward trajectories to show the outflows from Africa. With powerful computational capabilities, the HYSPLIT model is one of the most extensively used atmospheric transport and dispersion models (Fleming et al., 2012). Meteorological inputs to HYSPLIT are the NCEP reanalysis at a resolution of  $2.5^{\circ} \times 2.5^{\circ}$  (<http://ready.arl.noaa.gov/archives.php>). Six-day forward trajectories were calculated from 1987 to 2006 four times a day (00, 06, 12 and 18 UTC) at four sites, Cairo (31°E, 30°N, in North Africa), Abuja (7.4°E, 9°N, in West Africa), Dar es Salaam (39°E, 6.8°S, in East Africa) and Johannesburg (28°E, 26.2°S, in South Africa). The level at 1.5 km, 5.5 km, and 11 km are selected to represent the lower, middle and the upper troposphere, respectively. The results were clustered for each month.

## 2.3 Meteorological data

The meteorological data include the NCEP/NCAR reanalysis I (Kalnay et al., 1996). The daily wind fields are used to describe the climatology of atmospheric circulation during the study period from 1987 to 2006. The product is available on a  $2.5^{\circ} \times 2.5^{\circ}$  horizontal grid at 17 pressure levels from 1000 hPa to



10 hPa (<https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.html>).

Additionally, the monthly Outgoing Longwave Radiation (OLR) data from NCAR at  $2.5^{\circ} \times 2.5^{\circ}$  with temporal interpolation (Liebmann and Smith, 1996, [https://www.esrl.noaa.gov/psd/data/gridded/data.interp\\_OLR.html](https://www.esrl.noaa.gov/psd/data/gridded/data.interp_OLR.html)) are used to indicate the intensity of the convection over ITCZ. The dataset has been widely used for tropical studies on deep convection and rainfall (Mounier and Janicot, 2004).

### 3 Seasonal variation in imported African ozone over Asia

Because a substantial amount of ozone and ozone precursors are produced or emitted in Africa, tropospheric ozone in the other continents is largely influenced by ozone outflow from Africa (Williams et al., 2009; Zare et al., 2014). Fig. 2 shows seasonal variations of imported African ozone in the Asian troposphere, varying with latitude and altitude. The values are the 20-year means (1987-2006) from the GEOS-Chem simulation. The largest African influences appear in the Asian middle and upper troposphere, i.e., 2-16 ppbv at 500 and 700 hPa. African ozone in the Asian lower troposphere is reduced to between 2 to 10 ppbv at 700 hPa throughout the year. Unlike imported European and North American ozone that influences Asia mostly over high altitudes (north of  $30^{\circ}\text{N}$ ) (Wild et al., 2004; Holloway et al., 2008; Nagashima et al., 2010; Zhu et al., 2017b), imported African ozone prevails over lower latitudes, generally south to  $40^{\circ}\text{N}$ , considerably larger than European and North American ozone south of  $30^{\circ}\text{N}$  (Wild et al., 2004; Sudo and Akimoto, 2007; Zhu et al., 2017b). North of  $50^{\circ}\text{N}$ , African ozone influence is small, i.e., less than 4 ppbv and 2 ppbv, respectively, above and below the Asian middle troposphere. Seasonally, imported African ozone in the middle and upper troposphere peaks in March  $25^{\circ}\text{N}$  ( $\sim 16$  ppbv, Figs. 2a and 2b) and is at its minimum in July. Owing to the high radiative forcing efficiency, the change in ozone concentrations in the upper troposphere can impact climate more significantly than that in the lower troposphere (Lacis et al., 1990). Therefore, the influence of African ozone on the climate in southern Asia is likely larger than that of European and North American ozone



(Aghedo et al., 2007; Sudo and Akimoto, 2007). In the lower troposphere (Fig. 2c), between 10-40°N, African ozone is high in boreal winter (6-10 ppbv), while south of 15°N, African ozone peaks in boreal summer (~4 ppbv).

The strong seasonality of imported African ozone can also be shown vertically in Fig.3a, in which imported African ozone is averaged over Asia. The fractional contribution of imported African ozone to ozone in Asia is shown in Fig. 3b. In the upper troposphere, imported African ozone is the most during February-May (>10 ppbv) and the least during July-September (<6 ppbv). Slightly different from the situation in the upper troposphere, imported Africa ozone in the Asian middle troposphere is at a maximum (>10 ppbv, 14% in the fractional contribution) from December to April and at a minimum from June to September (~4 ppbv, 6% in the fractional contribution). In the lower troposphere, imported African ozone is the largest in boreal winter (~4 ppbv, 8% in the fractional contribution), lowest in boreal fall (~2 ppbv, 5% in the fractional contribution). Seasonal-altitude variations of imported ozone from NHAF and SHAF over Asia and the fractional contributions of NHAF and SHAF ozone to the total imported African ozone are shown in Fig. 4. Ozone from NHAF accounts for over 70% of the total imported African ozone in the Asian tropospheric column throughout the year, except for in the upper troposphere in boreal winter and the lower troposphere in boreal summer. The summertime transport of SHAF ozone to the Asian lower troposphere is also shown in a summer maximum of African ozone in Fig.2c (~4 ppbv at 700 hPa, south of 15°N). This was also reported by Lal et al. (2014), who attributed it to ozone transport from the Indian Ocean from the east coast of Africa.

As African ozone mainly peaks in the Asian middle and upper troposphere, the horizontal distributions of African ozone at 400 hPa in January, April, July, and October overlaid with winds are shown in Fig. 5 to further illustrate its seasonality and transport pathways. Driven by the upper tropospheric easterly jet around the equator (Diedhiou et al., 1999), high African ozone appears across the Atlantic, reaching the South America throughout the year. The westward transatlantic routes from



240 northern Africa are built by the easterly waves (Jones et al., 2003; Mari et al., 2008; Ben-Ami et al.,  
2009). In boreal winter, near the northern and southern borders of Africa, African ozone can be  
transported for a long distance along the subtropical westerly jets in the two hemispheres, reaching the  
western Pacific in the Northern Hemisphere and across Australia in the Southern Hemisphere,  
respectively. In boreal summer, the northern subtropical westerlies and tropical easterlies shift to their  
245 most northern positions; less African ozone can be transported to Asia than in the other seasons.  
However, around 40°N, the amount of African ozone being transported to Asia is similar to the other  
season (Fig. 2a).

#### 4 Possible mechanisms for the transport of African ozone to Asia

250 In this section, we analyze the transport pathways from Africa to Asia and associated underlying  
mechanisms so to cast some light on the spatial and seasonal variations in imported African ozone over  
Asia presented in section 3. Since Africa covers areas in both Northern and Southern Hemispheres with  
a large portion in the tropics, the atmospheric circulation over Africa experiences obvious seasonal  
changes induced by the seasonality of the ITCZ and the Hadley cell (Nicholson and Grist, 2003;  
255 Nicholson, 2008, 2009; Žagar et al., 2011; Suzuki, 2011). African ozone precursors are mainly from  
biogenic sources, biomass burning, and lightning NO<sub>x</sub> (Piketh and Walton, 2004; Thompson, 2004;  
Aghedo et al., 2007; Giglio et al., 2013; Monks et al., 2015). The seasonalities of emissions from  
biogenetic sources, biomass burning, lightning, and anthropogenic sources in Africa are characterized  
rather differently from the other continents (Williams et al., 2009; Guenther et al., 2012; Giglio et al.,  
260 2013; Albrecht et al., 2016).

##### 4.1 The ITCZ and ozone precursor emissions over Africa

Referred to Nicholson (2009), Suzuki (2011), and Žagar et al. (2011), the mean positions of the ITCZ in



Africa in the four seasons are approximately illustrated in Figs. 6a-6d. The seasonal migration of the ITCZ between latitudes is a robust phenomenon and varies with longitude (Nicholson, 2009; Suzuki, 2011). The latitudinal shift is wider in eastern Africa than in western Africa (Figs. 6a-6d). From boreal winter to summer, the center of the ITCZ swings from 10°N to 20°N in western Africa (Figs. 6a-6d), while in eastern Africa between 10°E and 40°E, ITCZ shifts between ~10°S in boreal winter and ~20°N in boreal summer (Figs. 6a-6d).

African ozone and its seasonality depend heavily on biogenic emissions, moderately on biomass burning and lightning emissions, and weakly on anthropogenic emissions. The anthropogenic emissions are small and have weak seasonality (Aghedo et al., 2007; Williams et al., 2009). The spatial distributions of isoprene emissions from biogenic sources, CO emissions from biomass burning, and NO<sub>x</sub> emissions from lightning are shown by season in Fig. 6. The normalized seasonal variations of these emissions averaged over Africa, NHAF and SHAF are shown in Fig. 7. The emission data are from the emission inventories in GEOS-Chem described in section 2.1. The normalized value equals to the original value minus the annual mean and then divided by the standard deviation.

As isoprene (C<sub>5</sub>H<sub>8</sub>) is the dominant non-methane volatile organic compound (NMVOC) emitted by vegetation (Marais et al., 2012) and biogenic isoprene emissions in Africa account for about 65% of the African upper troposphere ozone enhancement (Aghedo et al., 2007), isoprene emissions are shown in Figs. 6a-6d as a representative ozone precursor from biogenic sources. The maximum biogenic isoprene emissions are over central African rainforests throughout the year (Fig. 6a-6d). The seasonal cycle of biogenic isoprene over Africa peaks in boreal spring and fall (Fig. 7a). Plenty of non-methane VOCs, which may have relatively long lifetimes (such as methanol) or effective storage of NO<sub>x</sub> (such as PAN), are emitted from biogenic sources that can be uplifted by strong convection (Tie et al., 2003). Consequently, the contribution of biogenic emissions to ozone in the African upper troposphere is larger than biomass burning and anthropogenic emissions (Aghedo et al., 2007; Zare et al., 2014).



In boreal winter, fires in Africa are active in the Northern Hemisphere between 0-10°N and 15°W to 40°E (Sauvage et al., 2005). From boreal winter to fall, biomass burning regions shift southward from central Africa to southern Africa (Fig. 6, 2<sup>nd</sup> row, also see van der Werf et al., 2010; Giglio et al., 2013). In the Southern Hemisphere, fires are most active in boreal summer from the equator to 20°S. Therefore, the regional CO emissions from biomass burning peak in boreal winter in NHAF and in boreal summer in SHAF (Fig. 7b). Aghedo et al. (2007) stated that biomass burning has the largest impact on surface ozone in the vicinity of the African burning regions during the burning seasons.

In Africa, lightning NO<sub>x</sub> is produced mostly in the middle to upper troposphere (Figs. 6i-6p, also see Pickering et al., 1998). Miyazaki et al. (2014) estimated that the altitude where the annual lightning NO<sub>x</sub> emission maximizes is 11 km for northern Africa and 9.36 km for southern Africa. Therefore, lightning emissions mainly enhance ozone in the African middle and upper troposphere (Aghedo et al., 2007). Ascribe to the high efficiency of deep moist convection, frequent lightning activities occur in the ITCZ (Christian et al., 2003; Avila et al., 2010). Collier and Hughes (2011) suggested that the peak lightning activities are generally located on the southern border of the ITCZ in Africa. The seasonality of the locations for high lightning NO<sub>x</sub> emission clearly shows the influence of the solar declination and the ITCZ over Africa (Collier and Hughes, 2011). When the ITCZ reaches to its northernmost position in boreal summer (Fig. 6c), lightning NO<sub>x</sub> emission over the NHAF becomes the highest (Figs. 6, 7c and 7d). Similarly, the lightning NO<sub>x</sub> emission over the SHAF peaks in boreal winter (Figs. 6, 7c and 7d).

#### 4.2 Analysis of the transport pathways based on GEOS-Chem simulations

Fig. 8 shows the latitude-altitude cross sections of African ozone and wind fields averaged from 0 to 40°E. Fig. 9 is the same as Fig. 8 but for the longitude-altitude cross sections averaged from 25°N to 35°N. In Fig. 10, vertical distributions of African ozone fluxes along the western border and eastern border of Asia are shown by season. Apparently, the inflow fluxes are larger than the outflow fluxes



throughout the year. Controlled by the northern subtropical westerly jet, imported African ozone fluxes peak in the upper troposphere. The latitudes where African ozone flux peak vary with the seasonal swing of the northern subtropical westerlies.

315 In boreal winter, deep convection, strong convergence in the lower troposphere can be seen from 10°S to 20°S (Fig. 8a), indicating the position of the ITCZ in the Southern Hemisphere. The two cells of the Hadley circulation are clearly shown as well. Due to frequent lightning activities, the region with high ozone appears over the ITCZ in the African middle and upper troposphere in the Southern Hemisphere. In the lower troposphere, ozone concentrations are high from the equator to 10°N, 320 resulting from high biomass burning emissions as discussed in section 4.1 (Aghedo et al., 2007). This ozone is uplifted (Fig. 8a, 0-10°N) by the ITCZ in the Northern Hemisphere (Figs. 6a and 6e). Driven by the Hadley cell, ozone in NHAF is transported equatorward in the lower troposphere, upward over the ITCZ, poleward in the middle and upper troposphere. The northward outflow in the middle and upper troposphere gradually becomes weaker and weaker between 15-30°N (Fig. 8a). Eventually, 325 carried by the subtropical westerlies, African ozone is transported eastward to Asia (Fig. 9a, also see Fig. 12a in section 4.4). Consequently, the contribution of African ozone to Asia becomes the highest in the middle and upper troposphere (see Figs. 2b and 3a for ozone concentrations, and Fig. 10a for inflow ozone flux). During the season, the divergence outflow in the Southern Hemispheric upper troposphere carries African ozone (Fig. 8a, 500-200 hPa, 10-20°S) to the Asian upper troposphere, contributing to 2-330 3 ppbv ozone, which is 30% of the total imported African ozone over Asia (Figs. 4c and 4d).

In boreal spring, a region with high ozone concentrations (>40 ppbv) appears in higher altitudes and extends to a larger area in the middle and upper troposphere than in boreal winter (Figs. 8b and 9b) mainly due to the highest biogenic emissions in the NHAF (section 4.1, Figs. 6b and 7a). The region is also further north than in boreal winter under the influence of seasonal migration in the locations of the 335 ITCZ and consequently the lightning. Ozone in the African lower troposphere becomes lower than in



boreal winter, likely due to weaker biomass burning activities in the central Africa (Duncan et al., 2003). In general, the pathways for African ozone to be transported to Asia in boreal spring are similar to those in winter (Figs. 5a vs 5b, 8a vs 8b, 9a vs 9b, 10a vs 10b). In the upper troposphere, however, more ozone is transported to Asia in boreal spring than in boreal winter (Fig. 3a), mainly due to the seasonal increase of biogenic emissions (Fig. 7a).

In boreal summer, African ozone in the lower troposphere peaks from  $\sim 15^{\circ}\text{S}$  to the equator (Fig. 8c), corresponding to the biomass burning emissions (Figs. 6g and 7b). However, because the center of the ITCZ shifts to  $\sim 20^{\circ}\text{N}$  over the Sahara (Fig. 6c), ozone generated from biogenic emissions is uplifted less effectively than in the other seasons (Fig. 8c vs 8a, 8b, and 8d). Consequently, ozone in the African middle and upper troposphere south of  $15^{\circ}\text{N}$  is lower than in the other seasons (Fig. 8c). However, resulting from the northward migration of lightning activities, ozone in the middle and upper troposphere north of  $15^{\circ}\text{N}$  is higher than in the other seasons (Fig. 8c). The transport of African ozone to Asia in boreal summer differs from that in boreal winter. In boreal summer, the updrafts over the Tibetan Plateau are strong (Fig. 9c), which serves as a heat source for the Asian summer monsoon (Wu et al., 2012, 2015) and blocks the transport of African ozone to Asia (Fig. 9c). Furthermore, the northern westerly jet moves northward to around  $40^{\circ}\text{N}$  (Huang et al., 2012). The tropical easterlies also shift northward along (Fig. 5c) and further prevent African ozone from reaching Asia between  $10^{\circ}\text{N}$  and  $30^{\circ}\text{N}$  above the lower troposphere (see Figs. 2a-2d, and 3a-3b for imported ozone concentrations, Fig. 10c for inflow ozone flux).

In boreal autumn, the locations of the ITCZ and the Hadley cell are similar to these in boreal spring (Figs. 8b vs 8d). Ozone in the African middle troposphere is higher in boreal autumn than in boreal spring, attributed to stronger lightning  $\text{NO}_x$  emission (Figs. 7c and 7d). However, because of the weaker zonal winds (Figs. 9b vs 9d), the ozone inflow to Asia is smaller than in spring (Figs. 10b vs 10d).

Overall, the influence of African ozone on the Asian middle and upper troposphere is mainly



360 attributed to ozone transport driven by the Hadley circulation and subtropical westerlies. The seasonality of the ITCZ, the Hadley cell, and the westerlies can greatly influence the seasonality of the pathways of ozone transport from Africa to Asia.

### 4.3 Interhemispheric transport of ozone from the SHAF to Asia

365 In the upper troposphere, the horizontal distribution of SHAF ozone at 200 hPa in January is shown in Fig. 11a. SHAF ozone in Asia is 2-3 ppbv along 60°N-150°E between 0-30°N (Fig. 11a). Because of the ITCZ and the Hadley circulation, a part of SHAF ozone is inter-hemispherically transported to the Northern Hemisphere (Fig. 8a). Then, driven by northern subtropical westerly jet, this ozone can finally reach Asia (Fig. 11a). Over Asia, ozone from SHAF is the largest in boreal winter than in the other  
370 seasons in the upper troposphere, while in the lower troposphere, it is the largest in summer than in the other seasons (Figs. 4c and 4d). Fig. 11b shows SHAF ozone in July at 925 hPa, with concentrations of 2-6 ppbv over the Arabian Sea and the west coast of the Indian subcontinent. This ozone is transported to India in boreal summer by the Somalia cross-equatorial flow (Fig. 11b), which is the strongest seasonal cross-equatorial flow in the lower troposphere (Halpern and Woiceshyn, 1999, 2001). This is  
375 the reason for the summer maximum of African ozone (~4 ppbv) over the Asian lower troposphere south of 15°N (Fig. 2c). This transport is also captured in Fig. 10c as the influx of Africa ozone in the Asian lower troposphere south of 15°N.

### 4.4 Analysis of the transport pathways based on trajectory statistics

380 In this section, we use six-day forward trajectories to further analyze the pathways for ozone transport from Africa to Asia. Fig. 12 shows the clustered six-day forward trajectories during 1987-2006 starting from Cairo (31°E, 30°N), Abuja (7.4°E, 9°N), Dar es Salaam (39°E, 6.8°S), and Johannesburg (28°E, 26.2°S) in the lower, middle and upper troposphere, respectively. The four sites are chosen for different



latitudes in Africa.

385 In boreal winter, in NHAf, ~40% of the 6-days trajectories in the lower troposphere from Cairo flow  
to higher latitudes in Asia (Fig. 12c). In the middle troposphere, ~74% trajectories from Cairo reach  
Asia along the northern subtropical westerlies (Fig. 12b). Approximately, 10% trajectories from Abuja  
reach Asia following a pathway of upward along the northern Hadley cell and then eastward along the  
northern subtropical westerlies (Fig. 12b). In the upper troposphere, African ozone can be transported to  
390 Asia fast and with longer distances within 6 days (Fig. 12a). After reaching Asia (across 60°E), most of  
the trajectories in the middle and upper troposphere move downwards along the westerlies (Figs. 12a  
and 12b). Driven by the northern subtropical westerly jet, 98% trajectories from Cairo and about 41%  
trajectories from Abuja can reach Asia (Fig. 12a). Toward to further south, Dar es Salaam is mainly  
under the influence of the tropical easterlies and the south subtropical westerlies in the lower and  
395 middle troposphere so that the trajectories from this site are either westwards or eastwards along the  
westerlies in the Southern Hemisphere (Figs. 12b and 12c). However, in the upper troposphere, about 25%  
trajectories at the site flow to Asia (Fig. 12a) due to the convective divergence over the ITCZ. At  
Johannesburg in the southern Africa, nearly no trajectories can reach Asia (Figs. 12a-12c).

In boreal summer, induced by the northward shift of the northern subtropical westerlies (~40°N),  
400 fewer trajectories reach Asia than in winter (Figs. 12d-12f). Because of the northward shift of the  
Hadley cell and the tropical easterlies, trajectories from Abuja travel westward in the middle and upper  
troposphere, while in the lower troposphere, the trajectories run around locally (Figs. 12d, 12e, and 12f).  
Similar to winter, no trajectories from Johannesburg reach Asia because of the southern location of the  
site (Figs. 12d, 12e, and 12f). However, at Dar es Salaam in east Africa, there are ~14% trajectories in  
405 the lower troposphere that travel toward India (Figs. 12e and 12f) under the influence of Somali jet  
(Halpern and Woiceshyn, 1999, 2001; Lal et al., 2014). The Somali jet can transport ozone from the east  
coast of Africa to Asia, resulting in a summer maximum of African ozone over the Asian lower



410 troposphere south of 15°N (Fig. 2c for ozone concentration, and Fig 10c for the ozone inflow flux). Overall, the analysis of the trajectory statistic agrees well with the GEOS-Chem simulations discussed in section 4.2.

#### 4.5 Influences of the ITCZ on the interannual variation of African ozone transport to Asia in boreal winter

415 As known, the deep convection along the ITCZ can carry ozone precursors from biogenic, biomass burning, and anthropogenic emissions to upper levels. The ITCZ is also a zone with large lightning activities. The convective divergence in the upper troposphere over the ITCZ plays a significant role in ozone outflow and the interhemispheric transport between SHAF and NHAF. Therefore, the ITCZ can impact meteorology in Africa (Sultan and Janicot, 2000; Xie, 2004; Hu et al., 2007; Collier and Hughes, 2011; Suzuki, 2011) and the seasonality of ozone precursor emissions (for example, NO<sub>x</sub> from lightning). To explore the role that the ITCZ plays in ozone transport from Africa to Asia, we use the 420 OLR data as a proxy for convective activity in the tropics to describe the intensity of ITCZ, similar to previous studies (Waliser et al., 1993; Mounier and Janicot, 2004; Fukutomi and Yasunari, 2013, 2014). According to Waliser et al. (1993) and Fukutomi and Yasunari (2013), the number of the grid points with OLR  $\leq 260$  W/m<sup>2</sup> in the region of 15°W-45°E, 20°S-20°N can indicate the intensity of the 425 convection over the ITCZ in Africa.

Fig. 13 shows time series of ozone from Africa, NHAF and SHAF, averaged over Asia, against the intensity of the ITCZ over Africa (after normalization, i.e., the original value minus the mean that is divided by the standard deviation) in January from 1987 to 2006. The anomalies of imported ozone from SHAF are quite small in the lower and middle troposphere (Fig. 4), so the time series is not shown. 430 Positive correlations are found between the intensity of the ITCZ and anomalies of African ozone over Asia, with the correlation coefficient (*r*) being 0.61, 0.46, and, 0.64 for the Asian lower (975 hPa),



middle (600 hPa) and upper (200 hPa) troposphere, respectively, all statistically significant at the 95% level ( $p < 0.05$ ). Significant correlations are also found for NHAf and SHAF ozone in the upper troposphere (Fig. 13a). In boreal winter, the center of the ITCZ is located in the upper region of biogenic and biomass burning emissions (Figs. 6a and 6e). When the intensity of the African ITCZ is stronger, more ozone and ozone precursors are uplifted to the middle and upper troposphere and transported poleward and then eastward to Asia by the northern subtropical westerlies (Figs. 12a-12c). Additionally, driven by the enhanced convective divergence over the ITCZ, the interhemispheric transport from SHAF increases in the upper troposphere (Figs. 8a and 13a). Consequently, transport of ozone from Africa to Asia increases in the middle and upper troposphere. At the same time, carried by the downdrafts from Asian winter monsoon (Figs. 9a, 12a-12c), more ozone is transported to the surface in Asia (Zhu et al., 2017b).

## 5 An overview on the transport of African ozone to Asia

The processes discussed in section 4 on the transport of African ozone to Asia can be briefly summarized for boreal winter (Fig. 14a) and boreal summer (Fig. 14b), respectively, from perspectives of both the atmospheric circulations (Xie, 2004; Mari et al., 2008; Nicholson, 2009; An et al., 2015; Ding et al., 2015; Li et al., 2016; Nützel et al., 2016) and emissions of ozone precursors (Aghedo et al., 2007; Marais et al., 2012; Giglio et al., 2013; Miyazaki et al., 2014; Zare et al., 2014; Albrecht et al., 2016).

During boreal winter, the Hadley cell and the northern subtropical westerlies build the transport pathways from Africa to Asia. Formed at the ascending branch of the Hadley circulation, the location of the ITCZ over Africa shows different characteristics in Africa west of  $10^{\circ}\text{E}$  and Africa east of  $10^{\circ}\text{E}$  (Nicholson, 2009). Over the ITCZ, a great amount of latent heat is released by deep convection, which drives the ascending branch of the Hadley circulation (Xie, 2004). The biomass burning emissions are



mainly in the Northern Hemisphere (Fig. 6e), which contributes to the production of a significant amount of African ozone in the lower troposphere (Fig. 8a, also see Mari et al., 2008). The ozone precursors can be carried upward from the surface to the upper layers by the rising branch of the Hadley circulation.

460 In the African middle and upper troposphere, vigorous convection within ITCZ clouds results in strong thunderstorms, accompanied with much lightning (Avila et al., 2010). Based on the C-shaped vertical profile of the lightning-generated  $\text{NO}_x$ , lightning activities contribute to the generation of a large proportion of ozone in the African middle and upper troposphere (Fig. 8a, also see Pickering et al., 1998; Miyazaki et al., 2014). Then ozone generated from chemical reactions and convection uplifting is  
465 transported poleward by the convective divergence over the ITCZ and downward by the descending branch of the Hadley cell. The northern subtropical westerlies carry African ozone to Asia in the middle and upper troposphere. Overall, in boreal winter, African ozone is mainly produced from biogenic and lightning sources (Aghedo et al., 2007; Zare et al., 2014) and is transported to Asia mostly in the middle and upper troposphere, governed by the northern Hadley cell and subtropical westerlies.

470 In boreal summer, the ITCZ and the Hadley circulation move northward in response to the solar heating to the Northern Hemisphere. The African easterly jet, tropical easterly jet and northern subtropical westerly jet all shift northward as well. However, the fire areas shift southward to the Southern Hemisphere (Clain et al., 2009; Giglio et al., 2013). Therefore, the ITCZ can facilitate lightning  $\text{NO}_x$  production in NHAF but uplifts little ozone generated from biomass burning. In the  
475 upper troposphere, transport of African ozone to Asia is hampered by the SAH, a prevailing weather system over Eurasia in boreal summer (Nützel et al., 2016). The updrafts associated with SAH prevent African ozone from reaching Asia at low latitudes ( $<30^\circ\text{N}$ ) (Fig. 9c). In the meantime, the easterlies prevail south of  $30^\circ\text{N}$  so to block African ozone from reaching Asia. Overall, both the meteorology and emissions in boreal summer weaken the transport of African ozone to Asia in the middle and upper



480 troposphere. Noticeably, the Asian summer monsoon (Zhang et al., 2002; Ding et al., 2005) is in favor  
of transport of African ozone to Asia in the lower troposphere. Located north to the enhanced  
Mascarene Anticyclone, on the east coast of Somali, Somali cross-equatorial flow served as an essential  
component of the Asian monsoon system (Joseph and Sijikumar, 2004). Peaked at 925 hPa (Zhu, 2012),  
the Somali jet carries ozone in eastern Africa to India in the lower troposphere (Figs. 10c and 11b).

485

## 6 Conclusions

The transport of ozone originated in the African troposphere to Asia is investigated through the analysis  
of the simulations using a global chemical transport model, GEOS-Chem, and a trajectory model,  
HYSPLIT. This study shows that imported African ozone varies greatly with latitude, longitude, and  
490 altitude in the Asian troposphere, also with strong seasonality. In the Asian upper troposphere, imported  
African ozone is the largest during February-May ( $>10$  ppbv) and the lowest during July-September ( $<6$   
ppbv). In the middle troposphere, imported African ozone is at a maximum from December to April  
( $>10$  ppbv) and at a minimum from June to September ( $\sim 4$  ppbv), slightly different from the seasonality  
in the upper troposphere. In the lower troposphere, the African influence is small ( $<6$  ppbv), with a  
495 maximum from December to February ( $>6\%$ ) and a minimum from September to November ( $<5\%$ ).  
Overall, the maximum influence of African ozone is in boreal winter and early spring in the middle and  
upper troposphere of southern Asia ( $5^{\circ}\text{N}$ - $40^{\circ}\text{N}$ ). Ozone from NHAF makes up over 80% of the total  
imported African ozone in most seasons and layers in Asia, except over the upper troposphere in boreal  
winter and over the lower troposphere in summer, when the interhemispheric transport of ozone from  
500 SHAF becomes stronger than in the other seasons at the same layers.

The underlying mechanisms for these seasonal variations in the transport of African ozone to Asia are  
explored. In boreal winter, facilitated by ITCZ, ozone produced from its precursors from biogenic and  
biomass burning sources can be efficiently lifted up to high layers. The uplifted African ozone is



transported poleward by the Hadley circulation in the upper troposphere and then eastward by the  
505 subtropical westerlies in the middle and upper troposphere. The convective divergence over the ITCZ  
(the poleward branch of the Hadley circulation) in the Southern Hemisphere promotes the  
interhemispheric transport of SHAF ozone, resulting in a contribution of SHAF ozone of 2-3 ppbv in  
the upper troposphere, larger than in the other seasons. Under the influence of both the Hadley  
circulation and subtropical westerlies, African ozone peaks mainly in the Asian middle and upper  
510 troposphere. In boreal summer, as the northern westerlies shift northward, African ozone is mostly  
transported to the Asian upper troposphere around 40°N. In the lower troposphere, the Somali jet serves  
as another important pathway for the interhemispheric transport of SHAF ozone, carrying ozone from  
eastern Africa to India and thus forming a summer African ozone maximum in the lower troposphere  
over latitudes south of 15°N.

515 The interannual variation in African ozone transport to Asia is greatly influenced by the intensity of  
the ITCZ in boreal winter. Positive correlations are found between the intensity of the ITCZ in Africa  
and the interannual variations of imported African ozone over Asia in various layers ( $r = 0.64$  at 200 hPa,  
 $r = 0.46$  at 600 hPa, and  $r = 0.61$  at 975 hPa). Greatly influenced by the proximity of the ITCZ to the  
biogenic, biomass burning and anthropogenic emissions, the uplifting of ozone and its precursors from  
520 the surface to higher altitudes is much more effective in boreal winter than in boreal summer. The  
stronger the ITCZ in Africa is, the more ozone and its precursors from the surface emissions can be  
uplifted. In the meanwhile, lightning  $\text{NO}_x$  is produced in the middle and upper troposphere. Additionally,  
the convective divergence over the ITCZ in the upper troposphere is enhanced, resulting in increased  
interhemispheric transport from SHAF. Consequently, more African ozone can be transported to Asia by  
525 the subtropical westerlies in the middle and upper troposphere.



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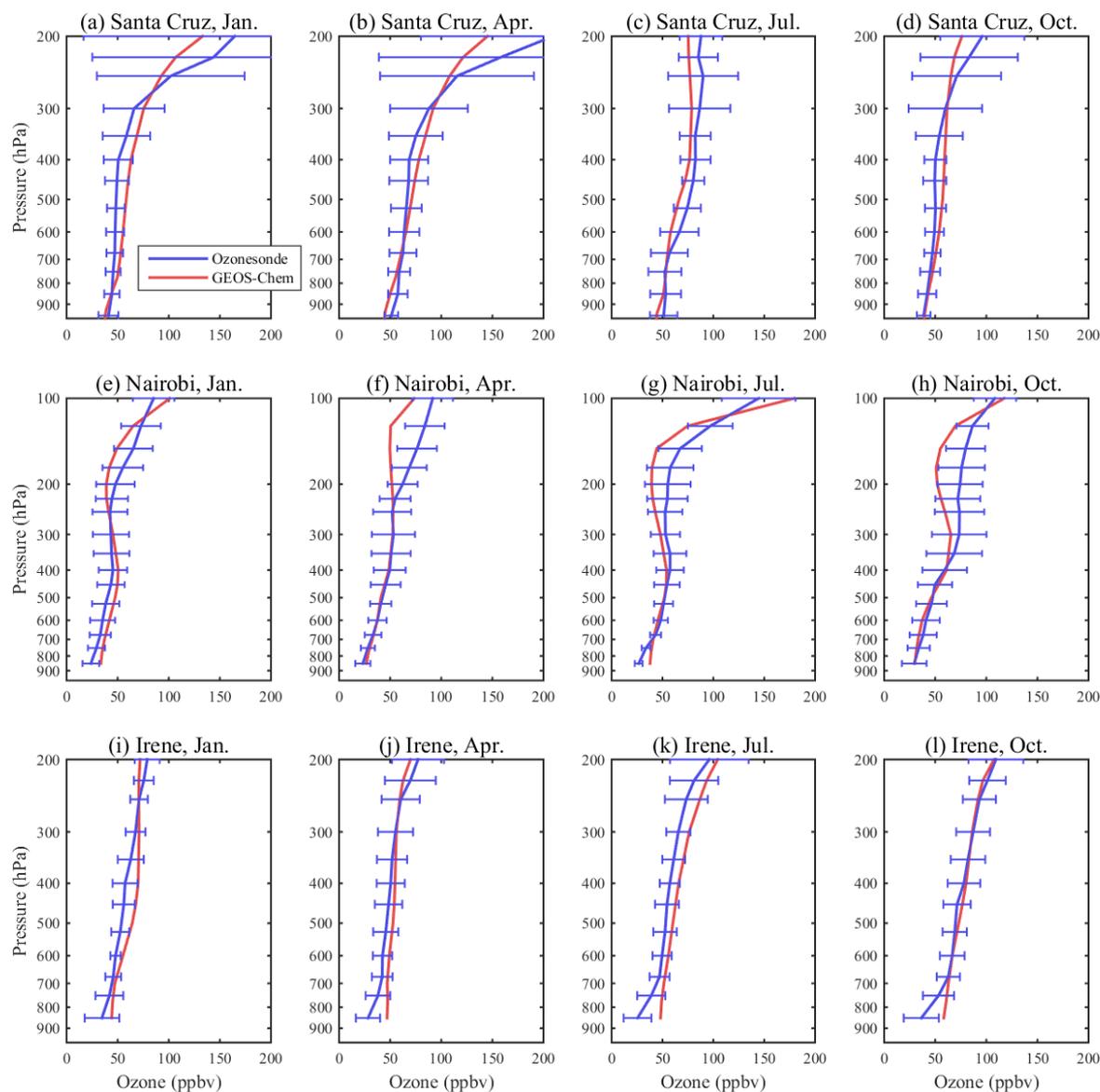


Figure 1. Comparison of the monthly mean ozone vertical profiles between the GEOS-Chem simulations (red line) and the ozonesonde measurements (blue line) at Santa Cruz (1st row), Nairobi (2nd row), and Irene (3rd row) in Africa, in January (1<sup>st</sup> col.), April (2<sup>nd</sup> col.), July (3<sup>rd</sup> col.), and October (4<sup>th</sup> col.), respectively. The horizontal bar indicates the standard deviation.

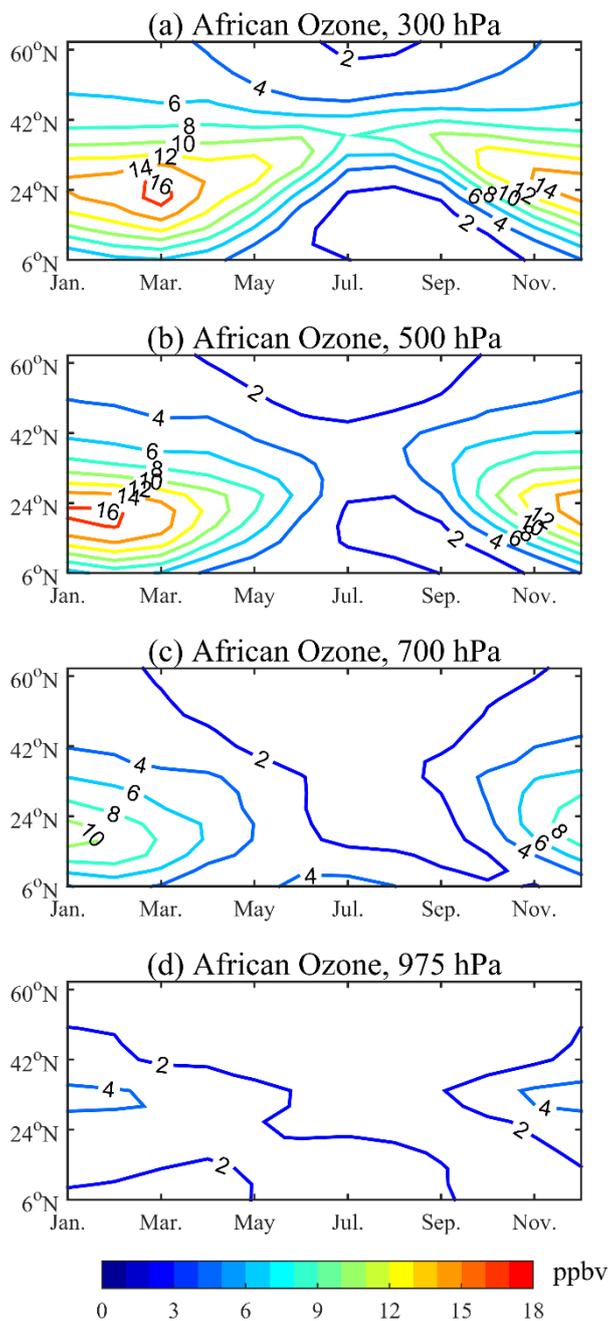


Figure 2. Seasonal variation of imported African ozone (in ppbv) over Asia varying with latitude at (a) 300 hPa, (b) 500 hPa, (c) 700 hPa, and (d) 975 hPa. The values are the 20-year means (1987-2006) from the GEOS-Chem simulation and averaged over 60-145°E.

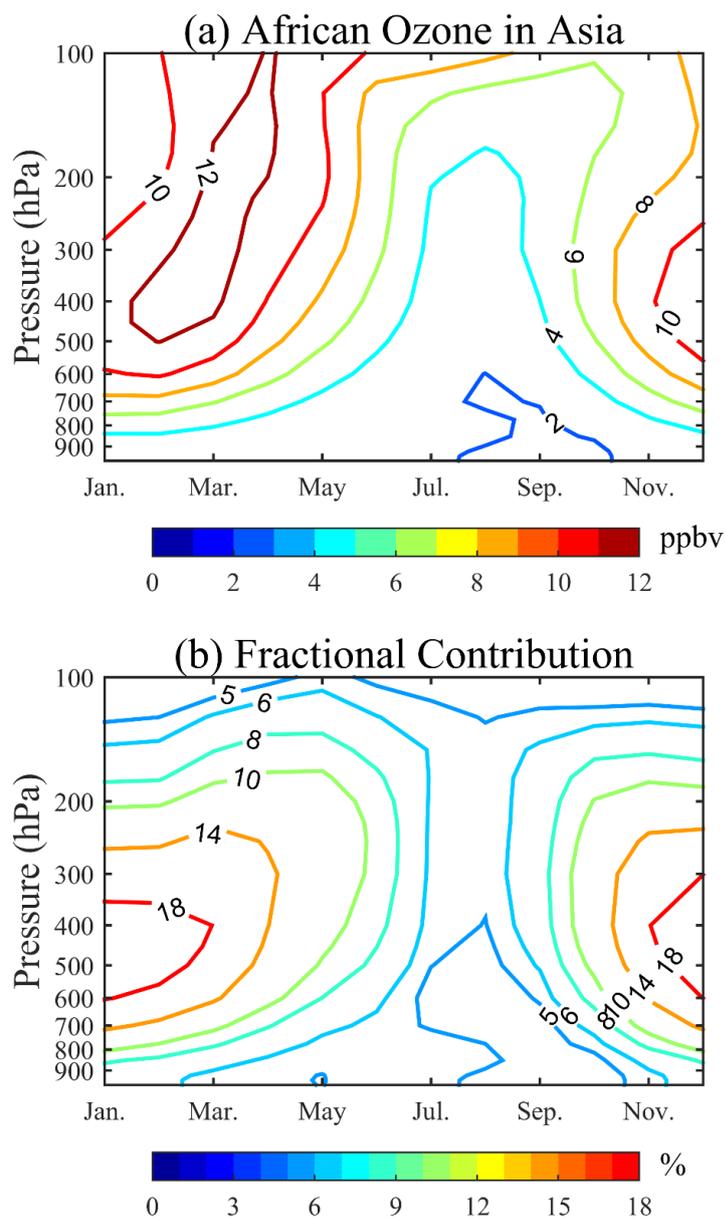


Figure 3. (a) Seasonal variation of imported African ozone (in ppbv) varying with altitude. (b) The same as (a) but for the corresponding fractional contribution (in %). The values are the 20-year means (1987-2006) from the GEOS-Chem simulation and averaged over Asia (60-145°E, 5-40°N).

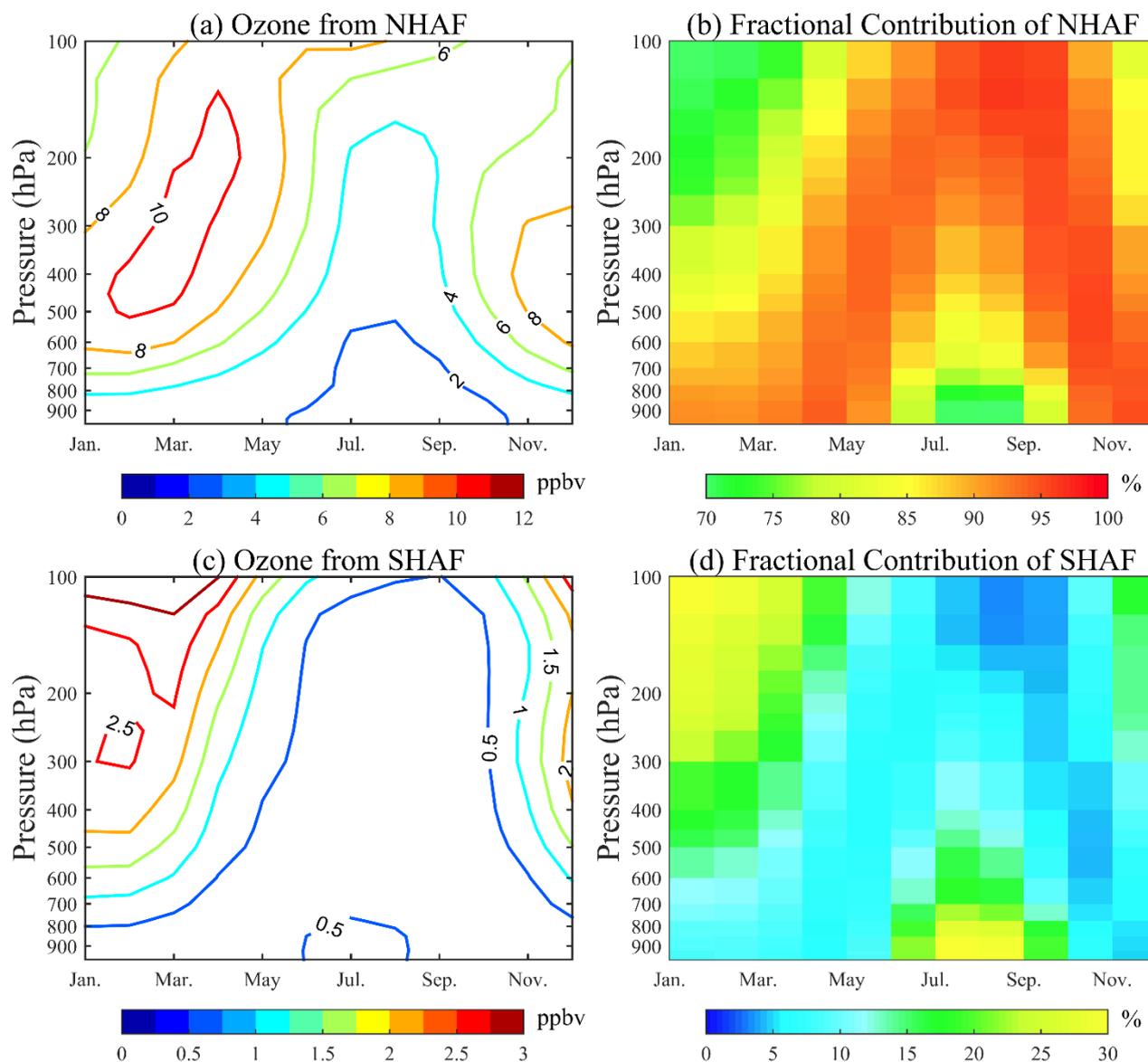


Figure 4. Seasonal-altitude variations of imported ozone (in ppbv) from (a) NHAF and (c) SHAF over Asia ( $60\text{-}145^{\circ}\text{E}$ ,  $5\text{-}40^{\circ}\text{N}$ ) and the fractional contribution of ozone from (b) NHAF and (d) SHAF to the total African ozone over Asia (in %). The ozone values are the means from the 20-year GEOS-Chem simulation.

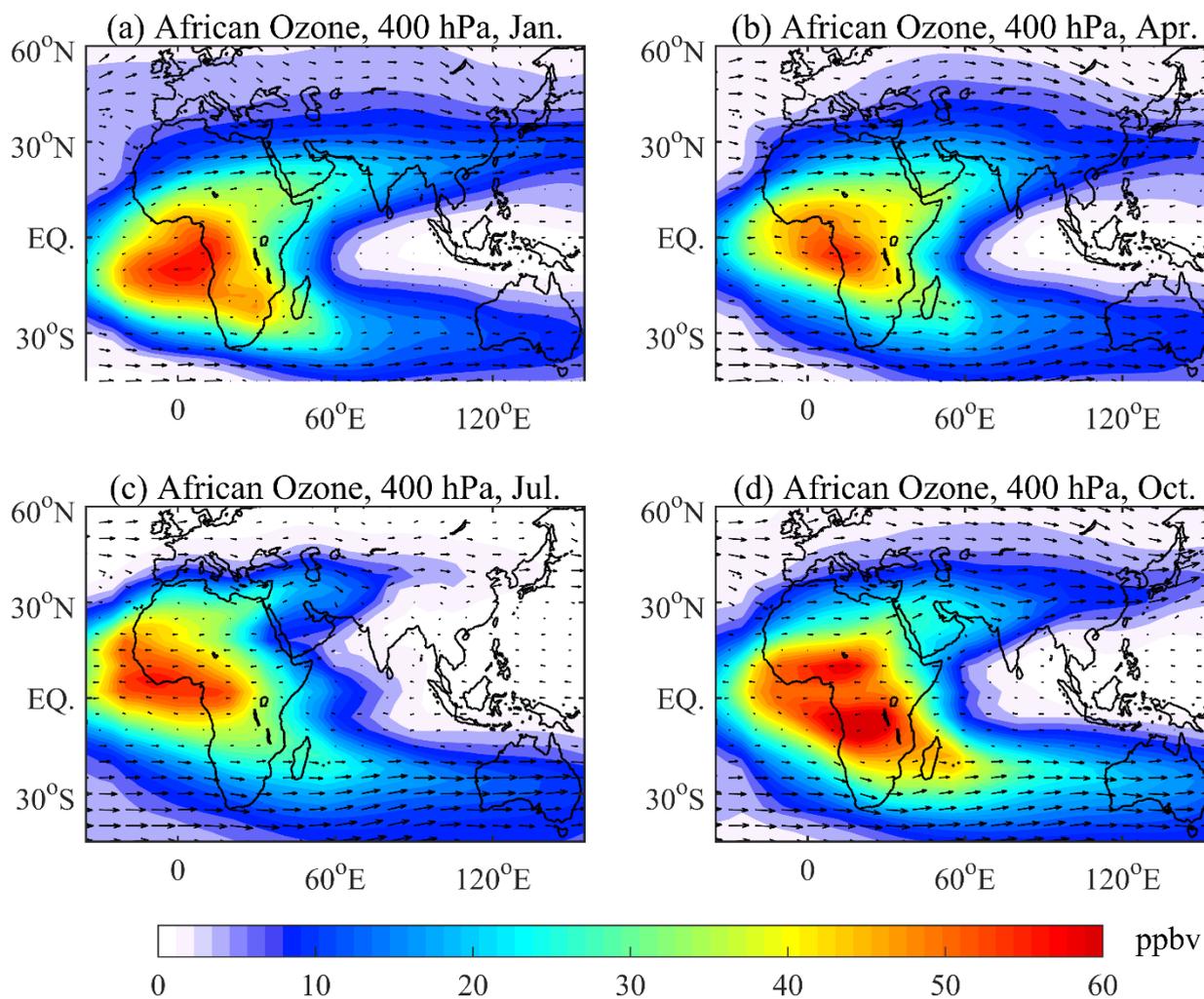


Figure 5. Horizontal distributions of African ozone (in ppbv, in color) overlaid with winds (in arrow) at 400 hPa in (a) January, (b) April, (c) July, and (d) October. The ozone values are the means from the 20-year GEOS-Chem simulation.

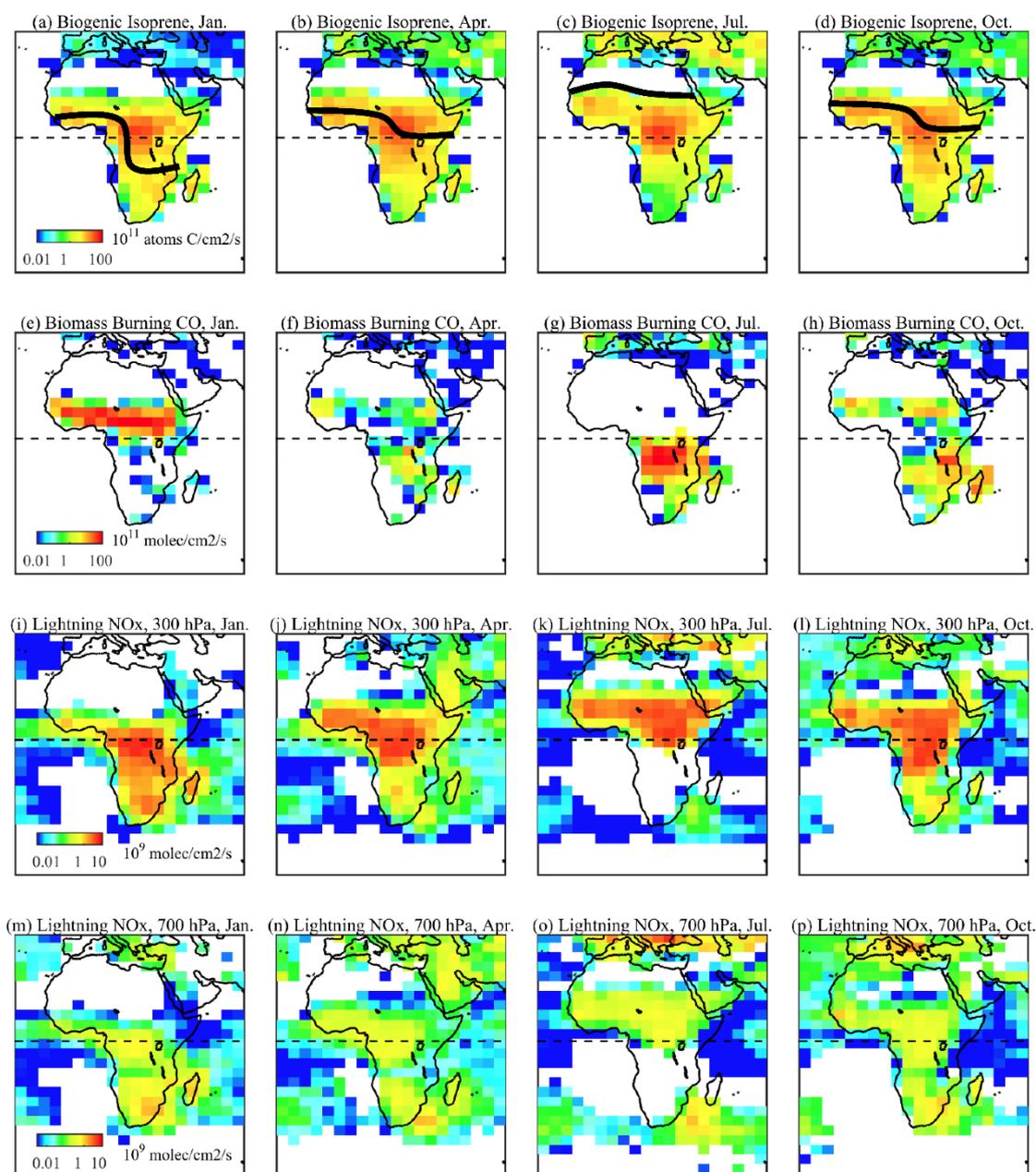


Figure 6. Distributions of isoprene emissions from biogenic sources (1<sup>st</sup> row), CO emissions from biomass burning (2<sup>nd</sup> row) and NO<sub>x</sub> emissions from lightning at 300 hPa (3<sup>rd</sup> row) and 700 hPa (4<sup>th</sup> row) by season in 2005. The data are based on the emission inventories in GEOS-Chem (see section 2.1 for details). The dash lines indicate the equator. The black solid lines in Figures 6a-6d indicate the location of the ITCZ.

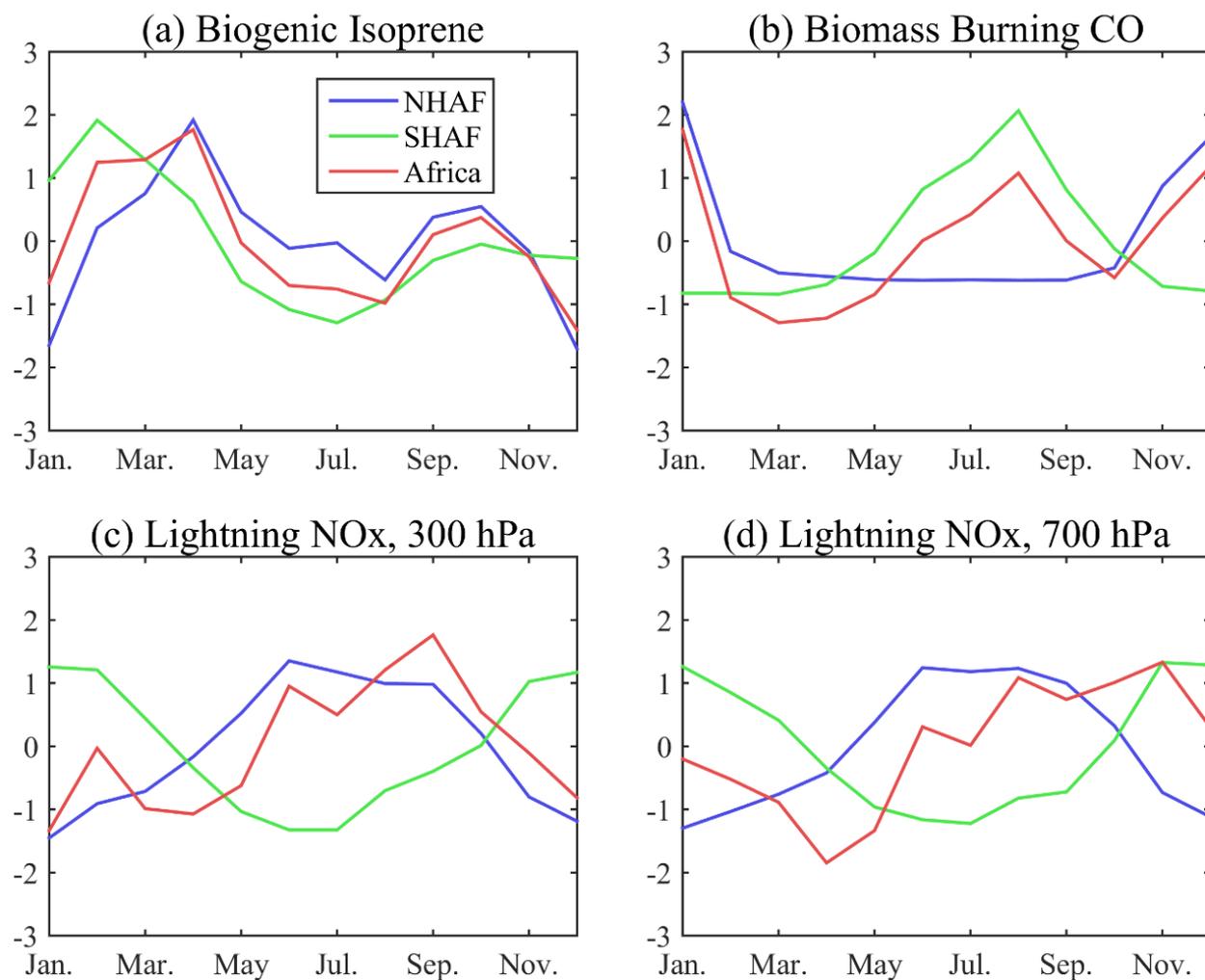


Figure 7. Seasonal variations of normalized values for isoprene emissions from (a) biogenic sources, (b) CO emissions from biomass burning, NO<sub>x</sub> emissions from lightning at (c) 300 hPa, and (d) 700 hPa averaged over Africa, NHAF and SHAF in 2005.

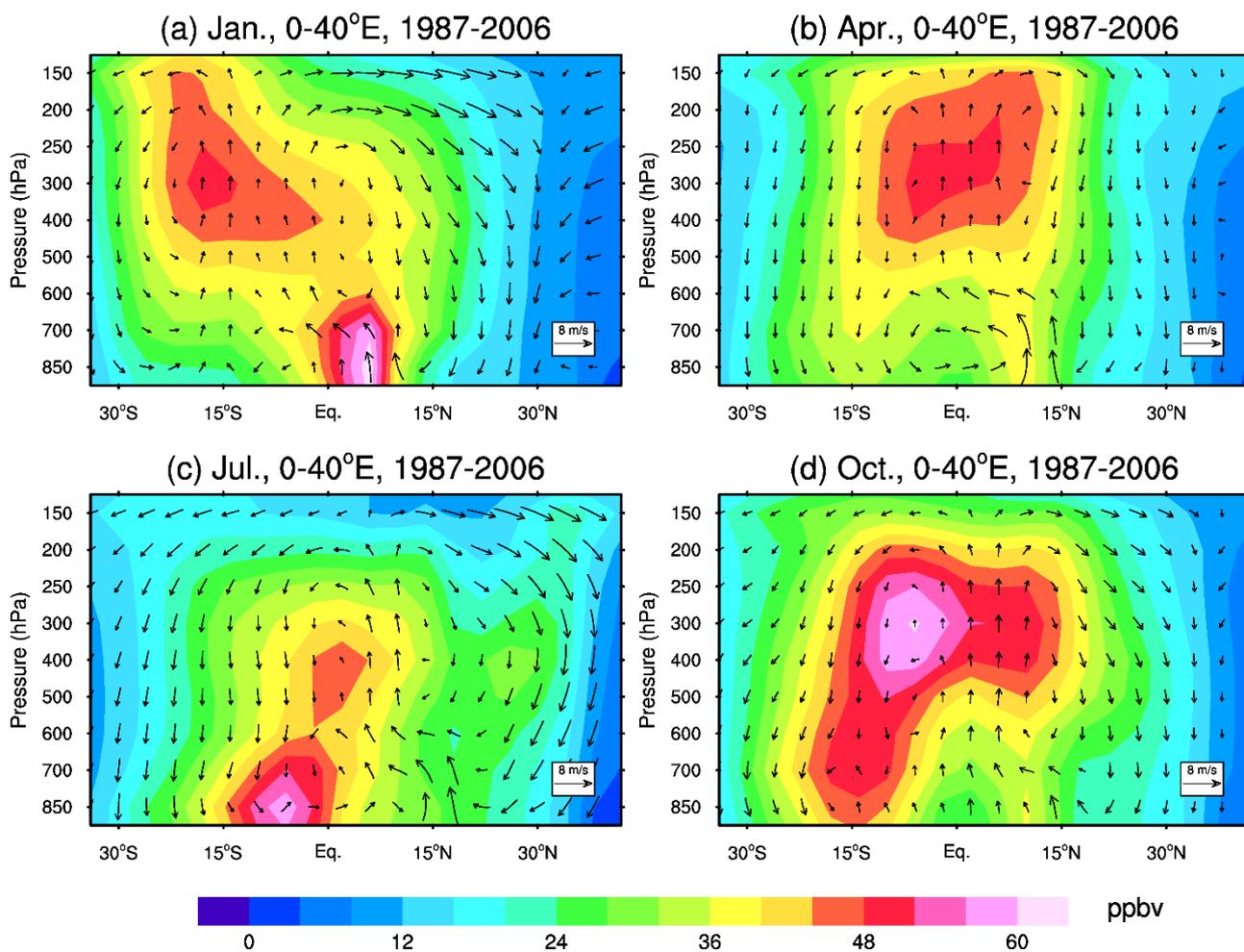


Figure 8. Latitude-altitude distribution of African ozone (in ppbv, in color), overlaid with winds (in arrow) in (a) January, (b) April, (c) July, and (d) October. The ozone values are the means over 0-40°E from the 20-year GEOS-Chem simulation. The vertical velocities in the p-coordinates are enlarged by 100 times for illustration purposes.

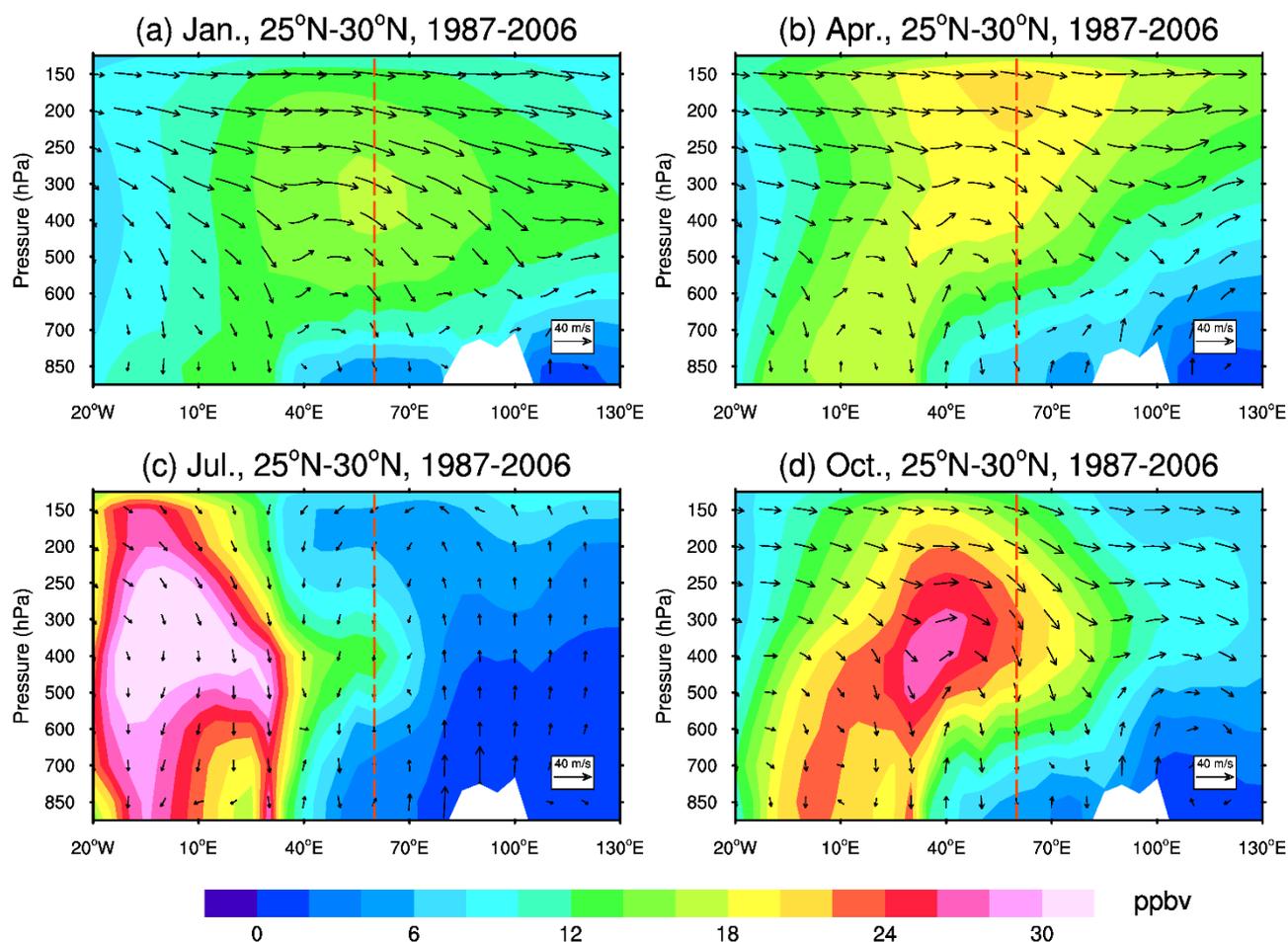


Figure 9. Longitude-altitude distribution of African ozone (in ppbv, in color), overlaid with winds (in arrow) in (a) January, (b) April, (c) July and (d) October. The ozone values are the means over 25°N-30°N from the 20-year GEOS-Chem simulation. White areas indicate topography. The red dash lines indicate the western border of Asia. The vertical velocities in the p-coordinates are enlarged by 100 times for illustration purposes.

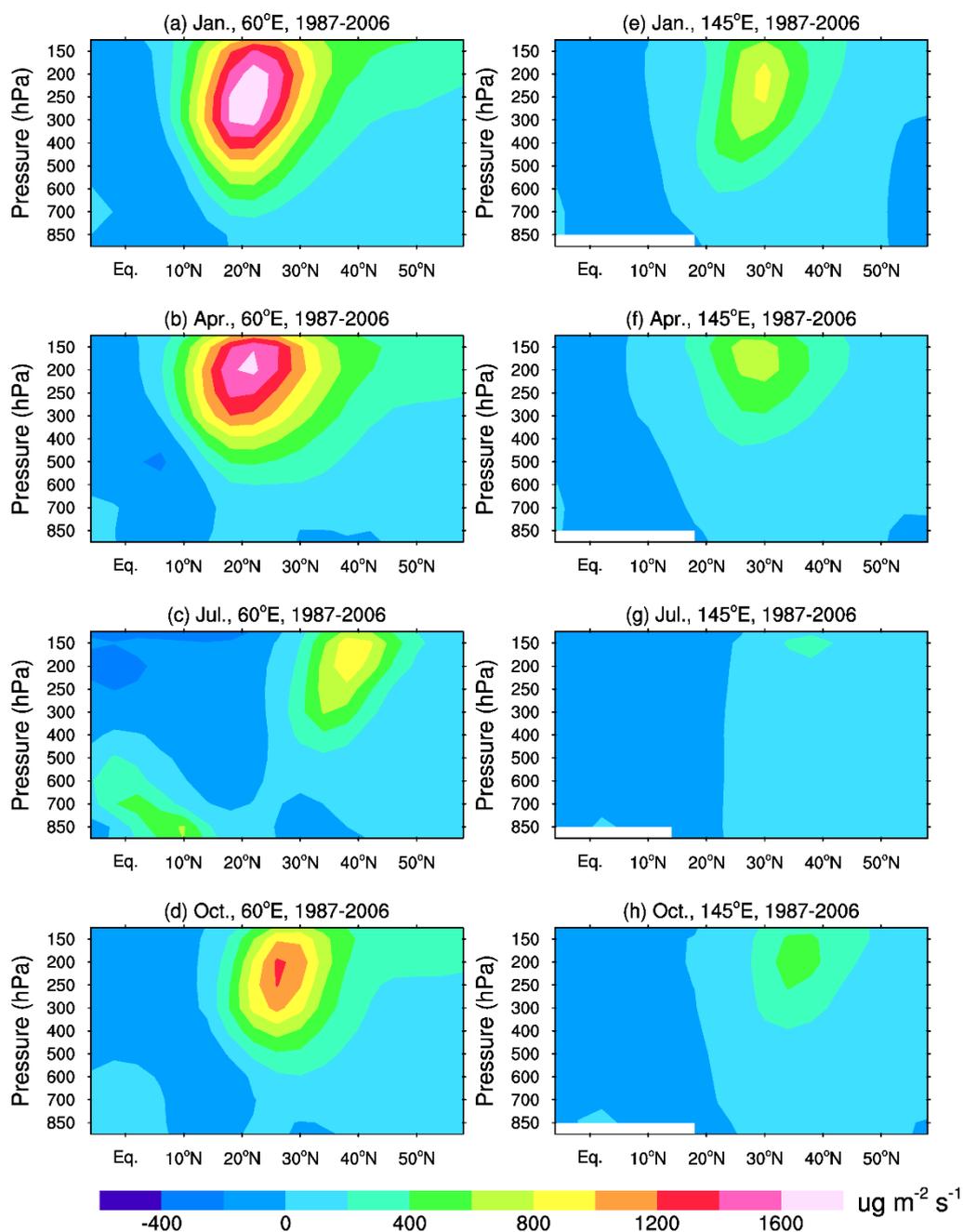


Figure 10. Latitude-altitude distribution of African ozone fluxes (in  $\mu\text{g m}^{-2} \text{s}^{-1}$ ) along the western border (60 °E, left panels) and eastern border (145 °E, right panels) of Asia in January (1<sup>st</sup> row), April (2<sup>nd</sup> row), July (3<sup>rd</sup> row) and October (4<sup>th</sup> row) averaged from 1987 to 2006.

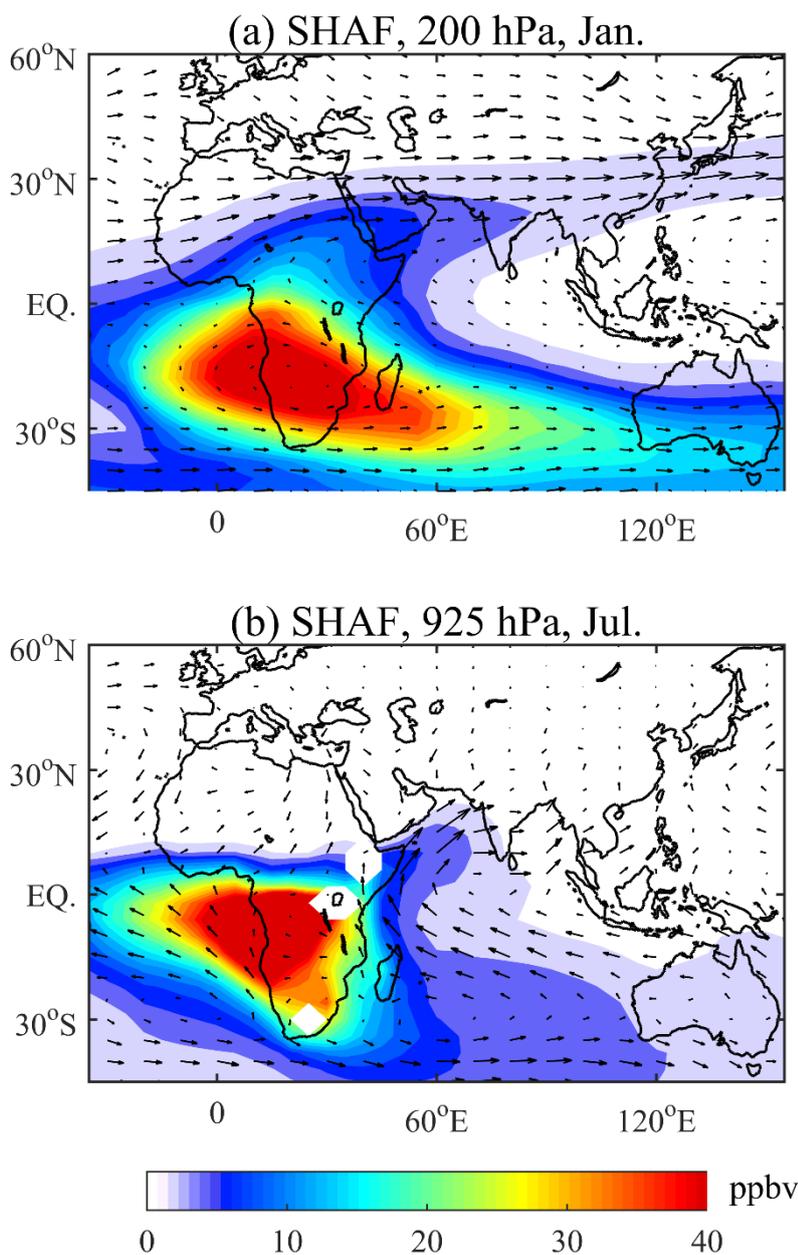


Figure 11. Distributions of ozone from SHAF (in ppbv, in color) overlaid with winds (in arrow) at (a) 200 hPa in January and (b) at 925 hPa in July. The ozone values are the means from the 20-year GEOS-Chem simulation.

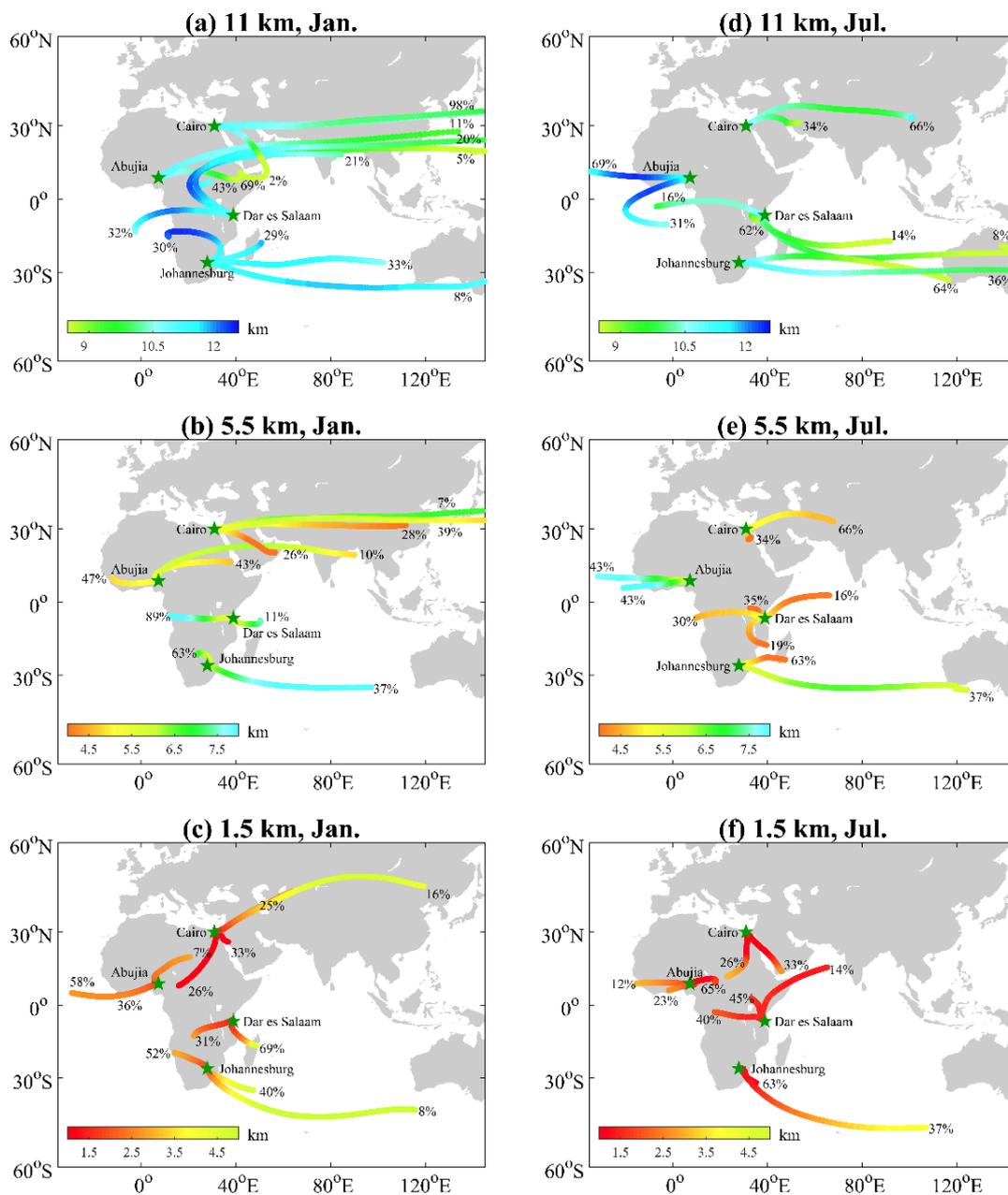


Figure 12. Six-day forward trajectories from Cairo (31°E, 30°N), Abuja (7.4°E, 9°N), Dar es Salaam (39°E, 6.8°S) and Johannesburg (28°E, 26.2°S) (stars) at 11 km (1<sup>st</sup> row), 5.5 km (2<sup>nd</sup> row), and 1.5 km (3<sup>rd</sup> row) in January (left panels) and July (right panels) clustered from 1987 to 2006.

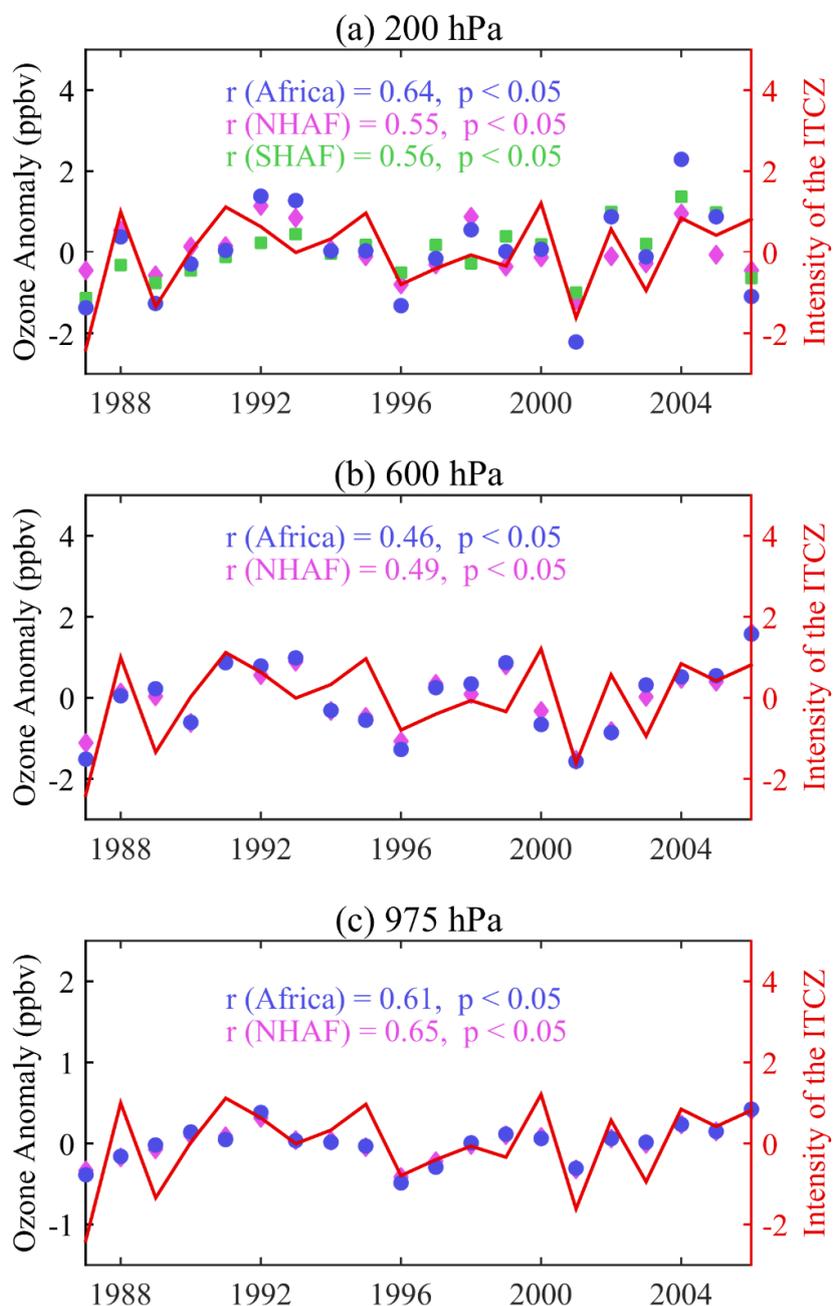


Figure 13. Interannual variation of the intensity of ITCZ (the values are normalized) over Africa and the anomaly of imported ozone from Africa, NHAF and SHAF over Asia from 1987 to 2006 at (a) 200 hPa, (b) 600 hPa, and (c) 975 hPa in January.

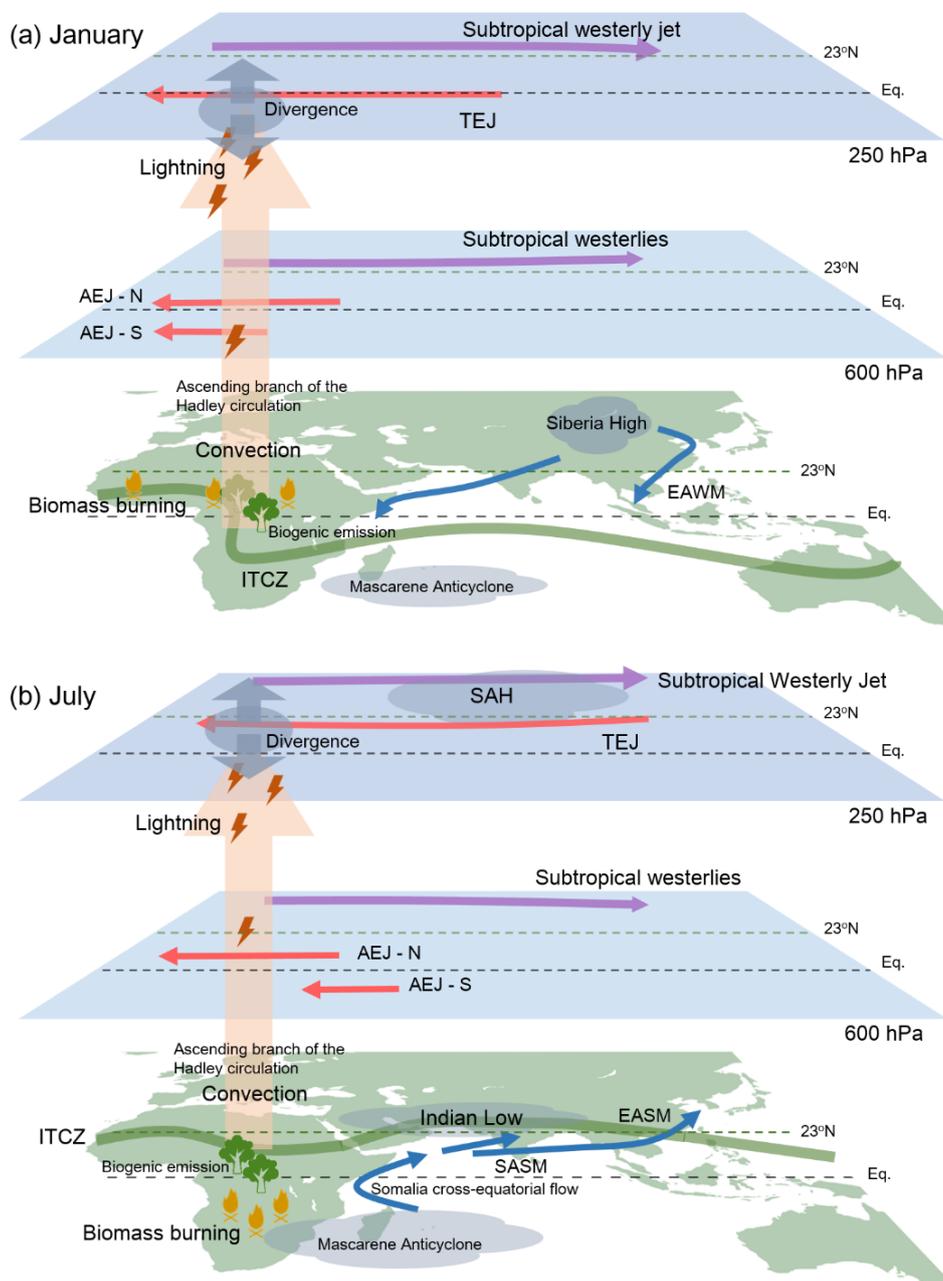


Figure 14. Schematic illustration of weather systems and the processes that impact the transport of African ozone to Asia in January (a) and July (b). TEJ stands for the tropical easterly jet. AEJ-N for the northern African easterly jet, AEJ-S for the southern African easterly jet, EAWM for the East Asian winter monsoon, EASM for the East Asian summer monsoon, SASM for the South Asian summer monsoon, SAH for the South Asian High, and ITCZ for the Intertropical Convergence Zone.