Tropical tropospheric ozone and carbon monoxide distributions: characteristics, origins and control factors, as seen by IAGOS and IASI

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Abstract. The characteristics and seasonal variability of the tropical tropospheric distributions of ozone (O_3) and carbon monoxide (CO) were analysed based on in situ measurements provided by the In-service Aircraft for a Global Observing System (IAGOS) program since 1994 and 2002 respectively, combined with observations from the Infrared Atmospheric Sounding (IASI) instrument on board the Met-op A satellite since 2008. The SOFT-IO model, which couples back trajectories with CO ~~emissions~~ emission inventories, was used to explore the origins and sources of the tropical CO observed by IAGOS. ~~The highest tropical~~

The highest O_3 and CO ~~maxima occur over Northern Tropical (NT)~~ mixing ratios occur over Western Africa in the low troposphere (LT~~;~~ : surface to 750 hPa) during the ~~dry~~ fire season (75 ppb of O_3 at 2.5 km and 850 ppb of CO at 0.3 km over Lagos in January)~~. Despite the active local fires, local~~ , mainly due to anthropogenic (AN) emissions~~(58 %) are dominant for the CO. The importance of the local AN emissions are highlighted over Central Africa, as they cause a persistent polluted surface layer during the transition seasons (40 % in October and 86 % in April). The second highest~~ , and a major contribution from fires. The secondary maxima are observed in Asia, in mid (MT: 750–300 hPa) and upper (UT: 300–200 hPa) troposphere in April for O_3 and in LT in January for CO, with larger contributions from AN emissions. The lowest O_3 and CO ~~maxima are observed over Asia. Local or regional Asian AN emissions cause the CO maximum in the LT(at 0.5 km) in January, and the O_3 maximum in above 6 km in the post-monsoon season (April). South China is the only Asian site where O_3 peaks in the LT (~~mixing ratios occur over Caracas.

In the tropical LT, the majority of the clusters are affected by local and regional AN emissions. The highest AN impact is found over Asia, Arabia and Eastern Africa, and South America (> 75 ~~ppb at 2.5 km), due to local fires (30 %)in addition to the local (52 %~~ of CO). Biomass burning (BB) emissions also originate from local or regional sources but with stronger seasonal dependence. The highest BB impact is found over Southern Tropical Africa (57–90 %), except in April, mostly due to local fires, but also from Northern Hemisphere Africa in January (45–73 %) and ~~regional (15 %) AN emissions~~Southern Hemisphere South America in October (29 % over Windhoek). ~~The~~

In the MT and UT, AN emissions are more important and dominate in the Eastern part of the Tropics (from Middle East to Asia). BB contributions are more important than in the LT, especially from the African fires in January and July, and from South-East and Equatorial Asia in April and October.

The overall highest amount of ~~transported CO originates from Africa. The~~ CO is exported from Africa, with main transport pathway ~~is~~ from the dry-season African regions towards the wet-season ones. ~~Contributions from Northern Hemisphere Africa are found over Arabia and Eastern Africa (up to 70 %) during the dry season and over South America all year long in the mid (MT; 750–300 hPa) and upper troposphere (UT; 300–200 hPa)(18–38 % over Caracas on annual basis).~~ In contrast, the impact of the Asian emissions in the LT and MT is limited on a local or regional scale. ~~Export~~ The transport of polluted Asian air masses is important in the UT during the Asian summer monsoon and post-monsoon seasons, when convection is active. ~~The AN Asian contributions are mostly found over Arabia and Eastern Africa (up to 80 %) during the Asian summer monsoon. During the post-monsoon, CO impacted by the Indonesian fires (resp. SouthEast Asian AN emissions) are transported towards Eastern Africa (64% and 16%) due to the Tropical Easterly Jet. The lowest O_3 and CO levels are observed over South America, due to less strong local emissions in comparison to Asia and Africa. The only important CO and O_3 enhancement is observed in the MT during the local fires (October), when O_3 and precursors impacted by the local AN and fire emissions are trapped in an anticyclone and transported towards Southern Africa (5–10 ppb from Northern and Southern Hemisphere South America respectively).~~

1 Introduction

Tropospheric ozone (O_3) and carbon monoxide (CO) are key components in the atmosphere. O_3 has a significant impact on human health close to the surface (Curtis et al., 2006; Jerrett et al., 2009) and on climate by being a powerful greenhouse gas (Gauss et al., 2003; IPCC, 2021). O_3 is a secondary pollutant produced by photochemical oxidation of precursors such as CO and volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x) (Logan et al., 1981). Its distribution is controlled by: ~~stratospheric transport (Stevenson et al., 2013); transport~~ transport processes at intercontinental and hemispheric scale (Wild et al., 2004); emissions of precursors (natural and anthropogenic)~~and~~ ; destruction processes (photochemical and depositional) (Monks et al., 2015) and stratospheric transport (Stevenson et al., 2013). Due to its longer lifetime, CO is considered a powerful pollution tracer of combustion products at a hemispheric level (Logan et al., 1981). CO impacts the oxidation capacity of the atmosphere by being the major sink of OH radicals in non polluted atmosphere (Lelieveld et al., 2016), and the climate by producing greenhouse gases, such as CO_2 and O_3, during its oxidation (Myhre et al., 2013). CO is primarily emitted by incomplete combustion, thus by anthropogenic (AN) (including biogenic fuels) and biomass burning (BB) (wildfires) sources (Galanter et al., 2000; Granier et al., 2011), with contributions between 450–600 and 350–600 Tg CO yr^{-1} respectively

(Lamarque et al., 2010; van der Werf et al., 2006). Its secondary sources include oxidation of VOCs and methane (450–1200 and 600– 1000 Tg CO y^{-1} resp.; ~~(Stein et al., 2014)~~Stein et al., 2014).

Recent studies (Gaudel et al., 2018, 2020; Zhang et al., 2016) have shown increasing tropospheric O_3 burden in the second half of the 20th century mostly due to increase of precursors in the ~~tropical regions~~Tropics. Based on aircraft observations, Bourgeois et al. (2020) recently presented a global-scale distribution of O_3 in the remote troposphere. However, uncertainties still remain in the global O_3 distribution and sources of precursors due to paucity of observations in the free troposphere, especially over developing countries in the ~~tropics~~ Tropics (Gaudel et al., 2018; Tarasick et al., 2019).

The ~~tropical region is~~ Tropics are of particular interest regarding tropospheric O_3 and CO. ~~It combines~~They combine: i) intense photochemistry due to high UV radiation and humidity, ii) large active natural sources of CO and other O_3 precursors through BB (Ziemke et al., 2009), biogenic (Aghedo et al., 2007) and lighting emissions (Sauvage et al., 2007b, c), iii) increasing AN due to rapid industrialisation ~~(Granier et al., 2011; Duncan et al., 2016)~~(Duncan et al., 2016; Granier et al., 2011), iv) large ~~ozone~~ O_3 net production potential because deep convection can transport surface emissions to higher altitudes, where their lifetime is increased due to lack of surface deposition and dilution with unpolluted background (Pickering et al., 1995) and v) dynamic processes capable of redistributing chemical species in a regional and global scale (Zhang et al., 2016). Thus, the ~~tropics~~ Tropics are a region where O_3 production is favoured.

Satellite observations from the OMI and MLS sensors (Ziemke et al., 2019) and simulations from the GEOS-Chem chemical transport model (Zhang et al., 2016, 2021) display the highest O_3 burden increase in the ~~tropical region~~ Tropics - mostly over India, East Asia and SouthEast Asia. Most studies tend to confirm an increase of O_3 in the ~~tropics~~ Tropics but they are mostly based on model simulations, sparse ground observations or satellite data with little consistency, and it is not clear what can cause such an increase. Indeed the trends are attributed to different factors such as BB (Heue et al., 2016), dynamics (Lu et al., 2019; Thompson et al., 2021), or AN (Zhang et al., 2016; Gaudel et al., 2020). Thus, further investigation based on ~~*in situ*~~ in situ observations is required in order to better ~~constraint~~ constrain models, validate satellite retrievals, and reduce the uncertainties in the quantification of O_3 and CO trends and source attribution over the ~~tropics~~Tropics.

Measurements of tropical O_3 and CO are available by satellite observations, but they have a coarse vertical resolution (e.g. Barret et al., 2008; Thompson et al., 2001). On short time scales, several field campaigns have been carried out in the ~~tropics~~Tropics, yielding measurements of various species over Africa (from TROPOZ 1987 to CAFE-Africa), Asia (from INDOEX to EMerge-Asia), South America (Cite-1/2/3, TROCCiNOX) and the tropical Pacific (from PEM-WEST-A/B to CAST/CONTRAST/ATTREX and Atom). These campaigns have provided invaluable insights on the atmospheric chemistry and dynamics of the ~~tropical region~~Tropics. On greater timescales, the Southern Hemisphere ADditional OZone Sounding (SHADOZ) program (Thompson et al., 2003a) provides long-term O_3 observations over the ~~tropics~~ Tropics using ozonesondes since 1998. These measurements have ~~offered~~ allowed a better understanding on the vertical distribution and trends of tropical O_3 (e.g. Thompson et al., 2021).

In a complementary way to these datasets, the IAGOS (In-service Aircraft for a Global Observing System~~;~~) (Marenco et al., 1998; Petzold et al., 2015; Thouret et al., 2022) program has provided O_3 and CO measurements over the ~~tropics~~ Tropics since 1994 and 2002 respectively. Using equipped commercial aircraft, IAGOS samples vertical profiles at take off and

landing, along with the lower part of the upper tropical troposphere at cruise altitude between 300 and 185 hPa (UT_{cruise}). Previous studies have documented the tropical composition over Africa ~~(Sauvage et al., 2005, 2007a, d; Lannuque et al., 2021)~~ (Lannuque et al., 2021; Sauvage et al., 2005, 2007a, d), South America (Yamasoe et al., 2015) and South Asia (Sahu et al., 2014; Sheel et al., 2014) using IAGOS data. However, they are focused on specific regions of the ~~tropics~~ Tropics and have limited temporal coverage, especially for CO as fewer measurements were available at this time. Thus, the O_3 and CO distributions and their interlocking in the entire ~~tropics~~ Tropics are still not well documented.

The SOFT-IO model (Sauvage et al., 2017) has been developed to supplement the analysis of the IAGOS dataset by estimating AN and BB contributions to the observed CO measurements. These measurements, along with the SOFT-IO output allow us to trace the CO origin over the ~~tropics~~Tropics. Further, global distributions provided by Infrared Atmospheric Sounding Interferometer (IASI)-Software for a Fast Retrieval of IASI Data (SOFRID) (Barret et al., 2011; De Wachter et al., 2012) retrievals since 2008 complement the O_3 and CO distributions provided by IAGOS. They allow us to understand the spatial extent of pollution plumes, and explore intercontinental transport patterns.

In this article we take advantage of the unique IAGOS database to (i) document the characteristics and seasonal variability of these two atmospheric species over the whole tropical band for the last decade, (ii) explore the origin of the observed CO anomalies, (iii) investigate transport processes driving the CO and O_3 ~~distribution in the tropics~~distributions in the Tropics.

The observational (IAGOS and IASI) and model based (SOFT-IO) datasets, and methodology are introduced in Sect. 2. In Section 3, the IAGOS observations are analysed to document O_3/CO vertical profiles, along with the upper tropospheric composition over the ~~tropics~~Tropics. In addition, the sources of observed CO are explored with SOFT-IO.

2 Data and Methods

2.1 IAGOS observations

The Research Infrastructure IAGOS (Petzold et al., 2015; Thouret et al., 2022) provides ~~*in situ*~~ in situ measurements of trace gases (O_3, CO, water vapour, NO_y between 2001 and 2005 (e.g. Gressent et al., 2014), and more recently NO_x, CH_4, CO_2 and cloud particles, see https://www.iagos.org/iagos-data/) and meteorological parameters (temperature and winds), using equipped commercial aircraft. Full description of the instruments can be found in Nédélec et al. (2015). O_3 (resp. CO) is measured using a dual-beam ultraviolet absorption monitor (resp. infrared analyser) with an overall uncertainty of ± 2 ppbv ± 2 % (resp. ± 5 ppbv ± 5 %) and a time resolution of 4 (resp. 30) seconds (Nédélec et al., 2015). IAGOS measures vertical profiles during ascend and descend phases, and the UT_{cruise} during cruise phases. Considering the aircraft speed (7–8 m s^{-1} during ascent/descent; 900 km h^{-1} during cruise), the time resolution of the instruments corresponds to a vertical resolution of 30 m (resp. 225 m), and a horizontal resolution of 1 km (resp. 7.5 km) for O_3 (resp. CO).

O_3 (resp. CO) observations have been collected since 1994 (resp. 2002) in the frame of the IAGOS Research Infrastructure and its predecessor MOZAIC (Marenco et al., 1998) program, based on the same instrument technologies. Good consistency in the measurements between the two programs (hereafter referred to as IAGOS) ~~(Nédélec et al., 2015; Blot et al., 2021)~~ (Blot et al., 2021; Nédélec et al., 2015) leads to IAGOS temporal coverage of 26 (resp. almost 20) years for O_3 (resp. CO), de-

pending on the availability of the flights. IAGOS data provides robust O_3 and CO climatologies, allowing studies of long-term trends (e.g. Cohen et al., 2018) along with validation of chemistry transport models ~~(e.g. Sauvage et al., 2007b; Gressent et al., 2016)~~ (e.g. Gressent et al., 2016; Sauvage et al., 2007b) and satellite data retrievals (e.g. De Wachter et al., 2012) on a global scale. To complement the IAGOS observations, we use the potential vorticity (PV) field, which is part of the ancillary data (https://doi.org/10.25326/3) from the IAGOS database. The PV is calculated from the European Centre for Medium-Range Weather Forecast's (ECMWF) operational fields (horizontal resolution 1°, time resolution 3 hours), interpolated along IAGOS trajectories.

2.1.1 Data treatment

The tropical zone can be defined in several ways, such as by meteorological characteristics (e.g. location of the subtropical jets), climatic elements (e.g. precipitation rates) or by the geographical extent. Following the latter way, in the Tropospheric Ozone Assessment Report, Phase II (TOAR-II; https://igacproject.org/activities/TOAR/TOAR-II) Ozone and Precursors in the Tropics working group, the ~~tropics~~ Tropics are defined between 20° S and 20° N. In our study, we consider the extended area between 25° S and 25° N, in order to investigate interactions of pollution and the transport of air masses between the ~~tropics and the subtropics~~Tropics and the Subtropics. Only tropospheric measurements are taken into account, by applying a PV filter of 2 PV units (pvu) for each measurement during cruise phase, and for the measurements between 20–25° N/S during ascend/descend. The UT_{cruise} climatologies are derived by averaging the cruise data on a 2 x 2.5° grid, for the period 1994–2020 (resp. 2002–2020) for O_3 (resp. CO). For the same time periods, the climatologies over the vertical are derived by averaging the data into 10 hPa pressure bins from the surface up to 200 hPa. ~~We also applied a distance criteria of 300 km around the IAGOS observational site~~Data are selected within a 300km radius circle centered on the airport location, similar to Petetin et al. (2016). This way we reduce uncertainties due to possible horizontal heterogeneity in the measurements, as the aircraft keeps moving in the horizontal plane during ascent and descent.

Table 1 shows the total number of profiles taken into account for this study for each ~~site~~ location for the whole IAGOS period. The temporal availability of the measurements differ for each ~~site~~ location and cluster, as it depends on the flight schedule of the aircraft (see ~~Figs. S4 and S5~~Fig. S1 and Fig. S2). For this reason, to determine a reliable climatological profile, we need to assess the statistical significance of the data. Similar to Logan (1999) and Sauvage et al. (2005), we compute the relative standard error (RSE) of the O_3 (CO) monthly mean, versus the number of flights per month. The RSE is defined as the ratio between the standard error ($SE = \frac{\sigma}{\sqrt{N}}$, with σ the square root of the sample variance and N the number of flights) and the O_3 (CO) monthly mean. The minimum number of flights required for statistical significance corresponds to the number above which $RSE \leq 10$ %. We choose RSE less than 10 %, because RSE depends not only on the number of measurements, but also on the O_3 (CO) variability which is high over the ~~tropics~~ Tropics (Thompson et al., 2003b). For each ~~site~~ location with an adequate number of flights per month, we consider an individual profile of O_3 (CO). Otherwise, we combine ~~sites~~ locations in clusters, in order to increase the amount of data and get statistically significant climatological profiles. Besides, the clusters can be useful for validation of models with a coarse horizontal resolution, because they represent a wider area as resolved by the models, which are not expected to capture small-scale variations in the ozone field (e.g. Emmons et al., 2010). For clustering,

the ~~sites~~ locations should be: i) in relatively close distance from each other, ii) governed by similar meteorological conditions, and iii) display similar characteristics in the vertical distribution of O_3 (CO) (see Sect. 3.1).

The meteorological conditions in the ~~tropics~~ Tropics are peculiar, with different seasonal patterns depending on the region. For instance, in Africa the main seasons are two (dry and wet) with two intermediate seasons passing from wet to dry and vice versa ~~(Sauvage et al., 2005; Lannuque et al., 2021)~~(Lannuque et al., 2021; Sauvage et al., 2005). On the other hand, in Asia the seasons are defined by the Asian monsoon phases: Asian summer monsoon (wet season); Asian winter monsoon (dry season) and post monsoon. Thus, we considered it more appropriate for our analysis to deviate from the classical definition of the seasons, which fits better to studies concerning higher latitudes. Instead, we analyse the O_3/CO profiles and horizontal distributions for months during the peak tropical seasons (January, April, July and October), to highlight seasonal patterns.

2.2 SOFT-IO model

The SOFT-IO (SOft attribution using FlexparT and carbon monoxide emission inventories for In-situ Observation database) tool (Sauvage et al., 2017; http://dx.doi.org/10.25326/2) has been developed to investigate the origin of the observed IAGOS-CO, by coupling FLEXPART 20-days backward transport simulations with emission inventories. For each point of IAGOS trajectory, SOFT-IO estimates the CO contribution coming from 14 different geographical regions (see Fig. 1), for AN and BB origin separately. We use Community Emissions Data System (CEDS2) AN emissions (McDuffie et al., 2020) and the Global Fire Assimilation System (GFAS) BB emissions (Kaiser et al., 2012) which include fire injection heights, to discriminate sources of CO anomalies over different regions of interest. For the calculations, the AN (resp. BB) emissions are updated on a monthly (resp. daily) basis.

SOFT-IO estimates the contribution to CO anomalies emitted by primary sources during the last 20 days, while it does not calculate the background CO. The background CO can be emitted by primary sources older than 20 days, and by secondary sources such as oxidation of methane and non-methane volatile organic compounds. The meteorological fields are based on 1° x 1° ECMWF analysis and forecast with a time resolution of 6 and 3h respectively.

Several studies (e.g. Cussac et al., 2020; Lannuque et al., 2021; Petetin et al., 2018b) have used SOFT-IO to track back the sources of CO measured by IAGOS. Sauvage et al. (2017) validated SOFT-IO performance against IAGOS CO observations for several regions and tropospheric levels. As detailed in their study, SOFT-IO uncertainties and biases are mostly due to uncertainties in emission inventories, and to a lesser extent to uncertainties concerning the meteorological fields and FLEXPART transport parameterizations (turbulence, convection). Their results show that SOFT-IO can simulate 95 % of the observed number of anomalies, without any strong dependence on altitude or region of the CO plume. SOFT-IO captures the intensity of CO anomalies with a bias lower than 10-15 ppb with respect to observed anomalies for most of the regions and tropospheric ~~levels~~layers. The bias is higher in extreme pollution events and might be related to uncertainties in the emissions inventories.

In our study, CO anomalies are defined as the positive difference between the observed and the background CO mixing ratio. ~~Background~~ The background CO mixing ratio represents a reference value, not affected by surface emission or pollution events. For this reason, it is computed as the monthly climatological median CO of a remote area away from polluted regions, in the UT_{cruise} (during the whole study period 2002–2020)~~(Sect. S4 for more details)~~. Overall in the Tropics, depending of the month

Table 1. Description of individual ~~sites~~ locations and clusters used in this study. The ~~location of the sites is~~ locations are displayed in Fig. 1. ~~NT and ST indicate Northern and Southern Tropical Africa respectively.~~ P O_3 and P CO indicate the total number of profiles for the IAGOS period.

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	Individual ~~sites~~locations/Clusters	IAGOS ~~sites~~ locations	P O_3	P CO
South America	South Brazil	Rio de Janeiro (Brazil), São Paulo (Brazil)	518	62
	Caracas	Caracas (Venezuela)	414	248
	Bogota	Bogota (Colombia)	190	142
~~NT Africa~~ Western Africa	Lagos	Lagos (Nigeria)	311	199
	Sahel	Abuja (Nigeria), Ouagadougou (Uganda), Niamey (Niger)	193	202
	Gulf of Guinea	Lome (Togo), Yaounde (Cameroon), Douala (Cameroon), Libreville (Gabon), Accra (Ghana), Abidjan (Ivory Coast), Malabo (Equatorial Guinea) Cotonou (Benin), Port Harcourt (Nigeria)	414	302
~~ST Africa~~ Southern Tropical Africa	Central Africa	Luanda (Angola), Brazzaville (Congo) Kinshasa (Democratic Republic of Congo)	169	89
	Windhoek	Windhoek (Namibia)	651	692
Arabia and Eastern Africa	AbuDhabi	Abu Dhabi (United Arab Emirates), Muscat (Oman)	118	56
	Khartoum	Khartoum (Sudan)	157	116
	Addis Ababa	Addis Ababa (Ethiopia)	121	110
	Jeddah	Jeddah (Saudi Arabia)	154	108
Asia	South China	Hong Kong (China), Guangzhou (China), Xiamen (China)	337	562
	Gulf of Thailand	Kuala Lumpur (Malaysia), Singapore (Singapore)	162	140
	Madras	Madras (India)	239	253
	Hyderabad	Hyderabad (India)	159	170
	Mumbai	Mumbai (India)	61	29
	Manila	Manila (Philippines)	66	101
	Bangkok	Bangkok (Thailand)	526	336
	Ho Chi Minh City	Ho Chi Minh City (Vietnam)	113	90

and region, a 80 ppb to 100 ppb background has to be added to SOFT-IO for direct comparison with IAGOS observations. We performed an evaluation of SOFT-IO for the ~~lower troposphere (LT,~~ low (LT: surface–750 hPa), ~~the mid troposphere (MT,~~ mid (MT: 750–350 hPa), and upper ~~troposphere (UT,~~ (UT: 300–200 hPa) troposphere. Our simulations detect CO anomalies at the same rates as Sauvage et al. (2017). On average, SOFT-IO underestimates the observed CO anomalies by 10 ppb in the MT and UT, and by 45 ppb in the LT. ~~A sensitivity test has shown absolute differences of 27~~When looking at the differences between IAGOS and SOFT-IO anomalies (taking into account the background not simulated by the model), SOFT-IO reproduces on average more than 93% of the observed anomalies for the 3 layers and 87% in the LT~~, 16% in the MT and 10% in the UT between~~ . April is the month where the model gives the largest underestimation, with 78% of the anomalies simulated on average over the 20 clusters (13% standard deviation). The lowest performance is in the LT of Sahel in January, with 52% of the anomaly simulated by SOFT-IO~~simulations using AN emissions from MACCity and from CEDS2. This clearly highlights the large uncertainty stemming from uncertainties in AN emissions.~~ . Another source of uncertainty comes from the definition of background CO. ~~In order to~~ To assess this source of uncertainty, we used ~~the 600–300 hPa~~ an alternative definition for the background calculated as the median CO mixing ratio ~~as background for each site~~between 600 and 300 hPa for each location. The differences between ~~the two backgrounds~~ this background and the one used in the study are within 2.5-60 ppbv. Nevertheless, using the alternative background did not make any difference in the anomaly source attribution and in the relative contributions.

2.3 IASI-SOFRID observations

The IASI sensor onboard MetOp-A (launched in 2006) has a 12 km footprint at nadir and a 2200 km swath allowing an overpass twice daily at 9:30 and 21:30 local solar time. IASI provides information for the atmospheric composition e.g. content of trace gases such as O_3 ~~(Eremenko et al., 2008; Barret et al., 2011; Boynard et al., 2016), CO (George et al., 2009; De Wachter et al., 2012)~~ (Barret et al., 2011; Boynard et al., 2016; Eremenko et al., 2008), CO (De Wachter et al., 2012; George et al., 2009) and N_2O (Barret et al., 2021). We use O_3 (v3.5) and CO (v2.1 ~~up to 2014,~~ and v2.2 ~~up to 2019~~after 2014) IASI retrievals performed with SOFRID (Barret et al., 2020; De Wachter et al., 2012).

SOFRID-O_3 v3.5 retrievals use a dynamical a priori profile based on latitude, season and the tropopause height (Barret et al., 2020). In the ~~tropics~~Tropics, where the surface temperature, thermal contrast and tropopause height are the highest, SOFRID-O_3 retrievals allow two independent pieces of information, one in the troposphere and one in the UTLS (Barret et al., 2020). Comparisons with ozonesonde measurements for the period 2008–2017, showed that SOFRID-O_3 is biased low in the tropical troposphere and UTLS, by 3 $\pm$ 16 % and 12 $\pm$ 33 % respectively in the Northern Tropics (0–30°N), and by 8 $\pm$ 14 % and 21 $\pm$ 30 % in the Southern Tropics (0–30°S) (Barret et al., 2020). The measurements above Northern Africa are erroneous, due to retrieval problems in the presence of desert ground with sand emissivity interfering with the O_3 signature (Boynard et al., 2018). The stripes along the 10°latitude bands in IASI O_3 maps (Fig. 2 e–h and q–t) are due to the use of a dynamical a priori profile, resulting in discontinuities between adjacent latitude bands with different a priori profiles. Nevertheless, the use of a dynamical a priori profile largely improves the retrieved O_3 profiles in terms of biases, variability and correlation relative to the previous version based on a single a priori profile (Barret et al., 2020).

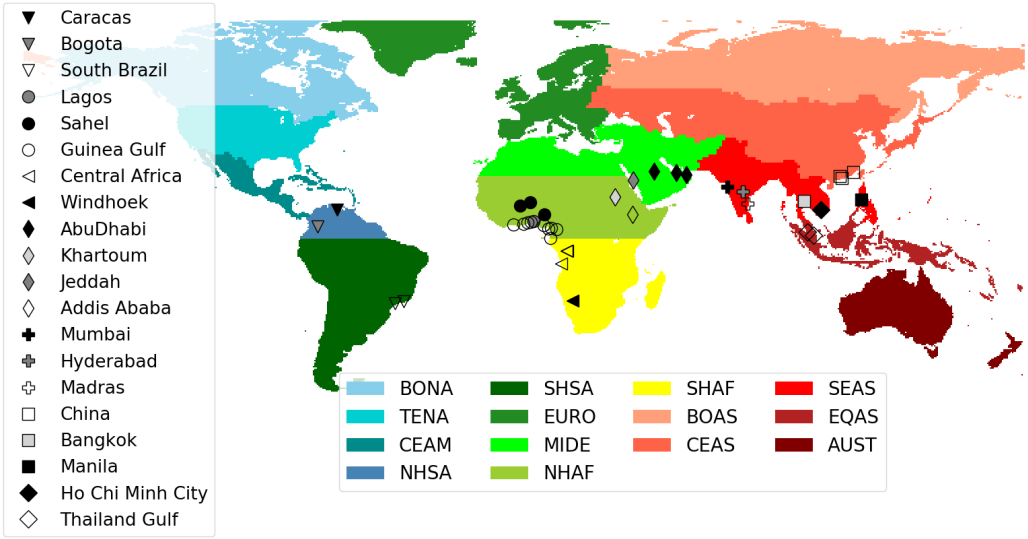

Figure 1. ~~Locations of tropical sites~~ Tropical locations served by IAGOS, and geographical source regions used in SOFT-IO model. BONA: Boreal North America; TENA: Temperate North America; CEAM: Central America; NHSA: Northern Hemisphere South America; SHSA: Southern Hemisphere South America; EURO: Europe; MIDE: Middle East; NHAF: Northern Hemisphere Africa; SHAF: Southern Hemisphere Africa; BOAS: Boreal Asia; CEAS: Central Asia; SEAS: SouthEast Asia; EQAS: Equatorial Asia; AUST: Australia and New Zealand.

For SOFRID-CO ~~v(2.1~~ (v2.1 and 2.2 after 2014), two independent pieces of information are provided in the low and upper troposphere (De Wachter et al., 2012). IASI correctly captures the seasonal variability of CO over southern Africa (Windhoek) and European mid-latitudes (Frankfurt) in the low (resp. upper) troposphere relative to IAGOS data (resp. correlation coefficients of 0.85 (0.70)). At Windhoek, SOFRID-CO is biased low in the low (resp. upper) troposphere by 13 $\pm$ 20 % (resp. 4 $\pm$ 12 %).

We use monthly averaged SOFRID-CO and O_3 retrievals on a 1° x 1° grid from 2008–2019. We focus on daytime measurements when larger thermal contrast between the surface and the atmosphere results in increased sensitivity of the instrument (Clerbaux et al., 2009) and on pressure levels corresponding approximately to the independent pieces of information: low troposphere defined between 900–700 hPa ($LT_{\sout{iasi}IASI}$), mid troposphere between 600–400 hPa ($MT_{\sout{iasi}IASI}$) and upper troposphere between 290–220 hPa ($UT_{\sout{iasi}IASI}$).

3 Results

3.1 Regional characteristics of tropical O_3 and CO

Figure 2 displays the horizontal distributions of CO in LT_{iasi} $_{IASI}$ (a–d)~~and UT_{iasi}~~ , UT_{IASI} (i–l~~and~~) and UT_{cruise} (q–t), and of O_3 in the MT_{iasi} $_{IASI}$ (e–h)~~and UT_{iasi}~~ , UT_{IASI} (m–p~~and~~) and UT_{cruise} (u–x). The results shown in Fig. 2 ~~(a–d)~~ motivated our choice in combining IAGOS ~~sites~~ locations in clusters when it is necessary to increase the number of measurements. ~~The LT_{iasi} CO maxima, like over the Gulf of Guinea, cover a wide area. Thus, cities located close to each other are likely to experience similar air masses. According to the wind maps they are also affected by similar meteorological conditions (Fig. S3).~~

Figures 3, 5, 6, 7 (panels 1 and 2) display the monthly average vertical distributions of O_3 and CO based on IAGOS data, for the African, Asian, South American, Arabian and Eastern African clusters. Panels 3 to 5 represent the mean ~~contribution to these~~ recent contribution to CO mixing ratios anomalies (above the background) from AN and BB emissions as estimated by SOFT-IO, with information about their geographical origin (see Sect. 2.2 and Fig. 1). To better ~~understand~~ summarise O_3 and CO anomalies, Fig. 4 displays the CO contributions for the most representative clusters in three tropospheric layers related to different dynamical regimes: LT corresponding roughly to planetary boundary layer; MT above, and UT corresponding to the beginning of convective detrainment.

3.1.1 Africa

The striking feature of CO and O_3 over the African ~~clusters is the LT~~ tropical clusters (Sahel, Gulf of Guinea, Lagos and Central Africa) is the low tropospheric maxima during the dry season (January for the Northern Hemisphere and July for the Southern Hemisphere) below 4km (Fig. 3 panels 1–2 ~~). CO~~ and Fig. S3 panels 1–2). Above CO and O_3 display mixing ratio close to the background. Enhanced O_3 and CO are confined in the low troposphere due to the stability of dynamical circulations which prevent vertical mixing (Sauvage et al., 2005). CO maximises close to the surface, with larger mixing ratios over Lagos (850 ppb) than Sahel (500 ppb)~~and~~ , Gulf of Guinea ~~and~~ or Central Africa (400 ppb). ~~The~~ O_3 ~~gradient close to the surface is~~ displays a vertical gradient from the minimum at the surface, likely related to surface deposition and titration by highly concentrated nitrogen oxide ~~(NO)(Monks, 2005)which is expected along the high CO concentration. The~~ (Monks, 2005), to the maximum near 2.5km, likely indicating chemically processed air masses. At Windhoek, O_3 ~~maximum and the elevated CO levels (exceeding 300–500 ppb) at 2.5 kmover the four clusters , likely indicate chemically processed air masses.~~ and CO have different vertical distributions, with maxima in October at the end of the Southern Hemisphere dry season (Fig. 3 panels 1c and 2c). Windhoek CO enhancement has the smallest magnitude among the African clusters (150 ppb at 4 km), while the O_3 peak is among the largest, reaching 80 ppb in the UT (11.5 km).

The annual minima of CO and O_3 over the African clusters occur during the transition from the wet to the dry season (April and October), when the local fires are minimum (Fig. S4b and d).

~~During the Northern Hemisphere dry season (January), the AN contribution dominates over Lagos (58%) and Sahel(57%), while BB slightly dominates over~~ Regional fires generally dominate CO contributions in the troposphere of Central Africa and Windhoek (Fig. 3 panel 4 and 5; Fig. 4 and Fig. S5). For the clusters located over Western Africa (Sahel, Gulf of Guinea ~~(53~~

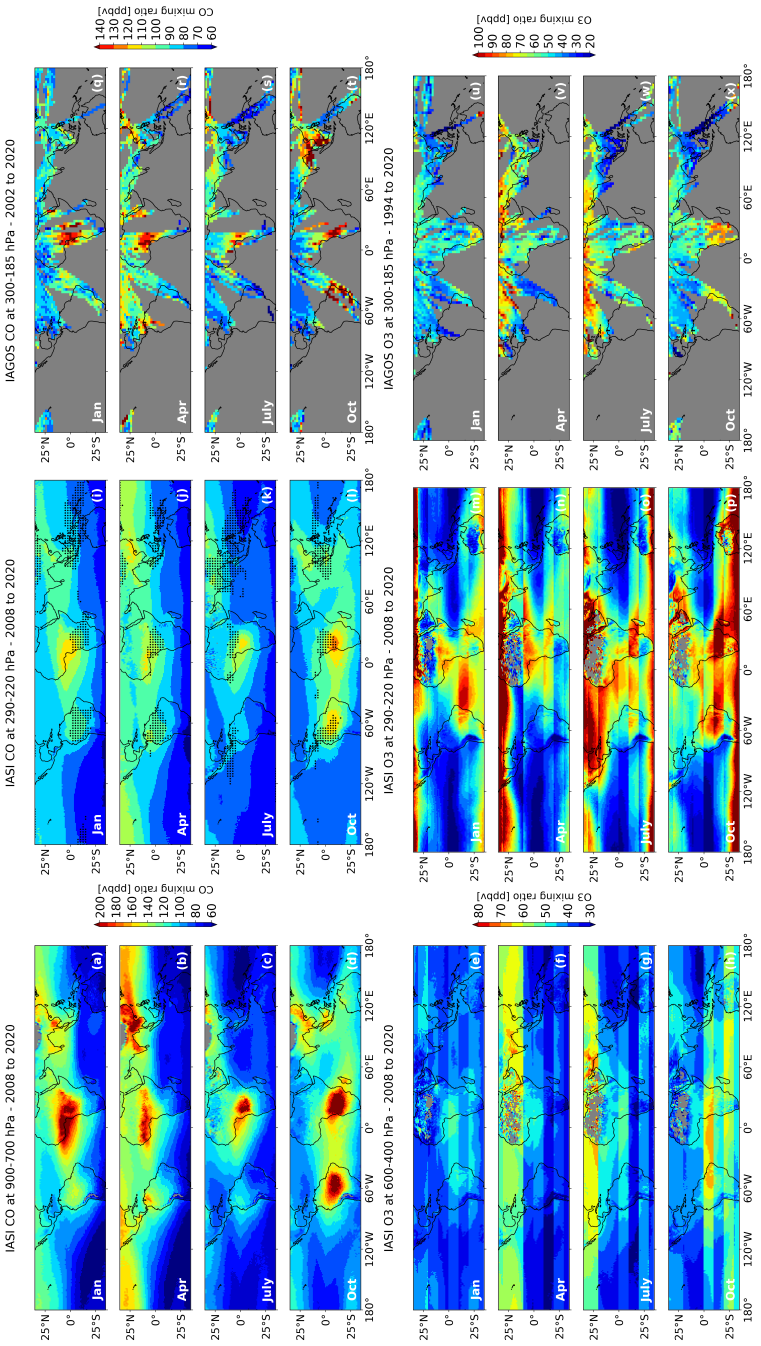

Figure 2. Monthly mean low tropospheric IASI CO distributions (900–700 hPa; a–d), mid tropospheric IASI O_3 distributions (600–400 hPa; e–h), upper tropospheric CO and O_3 distributions based on IASI (290–220 hPa; i–l and m–p resp.) and IAGOS (300–185 hPa; q–t and u–x resp.).

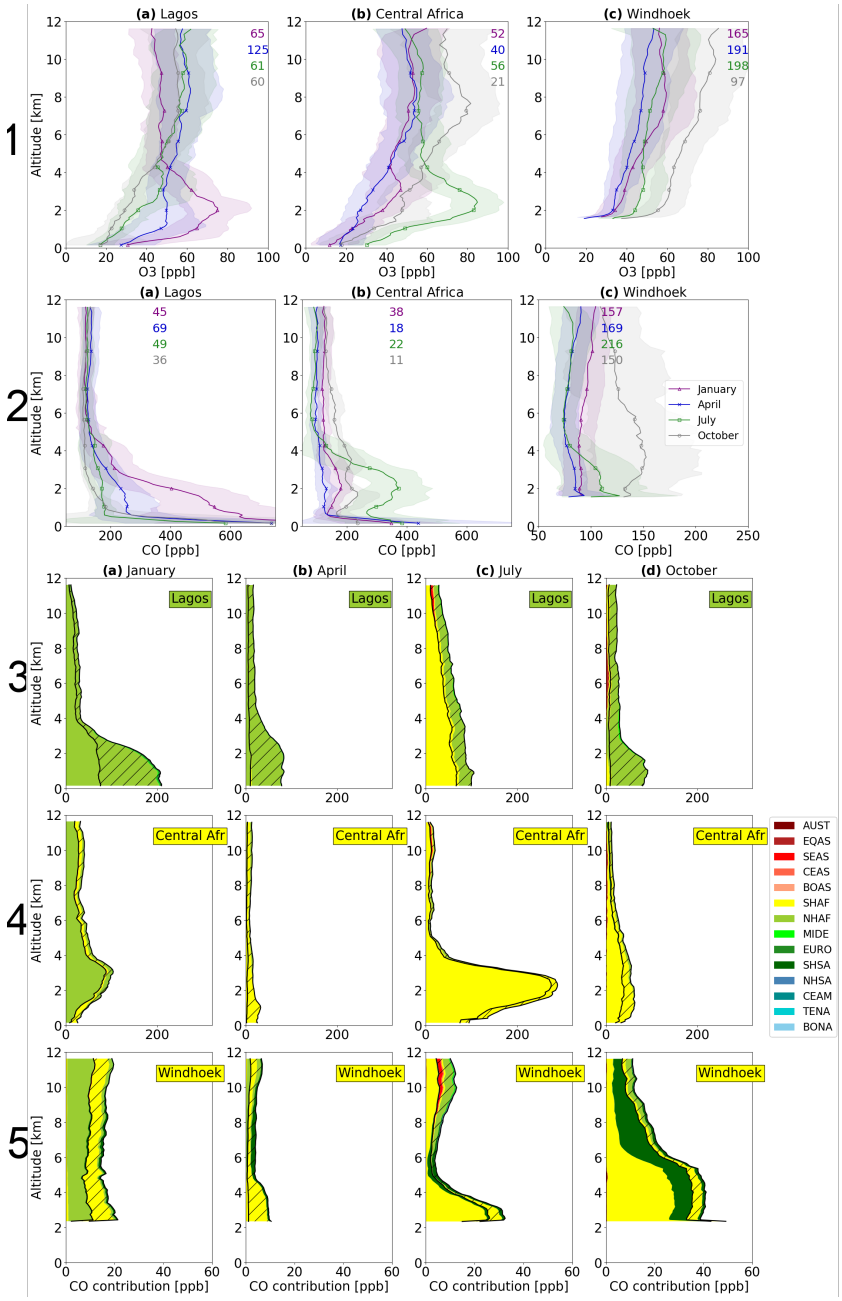

Figure 3. IAGOS monthly mean O_3 (panel 1) and CO (panel 2) vertical distributions. The annotated numbers correspond to the total number of flights per month for the IAGOS period, given in the same colour as in the legend. The shadowed part corresponds to ± 1 ~~one~~ standard deviation. Vertical distribution of CO contributions (in ppb) (panels 3 to 5) with the hatched part showing AN contribution, and the non-hatched part BB contribution. For clarity reasons the CO contribution for Sahel and Gulf of Guinea are displayed in Fig. ~~??~~S6.

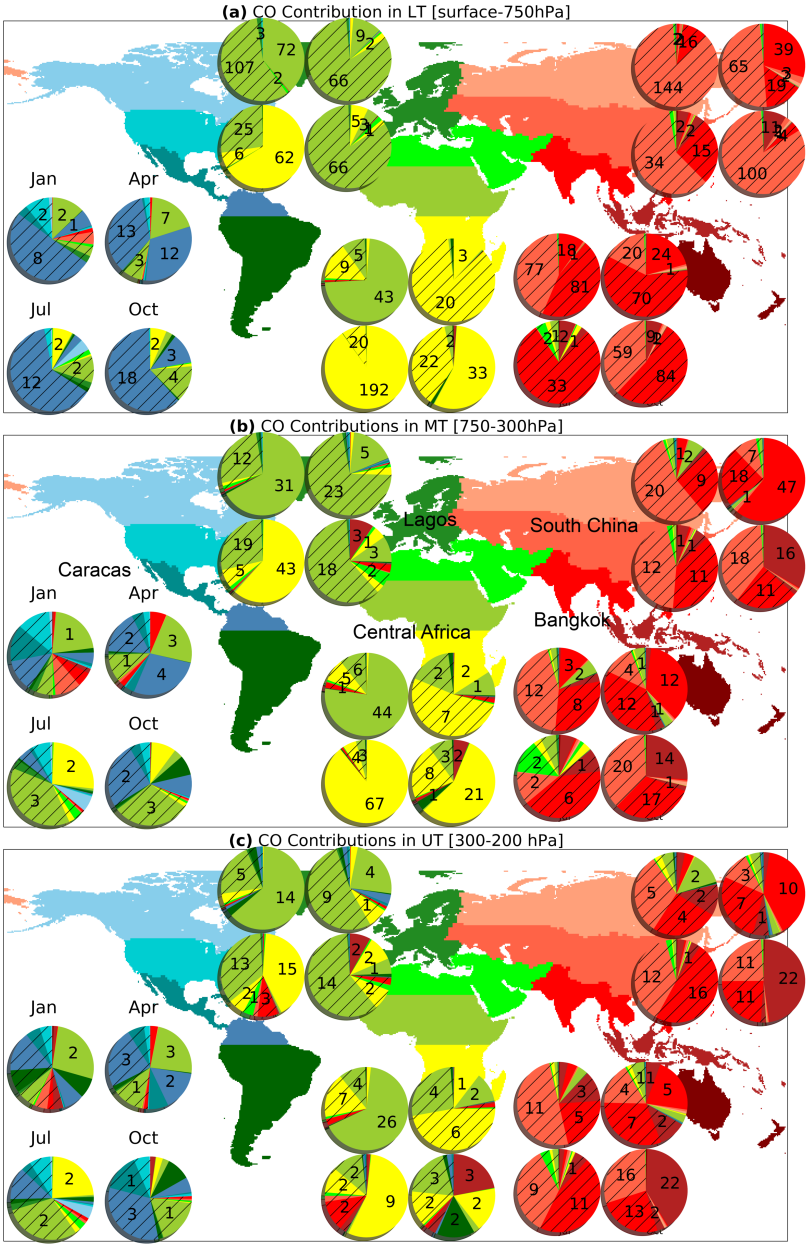

Figure 4. Mean SOFT-IO contributions (in ppb), averaged over all the positive CO anomalies for the tropical ~~sites~~ locations (Caracas, Lagos, Central Africa, South China and Bangkok) for LT (a), MT (b) and UT (c) for the AN (hatched) and BB (unhatched) contributions. Each pie corresponds to a different month and each group of four pies refer to a different ~~site~~ location (see panel b).

%) (Figsand Lagos), anthropogenic emissions dominate, but during fire seasons, the contribution from fires can be close to that from anthropogenic sources (for instance in January for the 3 clusters, in July for Lagos and Sahel) (Fig. 3 panel 3a; ?? panels 1a and 2a; 4a; ??a). Contribution from 3; Fig. 4 and Fig. S6 panels 1 and 2).

In January the Northern Hemisphere African fires is also found over Central Africa, where it intensifies and becomes the only important one contribution reaches 53 % over the Gulf of Guinea and also influences Central Africa with maximum contribution between 2 and 4 km (Fig. 3 panel 4a). During transportfrom Northern Hemisphere Africa4km. During transport, the air masses impacted by BB emissions are chemically processed resulting in the formation of an O_3 secondary maximum of 50 ppb coincident with the CO maximum (Fig. 3 panel panels 1d and 2d), as described in Sauvage et al. (2005). During the Southern Hemisphere dry season (July) , the local (Southern Hemisphere Africa) this month, the AN contribution dominates over Lagos (58%) and Sahel (57%).

In July the Southern Hemisphere African fires are responsible for the CO anomalies over Central Africa in the LT and MT by up to 90% (Fig. 4a and b). Impact of these fires is also found in the NT Africa clusters. CO over Lagos is mostly attributed to Southern Hemisphere African emissions (69 ppb) (Fig. 4a), from the surface to about 5 km (, and also influence Sahel, Gulf of Guinea and Lagos, accounting for up to 68 ppb in the LT of Lagos (Fig. S6 panels 1c and 2c; Fig. 4a and Fig. 3 panel 3c). Similar to Lagos, CO over the Gulf of Guinea and Sahel originates from local AN and Southern Hemisphere African BB emissions (Fig. ?? panels 1c and 2c) . O_3 LT mixing ratio is slightly higher over Central Africa (85 ppb) in July than over Northern Africa (in January) likely indicating rapid photochemical O_3 production by BB precursors (Singh et al., 1996) during the Southern Hemisphere fires.

The air masses above the fires in NT Africa (resp. ST Africa) (Fig. S1a and S1c resp.) during the respective dry seasons are transported from the continent During each previous dry season, BB air masses transport from the fire hemisphere (Fig. S3a and S3c resp.) S4a and c) (where the highest CO is measured) to the opposite hemisphere is allowed either by the north-easterly Harmattan flow (resp. January) or the south easterly winds and monsoon flow (July) (Sauvage et al., 2005). This result is clearly consistent transport is visible with IASI LT_{iasi} IASI CO (Fig. 2a and c) which shows transport from the fire region where the highest concentrations are detected towards Southern Africa in January (resp. the Gulf of Guinea and southern West Africa and seems to be more effective in the south-to-north direction (in July) . In the NT, the enhanced O_3 and CO are confined in the low troposphere due to the stability of the Harmattan flow and Saharan anticyclone which prevent vertical mixing (Sauvage et al., 2005). than in the opposite direction (in January).

Over the NT African clusters, secondary CO and O_3 maxima are observed below 4 km (Fig. 3 panels 1a–1c and 2a–2c)during the transition from the Northern Hemisphere dry to wet season (April), when the fires are suppressed (Fig. S1b). LT CO mainly comes from local AN emissions (Figs. 3 panel 3b; ?? panels 1b and 2b). The fact that SOFT-IO attributes approximately 80 ppbv of CO to local AN emissions (Figs. 3 panel 3b; ?? panels 1b and 2b), while the observed anomaly reaches 200–250 ppbv and no or few fires are detected by MODIS (Yamasoe et al. (2015); their Fig.7), indicates underestimation of the Northern Hemisphere African AN emissions. These high CO concentrations in April are detected by IASI in the LT_{iasi} (Fig. 2b) over the whole of West Africa indicating the large-scale extent of the impact of these emissions. The enhanced LT O_3 over the three clusters in April (Figs. 3a–c) indicates possible O_3 formation during the transport of the aforementioned emissions towards

Sahel and the Gulf of Guinea. A small O_3 enhancement is also detected by IASI in the MT_{iasi} over West Africa (Fig. 2f). During the dry season, nitrogen is accumulated in soils (Jaeglé et al., 2004). Enhanced NO_x concentrations, possibly due to soil emissions when rains start, also contribute to the O_3 increase over NT Africa in April (Saunois et al., 2009), especially over Sahel In October CO peaks over Windhoek because fires are localized further south in Africa during this month, contributing to 68 % versus 12 % for AN (Fig. 3 panel 1a–c)because of higher NO_2 concentrations above dry savannas than over wet savannas and forests (Southern Western Africa) (Adon et al., 2010). After excess nitrogen is consumed, the wet-season NO emissions decrease, contributing less to the local O_3 (Adon et al., 2010).

At Windhoek, O_3 and CO maximise in October after Southern Hemisphere dry season S5a), but also because of fire contribution from South America (20 %) (Fig. 3 panels 1e and 2e). This CO peak has the smallest magnitude among the African clusters (150 ppb at 4 km), while O_3 peak is among the largest, reaching 80 ppb in the UT (11.5 km). The LT CO anomalies over Windhoek in October are mainly caused by local BB emissions (68 % BB versus 12 % AN) (Fig. **??**apanel 5d and Fig. S5a). These high CO concentrations mixing ratios in October are detected by IASI in the LT_{iasi} $_{IASI}$ (Fig. 2d) over the whole of Southern Africa reflecting the large extent of the impact of these emissions. Using MOPPIT CO and MODIS fire count data, Edwards et al. (2006) also noticed the time lag between the peak of the fires and the CO concentration over Southern Africa. They attributed the lag to smoldering fires at the end of the burning season, characterised by low combustion efficiency and increased CO emissions factors (Zheng et al., 2018b). In addition, there is non-negligible influence from Southern Hemisphere South American emissions (20 % mostly BB)(Figs. 3 panel 5d and **??**a). The Southern Hemisphere South American BB The South American fires contribution increases with height (30 % contribution in MT and 50 % in UT) contributing to the O_3 maximum observed in the UT (Fig. **??**b S5b and c) (Sauvage et al., 2006).

An O_3 enhancement of 80 ppb is also observed , also observed at 8km over Central Africa at 8 km in October ((80 ppb) (Fig. 3 panel 1d)highlighting the stronger seasonal variability of MT and UT O_3 in ST than NT Africa related to the intense lightning activity in the Southern Hemisphere and the South Atlantic O_3 maximum (Sauvage et al., 2007b, c). The IASI UT_{iasi} O_3 distribution clearly shows that the O_3 maximum covers the entire region from South America to Africa south of the Equator 1b), both part of the South Atlantic maximum visible on IASI UT_{IASI} (Fig. 2l). p). This maximum is also explained by intense lightning activity (Sauvage et al., 2007b, c).

The annual minima of CO and O_3 over the African clusters occur In April, during the transition from the wet to the dry season (April for ST and October for NT)dry to wet season, when the local fires are suppressed fires are reduced (Fig. S1b and d). Over NT Africa, the S4b), secondary CO and O_3 minimum occurs below about 4km maxima (Fig. 3 panel 1a–c). The CO maximum mixing ratio below 1 km is due to local AN emissions (Figs. 3 panel 3d; **??** panels 1d and 2d). In contrast with the other months, the CO mixing ratio above the surface maximum decreases sharply with altitude showing low CO concentrations from panels 1 and 2 km to 12 km. Indeed, in October, the monsoon flow has disappeared and West Africa is impacted by the north easterly trade winds which block the transport of air masses impacted by BB from Southern Hemisphere Africa as is clearly visible on the low tropospheric CO distribution from IASI and Fig. S3) are observed below 4 km over the Western African clusters (Lagos, Sahel and Gulf of Guinea). These high CO mixing ratios in April are detected by IASI in the LT_{IASI}

(Fig. 2d) . This is confirmed by the predominant local (Northern Hemisphere Africa) origin of CO over Lagos (Fig. 3 panel 3d).

Interestingly, the b) over the whole of Western Africa. The annual CO surface maximum in Central Africa Occurs in April, before the beginning of the Southern Hemisphere fires, due occurs also in April. LT CO is attributed by more than 85% to local AN emissions (Fig. 3 panel 4b and Fig. 4a) when few fires are detected by MODIS (Yamasoe et al. (2015); their Fig.7). The measured CO maxima reaches 350 ppb anomaly reaches +200 ppb (after removing the background from the observed CO), while SOFT-IO attributes 40 ppb to the aforementioned sources. This means that Southern Hemisphere 80 ppb to AN. This indicates that African AN emissions are likely underestimated. Above 1km, in the absence of fire contributions, CO remains constant with 100 ppb which is the annual minimum, and the Omight be underestimated. The enhanced LT O_3 profile is characterised by a steep gradient and the lowest annual concentrations. IASI LT_{iasi} distribution indicates possible O_3 formation during the transport of the aforementioned emissions towards Sahel and the Gulf of Guinea, but could also be explained by enhanced NO_x mixing ratios, possibly due to soil emissions when rains start (Saunois et al., 2009), especially over Sahel (Fig. 2b) indicates that the CO minimum measured by IAGOS above 1km over Central Africa in April extends over the whole Central and Southern Africa3 panels 1a–c) because of higher NO_2 mixing ratios above dry savannas than over wet savannas and forests (Adon et al., 2010).

The classical increase of O

3.1.2 Asia

Similarly to Africa, Asian clusters present CO maximum in the LT, but below 2km, and with lower O_3 from the surface to the MT in October and April is due to photochemistry which changes from a net sink to a net source of ozone above 6km, depending on the NO_x concentration (Jacob et al., 1996). In the tropics, photochemical enhancement. Above 2km, CO is characteristic of the background, while O_3 destruction dominates the low troposphere (Archibald et al., 2020), where water vapour concentrations are high, presents a strong seasonal variability (Fig. 5 panels 1 and in highly polluted regions where there is direct removal by titration with NO (Monks et al., 2015). The vegetation can also act as a rapid sink for O_3 via dry deposition (Cros et al., 2000). The lack of these sinks in the MT and UT, coupled with lower water vapour concentrations leads to an increase of O_3 with altitude (Archibald et al., 2020). Lightning can also increase O_3 mixing ratios in the MT and UT (Barret et al., 2010)2 and Fig. S3).

Overall, AN emissions largely dominate CO contributions over fire ones, throughout the troposphere (Fig. 5 panels 3–5 and Fig. S6 panels 3–7). The dominant AN region is South-East Asia, followed by Central-East Asia which has a great influence over the South China cluster. Biomass burning from South-East and Equatorial Asia bring a substantial contribution in April and October respectively, but less important than the AN one.

3.1.3 Asia

Over the Asian clustersIn January, the annual CO maximum observed close to the surface occurs in January for all the clusters (except over Manila and the Gulf of Thailand)(, Fig. 5 panel 2), due to the lowest boundary layer height in winter, and is

mainly attributed to and Fig. S3 panel 2) are related to local AN emissions (Figs. 5 and ?? panels 3a, 4a, 5a) . Over the Indian and South China clusters, local AN emissions (SouthEast and Central Asian resp.) are dominant, with contributions from 85 to 95 % (Figs. 5 panels 3a and 4a; ?? panel 3a and 4a, and 4a). AN Central Fig. 5 panels 3a–5a and Fig. S6 panels 3a–7a) due to the low boundary layer height in winter. CO maxima are coincident with O_3 enhancements also observed by IASI (Fig. 2f), and due to the chemical ageing favoured by the confinement of the CO-rich air masses due to the large-scale subsidence (Lelieveld et al., 2001) and by the cloud free conditions. Central-East Asian emissions mainly control the LT CO anomalies over the Asian clusters (except Bangkok) in January for South China, Gulf of Thailand and Manila, from 52 % to 95 % (over Gulf of Thailand) to 75 % (over Manila) (Figs.4a, ??a and ??a)and South China resp., Fig. 4a; Fig. S5a and Fig. S7a), due to their advection by the northeasterly trade winds. The elevated CO mixing ratios below 2.5 km over the Asian clusters in January are coincident with O_3 enhancements (Figs. 5 panel 1)due to the chemical ageing favoured by: i) the confinement of the South-East Asian emissions dominate over the Indian clusters with an 85% contribution (Fig. S7a). As in Africa, the CO-rich air masses due to the large-scale subsidence (Lelieveld et al., 2001) and ii) the cloud free conditions. This accumulated in the LT are accompanied by a secondary LT O_3 maximum, significantly lower (40–60 ppb) than the Western African ones (65–75 ppb). It might be because the O_3 enhancement described in Barret et al. (2011) for South Asia during the post monsoon is also detected by IASI (Fig. 2f)over Asia is caused by AN-polluted air, while in Africa by mixed (AN and BB) polluted air. During the Atom campaign, Bourgeois et al. (2021) found that O_3 levels are more enhanced in mixed air pollution, because they are associated with greater NO_x and peroxy acyl nitrates (a NO_x reservoir compound), and thus increased O_3 production, in comparison to BB- or AN-polluted air alone.

During the pre monsoon season(April)In April during the pre-monsoon season, CO and O_3 are both enhanced above the boundary layer and below 4 to 6 km over most sites locations (Fig. 5 panels 1 and 2 and Fig. S3). Local AN emissions control the South-East Asian AN emissions dominate CO anomalies over the majority of the sites (Figslocations (Fig. 5 panels 3b–5b; ?? panels 3b–7b), while spring SouthEast Asian firesimpact 3–5 and Fig. S6). South-East Asian fires, especially located in the region of Myanmar, Northern Thailand and Laos (Fig. S4b), also contribute over South China, Bangkok, Ho Chi Minh City and Manila by at least 20–30 % in the LT and MT (Figs. 4 a and b; ??a and b). In spring the fires are mostly above East Asia and especially the region of Myanmar, Northern Thailand and Laos (Fig. S1b)and the corresponding large CO concentrations are Fig. 4 and Fig. S7), leading to large CO mixing ratios captured by IASI (Fig. 2b). The BB outflow towards South China and the Pacific ocean Ocean is due to westward low and mid tropospheric winds (Figs. 2b and S3b and S3fFig. S8b and f). The enhanced MT O_3 is attributable to the intense solar radiation associated with the important amounts of precursors from AN and BB emissions which were previously evidenced. This is in agreement with the observed O_3 maximum in spring over South China (Dufour et al., 2010) and Bangkok (Sahu et al., 2013). Yarragunta et al. (2019) found that local AN emissions are responsible for the CO and O_3 abundances over South India during the pre-monsoon season. This is in accordance with the SOFT-IO contributions over the Indian clusters (Figs. 5 panels 3b; ?? panels 3b and 4b; ??a). CO anomalies over Mumbai are also caused by transport Mumbai is the only location showing a more distant influence of AN emissions, from the Middle East (36%) in the LT, and Northern Hemisphere Africa (30%) in the MT (Fig. ??a and b). In the UT, the impact of Northern Hemisphere African and Northern Hemisphere Africa (AN and BB) emissions dominates over Mumbai in the UT (54 %) and

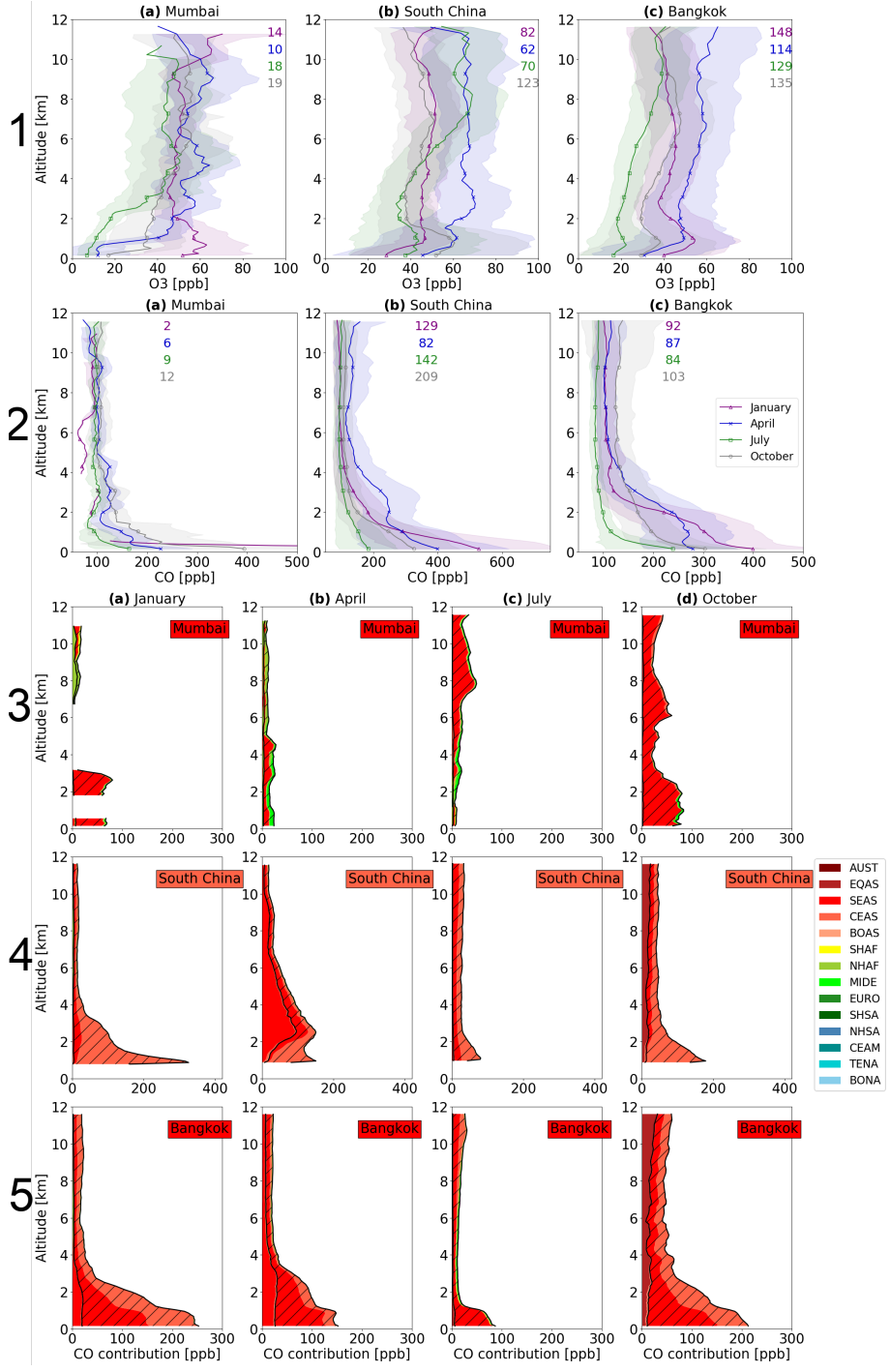

Figure 5. Same as Fig. 3 for the Asian clusters.

~~Hyderabad (50 %). It has to be noted that the number of profiles over Mumbai (6)and Hyderabad (19) are lower than the threshold established for representativeness (see Sect. 2.1.1). The UT_{iasi} CO and O_3 transport from Northern Hemisphere~~ (Fig. S7). This transport from Africa towards the Arabian ~~sea~~ Sea and South India is ~~also~~ captured by IASI (~~Figs~~Fig. 2j and ~~2~~n), ~~indicating~~ suggesting O_3 photochemical production during the transport.

~~The BB contribution is also important during the post monsoon season~~In October, during the post-monsoon season, either BB or AN can dominate depending on the cluster and the altitude. South-East Asian AN emissions dominate the entire troposphere of the Indian clusters (Fig. S7), while Central-East Asia AN emissions dominate over South China (~~October)because of active fires over Indonesia (~~Fig. ~~S1). The Gulf of Thailand cluster is the most affected, from the surface (600 ppb) to the UT (Fig.~~ 5 panel ~~1e). IASI CO data~~ 4d and Fig. 4). A mix of South-East and Central Asian AN emissions have the largest influence over Bangkok, Ho Chi Minh City (LT and MT) and Manila (LT) (Fig. 4 and Fig. S7). Equatorial Asian BB emissions, principally from Indonesia (Fig. S4d), dominate CO anomalies over the entire troposphere of the Gulf of Thailand (Fig. ~~2d)and wind fields (Fig. S3d)show that the LT_{iasi} CO-rich air masses impacted by the fires~~ S5), and also over Madras and Ho Chi Minh City (UT) and Manila (MT and UT) (Fig. S7). The same BB has a substantial influence (10 % on average) over the entire troposphere of South China and Bangkok (Fig. 4), and in the LT of Hyderabad and Manila (Fig. ~~S1d)are advected~~ S7). BB CO-rich air masses advection towards the South East Asian coastal clusters ~~(South China, Gulf of Thailand, Bangkok and Manila), as confirmed by the SOFT-IO (contributions of 10 % on average) (Figs. 5 panels 4d and 5d; **??** panels 6d and 7d; 4a; **??**a and **??**a~~is captured by IASI in the LT (Fig. 2d) and wind fields (Fig. S8d). The collocated O_3 enhancement (below 2 km) over these clusters (~~Figs. 5 pane 1c–1e~~Fig. 5 panel 1) indicates O_3 production ~~by BB and AN precursors~~during transport. The Equatorial Asian BB contribution generally intensifies in the UT ~~, over the SE Asian coastal clusters~~ (40–57 %) ~~, Madras (50 %) and Hyderabad (33 %) (Figs. 4c; **??**c and **??**c). The upper tropospheric CO maximum above the fires~~ (Fig. 4; Fig. S5 and Fig. S7), and is also captured by IASI and IAGOS (~~Figs. 2l; 2p and S1d). Based on MLS CO data,~~ Fig. 2). Livesey et al. (2013) also found an upper tropospheric CO maximum over Indonesia and attributed it to episodically strong convection, in agreement with the low OLR in Fig. 2p. In contrast, the UT_{iasi} ~~and MT_{iasi}~~ $IASI$ and MT_{IASI} O_3 ~~distribution~~ distributions show a SE-NW gradient (~~Figs. 2l and 2h~~Fig. 2) with lower O_3-levels over the Maritime continent and the southern Indian Ocean and higher ones over India and the Arabian ~~sea. This was reported by Barret et al. (2011)~~ Sea, as a result of convection over the first region and subsidence of precursor ~~enriched air masses~~ over the second one (Barret et al., 2011).

~~The~~ In July, during the monsoon season, LT O_3 and CO mixing ratios ~~over~~ are minima for all the Asian clusters ~~minimise during the summer monsoon (July) (~~(Fig. 5 panels ~~1–2~~1 and 2 and Fig. S3). The reversal of the north-easterly trades to the monsoon flow (Fig. ~~S3c~~S8c) results in the advection of O_3- and CO-poor air masses from the Indian ~~ocean~~ Ocean towards Asia. Furthermore, cloudy conditions reducing solar radiation and promoting the scavenging of O_3 precursors result in lower O_3 production than in clear sky conditions (Mari et al., 2000; Safieddine et al., 2016). The steep CO gradient close to the surface (below 1 km) indicates the convective uplift of polluted boundary layer air masses towards the UT. The resulting enhancement of CO in the UT within the Asian monsoon anticyclone (AMA) analysed in Park et al. (2008) and Barret et al. (2016) is clear from IASI (Fig. 2~~k~~). In contrast, the positive South-North O_3 gradient between the Maritime continent and north South Asia and the Middle East (~~Figs. 2o and w) is associated with: i)~~ Fig. 2) is likely associated with the photochemical ageing of air

masses while they are recirculating towards Middle East (Lawrence and Lelieveld, 2010)~~and ii)~~ , and with the high insolation over the Middle East (Barret et al., 2016).

3.1.3 South America

~~One common characteristic among the~~ South American clusters ~~is the increase in~~ present CO maximum in the LT, similarly to African and Asian ones (Fig. 6 panel 2). O_3 is increasing from the surface to the MT/UT (Fig. 6 panel 1)~~. This is~~ , probably due to the lack of depositional and chemical sinks above the ~~low troposphere~~LT, in combination with lower water vapour ~~concentrations~~mixing ratios, and lightning emissions, as discussed in Sec. 3.1.1. ~~In Caracas, O_3 shows one maximum in the LT and one in the MT in April. Over Bogotaand SBrazil O_3 peaks in October over the whole troposphere with mixing ratios reaching 45~~ Local AN emissions generally dominate the CO anomalies, either from North Hemisphere South America (Caracas and Bogota) or from South Hemisphere South America emissions (SBrazil) (Fig. 6 panels 3–5). Local (South America) and ~~60 ppb in the UT. For Bogota (resp. SBrazil) the tropospheric O_3 annual minimum occurs in July (resp. April). As IAGOS over SBrazil below 6 km, the IASI distributions over tropical South America(Fig. 2 e–h) display MT_{iasi} O_3 in January and April than in July and October~~intercontinental (Africa) emissions due to fires make a significant contribution, which can reach or exceed the one of AN emissions.

~~Another common characteristic among the three clusters is the LT CO maximum in April~~ In April CO is maximum in the LT (Fig. 6 panel 2)~~. The CO mixing ratios peak over Bogota below 1km (~~, reaching 350 ppb below 1.5 km over SBrazil, 400 ppb ~~) and over Caracas up to 2km (~~below 1km over Bogota and 200 ppb ~~). According to SOFT-IO, the CO maximum over Caracas is due to local~~ over Caracas below 2 km. For the last two locations CO anomalies are related to a mix of AN (35 %) and BB (32 %) emissions ~~(~~from Northern Hemisphere South America ~~) (Figs. ??, and 6 panel 5b). Similar contributions are found over Bogota ((~~Fig. 4 and Fig. 6 ~~panel 4). This local origin of emissions is~~ panels 3b and 4b), corroborated by the elevated IASI LT_{iasi} ~~CO mixing ratios~~ $_{IASI}$ CO (Fig. 2b) collocated with the strong AN emissions above Colombia and Venezuela, and active fires above the latter (~~Figs. S1b and S2b). Transport also plays an important role with~~ Fig. S4b and Fig. S9b). 20 % of the anomalies ~~caused by~~ are caused by transport of BB Northern Hemisphere African emissions (Fig. 4a). The O_3 maximum collocated with the CO one at 2 km over Caracas (Fig. 6 ~~panels 1a and 2a~~panel 1), indicates O_3 production during transport of Northern Hemisphere African air masses impacted by BB~~. The second O_3 maximum~~ , while the second one above 5 km ~~over Caracas is also noticed by Yamasoe et al. (2015) that attributed it~~ is attributed by Yamasoe et al. (2015) to local AN sources followed by lightning.

~~Over SBrazil, the~~ Contrary to Caracas and Bogota, SBrazil annual CO maximum ~~below 1.5 km in April (Fig. 6 panel 2c) is~~ is mostly due to local AN emissions ~~(Fig. 6 panel 5b)~~ located over the southern part of Brazil (Fig. ~~S2b). The observed CO enhancement reaches 350 ppb, while SOFT-IO attributes~~ S9b). Only 65 ppb ~~to the aforementioned emissions. This indicates~~ of the 260 ppb observed CO anomalies are simulated (Fig. 6 panel 5b), suggesting that Southern Hemisphere South American AN emissions are underestimated~~by SOFT-IO. The observed~~ . The CO enhancement at 1.5 km ~~is new compared to Yamasoe et al. (2015) and~~ (Fig. 6 panel 2c) not reported by Yamasoe et al. (2015) is related to increased local AN contributions for March–April–May 2014 relative to the previous years. This is in agreement with the ~~CEDS~~ CEDS2 inventory, which

shows a peak in AN emissions over South Brazil ~~(18–29° S and 35–52° W) in 2014, mostly coming from the transportation sector (Fig. not shown).~~ in 2014.

~~Same as Fig. 3 for the South American clusters.~~

~~The CO minima~~ In January CO is minimum over the three clusters ~~are observed in January, when CO concentrations are~~ with CO below 300 ppb (resp. 180 ppb) over Bogota (resp. Caracas and SBrazil) below 1km (Fig. 6 panel 2). ~~The CO mixing ratios decrease below 100 ppb above the polluted layers all year long, with exception of SBrazil, where a first maximum (150–200 ppb) occurs~~

In October, two CO maxima are observed over SBrazil, between 2–4 km ~~and a second one (200–250 ppb)~~(150–200 ppb), and above 8 km ~~in October. IASI clearly~~ (200–250 ppb) (Fig. 6 panel 2c). IASI detects the $MT_{\sout{iasi}}$ ~~and $UT_{\sout{iasi}}$ (Fig. 2d and p resp.)~~ $_{IASI}$ and UT_{IASI} maxima over most of tropical ~~south America in October.~~

~~From SOFT-IO we can see that, over SBrazil in October,~~ South America (Fig. 2). At the same time O_3 peaks above 2km over Bogota and SBrazil, reaching 45 and 60 ppb in the UT (Fig. 6 panel 2). While CO enhancement below 1 km of SBrazil is caused by local AN (52 %) and BB (44 %) emissions (Fig. 6~~panel 5d). In the MT and UT, the~~), BB contribution exceeds 80 % in the MT and UT because of the strong convection moving over the BB regions (Liu et al., 2010), documented by low OLR (Fig. 2l). The uplifted BB products are trapped in an anticyclonic circulation developed over Central South America (Fig. ~~S3l~~S8l). CO from Southern Hemisphere South American fires are transported over Bogota, at the edge of the anticyclone, but does not reach Caracas which lies outside of the anticyclone (Fig. 6 panels 3d and 4d). Their photochemical processing contributes to the seasonal O_3 enhancement over South America ~~which is the western part of the wave-one pattern (Thompson et al., 2003b; Sauvage et al., 2006). This is~~ (Thompson et al., 2003b; Sauvage et al., 2006), highlighted by the collocation of IASI $UT_{\sout{iasi}}$ $_{IASI}$ CO (Fig. 2l) and O_3 (Fig. 2p) maxima~~within the anticyclone~~.

3.1.4 Arabia and Eastern Africa (AEA)

~~The striking feature of the AEA clusters is the elevated O_3 centered at around 8 km (70 ppb on average) for all the clusters during April and for the northern clusters of Jeddah and Abu Dhabi during July (Fig. 7 panel 1 a–d). The particularly low CO mixing ratio accompanying the O_3 enhancements~~ AEA clusters present CO maximum in the LT (Fig. 7 panel ~~2a–d) points to a dynamical origin of O_3. Note that only limited number of profiles are available over Abu Dhabi above 10 km and Khartoum in April. The O_3 enhancements over the 4 sites of AEA and the anticorellation with CO, are also detected by IASI in the $MT_{\sout{iasi}}$ and $UT_{\sout{iasi}}$ (Fig. 2j, k, n and o)~~2 and Fig.

~~Same as Fig. 3 for the Arabian and Eastern African clusters.~~

~~Tropopause foldings in the vicinity of the subtropical jet stream are associated with downward transport of stratospheric ozone (Stohl et al., 2003; Lelieveld et al., 2009; Safieddine et al., 2014) resulting in a tropospheric O_3 enhancement during spring and summer (Tang et al., 2011). This is in agreement with Cohen et al. (2018) that found the maximum O_3 to CO ratio over the Arabian peninsula for the same seasons (their Fig. A1), using IAGOS data for the period 1994 to 2013. Also, large O_3 regional enhancements are detected by IASI over the Arabian sea similarly to Jia et al. (2017) based on TOC from OMI/MLS. Jia et al. (2017) attributed these O_3 enhancements to emissions from India (50 %), with smaller contributions from the Middle~~

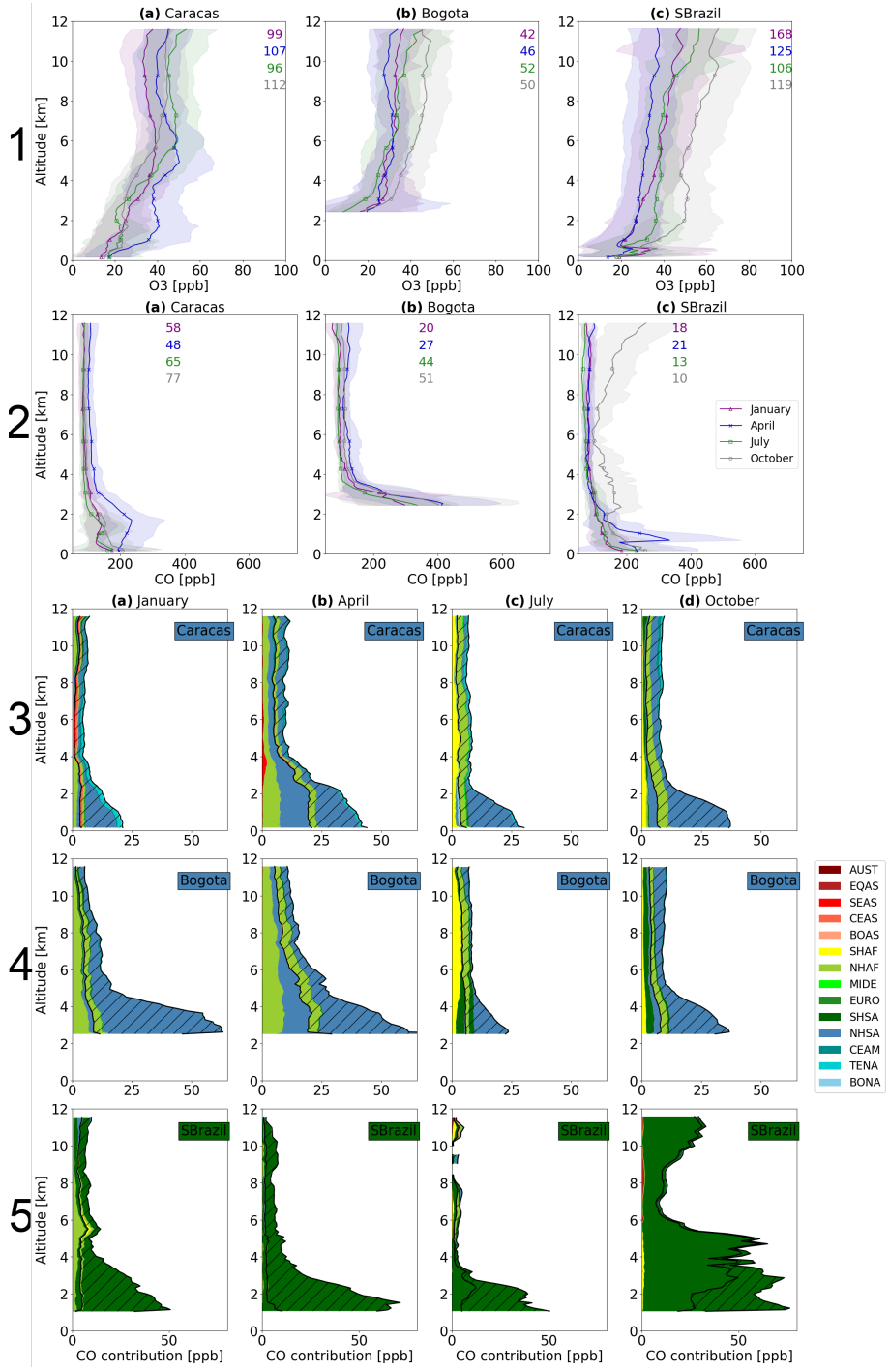

Figure 6. Same as Fig. 3 for the South American clusters.

East and AfricaS3 panel 2e), similar to other tropical locations mostly due to AN local (Middle East or Africa) emissions, except Addis Ababa and Khartoum in July, showing substantial African BB influence (30 %) . This is in agreement with SOFT-IO, which shows a significant contribution from SouthEast Asia over Jeddah (29 %) and a lower one over Addis Ababa (7%) (Figs. 7 panels 4b and 5b, and ??a). SOFT-IO also attributes large contributions from Northern Hemisphere African AN and BB emissions over Abu Dhabi and to a lesser extent over Khartoum (Fig. 7 panels 3b and 6b). The contribution of American sources over Abu Dhabi indicates eastward transport, which is not present in the rest of the AEA clusters because Abu Dhabi is affected by the subtropical westerly jet in the UT. In contrast the rest of the AEA clusters are affected by the tropical easterly jet which brings CO from Asian regions. 3–5 and Fig. S6 panel 8).

In July, the Middle East summer O_3 maximum is also partly related to subsidence of AMA air masses which brings O_3 produced from South Asian AN and $LiNO_x$ emissions (Barret et al., 2016). The polluted air masses from South and SouthEast Asia uplifted by monsoon deep convection are trapped in the AMA which extends westward to Northeast Africa and the Middle East (Barret et al., 2016; Park et al., 2007). Over Khartoum and Jeddah (resp. Addis Ababa and AbuDhabi) 20 ppb (resp. 10 ppb) of CO originates from SouthEast Asia at 6–12 km. The impact of the SouthEast Asian emissions is stronger over Jeddah (78 %) than over Khartoum (60 %) and Addis Ababa (46 %) (Fig. ??c) which are outside of the AMA (Fig. 7 panel 1). Furthermore, the O_3 minimum over Addis Ababa (45–50 ppb) is related to the ITCZ located between 5° N and 10° N during the Northern Hemisphere wet season (Lannuque et al., 2021). The UT_{iasi} O_3 enhancement over Arabia and the Arabian sea, and the transition to lower concentrations south of the tip of Arabia are also clear with IASI (Fig. 2c). The O_3 minimum over Africa is caused by uplift of local African O_3-poor air masses from the surface to the ITCZ (Lannuque et al., 2021). The increase of O_3 northwards (such as over Khartoum with 60 ppb; Fig. 7 panel 1d) is due to the O_3 production within uplifted CO-rich air masses, transported away from the ITCZ by the upper branches of the Hadley cell (Lannuque et al., 2021).

One common characteristic among the AEA clusters is the elevated CO mixing ratio in the surface layer (below 1km) all year long (Fig. 7 panel 2). The surface maximum is larger over Addis Ababa (700 ppb in July) and Khartoum (350 ppb in April), than in Jeddah and AbuDhabi (<250 ppb). Over the East African sites (Khartoum and Addis Ababa), a layer of enhanced CO is observed below 4 km, in January and April. This winter to spring high CO layer in the LT_{iasi} over Eastern Africa is detected by IASI which clearly shows that it does not reach Arabia (Fig. 2a–b).

In January, the surface CO maximum is mostly controlled by local AN emissions over the AEA clusters (Figs. 7 panels 3a to 6a). Strong AN emissions strong AN emissions from Northern Hemisphere Africa control the LT CO anomalies over Addis Ababa with contribution of 71 % in the LT and 58 % in the MT (Fig. ??aS5). Influence from the Northern Hemisphere African fires is also evident (12 % in the LT and 20 % in the MT)(Fig. ??a). . The impact from these fires intensifies over Khartoum and Jeddah between 2 and 4 km with contributions of 58 % and 53 % respectively around 55 % (Fig. ??a). The effect of the Northern Hemisphere African emissions towards Eastern Africa and Jeddah is also detected by IASI (Fig. 2a), which shows a negative eastward CO gradient. The co-occurring O_3 enhancement (Fig. 2e) over Khartoum and Jeddah below 4 km reflects O_3 formation during transport from the fires (Fig. 7 panels 1b and 1d7 panels 4a and 5a).

In July, the CO surface LT maximum is again caused by local AN emissions(Fig. 7 panels 3c–5c), except over Khartoum where air masses from Southern Hemisphere African fires are the dominant source of CO (Fig. 7 panel 6c5c). The combination

of local AN (70 %) and Southern Hemisphere African BB (23 %) emissions is responsible for the annual CO maximum at the surface over Addis Ababa (~~Figs. 7 panel 5c, and ??a). Interestingly, the impact of the Southern Hemisphere African fires below 4km over Khartoum and Addis Ababa is stronger than the impact of local fires during the respective dry season (Figs. 7 panels 5ac and 6ac). The~~ Fig. S6 panel 8c and Fig. S5).

The striking feature is the elevated O_3 around 8 km (70 ppb on average) for all the clusters during April, and for Jeddah and Abu Dhabi during July (Fig. 7 panel 1 and Fig. S3 panel 1e). Low CO accompanying the O_3 ~~enhancement below 4 km over the Jeddah, Khartoum and Addis Ababa indicates~~ enhancements (Fig. 7 panel 2 and Fig. S3 panel 2e) points to a dynamical origin of O_3~~production during the transport of CO-rich air masses impacted by the Southern Hemisphere African fires~~ . In July these features are also detected by IASI in the MT_{IASI} and UT_{IASI} (Fig. ~~7 panels 1 jto l) . In contrast, over Abu Dhabi the O_3 enhancements at around 7 and 10 km~~ 2j, k, n and o) and can be due to either i) tropopause foldings in the vicinity of the subtropical jet stream associated with downward transport of stratospheric ozone (Stohl et al., 2003; Lelieveld et al., 2009; Safieddine et al. or ii) emissions from South-East Asia (50 %), with smaller contributions from the Middle East and Africa (30 %) (Jia et al., 2017) trapped in the AMA (Barret et al., 2016; Park et al., 2007). This last influence is in agreement with SOFT-IO, showing a significant contribution from South-East Asia over Jeddah (29 % to 78 %), Khartoum (60 %) and Addis Ababa (46 %) (Fig. S5), affected by the tropical easterly jet transport. SOFT-IO also attributes large contributions from Northern Hemisphere African AN and BB emissions, and also from American sources over Abu Dhabi (Fig. 7 panel ~~1a)are likely related to transport of CO-rich air masses from Boreal Asia (Fig~~3a), related to eastward transport by the subtropical westerly jet in the UT, not present in the rest of the AEA clusters.

In July O_3 is minimum over Addis Ababa (45–50 ppb) (Fig. S3 panel 1e), related to the Inter Tropical Convergence Zone located between 5° N and 10° N (Lannuque et al., 2021). ~~7 panel 3e).~~

In October, in the LT the ~~long range~~ long-range transport from Asia (~~SouthEast~~ South-East AN and Equatorial BB) plays a significant role in CO anomalies over the AEA ~~sites (Figs~~locations (Fig. 7 panels ~~3d–6d~~3d–5d and Fig. S6 panel 8d), especially over Addis Ababa and Jeddah. In the $LT_{\sout{iasi}\ IASI}$, IASI captures the northeasterlies (Fig. ~~S3d) transport~~ S8d) transport of CO-rich air masses from Asia towards eastern Africa ~~as detected by IASI~~ (Fig. 2d).

~~Above 4km in October~~Above 4 km, O_3 enhancements are observed over the AEA ~~sites~~ locations, especially over Abu Dhabi which is the easternmost ~~site~~ location of the AEA region (Fig. 7 panel 1). IASI detects a $MT_{\sout{iasi}\ IASI}$ O_3 increase above the Arabian ~~sea~~ Sea and Northern India (Fig. 2h)~~. The O_3 enhancement in the MT over the Arabian sea~~ , previously detected with ozone soundings during the INDOEX campaign (1999–2000) ~~has been~~ and attributed to Indian sources uplifted over the marine boundary layer by the sea breeze circulation ~~in Lawrence and Lelieveld (2010) and was further analysed and documented with IASI data by Barret et al. (2011)~~(Lawrence and Lelieveld, 2010; Barret et al., 2011). The O_3-rich air masses are further transported towards Eastern Africa by the prevailing northeasterlies (Fig. ~~S3h~~S8h) as documented by the predominant ~~SouthEast~~ South-East Asian origin of MT and UT CO over the AEA ~~sites (Figs. 7panels 3d and 6d~~locations (Fig. 7).

3.2 Control factors of tropical O_3 and CO

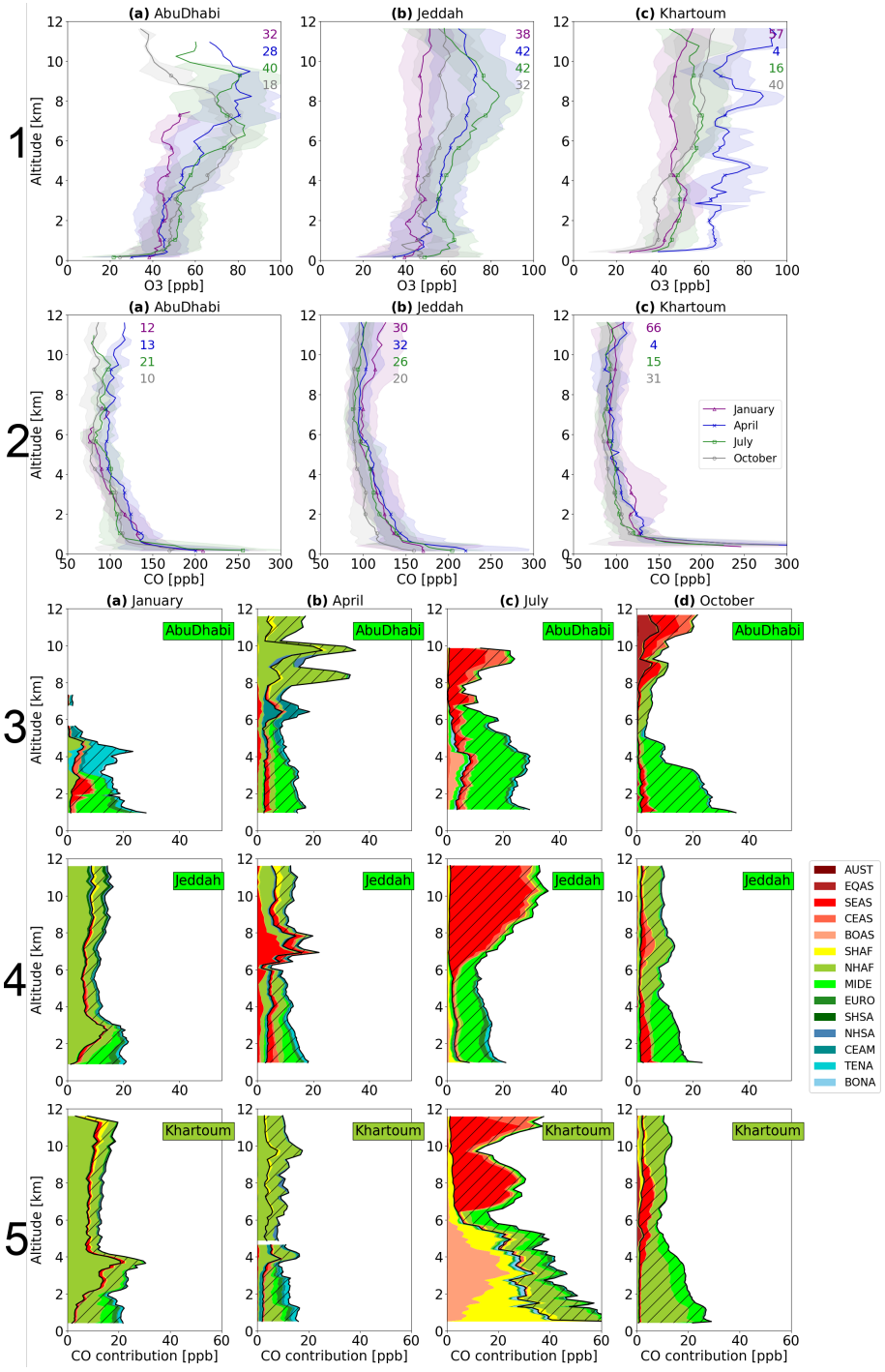

Figure 7. Same as Fig. 3 for the Arabian and Eastern African clusters.

First, we give an overview of the Tropics, focusing on the extrema of O_3 and CO of all clusters, then on AN and BB contributions in the upper troposphere. Finally, we look at tropical connections between emission source regions and CO and O_3 distributions. Figure 8 displays the annual ~~maximum~~maxima/~~minimum~~ minima of O_3 (a) and CO (b) mixing ratios and their corresponding mean height. The annual maxima/minima are calculated based on monthly averaged mixing ratios over vertical layers with 40 hPa thickness. ~~Figure 10 displays the transport pathways of CO emissions from the African, South American and Asian source regions, towards the 20 tropical sites in the LT (a), MT (b) and UT (c). We show the source regions and the months corresponding to the largest amounts of transported CO (in ppb). Figure 9 displays the AN and BB contribution to CO anomalies (in ppb) over the tropical UT_{cruise}.~~

Overall, the CO profiles ~~above all tropical clusters~~ display an annual maximum ~~above the surfacelayer (~~close to the surface, approximately at 0.5 km ~~)~~ (Fig. 8b) ~~. This is also valid for Caracas, Bogota, Windhoek and Addis Ababa which are located at high altitude above the sea surface (with a mean elevation of 0.9 km, 2.6 km, 1.6 km and 2.3 km respectively). For all the clusters located in the NT (except the Gulf of Guinea and Caracas), the CO-polluted boundary layer, is~~ with strong magnitude variations. The minima are generally located in the free troposphere (MT or UT), in the absence of recent pollution and where CO is chemically destroyed, and are uniform in terms of mixing ratios, close to the background levels. Both maxima and minima present strong seasonality related to surface emissions and meteorological conditions.

For the majority of the Northern Tropical locations the CO maxima are mainly attributed to local AN emissions~~, even for Lagos and Sahel, where BB is expected to be of great importance (Reeves et al., 2010; Mari et al., 2008). This finding~~ . However, for African locations and Caracas, BB contribution is almost similar to or greater than AN (Fig. 3 panel 3; Fig. S6 panels 1 and 2 and Fig. 6 panel 3). This confirms the key role of AN emissions in the Northern Hemisphere, related to a larger population compared to the Southern Hemisphere, and enhanced AN urban and industrial activity. ~~Concerning the ST, the~~ The surface-layer pollution of the Southern Tropics is predominantly caused by BB over ~~ST~~ Africa (Central Africa and Windhoek) during the dry season~~(Figs. 4, **??**), ~~, and by AN over ~~the~~ SBrazil (Fig. **??**3 panels 4 and 5 and Fig. 6 panel 5). The CO maxima over the latter ~~occurs~~ occur before the burning season~~. This is ~~, in accordance with previous studies suggesting fossil fuels as the main CO source over Sao Paulo and Rio de Janeiro (Alonso et al., 2010), and decreasing BB over South America (Andela et al., 2017; Deeter et al., 2018) by more than 40 % due to the long-term declining deforestation rates, especially over forested areas ~~(≈ 54 %)~~ and over savanna and shrublands ~~(≈ 39 %) (Naus et al., 2022). The importance of the AN emissions is also evident over Central Africa, where a polluted surface layer is present all year long and during the transition seasons, when the fires are suppressed, it is largely caused by AN contributions (40 % and 86 % in April and October; Fig. 4a). Thus, the impact of the AN emissions is also important in the ST.~~ (Naus et al., 2022).

~~Total (AN + BB) CO emission rates (in 10^{-10} kg m^{-2} s^{-1}) based on CEDS and GFAS emission inventories over West Africa (10° W–12.5° E; 0–12.5° N), Central Africa (10–35° W; 2.5–20° S), East Asia (92.5–110° E; 10–27° N), Maritime Continent (93–121° E; 10° S–10° N), South Brazil (35–50° W; 0–20° S) and Arabia and Eastern Africa (30–60° E; 5–25° N). West Africa Central Africa East Asia India Maritime Continent South Brazil AEA January 6 1 4 3 1.5 1 1.5 April 3 1 11 3.5 1.5 0.5 1 July 2.5 10 3 3 2 1 0.5 October 2.5 3 3 3 6 4 1 Annual 3.5 3.7 5.5 3.1 3 1.5 1~~

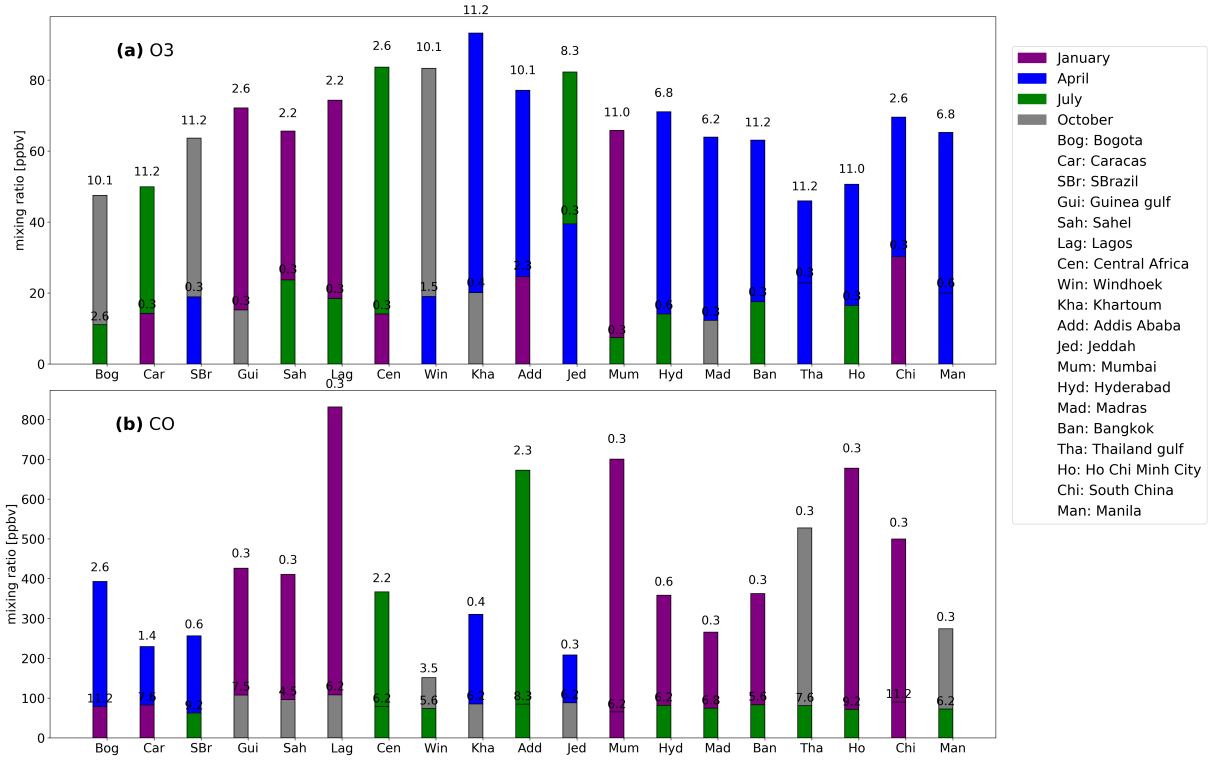

Figure 8. O_3 (a) and CO (b) annual maximum (higher bar) and minimum (lower bar) mixing ratio observed over the tropical clusters. The annotated number on top of each bar indicates the altitude (in km) of the observed annual maximum/minimum mixing ratio. The colour in the bar indicates the month of the maximum/minimum.

~~The CO maxima show strong variations in terms of magnitude and season among the tropical clusters because they are mostly caused by local emissions with varying intensity and seasonal pattern, depending on the region. In contrast, the CO minima are uniform in terms of intensity levels of mixing ratios, close to the CO background levels, due to mixing and transport over the lifetime of CO. As expected, they occur in the MT or UT, in the absence of the emissions and where CO is chemically destroyed. As for the CO maxima, their strong seasonality is related to the seasonality of the surface emissions and the meteorological conditions, which differ over each region. Further discussions on the magnitude and the seasonality of the CO maxima and minima will follow later.~~

Because of its complex chemistry, the situation for O_3 is more complicated. Africa is the only region where the annual O_3 maximum occurs in the LT (at 2.5 km) during the dry season (Fig. 8a Sahel, Guinea Gulf, Lagos and Central Africa). The co-occurrence of maximum O_3 with ~~the maximum in~~ enhanced CO over Africa ~~during the local fires indicates~~ indicates the stronger dependency of O_3 on the surface AN and BB CO emissions for these regions, in agreement with Sauvage et al. (2007b). South China is the only Asian cluster where the annual O_3 maximum is observed in the LT (at 2.6 km) during the active local fires (April). In contrast, over the other regions, the annual O_3 maximum is observed above 6 km (Fig. 8a). This

Table 2. Total (AN + BB) CO emission rates (in 10^{-10} kg m^{-2} s^{-1}) based on CEDS and GFAS emission inventories over West Africa (10° W–12.5° E; 0–12.5° N), Central Africa (10–35° W; 2.5–20° S), East Asia (92.5–110° E; 10–27° N), Maritime Continent (93–121° E; 10° S–10° N), South Brazil (35–50° W; 0–20° S) and Arabia and Eastern Africa (30–60° E; 5–25° N).

	West Africa	Centr
January	6	
April	3	
July	2.5	10 ppb) among the NT African clusters. In contrast to CO, the O_3 enhancement does not strongly depend on the proximity t
October	2.5	
Annual	3.5	

indicates that O_3 is likely associated with larger ozone O_3 production efficiency in the MT and UT (Sauvage et al., 2007c). In regions such as Arabia (Jeddah and Abu Dhabi), the lack of CO enhancement in the UT indicates dynamical origin of O_3 (e.g. stratospheric influence and transport of O_3 and precursors from Asia; see Sec. 3.1.4). In contrast, in regions such SBrazil and Windhoek in October, the co-occurrence of O_3 and CO enhancement in the MT and UT indicates O_3 production from surface sources (e.g. fires). In addition, LiNOx emissions can contribute to O_3 production at higher altitudes with LiNOx emissions (Secs. 3.1.1 and 3.1.3).

The annual O_3 minima for all the tropical clusters are observed close to the surface (below 0.5 km on average)(Fig. 8a). This is , likely related to deposition and titration by NO(see Sect. 3.1.1 for more details, or because of lower O_3 production in the presence of convective clouds relative to clear sky conditions (Sect. 3.1.2).

The Throughout the Tropics, the highest CO and O_3 maxima among all the tropical clusters occur over NT occur over Western Africa in the LT (at 0.3 km for CO and 2.5 km for O_3Fig. 8) during the dry season (January)mostly due to local AN emissions (over Lagos and Sahel) and BB (over Guinea Gulf). Table 2 displays the total (AN and BB) CO emissions rates over several regions of interests interest based on the sum of CEDS CEDS2 and GFAS emission inventories. The NO_x-limited O_3 production regime over Western Africa (Saunois et al., 2009; Zhang et al., 2016) likely explains the O_3 maxima when the local emissions, and thus the NO_2 concentration (Jaeglé et al., 2004), intensify mixing ratios increase in the region (Jaeglé et al., 2004). The largest O_3 and CO mixing ratio over Lagos (Fig. 8) is due to its proximity to the strong Nigerian AN emissions and its location downwind of fire plumes through Harmattan, as confirmed by SOFT-IO (see Sect. 3.1.1). The O_3 maxima show smaller variations (of approximately

Transport of CO (AN+BB) emissions from the African, South American and Asian source regions towards the 20 tropical sites taken into account for this study. The colorbar shows the amount of CO transported in ppb.

The second highest CO and O_3 maxima over the tropical regions Tropics are observed over Asia (Fig. 8) in January, mostly due to AN emissions. According to Table 2, the CO emissions over East Asia and India are lower than the ones over Western West Africa in January. This explains , explaining the lower CO mixing ratio ratios over Asia than over Lagos. SOFT-IO seems to represent represents better the Asian contributions than the African ones. As mentioned in Sect. 3.1.1, the AN emissions over

NH and SH emissions over Africa are likely underestimated by the SOFT-IO computations(Fig. 3 panels 3b and 4b). This is confirmed by that fact that CO mixing ratio is higher over Africa than over Asia, in contrast to the CO contributions estimated by SOFT-IO.

Previous studies have already found concentrations found mixing ratios of pollutants in West Africa (e.g. Lagos, Abidjan, Cotonou) comparable to those observed over Asian megacities (Assamoi and Liousse, 2010; Adon et al., 2016; Sauvage et al., 2007b). The rapid growth over of African megacities is indeed responsible for increasing emissions from diffuse and inefficient combustion sources (Marais and Wiedinmyer, 2016), such as residential sources mainly for cooking and heating (Zheng et al., 2019), and traffic emissions (related to a large number of two-stroke vehicles, poor fuel quality and poorly-maintained engines) (Assamoi and Liousse, 2010). In contrast, Eastern China has had one of the largest decreases in CO emissions (Hedelius et al., 2021) due to technological changes with improved combustion efficiency (Zheng et al., 2018a), such as replacing residential coal use with electricity and natural gas (Buchholz et al., 2021), reduction of NO_x emissions from coal power plants since 2010 (Wang et al., 2017), and implementation of Clean Air Policies (van der A et al., 2017) around 2010. In India, on the other hand, there are no regulation in the emissions, and this explains the highest CO mixing ratios among the Asian clusters (Fig. 8). Previous studies have already reported increasing CO emissions over India from 1996–2015, due to several factors such as increases in residential and agricultural sources (Pandey et al., 2014) and to power production and transport activities (Sadavarte and Venkataraman, 2014).

As in NT Africa, the CO-rich air masses accumulated in the LT over the Asian clusters in January are accompanied by a secondary LT O_3 maximum. However, these maxima are significantly lower (40–60 ppb) (Sect. 3.1.2) than the NT African ones (65–75 ppb) (Fig. 8a; Sect. 3.1.1), even for clusters with similar LT CO mixing ratios (e.g. Sahel and South China) (Fig. 8a). This is because: i) the CO emissions are less strong over the Asian clusters, as mentioned before, and ii) the O_3 enhancement over Asia is caused by AN-polluted air, while in NT Africa by mixed (AN and BB) polluted air. During the Atom campaign, Bourgeois et al. (2021) found that O_3 levels are more enhanced in mixed air pollution, because they are associated with greater NO_x and peroxy acyl nitrates (a NO_x reservoir compound), and thus increased O_3 production, in comparison to BB- or AN-polluted air alone. This is in agreement with the O_3 annual maximum in April over East Asia (Fig. 8a), over clusters such as South China and Bangkok, which are affected by the local fires.

The highest emission rates over East Asia and India are observed in April (Table 2). In the absence of the stability of the north easterlies, the air masses are not confined close to the surface like in January. Over East Asia, the contribution of the local fires is present in addition to the local AN emissions. The impact of the fires dominates in the MT in clusters such as South Chinaand Bangkok, and is evident over Manila and Ho Chi Minh City (see Sect. 3.1.2). Interestingly, the Northern Hemisphere African fires in January correspond to 72 % of the global burned area, whereas the Northern Hemisphere Asian fires only to the 2.5 % (Van der Werf et al., 2010). However, both regions contribute significantly to the global CO concentrations (44 % for Africa and 22 % for Asia) because of more complete oxidation, and thus reduced CO production, over grass fires (Africa savannas), relative to fires in forests and peatlands (deforestation and peatland fires over Asia) (Van der Werf et al., 2010). The large extend of the impact of the Northern Hemisphere Asian fires is displayed in IASI, with an outflow towards SE Asian

coast and the Pacific (Fig. 2f). The stronger winds in April than in January and the eastwards transport pathway (Fig. 10 panel 1 a–b) leads to lower CO mixing ratio in April, despite the higher emission rates (Table 2).

Concerning India, local AN emissions are responsible for the CO enhancement in April, with negligible BB contribution (Figs. 5 panel 3a and ?? panels 3a and 4a). According to IAGOS and IASI (Figs. 2a–d, 5 panel 1ab), the LT CO over India shows strong seasonal variability which cannot be explained by the weak variations of the Indian emissions (Table 2). This indicates that LT CO over India is rather linked with seasonal changes in the meteorological circulation. Similarly to East Asia, during January the air masses are transported southward due to the north easterlies, while the reversal of the winds to southwesterlies in July results in northward transport (Figs. 2 and S3) (Lawrence and Lelieveld, 2010). Because of this circulation pattern in July, the oceanic influence brings clean air masses over the Asian clusters resulting in an annual CO minimum during the Asian summer monsoon (Fig. 8). The CO-rich air masses for the surface are uplifted in the upper troposphere due to deep convection over the area (Sect. 3.1.2).

As for CO, O_3 seasonality is also linked with the seasonality of the meteorological conditions and dynamics over Asia. The O_3 maximum in April is attributed to the intense solar radiation associated with important amount of precursors from mostly AN emissions, except for South China where BB emissions dominate. The O_3 minimum occurs during the Asian summer monsoon (July), because of lower Over South China, CO emissions have also been reduced in the last years, but the O_3 production in the presence of convective clouds relative to clear sky conditions (Sect. 3.1.2).

Despite the CO emissions reductions over South China, the O_3 levels remain relatively high (Fig. 8a). This is because the O_3 production regime over South China is VOCs-limited (Li et al., 2013), and the total NMVOCs emissions increased by a factor of 3.5 (1997–2017) because of activity increases in the solvent, energy, and industry sectors (Zheng et al., 2018a). Despite the successful controls of NO_x emissions from coal fired power plants since 2010 over Eastern China (Wang et al., 2017), it is recommended to apply controls over VOCs emissions as they control the local O_3 distribution. In contrast, over India the O_3 production regime is NO_x-limited (Kumar et al., 2012), as the local In India, on the other hand, the lack of efficient pollution regulations explains the highest CO mixing ratios among the Asian clusters (Fig. 8). Previous studies have already reported increasing CO emissions over India from 1996–2015, due to several factors such as increases in residential and agricultural sources (Pandey et al., 2014), power production and transport activities (Sadavarte and Venkataraman, 2014). Furthermore, Indian emissions are mostly associated with incomplete combustion processes by biofuel burning , and thus higher NMHC processes from biofuel burning (Kumar et al., 2012) and are therefore characterised by higher NMVOCs to NO_x emission ratio as compared to other regions of the Northern Hemisphere (Lawrence and Lelieveld, 2010).

Concerning Central Africa, the O_3 and CO maximum in the LT during the dry season, indicates the strong dependence of the CO and O_3 distribution on the surface emissions, as over NT Africa. The CO magnitude over Central Africa is similar to the one over Sahel and Guinea Gulf during the respective dry season, even though the emissions rates are higher over the former (Table 2). This is because higher amount of CO impacted by the Southern Hemisphere African fires is transported towards the NT Africa due to the trade winds, relative to the respective southward transport during the Northern Hemisphere dry season (Fig. 10). In addition, the ratios than other Asian regions leading to a NO_x-limited O_3 mixing ratio is slightly higher over

~~Central Africa (85 ppb) likely indicating rapid photochemical O_3 production by BB precursors (Singh et al., 1996) during the Southern Hemisphere fires~~production regime (Lawrence and Lelieveld, 2010).

The smallest LT CO ~~maximum over the NT~~ maxima over the Northern Tropics are observed over ~~Arabia and East Africa~~ the AEA clusters and South America (Fig. 8) because of the smallest emissions rates among the ~~tropics~~ Tropics (Table 2). The ~~CO emissions over Middle East~~ Middle East CO emissions are mainly related to electricity generation, water desalination, and industry supplied by oil and gas deposits with cheap but relatively clean fuels (Krotkov et al., 2016). ~~In addition, because of its location between the two highest emittors (Asia and Africa), transport plays a significant role in CO enhancements over AEA, especially in~~ O_3 maxima in the MT and UT ~~where long range transport of emissions is favoured (Figs. 10 panels 1 b–c; 2 a–c and 2b–c). This transport from Asia and Africa over AEA clusters determines the O_3 maxima over the AEA clusters (Sect. 3.1.4). Similarly, over NT South America, the local AN contributions are much smaller than the respective local Asian of African ones, indicating lower pollution levels over South America than Asia and Africa. The O_3 maximum is controlled by~~ of the AEA and South American clusters are related respectively to pollution transport and $LiNO_x$ emissions at higher altitudes ~~.~~ (Sects. 3.1.4 and 3.1.3).

From the previous analysis, all the tropical clusters ~~and the associated CO source regions exhibit primarily local influence , in the proximity of the region where they are emitted. However, CO transport plays also~~ are primarily influenced by local sources of CO. However, AN and BB emissions can be redistributed vertically and then carried horizontally aloft by zonal winds to influence the tropical upper troposphere. Figure 9 displays the AN and BB contributions to the CO anomalies (in ppb) over the tropical UT_{cruise}. AN emissions have the largest influence (Fig. 9) especially in the Eastern part, from the Middle East to Asia in July and October. BB has a marked seasonal influence, less intense than the AN one. Fires strongly influence deep convection outflow regions, above the Inter Tropical Convergence Zone over Africa in January and July, above the South Atlantic Convergence over South America and the South Pacific Convergence Zone above Asia in October. For the latter, the influence of fire is of the same order as that of AN emissions (24 ppb simulated).

In addition to the local influence, long-range transport plays an important role in the ~~CO distribution over the tropics.~~ redistribution of these AN and BB emissions and, consequently, in the distribution of CO in the Tropics. To illustrate this, Figure 10 displays the transport pathways of CO emissions from the African, South American and Asian source regions, towards the 20 tropical IAGOS locations in the LT (a), MT (b) and UT (c). Only the source regions and the months corresponding to the largest amounts of transported CO (in ppb) are depicted.

CO sources located over Africa show the ~~maximum~~ largest influence on the regional tropical CO. ~~The highest impact of the African emissions is found at an inter-hemispheric scale, where CO~~ Large amounts of CO are indeed transported inter-hemispherically from the dry-season ~~African regions is transported~~ region towards the wet-season ~~African~~ region (Fig. ~~10 panels 1 and 2)~~ 9 and Fig. 10) from the LT with advection by the trades winds to the UT with the Hadley cells after vertical convective uplift in the convergence zone. As a result, CO contributions of 45–50 ppb (resp. over 50 ppb) from Northern Hemisphere (resp. Southern Hemisphere) Africa ~~is~~ are found over Southern (resp. Northern) Africa during the respective dry season in the ~~LT and MT. Also, Northern Hemisphere African emissions are transported towards South America (10–15 ppb in MT;~~

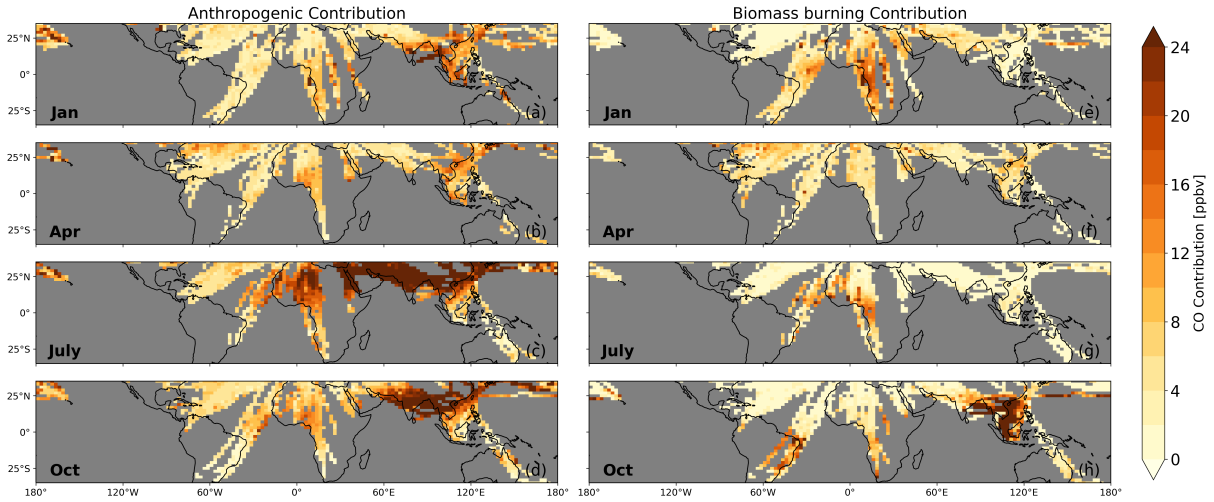

Figure 9. Mean AN (a–d) and BB (e–h) contributions in ppb between 300–185 hPa from 2002–2019.

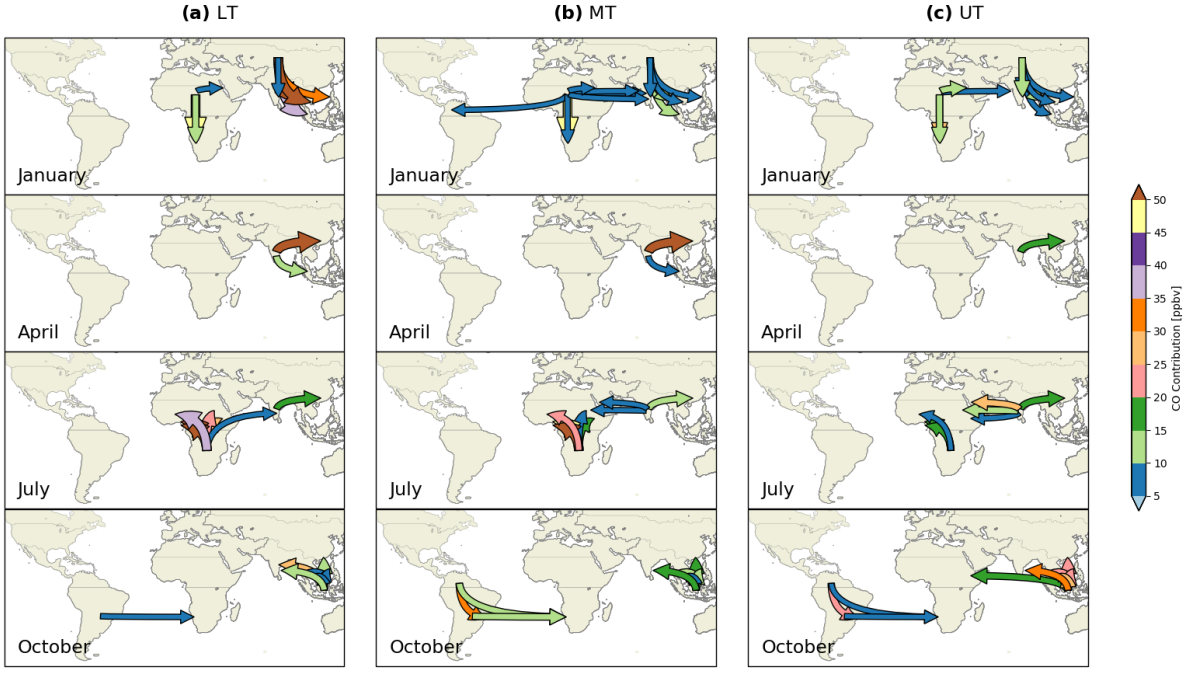

Figure 10. Transport of CO (AN+BB) emissions from the African, South American and Asian source regions towards the 20 tropical locations taken into account for this study. The colorbar shows the amount of CO transported in ppb.

5–10 ppb in the rest) in April (Fig. not shown) and significantly contribute to the local South American annual maximum (30 % and 50 % of CO anomalies over Caracas (LT and MT resp.) (Fig. 4a–b).

During the transport of the Southern (resp. Northern) Hemisphere African emissions towards the ITCZ location in the Northern (resp. Southern) Africa, the air masses reach convective regions and are injected in the UT_{cruise} (Fig. 9). This entire troposphere. In the UT this explains why the Southern (resp. Northern) Hemisphere African emissions are dominant in the wet-season hemisphere during July (resp. January) (FigsFig. 9 and resp. ?? and ??). Nevertheless, the Northern Hemisphere African contribution in the UT_{cruise} CO anomalies is present on a local scale all year long, above NT Africa and South Atlantic. Fig. S12 and Fig. S10). During the dry season, the impact of the Northern Hemisphere African emissions is stronger and extends to a wider area over South America, Middle East, the Middle East and South Asia (Figs. ?? Fig. S10 NHAF). Also, Northern Hemisphere African emissions are transported towards South America (10–15 ppb in MT; 5–10 ppb in the rest) in April (Fig. not shown) and significantly contribute to the local South American annual maximum (30 % and 50 % of CO anomalies over Caracas, LT and MT resp.) (Fig. 10 and Fig. 4a–b).

Mean AN (a–d) and BB (e–h) contributions in ppb between 300–185 hPa from 2002–2019.

The contribution of Asian emissions in the tropical LT is limited to a regional or local scale, as they are mostly impact impacting neighbour Asian regions (Fig. 10a)(see Sect. 3.1.2 for more details). . CO export from Asia is favoured enhanced during the Asian summer monsoon and post monsoon post-monsoon (July and October) in the UT, where the transport is favoured due to stronger winds relative to the surface (Fig. S3).

During the Asian summer monsoon, the CO-rich (and O_3-poor) air masses from the boundary layer (Fig. 2c and g) are convectively uplifted in to the UT_{cruise} (FigsFig. 9c and ??Fig. S12), and trapped in the AMA circulation (see Sect. 3.1.2). Asian emissions are transported towards Arabia (25–30 ppb) and Eastern Africa (10–15 ppb) in the UT_{cruise} (FigsFig. 9c; Fig. 10 panel 3c and ?? Fig. S12 SEAS and CEAS). Subsidence of air masses from AMA above Arabia are responsible for is contributing to an O_3 maximum above AEA (Fig. 8a) (Sect. 3.1.4). During the post monsoon post-monsoon season (October), sporadic convection uplifts CO emitted by the Equatorial Asian fires in the UT_{cruise} (FigsFig. 9g , ?? and Fig. S13 EQAS) (see Sect. 3.1.2). At the same time, convection over continental Asia uplifts SouthEast South-East Asian AN emissions (FigsFig. 9d and ?? SEAS) in Fig. S13 SEAS) to the UT_{cruise}. As a result, CO from the Asian emissions impacts Asian emissions impact CO anomalies in the UT over Eastern Africa (15–20 ppb from Equatorial Asia and 5–10 ppb from SouthEast South-East Asia; Fig. not shown).

South American emissions mostly influence the troposphere over the South Atlantic and South America in October (Fig. 10) through the South Atlantic Anticyclone circulation, accounting for around 25 % of the observed CO.

4 Summary and conclusions

IAGOS O_3 and CO observations since 1994 and 2002 respectively, were used (when available) in order to analyse vertical to analyse tropospheric profiles over 20 tropical siteslocations, along with the UT_{cruise} . One limitation of our study is the rather limited spatial coverageof IAGOS profiles to limited locations. However, according to Petetin et al. (2018a), a few hundreds

meters above the ground, these measurements are representative of distributions. Despite their sparse spatial coverage, these observations provide a good overview of the O_3 and CO distributions in most tropical regions. They are indeed representative of both the urban background close to the ground and of the regional scale at higher altitudes in the lower troposphere starting a few hundred meters above the surface (Petetin et al., 2018a). Furthermore, IASI, which provides global daily global IASI-SOFRID O_3 and CO distributions with a coarse vertical resolution, allows allow us to complement IAGOS observations on the global scale over the data sparse over the whole tropical band. Throughout the paper we have shown that the anomalies detected by IAGOS are often also detected by IASI at the regional scale.

In the LT, The results of the study indicate that the highest O_3 and CO mixing ratios are observed over Africa during the fire season in January, with anomalies located in the LT (75 ppb at 2.5 km for O_3 and 800 ppb at 0.3 km for CO over Lagos), and explained mainly by anthropogenic emissions, but with a strong contribution from fires. The secondary O_3 and CO maxima are observed over Asia, in the MT-UT in April for the first and in the LT in January for the second, with a larger contribution from AN emissions. The Asian distributions are largely controlled by meteorological conditions associated with the Asian monsoon phases. The lowest anomalies are measured over Caracas.

More specifically in the tropical LT, where the CO is maximum, the CO anomalies are caused by a combination of AN and BB emissions. In the majority of the clusters , local AN contributions are dominant are mostly influenced by local and regional AN contributions all year long. The BB contribution increases or dominates over some clusters, when the regional or local fires are active. Local In particular, AN emissions have the greatest impact over Asia, Arabia and Eastern Africa and South America, where they account for more than 80 75 % of the CO. The BB impact increases These results highlight the importance of the AN emissions over the tropical locations, in accordance with the global decreasing trends of BB and the increasing AN emissions. The impact of BB in the LT has also a predominantly local and regional influence with a strong seasonal dependence. BB contributions increase over South China (35 % in April) , and dominates and dominate over the Gulf of Thailand (90 % in October) during the local fires (SouthEast fire season (South-East and Equatorial Asian resp.). Over NH NT Africa, with contributions in the range of 57-85% local AN emissions largely dominate the CO anomalies all year long. There are a few exceptions of larger BB contributions in January over Guinea Gulf (53%) and in Julyover Lagos (53%) and Guinea Gulf (66%) during Northern and Southern Hemisphere African BB seasons. Similar impact of the Southern Hemisphere African fires is found over Khartoum in July. In contrast, the rest of the AEA clusters are impacted by local ANemissions all year long (70–95 %). Over South America, stronger local AN contribution are found over the ST (81–94 % over SBrazil) than in the NT (75–80 % Caracas and Bogota), while the BB contributions are similar (51 % over Caracas in Apriland 53 % over SBrazil in In contrast to the other continents, African LT with the highest O_3 and CO measured, is more impacted by BB during the dry seasons (January and July) with an influence of the same order as AN. This is also the case over Caracas (April) and SBrazil (October). The highest BB impact is found over ST Southern Hemisphere Africa all year long (57%–90%57–90%), except in April, with BB emissions mostly originating from local fires, but also from Northern Hemisphere Africa in January (45-73 %) and Southern Hemisphere Southern America in October (29 % over Windhoek). During the transition periods, the local

In the MT and UT, AN contributions are ~~larger (46 and 80 %). Our results highlight the importance of the AN emissions over the tropical sites, even in the ST. This is in accordance with the global decreasing trends of BB and the increasing AN emissions.~~

~~In the MT and UT, the BB~~ more important and dominate in the Eastern part of the Tropics, from the Middle East to Asia. BB contributions are increased compared to the LT ~~, and their effect dominates over more clusters. Also, the contribution of the transport is more important than in the LT, where mostly local emissions dominate. Over NT Africa, the BB dominates twice a year, during the Northern and Southern Hemisphere dry seasons, because of local and Southern Hemisphere African fires respectively. In NT Africa , as in the LT, BB dominates all year long except April. In addition to the African BB, AN SouthEast Asian and BB Southern Hemisphere South American contributions are found in the MT and UT. Over Asia, SouthEast Asian BB contributions dominate~~ and are higher in the outflow of the deep convection regions, especially African fires over Africa in January and July, and South-East and Equatorial Asian fires over the South East Asian coast (South China, Manila, Ho Chi Minh City) in April ~~in the MT. In contrast, the impact of the Equatorial Asian BB is stronger in the UT, and has a largest spatial extent over the South East Asian coast (China, Ho Chi Minh City, Manila), India (Madras) and Eastern Africa (Addis Ababa).~~

~~Over Africa, the O_3 and CO maxima are observed in the LT during the respective dry season. The role of the local AN emissions are~~ or October. Also, the contribution of long-range transport is more important than ~~previously documented as: i) local AN emissions control the CO anomalies over Lagos and Sahel, and ii) the persistent CO-rich surface layer in Central Africa is caused by local AN emissions (40 and 86 %) in the absence of local fires. Africa is also the most important~~ in the LT and allows connections between source regions and other continents.

Africa is the leading tropical region in terms of ~~export of emissions in~~ emissions export to the tropical troposphere. ~~According to IASI, the~~ The main export pathway is the inter-hemispheric transport of O_3 and ~~precursors~~ CO from the dry-season African regions to the wet-season ones~~(≈ 50 ppb), confirmed by SOFT-IO. During the dry season, the~~ . Northern (resp. Southern) Hemisphere African fires are also the dominant source of CO over ~~AEA~~ Arabia and Eastern Africa (resp. Khartoum and Jeddah) in the MT and UT, and they also reach ~~India accounting for 5–10 ppb in~~ the MT and UT of India, accounting for 20% of the observed CO. Transport of ~~mostly BB~~ African emissions from Africa occurs all year round towards northern South America in ~~all tropospheric layers. The highest Northern Hemisphere African regional impact is found~~ the entire troposphere, with the largest impact over Caracas in the MT and UT (30 % on average).

In contrast, the impact of Asian emissions ~~,~~ is mostly limited on a regional or local scale, especially in the LT and MT. ~~The transport of the~~ Nevertheless, long-range transport of Asian emissions is ~~important only during the Asian summer monsoon in the UT towards Arabia and Eastern Africa.~~

~~The highest O_3 and CO maxima among the tropical clusters occur in the LT of NT Africa in January (75 ppb at 2.5 km for O_3 and 800 ppb at 0.3 km for CO over Lagos). This is largely a result of the local AN emissions as suggested by the co-occurrence of the peaks of O_3 and CO in the LT. In contrast over Asia, the second most polluted region, the distributions are mostly controlled by meteorological conditions associated with the Asian monsoon phase. The CO maximum occurs in the LT during January, due to the stability of the northeasterlies which confine the CO-rich air masses to the LT. The annual maximum of O_3 occurs during the pre monsoon season (April) when the increased solar radiation favours O_3 production.~~ responsible for

60 to 80% of the CO anomalies in the UT over AEA in July and October. During the Asian summer monsoon ~~, O_3 and CO mixing ratio minimize in the LT because of : i) transport of clean oceanic air above continental Asia, ii) reduced photochemical O_3 production due to cloudy conditions, and iii) convective uplift of CO-rich air masses from the surface towards the Asian upper troposphere.~~

~~Over Asia , the LT and MT CO and O_3 anomalies are mostly impacted by regional or local Asian emissions of AN origin. The BB contribution is important during April and significantly contributes to CO anomalies over South China. According to IASI, the BB impact extends over the tropical Pacific. The export of the AN Asian emissions is important only in the UT during the Asian summer monsoon and post monsoon season (July and October). According to IASI, the polluted air masses from the surface are uplifted in the UT_{cruise} in July and are trapped in the AMA. These air masses are transported over AEA (CO contributions of 25–30 ppb and 10–15 ppb) causing the annual O_3 maxima due to subsidence and high isolation over the regions. This highlights the importance of long range transport for the air quality in the UT over Arabia, which shows the lowest CO local contribution and the highest O_3 levels among the tropical clusters. The CO transport towards Eastern Africa in the UT by the Tropical Easterly Jet, is found in October when the~~ CO from South Asia is transported westwards within the AMA while in October, the tropical easterly jet transports air masses impacted by ~~the Indonesian fires , and the AN continental source are uplifted in the UT, and transported towards Eastern Africa(CO contributions of 15–20 ppb and 5–10 ppb respectively)~~Indonesian fires towards Africa.

Last, over South America the local CO contributions at the surface level are as low as over ~~Arabia and Eastern Asia~~AEA. During the dry season (October), when the convection moves over the South American fires, CO and precursors are trapped in an anticyclonic circulation developed over Central South America, resulting in the annual local maxima of O_3 and CO. ~~The transport of O_3 and precursors over Atlantic can be seen by IASI and this contributes to the O_3 wave-one pattern. This is confirmed by SOFT-IO which calculates~~ As a result, Northern and Southern Hemisphere South American ~~contribution of 10–15 ppb each towards Windhoek, in the altitude of the anticyclone(MT).~~

5 ~~SOFT-IO CO contributions~~

4.1 ~~Vertical profiles~~

~~Same as Fig. 3 (panel 3) for CO contributions over Sahel (1), Gulf of Guinea (2), Madras (3), Hyderabad (4), Ho Chi Minh City (5), Gulf of Thailand (6) and Manila (7).~~

4.1 ~~Low, mid and upper troposphere~~

~~Same as Fig. 4 for CO contributions over Windhoek, Addis Ababa, Khartoum, Jeddah and Gulf of Thailand.~~ ~~Same as Fig. 4 for CO contributions over Madras, Mumbai, Hyderabad, Ho Chi Minh City and Manila.~~

4.1 ~~Upper troposphere~~

~~Mean CO contribution (in ppb) per source region in the tropical UT_{cruise} (300–185 hPa) averaged from 2002–2020 for January. The hatched part indicates BB as the dominant source of CO. Same as Fig. ?? for April. Same as Fig. ?? for July. Same as Fig. ?? for October.~~

emissions contributed to 30 % of the CO observed in the MT over Windhoek, after transport in the South Atlantic anticyclone.

Data availability. The IAGOS data are available on the IAGOS data portal (https://doi.org/10.25326/20). The SOFT-IO v1.0 products are part of the ancillary products of IAGOS central database (https://doi.org/10.25326/2; https://doi.org/10.25326/3). The SOFRID-O_3 data are freely available on the IASI-SOFRID website (http://thredds.sedoo.fr/iasi-sofrid-o3-co/, last access: 8 June 2022; SEDOO, 2014).

Author contributions. MT, BS and BB designed the research. All the co-authors contributed to acquisition of data. MT analysed the data. MT, BS and BB interpreted the data. MT drafted the article. MT, BS and BB revised the article. VT and HC commented the article. ELF and BB are responsible for the SOFRID retrieval software. ELF is in charge of the production and quality control of SOFRID data.

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

Acknowledgements. We acknowledge the strong support of the European Commission, Airbus and the airlines (Deutsche Lufthansa, Air France, Austrian, Air Namimbia, Cathay Pacific, Iberia, China Airlines and Hawaiian Airlines) that have carried the MOZAIC or IAGOS equipment and performed the maintenance since 1994. IAGOS has been funded by the European Union projects IAGOS–DS and IAGOS–ERI. Additionally, IAGOS has been funded by INSU-CNRS (France), Météo-France, Université Paul Sabatier (Toulouse, France) and Research Center Jülich (FZJ, Jülich, Germany). The IAGOS database is supported in France by AERIS (https://www.aeris-data.fr). IASI is a joint mission of EUMETSAT and the Centre National d'Etudes Spatiales (CNES, France). The authors acknowledge the CNES for financial support for the IASI activities.

Financial support. This research has been supported by Bonus Stratégique programme at Université Paul Sabatier Toulouse III who funded the first author's doctoral position.

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