

An upper tropospheric ‘ozone river’ from Africa to India during the 2008 Asian post-monsoon season

By FLORE TOCQUER^{1,2*}, BRICE BARRET^{1*}, CÉLINE MARI¹, ERIC LE FLOCHMOËN¹, JEAN P. CAMMAS^{3,4} and BASTIEN SAUVAGE¹, ¹Laboratoire d’Aérodynamique, Observatoire Midi-Pyrénées, Université Paul Sabatier, CNRS, Toulouse, France; ²Météo-France, Toulouse, France; ³LACy (Laboratoire de l’Atmosphère et des Cyclones), UMR8105, CNRS – Université de la Réunion – Météo-France, Saint Denis de la Réunion, France; ⁴UMS3365 de l’OSU-Réunion, CNRS – Université de la Réunion, Saint Denis de la Réunion, France

(Manuscript received 2 July 2014; in final form 23 January 2015)

ABSTRACT

We have used ozone data from the Infrared Atmospheric Sounding Interferometer to follow an event of ozone-enriched air-masses in the upper troposphere from eastern Africa to northern India. The ozone transport (hereafter called ‘ozone river’ or O3R) occurred during the Asian post-monsoon season in 2008 and was associated with Rossby wave propagation. The persistence of the O3R in a narrow channel was confirmed by MOZAIC airborne data over the northwestern Indian coast. The regions of origin of the O3R were identified by a transport analysis based on the Lagrangian model FLEXPART. The Lagrangian simulations combined with potential vorticity fields indicate that stratospheric intrusions are not likely to be the most important contributor to the observed O₃ enhancements. A high-resolution Eulerian model, Meso-NH, with tagged tracers was used to discriminate between African biomass burning, lightnings and Indian anthropogenic pollution as potential sources of precursors for the O3R. Lightning NO_x emissions, associated with convective clouds over Africa, were found to be the principal contributor to the ozone enhancement over the Indian Ocean taking advantage of a northeastward jet. This case study illustrates African lightning emissions as an important source for enhanced O₃ in the upper troposphere over the Indian Ocean region during the post-monsoon season.

Keywords: ozone, Indian Ocean, upper troposphere, lightning

This paper is part of a Special Issue on MOZAIC/IAGOS in Tellus B celebrating 20 years of an ongoing air chemistry climate research measurements from airbus commercial aircraft operated by an international consortium of countries. More papers from this issue can be found at <http://www.tellusb.net>

1. Introduction

Ozone (O₃) is one of the most important greenhouse gases contributing to global warming. The net radiative forcing resulting from changes in tropospheric O₃ during the next century is particularly important within the northern tropics from Central America to South Asia and the Indian Ocean (Shindell et al., 2013). In the tropics, the upper tropospheric (UT) O₃ is characterised by a global wave-

number-one zonal variability with the highest O₃ concentrations over the Atlantic and the lowest O₃ concentrations over the Western Pacific (Fishman et al., 1990; Thompson et al., 2003). As part of this global pattern, UT O₃ is low over the whole Indian Ocean region while it is much higher over Africa, where it benefits from a process of injection of O₃ precursors followed by zonal O₃ build-up inside Walker cells (Sauvage et al., 2007b).

Nevertheless, O₃ profiles measured during the Pre-INDOEX (1995–1998) and INDOEX (1999) campaigns in the northern Indian Ocean region revealed the frequent occurrence of elevated O₃ concentrations in the mid- to upper-troposphere during the winter season (Zachariasse

*Corresponding authors.
email: flore.tocquer@aero.obs-mip.fr; brice.barret@aero.obs-mip.fr
Responsible Editor: Leonard Barrie, Stockholm University, Sweden.

et al., 2000, 2001; De Laat, 2002; Chatfield et al., 2007; Lawrence and Lelieveld, 2010). In particular, O_3 peaks reaching 120 ppbv were often found in the UT within layers 1–2 km thick just below the tropopause (De Laat et al., 1999).

Several sources and transport processes were put forward to explain these high concentrations of O_3 in the mid-upper troposphere over the Indian Ocean. Using ozonesondes launched in winter 1998 and back-trajectories analysis, Zachariasse et al. (2000) demonstrate that O_3 laminae in the UT originate from stratosphere to troposphere exchange (STE) along the subtropical westerly jet (SWJ). This direct contribution of O_3 by isentropic intrusion of stratospheric air masses is characterised by high O_3 concentrations associated with low humidity. Based on O_3 observations from radiosondes during INDOEX 1999, satellite measurements and back-trajectories analysis, Chatfield et al. (2007) attribute O_3 peaks in the mid-troposphere primarily to STE and convective lofting of pollution from the Indian continental outflow in the intertropical convergence zone (ITCZ).

Furthermore, Chatfield et al. (2004, 2007) attribute east–west streaks of elevated mid-tropospheric O_3 observed from space over the Indian Ocean to high-level westward transport of pollution potentially contributing to the south Atlantic O_3 maxima in boreal winter. Indeed, the biomass burning (BB) season in Africa in the Northern Hemisphere takes place from October to January (Cooke et al., 1996). Transport of BB emissions in the free troposphere is observed and described in Edwards et al. (2003), which explains the inter-hemispheric transport of African O_3 precursors. Through the analysis of modelled CO (carbon monoxide) and O_3 distributions, De Laat (2002) explains winter mid-tropospheric O_3 peaks observed from radiosondes over the Indian Ocean with the eastward advection of air masses impacted by African BB emissions caused by the propagation of waves along the SWJ. It must be emphasised that in their review of the outflow of atmospheric pollution from south Asia, Lawrence and Lelieveld (2010) point to some possible inconsistency in De Laat (2002) and rather support the scheme of Chatfield et al. (2007) to explain the elevated layers of O_3 in the Indian Ocean mid-troposphere.

Another significant source of O_3 precursors over this equatorial region is lightning. Deep convection that characterises central Africa during austral summer is both promoting vertical mixing of air masses but also a high electrical activity. From spaceborne data Bond et al. (2002) present an overview of the distribution of nitrogen oxides (NO_x) produced by lightning ($LiNO_x$) with a peak production between September and November and show the importance of these emissions relative to other sources of NO_x in this region. Based on simulations from a chemical transport model (CTM) and airborne observations, Barret et al. (2010) demonstrate the significant impact of $LiNO_x$ on the

middle to upper troposphere over Africa and the tropical Atlantic during the West African Monsoon. Indeed, $LiNO_x$ emitted directly into the UT have a longer life-time and can therefore travel longer distances increasing their impact on tropospheric chemistry (Labrador et al., 2005; Sauvage et al., 2007b). The tropospheric O_3 distributions over the Indian Ocean between 2005 and 2009 have been examined in a study by Zhang et al. (2012). Based on spaceborne observations they document an annual maximum of O_3 in the mid- and upper-troposphere of the equatorial southern Indian Ocean during May and conclude that this maximum results from lightning NO_x emissions. Referring to Zhang et al. (2011) they explain the important O_3 maximum detected in the mid- and upper-troposphere over the Indian Ocean during autumn 2006 by the strong El-Nino event. This event was indeed responsible for large-scale fires in equatorial Asia and perturbations of the tropospheric circulation that resulted in the transport of BB impacted air masses to the Indian Ocean.

This study focuses on an event of O_3 elevated air-masses transport from Africa to northern India during early winter 2008 identified from spaceborne Infrared Atmospheric Sounding Interferometer (IASI) observations, hereafter referred to as ‘ozone river’ (O3R). The high spatio-temporal resolution of IASI enabled the observation of such an event for the first time to our knowledge. This observed episode provides an opportunity to better understand the transport processes responsible for O_3 enrichments within the Indian Ocean troposphere. Section 2 describes the spaceborne and airborne observations and the models used in the present study. The chemical and meteorological context associated with the O3R is described in Section 3. In Section 4, we use Lagrangian and Eulerian simulations to provide insights into the emission sources of the O3R and their transport pathways.

2. Data and models description

2.1. IASI spaceborne data

The Metop-A/IASI sensor measures the Earth–atmosphere radiance in thermal infrared (range $645\text{--}2760\text{ cm}^{-1}$) with a spectral resolution of 0.5 cm^{-1} . With a 2200 km swath across the flight track (12 km footprint at nadir), IASI provides a global coverage twice daily. Designed to measure atmospheric temperature and water vapour, IASI quantifies atmospheric abundances of a number of chemical species such as CO (George et al., 2009; De Wachter et al., 2012) and O_3 (Barret et al., 2011; Dufour et al., 2012).

The O_3 tropospheric profiles used in the present study were retrieved from IASI radiances with the Software for a Fast Retrieval of IASI Data (SOFRID) described in Barret et al. (2011). IASI–SOFRID O_3 data are validated

on the global scale (Barret et al., 2011; Dufour et al., 2012) with ozonesonde data and over the Indian region with Measurements of OZone, water vapour by in-service Airbus airCraft (MOZAIC) airborne data (Barret et al., 2011). The retrieved O_3 profiles contain between 1.8 and 2.5 independent pieces of information in the tropics, which is equivalent to an 8-km resolution. They are particularly suited to study the upper troposphere–lower stratosphere (UTLS) O_3 because the sensitivity of the O_3 retrievals is the highest in this altitude range ($R > 0.91$) (Barret et al., 2011; Dufour et al., 2012). Furthermore, the retrieval error of the IASI–SOFRID UTLS columns is estimated to be less than 15% for O_3 . Nevertheless, according to Dufour et al. (2012) the three different IASI retrievals investigated are overestimating ozonesondes UT columns by 11–24% when smoothing by the retrievals averaging kernels are taken into account. The bias is larger (20–40%) for UT mixing ratios when the ozonesonde profiles are not smoothed by the averaging kernels (Dufour et al., 2012).

2.2. MOZAIC airborne data

The MOZAIC programme gathers concentrations of trace gases mainly in the UTLS between 9 and 12 km from measurements aboard commercial aircrafts (www.iagos.fr and Marengo et al., 1998). The vertical resolution is about 20–30 m and the horizontal resolution is about 1 km. The airborne *in-situ* measurements of CO and O_3 used in this study were performed from Hyderabad (17.2°N, 78.3°E, India) to Frankfurt (50°N, 8.6°E, Germany). Based on the dual-beam UV absorption principle, the ozone measurement accuracy is estimated to ± 2 ppbv ($\sim 2\%$) for a 4 seconds response time (Thouret et al., 1998). Based on an infrared analyser, the CO measurement accuracy is estimated to be ± 5 ppbv ($\sim 5\%$) for a 30 seconds response time (Nedelec et al., 2003). In the present study, we have used the data from the four Hyderabad–Frankfurt (HY–FR) flights that took place during the O3R event from 27 November to 3 December, that is, two flights on 27 November and two flights on 3 December.

2.3. FLEXPART Lagrangian particle dispersion model

In order to determine the geographical regions influencing the O_3 concentrations in the UT over the Arabian Sea, simulations were performed with the FLEXPART Lagrangian particle dispersion model (version 6.2) (Stohl et al., 1998, 2005). FLEXPART simulates the transport and dispersion of linear tracers and treats advection and turbulent diffusion by calculating the trajectories for a multitude of particles. Stochastic fluctuations, obtained by solving Langevin equations (Stohl and Thomson, 1999), are superimposed on the grid-scale winds from the European Centre

for Medium-Range Weather Forecasts (ECMWF) data set to represent transport by turbulent eddies. The spatial and temporal resolutions of ECMWF data are respectively $1^\circ \times 1^\circ$ and 3 hours. FLEXPART enables establishing a relationship between a source which may be a region characterised by pollutant emissions and a receptor which may be a location impacted by these emissions. A large number of particles (not necessarily representing real particles, but infinitesimally small air parcels) is released from the receptor location and transported backward in time. For the present work a large number of so-called particles are released from three different boxes over the Arabian Sea between 9 and 12 km, and located within and on either side of the O3R in UT (rectangles on the Arabian Sea in Fig. 5) (see Section 4.1 for further details on the boxes definition). Particles are released during 4 d between 29 November 2008, 12 UTC, and 2 December 2008, 12 UTC, which corresponds to the period with the highest O_3 concentration according to IASI. The time step for backward model calculation is 3 hours. Back trajectories calculation period is set to 20 d. FLEXPART outputs the residence time of all the particles that represents the sensitivity of the source to the receptor. Output is produced every 3 hours (particles position and residence time).

2.4. Meso-NH chemistry and meteorology model

The French research Mesoscale Non-Hydrostatic model (Meso-NH) used in this study is a grid point Eulerian model based on a non-hydrostatic system of equations. It couples meteorology and chemistry and allows to simulate atmospheric motions ranging from the synoptic scale to large-eddy simulations. A full description of the model capabilities is available on <http://www.aero.obs-mip.fr/observation/mesonh> (Lafore et al., 1998). The model is used to investigate a large domain covering the Indian Ocean, central and eastern Africa and south-west Asia [20°S – 40°N ; 4° – 85°E]. The simulation was conducted on one single domain with a horizontal dimension of 6700 km by 9300 km at 15 km horizontal resolution and a vertical grid of 72 levels, with a stretched resolution ranging from 60 m at the lowest level to 600 m at the highest. The altitude of the model top is at 30 km.

The atmospheric model (version 4.8) was run from 20 November 2008 to 15 December 2008 with a time step of 15 seconds. The initial and lateral boundary conditions were obtained from ECMWF analyses. The physics of the model included the prognostic calculation of the turbulence and a convection scheme based on mass-flux calculations: the Kain–Fritsch parametrisation scheme (Bechtold et al., 2001). The exchanges of energy at the surface are represented by four possible surface types (natural surfaces, urban areas, oceans and lakes) in the SURFEX model coupled with

Meso-NH (Masson et al., 2012). Shallow convection is parameterised according to Pergaud et al. (2009). A mixed-phase microphysics (Pinty and Jabouille, 1998) and the subgrid cloudiness (Chaboureaud and Bechtold, 2002) are available for these simulations. The radiation fluxes are provided by the ECMWF scheme (Gregory et al., 2000). Three passive tracers were used to mimic O_3 precursors and assess the potential contribution of three important pollution sources in this region: BB over Africa between 5° and 15° N, lightning produced NO $_x$ associated with the convective clouds within the ITCZ; and anthropogenic pollution outflow from India. The tracers have a fixed lifetime of 10 d and are initialised to zero. The first tracer represents the NO $_x$ produced by the combustion of biomass burning (hereafter BBTR). Monthly BB emissions were derived from the GFEDv3 inventory (Giglio et al., 2010; Van der Werf et al., 2010). The second one represents the NO $_x$ produced by lightning activity (hereafter LITR). Its source is directly calculated in the deep convection scheme of the model following the parametrisation of Mari et al. (2008). The last tracer represents the CO produced by human activities (hereafter ANTR). Monthly emissions are taken from the MACCity inventory (Granier et al., 2011).

Both models are used to provide complementary views on the processes responsible for the O3R. First, FLEX-PART Lagrangian simulations enable the determination of the broad geographical domain influencing the O3R. Then, Meso-NH simulations carried out over this domain are used to determine the O_3 precursor sources impacting the O3R.

3. Observations and analysis

3.1. IASI UT ozone

The mean O_3 distribution observed by IASI in the UT (190–290 hPa) from Africa to South Asia during the post-monsoon season of 2008 (October–December) is marked by high concentrations of O_3 over Africa and low concentrations over the Indian Ocean (Fig. 1). This feature is the regional part of the tropospheric ozone zonal wave-one initially detected with satellite data by Fishman et al. (1990) and characterised by tropospheric columns of O_3 minima over the western Pacific and maxima over the tropical Atlantic. The zonal wave-one was later documented by Thompson et al. (2003) with tropical ozonesonde data, completed over continental Africa by Sauvage et al. (2006) with MOZAIC data and globally by Sauvage et al. (2007b) with satellite data and model simulations. The minima are a result of convective mixing over the Indian Ocean and the western Pacific that injects O_3 poor air masses from the marine boundary layer (BL) into the UT [see Lawrence et al. (2003) for further details about the impact of convec-

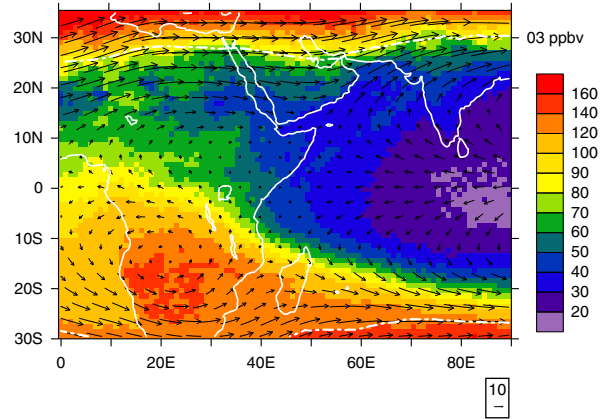


Fig. 1. IASI-SOFRID O_3 UT (190–290 hPa) for 3 months average (October–December 2008). 200–300 hPa horizontal wind from ECMWF reanalysis are represented as black arrows. The dashed white line represents the ± 1.5 PV units surface from ECMWF reanalysis at 200 hPa for the same period.

tion on tropospheric O_3]. The primary source responsible for the maxima that extend from Africa to South America is LiNO $_x$ produced over these continents (see e.g. Sauvage et al., 2007b). The UT O_3 concentrations from IASI over Africa are clearly overestimated with values reaching 120 ppbv which is 40 ppbv higher than the highest values documented by Sauvage et al. (2006) for southern Africa during SON. This is probably a result of the 20–40% IASI O_3 mixing ratios overestimation in the UT described in Section 2.1.

At the end of November 2008, the UT O_3 distribution observed by IASI shows a striking evolution with a channel of elevated O_3 that starts from the coast of Somalia around 26 November (Fig. 2a), crosses the Arabian Sea and northern India (Fig. 2b, c) and recedes after 4 December. We have called this phenomenon O3R. During this period, the region encompassing the Bay of Bengal, south India and the southeastern Indian Ocean from 20° S to 20° N remains little perturbed with a persistent O_3 minimum in the UT. The high O_3 concentrations following the undulations of the SWJ correspond to extratropical stratospheric air masses as will be discussed further in the text.

3.2. MOZAIC ozone observations

The MOZAIC HY-FR observations are used to corroborate the occurrence and composition of the O3R over the Arabian Sea. Figure 3 shows MOZAIC in-flight observations off the western coast of India and IASI O_3 pressure–longitude cross-sections along MOZAIC flight tracks during the O3R period. The corresponding flight tracks are displayed on top of the IASI O_3 distribution in Fig. 2a, c. On 27 November, IASI and MOZAIC ozone mixing ratios

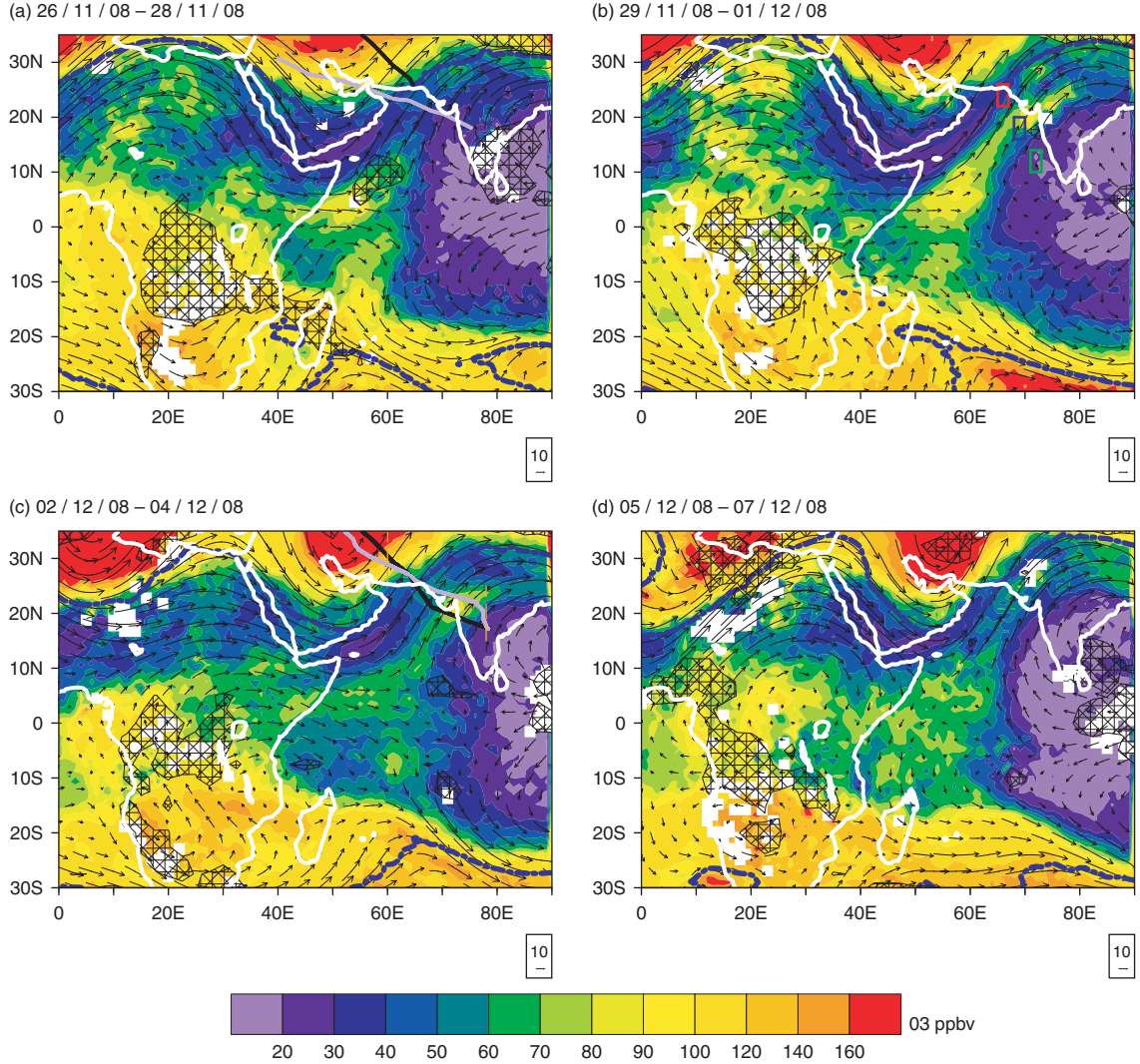


Fig. 2. IASI-SOFRID UT O_3 (190–290 hPa average), in ppbv, for (a) 26–28 November, (b) 29 November–1 December, (c) 2–4 December 2008, (d) 5–7 December 2008. 200–300 hPa averaged horizontal wind from ECMWF reanalysis are represented as black arrows. The blue line represents the ± 1.5 PV units surface from ECMWF reanalysis at 200 hPa. Black hatched contours indicate OLR below 220 W/m^2 . The black and grey lines in (a) and (c) indicate the flight track for MOZAIC observations, Hy–Fr and Fr–Hy respectively. The coloured boxes in (b) over Arabian Sea shows the starting boxes of particulate emissions for FLEXPART simulations.

were in good agreement showing both a longitudinal gradient from low O_3 concentrations typical of the tropical oceanic UT to enhanced O_3 concentrations when the aircraft crossed the SWJ and entered the extratropical stratosphere. On 3 December, the O_3 features observed during the outward and return MOZAIC flights between 70° and 80°E are well captured by IASI. The correlation coefficient between IASI and MOZAIC data for this episode taking all the observations displayed in Fig. 3 into account, is $R=0.85$, which shows that IASI is able to capture the variability of UT O_3 . This comparison remains qualitative as IASI profiles have a vertical resolution of about 8 km in the UT while MOZAIC data are *in-situ*.

3.3. Meteorological situation

The horizontal 200 hPa winds and the 1.5 PVU contours (in absolute values) from ECMWF reanalyses are displayed in Fig. 2 for the O3R period. Prior to the O3R event, between 6 and 25 November (not shown), the stratosphere–troposphere limit (1.5 PVU) lies between 30° and 20° in both hemispheres. During the O3R period, strong winds lie along the Northern Hemisphere SWJ and result from midlatitude Rossby waves propagating into the tropics, over the Arabian Peninsula. A number of studies show that Rossby waves tend to propagate through an equatorial westerly duct in the UT (e.g. Webster and Holton, 1982;

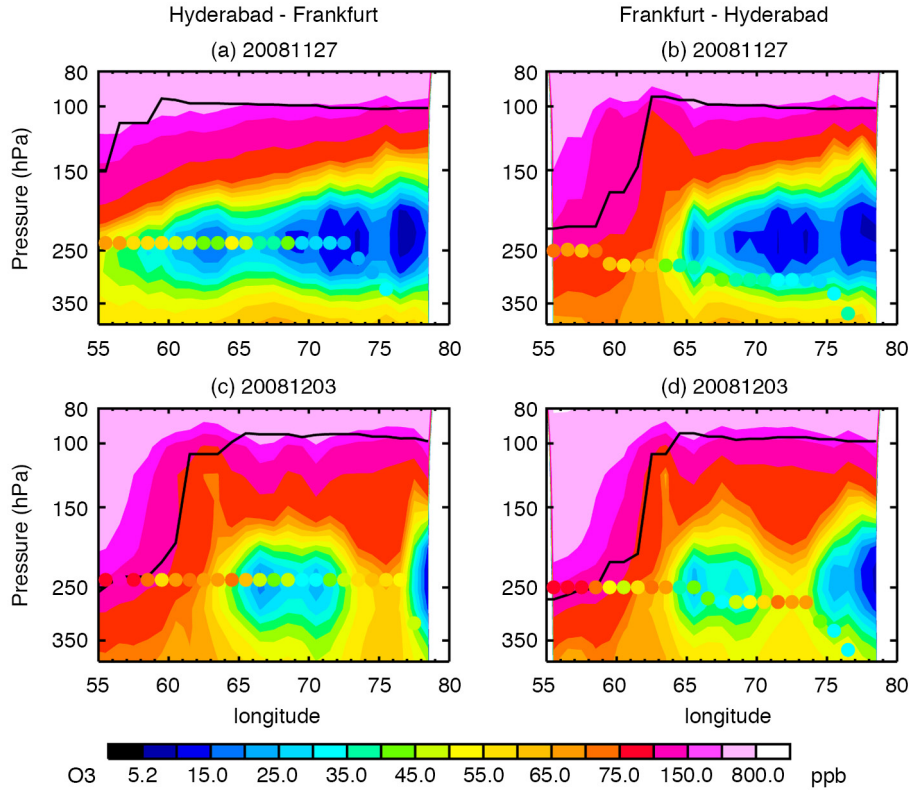


Fig. 3. Longitude–pressure cross-sections of IASI O₃ volume mixing ratios vertical profiles along four MOZAIC flight tracks for (a, b) 27 November, (c, d) 3 December 2008. The IASI data are averaged over (a, b) 26–28 November (c, d) 2–4 December 2008. The coincident ozone MOZAIC observed volume mixing ratios are displayed with the colour-coded circles. The black solid line is the tropopause derived from NCEP data.

Yang and Hoskins, 1996; Homeyer and Bowman, 2013). A climatology of westerly ducts in the Northern Hemisphere based on 20 yr (1980–1999) of meteorological reanalyses by Waugh and Polvani (2000) show an autumn–winter occurrence peak. An analysis of the wind anomalies conducted over 4 months (October 2008–January 2009; not displayed) over the Indian Ocean also shows the same abnormal flow to the NE across the equatorial band. On 26–28 November 2008, the westerly duct lies across the equator at about 50°E (Fig. 2) and encompasses regions from the Arabian Sea in the Northern Hemisphere to the northwest of Madagascar in the Southern Hemisphere. The westerly duct leads to the formation of a strong jet over the equatorial Indian Ocean with a northeastward branch over the Arabian Sea that marks the O3R. It is also important to note that during the O3R period, in the Southern Hemisphere stratospheric air masses remain south of 20°S except some very limited incursions over the Mozambic channel (Fig. 2a) around 27 November and over the centre of Madagascar (Fig. 2b) around 30 November. Deep convective areas corresponding to OLR (Outgoing Longwave Radiation) below 220 W/m² (Park et al., 2007)

are displayed in Fig. 2 in black hatched contours. These convective areas are located over central Africa and the convective outflow will probably impact the air masses transported by the westerly duct. After 4 December, only weak winds remain over the Arabian Peninsula and the westerly duct has disappeared from the Indian Ocean. Extended clusters of deep convective storms are observed over southern India and the Bay of Bengal in Fig. 2a and over the Bay of Bengal in Fig. 2c, d. The first one corresponds to cyclonic storm Nisha (Barret et al., 2011) which formed over Sri Lanka on 26 November and died over central India on 28 November. The second convective cluster is an important deep depression which lasted from 4 to 7 December 2008 over the southern Bay of Bengal within the ITCZ (hereafter BOB08) and propagates westward.

4. Origin of the O3R

4.1. MOZAIC air-mass characterisation

In this section, we use O₃ and CO observations from the MOZAIC programme to better characterise the composition and the possible origin of the air masses during the

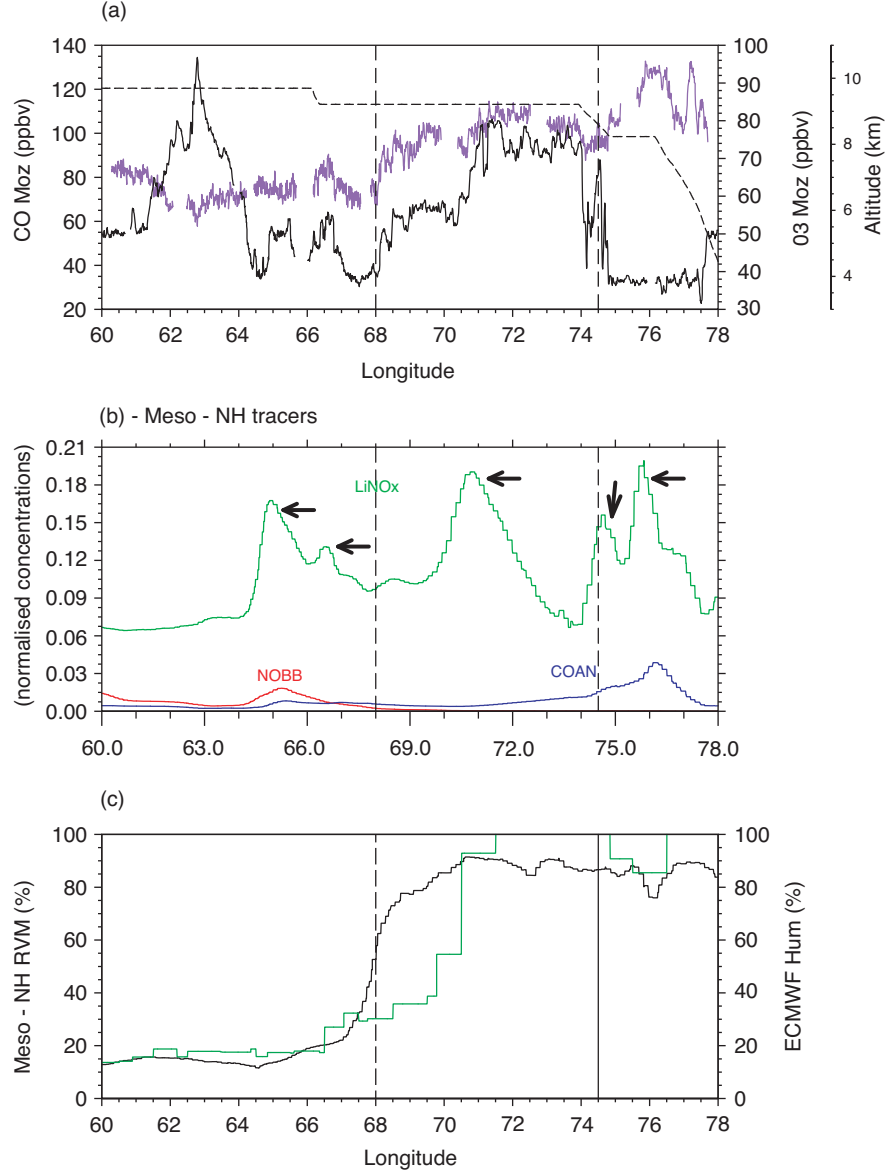


Fig. 4. (a) Longitudinal profiles of MOZAIC O_3 (black line) and CO (violet line) mixing ratios along a Hyderabad Frankfurt flight on 3 December 2008 with the altitude of the aircraft (dashed black line). (b) Meso-NH passive tracers normalised concentrations along the MOZAIC flight at 258 hPa. Concentrations have been normalised relative to their respective maxima over the whole simulation domain. The red curve corresponds to BBTR, the green to LITR and the blue to ANTR. (c) Meso-NH and ECMWF relative humidity along the MOZAIC flight.

O3R event. The O_3 and CO concentrations observed along the MOZAIC HY–FR flight of 3 December are displayed in Fig. 4a. Four distinct air masses are observed. On the southeast side of the flight, CO concentrations reached their maximum values (100–140 ppbv) while the O_3 concentrations remained low (50 ppbv) and stable. This O_3 –CO relationship is typical of fresh pollution that has not yet been photochemically processed (Chin et al., 1994). The sampled polluted air-masses were probably recently uplifted in the UT through convection. Within the O3R (68°–

74.5°E), O_3 and CO are both elevated and highly correlated ($R=0.76$) with an almost linear relationship. The slope $\Delta O_3/\Delta CO = 0.98$ ppbv/ppbv is larger than climatological values (0.20–0.27) computed for the tropical UT during the autumn winter season from CARIBIC data (Zahn et al., 2002). The $\Delta O_3/\Delta CO$ measured within the O3R corresponds to photochemically aged tropospheric air. Further northwestwards (60°–64.5°E near the Northern Hemisphere SWJ) O_3 CO are highly anticorrelated ($R = -0.73$) with an almost linear relationship and a large negative

slope (-1.3 ppbv/ppbv) indicating the import of stratospheric air-masses as the most likely dominant process controlling the O_3 distribution (Zahn et al., 2002; Fadnavis et al., 2010). In between (from 64.5° to 68° E), the air masses are characterised by intermediate values of O_3 and CO typical of the tropical UT with moderate O_3 peaks, whose origins are investigated in Section 4.3.

4.2. Simulation of retroplumes

The analysis of MOZAIC observations showed that the air-masses involved in the O3R region have different chemical signatures which probably correspond to different origins from the midlatitude stratosphere to the marine BL. The aim of the present section is to identify the potential sources of O_3 precursors which are responsible for the O_3 enrichment in the O3R. The IASI observations are used to define three boxes between 9 and 12 km over the Arabian Sea just off the west coast of India (see Figs. 2 and 5). The red box (22° – 26° N, 65° – 67° E) and the green box (10° – 14° N, 71° – 73° E) are located respectively northwestward and southeastward of the O3R and the blue box (16° – 20° N, 68° – 70° E) is located within the O3R (see Fig. 2b). FLEXPART is run following the methodology described in Section 2.3. Figure 5 shows the regions which encompass 70% of the mean residence times within the BL (0–3 km, Fig. 5a) and within the UTLS (9–15 km, Fig. 5b) for the corresponding three boxes.

November–December corresponds to the dry season over the African continent north of the Equator. During this period, BB emissions occur over a large zonal band extending from 5° to 15° N that roughly corresponds to African savannah and forests (Cooke et al., 1996) (Fig. 5a). Anthropogenic emissions are high all over India but more particularly over the Indo Gangetic Plain in Northern India (Fig. 5a). From Fig. 5a, it can be seen that air masses, which reach the three boxes, have spent time in the BL mostly over the northern Indian Ocean region. The contours are shifted in latitude according to the position of the box. It is interesting to note that none of the three BL contours are overlapping the region of African BB emissions that have therefore probably little impact on the observed UT composition. On the contrary, the three contours are overlapping regions of large anthropogenic emissions from the Indian continent. The green box is the most impacted by BL air masses with the 0–3 km layer contributing to 16% of the total residence times compared to 5% for the blue box and 3% for the red one. Convective uplift of these air-masses was probably favoured by the crossing of cyclonic storm Nisha from the Bay of Bengal to central India at the end of November (Barret et al., 2011).

In order to determine the region with LiNOx emissions, we are using the High Resolution Monthly Climatology

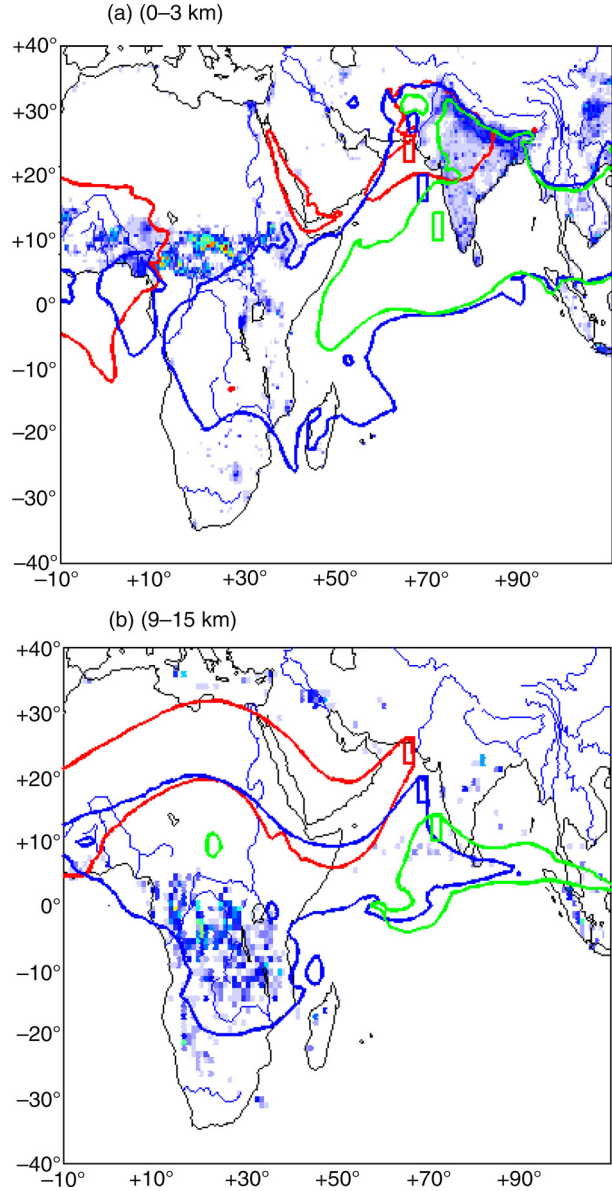


Fig. 5. The colour coded solid contours represent the area encompassing 70% of the FLEXPART total retroplume residence times for particles released between 29 November 12 UT and 2 December 12 UT from the corresponding colour coded boxes (the blue rectangle is within the O3R) (a) residence times within the BL (0–3 km) with monthly GFED3 fire data and monthly MACCity anthropogenic emissions (fluxes are normalised to their respective maxima) and (b) residence times within the UT (9–15 km) with LIS/OTD HRMC flash rates (values given in $f/km^2/yr$ are normalised to their maximum) for November 2008.

(HRMC, <ftp://ghrc.nsstc.nasa.gov>) from the Lightning Imaging Sensor (LIS) and the Optical Transient detector (OTD) onboard the TRMM platform (Christian et al., 2003). The HRMC product is a $0.5^\circ \times 0.5^\circ$ gridded composite of total (Intra-Cloud + Cloud-Ground) lightning bulk production,

expressed as a flash density ($\text{fl}/\text{km}^2/\text{yr}$). In November 2008 most of lightnings occur over central and southern Africa between equator and 15°S corresponding to the rainy season. Air masses reaching the red box are mostly originating from the tropical UT over west and north Africa and the Arabian peninsula (Fig. 5b). They are advected by the strong winds following the Rossby waves south of the tropopause (see Fig. 2 for the location of the tropopause). They are therefore characterised by lower concentrations of O_3 than the lower stratospheric air masses to the north and then the O3R to the south as detected by IASI and MOZAIC (Fig. 2). The green contour, centred over the Indian Ocean at the southern tip of India is probably the signature of cyclonic storm Nisha. The blue contour is the only one to extend southwestward into Africa and overlap with the region where LIS detects most lightnings. Lightnings are thus a distinct source of ozone in the O3R compared to the two bordering regions.

As mentioned in the introduction, previous studies (Zachariasse et al., 2000, 2001; Chatfield et al., 2007) have shown that stratospheric intrusions could play a role in O_3 increase in the mid- and UT over the Indian Ocean. Our Lagrangian simulations provide first evidences to rule out STE as the primary source of O_3 enhancement within the O3R. Indeed, the blue contour displayed in Fig. 5b concerning residence times within the 9–15 km layer is confined within the tropical band (20°S – 20°N) while, as discussed in Section 3.3, at 200 hPa the stratosphere is outside the 20°S – 20°N from 6 November to 7 December period (see the 1.5 PVU limit in Fig. 2 for the O3R period). Furthermore, as discussed above (Section 4.1), MOZAIC data indicate photochemically aged air-masses within the O3R.

FLEXPART simulations point out that African BB emissions have probably little impact upon the O_3 or CO increase over the Arabian Sea during the O3R period. On the contrary, the backtrajectories highlight the potential contribution of LiNOx emissions from southern Africa to the O3R. High CO over the Arabian Sea probably originates from Indian polluted air masses freshly uplifted by cyclonic storm Nisha. To confirm the assumptions on the sources and the transport pathways responsible for the O3R, we have performed an onward simulation with the Eulerian Meso-NH model over a domain encompassing the main sources identified with FLEXPART.

4.3. Tagged simulation at mesoscale

A high-resolution forward simulation was performed with the mesoscale meteorological model Meso-NH from 20 November to 15 December 2008 (see Section 2.4). The domain was chosen in order to represent the O_3 precursors sources identified with the FLEXPART backward simulations. Three passive tracers were implemented to mimic

African BB emissions (BBTR), anthropogenic emissions (ANTR) and emissions from lightning (LITR) (see Section 2.4 for details). In the following, the transport pathways of these tracers are illustrated with concentrations normalised by their respective maximum values on all vertical levels.

Figure 6 shows the contours of the forward normalised plumes for the three passive tracers from 26 November to 7 December 2008 at 210 hPa. Surface emissions (BBTR and ANTR) are uplifted to the UT. The two tracers originating from the African continent (BBTR, LITR) experience a northeastward transport by the southwesterly duct. ANTR higher concentrations are located over the Bay of Bengal and central India south of the O3R. The tracers thus have three quite distinct transport pathways. During the studied period the most intense convective systems occur in the region of central Africa where LIS detected the largest flash frequencies (Fig. 5b). The largest lightning frequencies simulated by Meso-NH (not displayed) are in good agreement with the LIS observations in capturing the central African maximum. Figure 6 shows that, once emitted in the central African UT, the LITR is rapidly transported eastward over the Indian Ocean and northeastward within the southwesterly duct. Throughout the studied period the LITR plume is the one that best fits within the blue contour that corresponds to the O3R (Fig. 6b). The maximum of LITR concentration over the south-western part of the Indian Ocean (Fig. 6) is also in good overlap with the UT O_3 high concentrations observed by IASI in this region (Fig. 2). Nevertheless, after 1 December the LITR recedes faster over the Arabian Sea than the O_3 enhanced concentrations and both remain high above the western equatorial Indian Ocean (Fig. 6c). The NOx lifetime range from 4 d to 1 week in the UT (Brasseur et al., 1999) and we have chosen a longer lifetime (10 d) for our LITR. Tropospheric O_3 has a lifetime of more than 20 d (Stevenson et al., 2006) which makes O_3 produced from LiNOx able to travel on larger distances than LiNOx themselves. This probably explains the inconsistency between the LITR and O_3 enhancements in Fig. 6c. Deep convective systems associated with storm Nisha over India have also produced LITR but east of the O3R observed by IASI (Fig. 6b). The simulated south-western branch of BB transport was described by Edwards et al. (2003). BB emissions from eastern Africa are uplifted to the free troposphere by pyroconvection and advected southwestward by the prevailing Harmattan winds. When they reach the ITCZ, the polluted air masses are lofted on top of the incoming cool ‘monsoon’ flow from the Atlantic and end-up over southern central Africa or over the gulf of Guinea. In our case study, the transport of BB is noteworthy because it is characterised by an UT circulation which then favours a northeastward transport by the southwesterly duct to end up over the Indian continent.

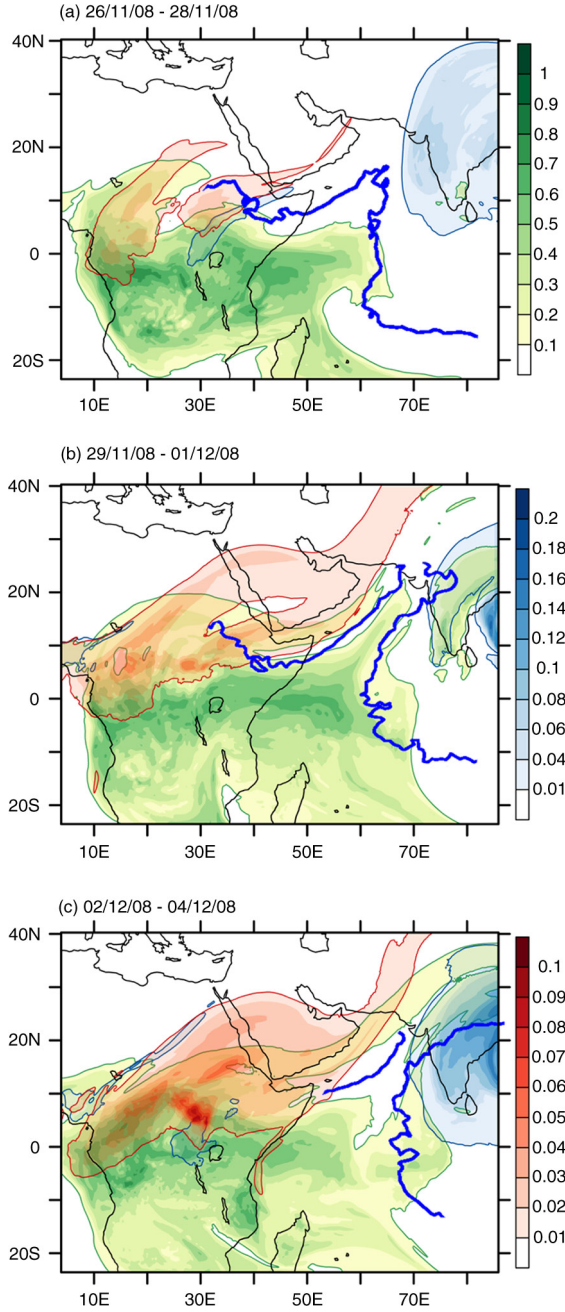


Fig. 6. Meso-NH passive tracers distributions at 210 hPa from 26 November to 4 December 2008 averaged over 3 days. (a) 26–28 November 2008, (b) 29 November to 1 December 2008, (c) 2–4 December 2008. Concentrations were normalised relative to the maxima values over all vertical levels. The red plume corresponds to BBTR, the green one to LITR and the blue one to ANTR. And the blue isocontour corresponds to 60 ppbv O_3 from IASI and delimits the position of the O3R.

The simulated BBTR plume overflows the Asian continent but north of the Arabian Sea which excludes a potential contribution to the O3R.

According to Barret et al. (2011), the cyclonic storm Nisha formed over Sri Lanka on 26 November and crossed the south-east Indian coast in the morning of 27 November and headed northwest.

Figure 7 displays the Brightness Temperature (BT) at $10.8 \mu\text{m}$ as simulated by the Meso-NH model (Fig. 7b) and as observed by the VIRS sensor onboard the TRMM satellite (Barnes et al., 2000) (Fig. 7a). The position of the storm modelled by Meso-NH is in good agreement with that measured by VIRS. Meso-NH slightly overestimates the BT, and therefore underestimates the cloud top. Both model and observations give BT below 220°K within the storm corresponding to a cloud top above 12 km. The cyclonic circulation associated with Nisha (see Fig. 7b) is responsible for the transport of polluted air masses from northern and central India to the centre of the storm located around 80°E and 12°N on 26 November. The vertical transport of the ANTR tracer by storm Nisha is highlighted by Fig. 7c which displays the longitude–pressure cross section of ANTR at 12°N averaged from 26 to 28 November and normalised by its maximum value at this latitude. The ANTR tracer reaches the UT and then spreads westward towards the Arabian Sea where it is transported north-eastward on the southern edge of the southwesterly duct described earlier. The uplift of air-masses from the BL to the UT associated with the storm is responsible for the low O_3 concentrations around the storm (see Fig. 2a with the storm represented by the hatched contour at the southern tip of India) and for the high concentrations of ANTR (see Fig. 6a). Cyclonic storm BOB08 is responsible for very similar vertical transport of Indian BL pollution to the UT in early December. The ANTR transport to the UT by BOB08 is clear in Fig. 6c and its chemical signature is observed by MOZAIC flight of 3 December 2008 with low O_3 and high CO south-east of the O3R (Fig. 4a).

Figure 4b shows the three simulated tracers along the MOZAIC flight track on 3 December. In the following, we analyse the MOZAIC and coincident Meso-NH longitudinal transects from west to east. The large O_3 maximum detected by MOZAIC west of 64°E (Fig. 4b) is associated with low concentrations of the LITR which was expected due to its stratospheric characteristics. From 64° to 75°E , we can identify five relative maxima of the LITR associated with relative O_3 maxima observed by MOZAIC. The four westernmost LITR maxima are associated with negligible ANTR concentrations and are therefore most likely of African origin. The westernmost maximum is even associated with a low maximum of the BBTR indicating the convective lifting of a small fraction of African BB emissions further transported by the southwesterly duct as discussed previously (see Fig. 6c). The two easternmost LITR maxima are noteworthy because they are only separated by about 1.5° but their analysis demonstrates different origins. Firstly,

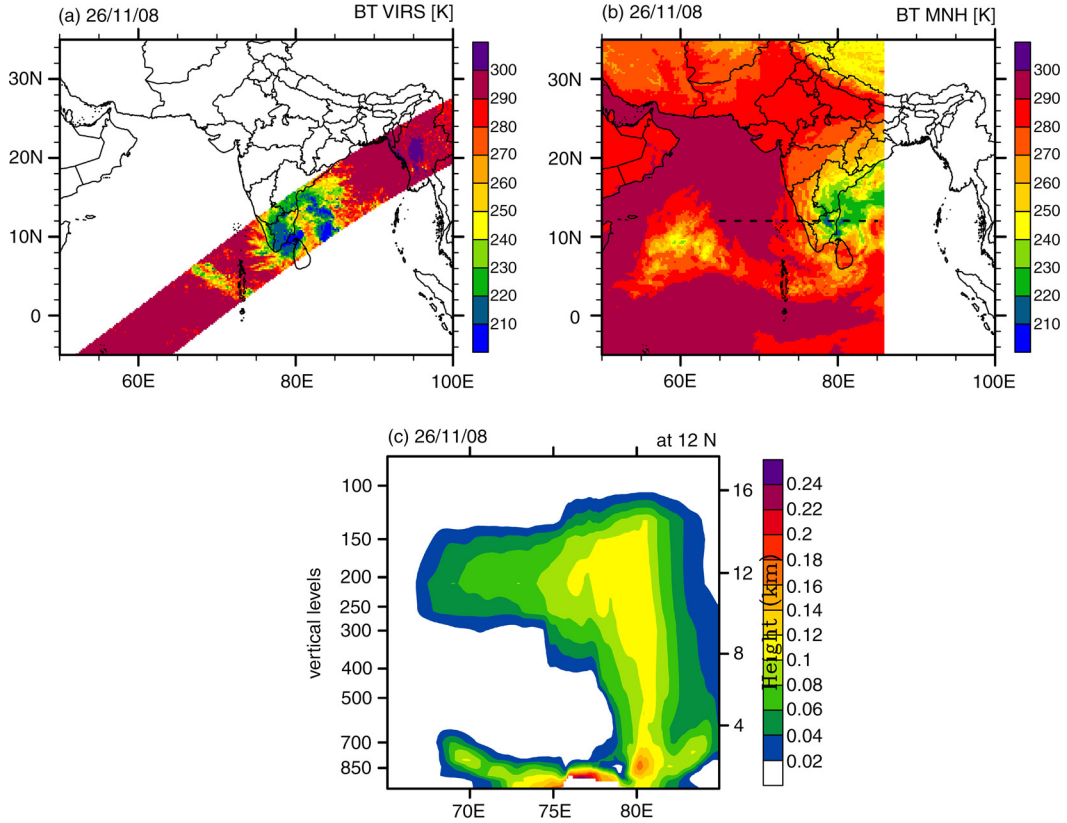


Fig. 7. 26 November 2008 $10.8 \mu\text{m}$ BT ($^{\circ}\text{K}$) (a) measured by TRMM/VIRS and (b) simulated by Meso-NH. (c) Cross-section of Meso-NH simulated anthropogenic tracer ANTR at 12°N with concentrations normalised by their maximum value at this latitude. The corresponding section is represented by a black line on the plot (b).

the maximum at 74.5°E is associated with a sharp O_3 maximum from MOZAIC corresponding to ‘old’ LiNOX from African origin responsible for an important O_3 production. Secondly, the large LITR maximum at 76°E is associated with the lowest MOZAIC O_3 concentrations and both with the maximum of ANTR and of MOZAIC CO concentrations all indicating fresh convective outflow from the BOB08 storm. These model results indicate that elevated O_3 in the O3R is most consistent with African LiNOx. Their analyses have also shown that the Meso-NH simulation was able to explain qualitatively most of the structure of the high-resolution MOZAIC O_3 and CO transect.

5. Discussion

As shown in the previous sections, MOZAIC observations, FLEXPART Lagrangian and Meso-NH tagged simulations tend to indicate African LiNOX as the most important source to build up the O3R. Nevertheless, most INDOEX studies (Zachariasse et al., 2000, 2001; Chatfield et al., 2007) related O_3 UT enhancements over the Arabian sea to STE. Because of the southwesterly duct originating around the equator, our case seems most similar to the cases of inter-

hemispheric STE discussed by Zachariasse et al. (2001). The first point that contradicts the hypothesis of STE for the O3R is that during the period from 6 to 25 November, the 1.5 PVU contour is confined south of 20°S with some limited incursions of stratospheric air masses around Madagascar afterwards. FLEXPART 20d retroplume simulations show that UT air-masses reaching the O3R are coming from the tropics, between 20°S and 20°N and mostly from the African continent (Fig. 5b). Nevertheless it is not possible to completely rule out the fact that part of the stratospheric air-masses that reached Madagascar on two occasions could be entrained into the southwesterly duct and marginally participate to the O3R. This is very different from the main case of interhemispheric STE documented by Zachariasse et al. (2001) who state: ‘some trajectories [...] move from the STJ at roughly 35°S to as far north as 10°N ’. Moreover, in their case there is a continuous ‘flow regime from the Southern to the Northern Hemisphere along Madagascar and the east coast of Africa’. In the case of the O3R, there is a strong westerly duct across the Equator which prevent any interhemispheric transport over the Indian Ocean such as described in Zachariasse et al. (2001) (see Fig. 2a–c).

Furthermore, the tagged Meso-NH simulations clearly indicate a strong correlation between LITR and O_3 maxima with some limitations linked to the different lifetimes as discussed above. Finally, water vapour can be used as another indicator to discriminate stratospheric and tropospheric air masses. Unfortunately, the MOZAIC water vapour sensor was out of order during the flights that document the O3R. Nevertheless, we have access to the relative humidity from both Meso-NH and ECMWF whose values are plotted along the MOZAIC flight track of 3 December in Fig. 4c. Relative Humidity from Meso-NH is below 20% west of 66°E, sharply increases between 66°E and 68°E and remains higher than 80% east of 68°E. ECMWF relative humidity has a very similar behaviour with lower values between 68°E and 70°E. Relative humidity from both Meso-NH and ECMWF is therefore higher than 80% within most of the O3R which is another evidence against the inter-hemispheric STE. Stratospheric air masses are indeed very dry and in Zachariasse et al. (2000, 2001) STE events over the INDOEX domain are always associated with air masses with relative humidity below 20%.

Even if, according to what has been discussed above, STE cannot be the main source of O_3 enrichment in the O3R, it is necessary to discuss the ability of LiNOx to produce the observed enhancements of O_3 . According to IASI retrievals, the O_3 mixing ratios reach 70 ppbv within the O3R and are below 30 ppbv on both sides of the O3R (2). MOZAIC measurements give mixing ratios of about 40 ppbv around the O3R and of 70–75 ppbv within the O3R (Fig. 4a) which is in qualitative agreement with IASI. The underestimation of IASI relative to MOZAIC on both sides of the O3R is related to a retrieval problem documented in Dufour et al. (2012). They show that tropical S-shape O_3 profiles retrieved by IASI are characterised by too low O_3 at the level of the UT minimum. We can therefore give 30 ppbv as a reasonable value for the O_3 enrichment within the O3R. The level of O_3 enrichments produced by LiNOx has been largely documented in previous studies led by one of the authors of the present study. Firstly, MOZAIC observations have shown that during boreal autumn, the African UT is characterised by a large meridional gradient of O_3 with values of 40–50 ppbv over the north and 70–80 ppbv over the south impacted by convective activity (Sauvage et al., 2006). Based on the same MOZAIC observations, Sauvage et al. (2007a) have roughly estimated that LiNOx are responsible for the production of 1.6 ppbv O_3 per day in the African UT during the monsoon season. Therefore, from 40 to 50 ppbv background O_3 it is possible to reach 70–80 ppbv in 20 d from LiNOx production. Finally, based on GEOS-Chem simulations Sauvage et al. (2007b) have shown that the meridional O_3 gradient over Africa and the South Atlantic was mainly driven by LiNOx which contribution can reach 30 ppbv in the

middle and upper troposphere of southern tropical Africa. Therefore, LiNOx is a perfectly realistic source for the 30 ppbv O_3 enrichment observed within the O3R.

6. Conclusions

With its high spatio temporal resolution, IASI enables studying UT transport and chemistry processes at a day to weekly time scale. In the present study, we have focused on a case of O_3 enriched air masses flowing from Africa to northern India during the 2008 post-monsoon season, the O3R. This transport event is related to Rossby waves propagating into the tropics and leading to the formation of an equatorial south-westerly duct over the Arabian Sea. The MOZAIC airborne observations have confirmed the crossing of the O3R over the northwestern coast of India during our period of analysis. Moreover, MOZAIC *in-situ* CO observations enabled us to determine that air-masses involved in the O3R were of tropospheric rather than stratospheric origin. Lagrangian simulations with the FLEXPART model were used to identify the regions of the atmosphere through which air masses reaching the O3R had recently passed. They show that air masses that fed the O3R mostly originate from the tropical African UT. In order to have a more accurate determination of the sources and transport pathways of the emissions we have performed meso-scale high-resolution simulations with the Meso-NH model. The simulation has been set-up over a large domain encompassing the source regions identified with FLEXPART and with passive tracers implemented for the O_3 precursors sources anticipated with the Lagrangian analyses. The Meso-NH simulations have shown that the elevated O_3 values within the O3R over the Arabian Sea are mostly resulting from the LiNOx emissions from central Africa and subsequent transport within the south-westerly duct. The arguments described previously together with the high relative humidity documented by both Meso-NH simulations and ECMWF reanalyses rule out STE as a large contributor to the O3R. This transport pathway therefore complement the previous studies by Chatfield et al. (2007), Zachariasse et al. (2000, 2001) that have mostly emphasised STE as a source of enhanced UT O_3 in this region during the winter season. African BB has been shown to play a negligible role in the formation of the O3R and this case therefore does not support the hypothesis of De Laat (2002). On the north-eastern edge of the O3R, we have shown that the UT composition is impacted by Indian pollution that has been uplifted by the tropical storms Nisha and BOB08 resulting in the low (resp. high) O_3 (resp. CO) concentrations observed by MOZAIC. This particular transport pathway agrees with the results of Chatfield et al. (2007) who have identified the uplift of Indian pollution as one of the sources for enhanced mid- to upper- tropospheric O_3 over the Arabian sea.

7. Acknowledgements

This subject was part of the Flore TOCQUER’s PhD thesis, cofinanced by CNES and Météo–France. IASI, developed and built under the responsibility of the Center National d’Etudes Spatiales (CNES, France), is flown onboard the MetOp satellites as part of the EUMETSAT Polar System. IASI L1c and L2–EUMETSAT data have been downloaded from the Ether French atmospheric database (<http://ether.ipsl.jussieu.fr>). The research concerning IASI is conducted with the financial support of CNES (TOSCA–IASI project). MOZAIC is presently funded by INSU–CNRS, Météo–France, and FZJ (Forschungszentrum Julich, Germany). Interpolated OLR data are provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their website at <http://www.cdc.noaa.gov>. Meso-NH model jointly developed by the Laboratoire d’Aérodynamique (UMR 5560 UPS/CNRS) and by CNRM–GAME (URA 1357 CNRS/Météo–France). Computer time has been provided by the scientific group Calculation in Midi-Pyrenees [CALMIP (grant P12171)].

References

- Barnes, W., Lyu, C. and Gales, M. 2000. An overview of the visible and infrared scanner radiometric calibration algorithm. *J. Atmos. Oceanic Technol.* **17**, 395–405.
- Barret, B., Le Flochmoën, E., Sauvage, B., Pavelin, E., Matricardi, M. and co-authors. 2011. The detection of post–monsoon tropospheric ozone variability over south Asia using IASI data. *Atmos. Chem. Phys.* **11**, 9533–9548.
- Barret, B., Williams, J. E., Bouarar, I., Yang, X., Josse, B. and co-authors. 2010. Impact of West African Monsoon convective transport and lightning NO_x production upon the upper tropospheric composition: a multi–model study. *Atmos. Chem. Phys.* **10**, 5719–5738.
- Bechtold, P., Bazile, E., Guichard, F., Mascart, P. and Richard, E. 2001. A mass–flux convection scheme for regional and global models. *Q. J. Roy. Meteorol. Soc.* **127**, 869–886.
- Bond, D. W., Steiger, S., Zhang, R., Tie, X. and Orville, R. E. 2002. The importance of NO_x production by lightning in the tropics. *Atmos. Environ.* **36**, 1509–1519.
- Brasseur, G., Orlando, J. and Tyndall, G. 1999. *Atmospheric Chemistry and Global Change*. Topics in Environmental Chemistry. Oxford University Press, ISBN 0-19-510521-4.
- Chaboureaud, J.-P. and Bechtold, P. 2002. A simple cloud parameterization derived from cloud resolving model data: diagnostic and prognostic applications. *J. Atmos. Sci.* **59**, 2362–2372.
- Chatfield, R. B., Guan, H., Thompson, A. M. and Smit, H. G. J. 2007. Mechanisms for the intraseasonal variability of tropospheric ozone over the Indian Ocean during the winter monsoon. *J. Geophys. Res.* **112**, D10303.
- Chatfield, R. B., Guan, H., Thompson, A. M. and Witte, J. C. 2004. Convective lofting links Indian Ocean air pollution to paradoxical South Atlantic ozone maxima. *Geophys. Res. Lett.* **31**, L06103.
- Chin, M., Jacob, D. J., Munger, J. W., Parrish, D. D. and Doddridge, B. G. 1994. Relationship of ozone and carbon monoxide over North America. *J. Geophys. Res.* **99**, 14565–14573.
- Christian, H. J., Blakeslee, R. J., Boccippio, D. J., Boeck, W. L., Buechler, D. E. and co-authors. 2003. Global frequency and distribution of lightning as observed from space by the Optical Transient Detector. *J. Geophys. Res.* **108**, 4005.
- Cooke, W. F., Koffi, B. and Grégoire, J.-M. 1996. Seasonality of vegetation fires in Africa from remote sensing data and application to a global chemistry model. *J. Geophys. Res.* **101**, 21051–21065.
- De Laat, A. T. J. 2002. On the origin of tropospheric O₃ over the Indian Ocean during the winter monsoon: African biomass burning vs. stratosphere–troposphere exchange. *Atmos. Chem. Phys.* **2**, 325–341.
- De Laat, A. T. J., Zachariasse, M., Roelofs, G. J., Van Velthoven, P., Dickerson, R. R. and co-authors. 1999. Tropospheric O₃ distribution over the Indian Ocean during spring 1995 evaluated with a chemistry–climate model. *J. Geophys. Res.* **104**, 13881–13893.
- De Wachter, E., Barret, B., Le Flochmoën, E., Pavelin, E., Matricardi, M. and co-authors. 2012. Retrieval of MetOp–A/IASI CO profiles and validation with MOZAIC data. *Atmos. Meas. Tech.* **5**, 2843–2857.
- Dufour, G., Eremenko, M., Griesfeller, A., Barret, B., Le Flochmoën, E. and co-authors. 2012. Validation of three different scientific ozone products retrieved from IASI spectra using ozonesondes. *Atmos. Meas. Tech.* **5**, 611–630.
- Edwards, D. P., Lamarque, J. F., Attie, J. L., Emmons, L. K., Richter, A. and co-authors. 2003. Tropospheric ozone over the tropical Atlantic: a satellite perspective. *Atmos. Chem. Phys.* **108**, 4237.
- Fadnavis, S., Chakraborty, T. and Beig, G. 2010. Seasonal stratospheric intrusion of ozone in the upper troposphere over India. *Ann. Geophys.* **28**, 2149–2159.
- Fishman, J., Watson, C. E., Larsen, J. C. and Logan, J. A. 1990. Distribution of tropospheric ozone determined from satellite data. *J. Geophys. Res.* **95**, 3599–3617.
- George, M., Clerbaux, C., Hurtmans, D., Turquety, S., Coheur, P.-F. and co-authors. 2009. Carbon monoxide distributions from the IASI/METOP mission: evaluation with other space–borne remote sensors. *Atmos. Chem. Phys.* **9**, 8317–8330.
- Giglio, L., Randerson, J. T., van der Werf, G. R., Kasibhatla, P. S., Collatz, G. J. and co-authors. 2010. Assessing variability and long-term trends in burned area by merging multiple satellite fire products. *Biogeosciences*. **7**, 1171–1186.
- Granier, C., Bessagnet, B., Bond, T., D’Angiola, A., van der Gon, H. D. and co-authors. 2011. Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period. *Clim. Change*. **109**, 163–190.
- Gregory, D., Morcrette, J. J., Jakob, C., Beljaars, A. C. M. and Stockdale, T. 2000. Revision of convection, radiation and cloud schemes in the ECMWF Integrated Forecasting System. *Q. J. Roy. Meteorol. Soc.* **126**, 1685–1710.
- Homeyer, C. R. and Bowman, K. P. 2013. Rossby wave breaking and transport between the tropics and extratropics above the subtropical jet. *J. Atmos. Sci.* **70**, 607–626.

- Labrador, L. J., von Kuhlmann, R. and Lawrence, M. G. 2005. The effects of lightning-produced NO_x and its vertical distribution on atmospheric chemistry: sensitivity simulations with MATCH-MPIC. *Atmos. Chem. Phys.* **5**, 1815–1834.
- Lafore, J. P., Stein, J., Asencio, N., Bougeault, P., Ducrocq, V. and co-authors. 1998. The Meso-NH atmospheric simulation system. Part I: adiabatic formulation and control simulations. *Ann. Geophys. Atmos. Hydrosph. Space Sci.* **16**, 90–109.
- Lawrence, M. G. and Lelieveld, J. 2010. Atmospheric pollutant outflow from southern Asia: a review. *Atmos. Chem. Phys.* **10**, 11017–11096.
- Lawrence, M. G., von Kuhlman, R. and Salzmann, M. 2003. The balance of effects of deep convective mixing on tropospheric ozone. *Geophys. Res. Lett.* **30**, 1940. DOI: 10.1029/2003GL017644.
- Marenco, A., Thouret, V., Nedelec, P., Smit, H., Helten, M. and co-authors. 1998. Measurement of ozone and water vapour by Airbus in-service aircraft: the MOZAIC airborne program, an overview. *J. Geophys. Res.* **103**, 25631–25642.
- Mari, C., Cailley, G., Corre, L., Saunois, M., Attié, J. L. and co-authors. 2008. Tracing biomass burning plumes from the Southern Hemisphere during the AMMA 2006 wet season experiment. *Atmos. Chem. Phys.* **8**, 3951–3961.
- Masson, V., Le Moigne, P., Martin, E., Faroux, S., Alias, A. and co-authors. 2012. The SURFEXv7.2 land and ocean surface platform for coupled or offline simulation of Earth surface variables and fluxes. *Geosci. Model Dev. Discuss.* **5**, 3771–3851.
- Nedelec, P., Cammas, J.-P., Thouret, V., Athier, G., Cousin, J. M. and co-authors. 2003. An improved infrared carbon monoxide analyser for routine measurements aboard commercial Airbus aircraft: technical validation and first scientific results of the MOZAIC III programme. *Atmos. Chem. Phys.* **3**, 1551–1564.
- Park, M., Randel, W. J., Gettelman, A., Massie, S. T. and Jiang, J. H. 2007. Transport above the Asian summer monsoon anticyclone inferred from Aura Microwave Limb Sounder tracers. *J. Geophys. Res.* **112**, D16309.
- Pergaud, J., Masson, V., Malardel, S. and Couvreux, F. 2009. A parameterization of dry thermals and shallow cumuli for meso-scale numerical weather prediction. *Bound. Layer Meteorol.* **132**, 83–106.
- Pinty, J.-P. and Jabouille, P. 1998. A mixed-phase cloud parameterization for use in mesoscale non-hydrostatic model: simulations of a squall line and of orographic precipitations. In: *Proceedings of Conference of Cloud Physics*, Everett, WA, USA, American Meteorological Society, 132, 217–220.
- Sauvage, B., Martin, R. V., van Donkelaar, A. and Ziemke, J. R. 2007a. Quantification of the factors controlling tropical tropospheric ozone and the South Atlantic maximum. *J. Geophys. Res.* **112**, D11309.
- Sauvage, B., Thouret, V., Cammas, J. P., Brioude, J., Nedelec, P. and co-authors. 2007b. Meridional ozone gradients in the African upper troposphere. *Geophys. Res. Lett.* **34**, L03817.
- Sauvage, B., Thouret, V., Thompson, A. M., Witte, J. C., Cammas, J.-P. and co-authors. 2006. Enhanced view of the ‘tropical Atlantic ozone paradox’ and ‘zonal wave one’ from the *in situ* MOZAIC and SHADOZ data. *J. Geophys. Res.* **111**, D01301.
- Shindell, D. T., Lamarque, J.-F., Schulz, M., Flanner, M., Jiao, C. and co-authors. 2013. Radiative forcing in the ACCMIP historical and future climate simulations. *Atmos. Chem. Phys.* **13**, 2939–2974.
- Stevenson, D. S., Dentener, F. J., Schulz, M. G., Ellingsen, K., van Noije, T. P. C. and co-authors. 2006. Multimodel ensemble simulations of present day and near-future tropospheric ozone. *J. Geophys. Res.* **111**, D08301.
- Stohl, A., Forster, C., Frank, A., Seibert, P. and Wotawa, G. 2005. Technical note: the Lagrangian particle dispersion model FLEXPART version 6.2. *Atmos. Chem. Phys.* **5**, 2461–2474.
- Stohl, A., Hittenberger, M. and Wotawa, G. 1998. Validation of the Lagrangian particle dispersion model FLEXPART against large-scale tracer experiment data. *Atmos. Environ.* **32**, 4245–4264.
- Stohl, A. and Thomson, D. J. 1999. A density correction for Lagrangian particle dispersion models. *Bound. Layer Meteorol.* **90**, 155–167.
- Thompson, A. M., Witte, J. C., Oltmans, S. L., Schmidlin, F. J., Logan, J. A. and co-authors. 2003. Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology – tropospheric variability and the zonal wave-one. *Atmos. Chem. Phys.* **108**, 8241.
- Thouret, V., Marenco, A., Logan, J. A., Nedelec, P. and Grouhel, C. 1998. Comparisons of ozone measurements from the MOZAIC airborne program and the ozone sounding network at eight locations. *J. Geophys. Res.* **103**, 25695–25720.
- Van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M. and co-authors. 2010. Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009). *Atmos. Chem. Phys.* **10**, 11707–11735.
- Waugh, D. W. and Polvani, L. M. 2000. Climatology of intrusions into the tropical upper troposphere. *Geophys. Res. Lett.* **27**, 3857–3860.
- Webster, P. J. and Holton, J. R. 1982. Cross-equatorial response to middle-latitude forcing in a zonally varying basic state. *J. Atmos. Sci.* **39**, 722–733.
- Yang, G.-Y. and Hoskins, B. J. 1996. Propagation of Rossby waves of nonzero frequency. *J. Atmos. Sci.* **53**, 2365–2378.
- Zachariasse, M., Smit, H. G. J., Van Velthoven, P. F. J. and Kelder, H. 2001. Cross-tropopause and interhemispheric transports into the tropical free troposphere over the Indian Ocean: Indian Ocean experiment. *J. Geophys. Res.* **106**, 28441–28452.
- Zachariasse, M., van Velthoven, P. F. J., Smit, H. G. J., Lelieveld, J., Mandal, T. K. and co-authors. 2000. Influence of stratosphere–troposphere exchange on tropospheric ozone over the tropical Indian Ocean during the winter monsoon. *J. Geophys. Res.* **105**, 15403–15416.
- Zahn, A., Brenninkmeijer, C. M., Asman, W. H., Crutzen, P. J., Heinrich, G. and co-authors. 2002. Budgets of O₃ and CO in the upper troposphere: CARIBIC passenger aircraft results 1997–2001. *J. Geophys. Res.* **107**, 4337.
- Zhang, L., Li, Q. B., Jin, J., Liu, H., Livesey, N. and co-authors. 2011. Impacts of 2006 Indonesian fires and dynamics on tropical upper tropospheric carbon monoxide and ozone. *Atmos. Chem. Phys.* **11**, 10929–10946.
- Zhang, L., Li, Q. B., Murray, L. T., Luo, M., Liu, H. and co-authors. 2012. A tropospheric ozone maximum over the equatorial Southern Indian Ocean. *Atmos. Chem. Phys.* **12**, 4279–4296.