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Impact of deep convection on the tropical tropopause layer composition in Equatorial Brazil

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Abstract

This paper documents measurements of carbon monoxide (CO), ozone (O_3) and temperature in the tropical tropopause layer over Equatorial Brazil for the first time. These measurements were sampled by the balloon-borne instrument SPIRALE (Spectroscopie Infa-Rouge par Absorption de Lasers Embarqués) in June 2005 and in June 2008, both at the transition period from wet to dry season. The height of the Tropical Tropopause Layer (TTL) top and bottom determined from the chemical species profiles are similar for the two flights. Nevertheless the measured profiles of ozone and CO are different in their volume mixing ratio and shape. The larger CO values measured in the TTL in 2005 can be linked to a more intense biomass burning activity in 2005 than in 2008. We also show that both measured profiles are influenced by convection but in different ways leading to different shapes. The CO profile in 2005 is characterised by a generally smooth decrease in the TTL from tropospheric to stratospheric conditions, except for two layers of enhanced CO around 14.2 (>100 parts per billion by volume = ppbv) and 16.3 km altitude (>85 ppbv). Backward trajectories indicate that these layers come from the vertical transport by remote deep convection occurring 2 and 3 days prior to the flight, respectively. This shows that the transition period from wet to dry season is favourable for the transport of significant amounts of CO in the TTL, sometimes above the level of zero radiative heating, because of increasing biomass burning together with decaying but still important convective activity. In 2008 we focus our analysis on a 1 km deep layer, between 17 and 18 km, where both the temperature and the ozone profiles are uniform in the vertical, corresponding to a layer of well-mixed air. We show that this unusual behaviour is indirectly related to the interaction between convection and the Quasi-Biennial Oscillation (QBO), through vertically propagating gravity waves. Quasi-stationary gravity waves are likely to be produced by convective systems and certainly break in the intense wind shear that imposes the QBO at these altitudes. This conclusion is supported by the fact that the 16–18 km layer is devoid of ice particles (hence the mixing is not convective) and from backward trajectories that

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point towards a convective region as the origin of the air masses in this layer.

1 Introduction

The tropical tropopause layer (TTL) is the transition layer between the troposphere and the stratosphere in the tropics. The TTL has been the focus of many studies, particularly in the past 10 years (e.g. review by Fueglistaler et al., 2009), because it is the main pathway for the entry in the stratosphere of tropospheric sources of chemical species and aerosols. Therefore the understanding of its composition and the quantification of transport processes into and from the TTL are necessary steps for stratospheric ozone prediction at the global scale. Deep convection is a key process affecting tropospheric and stratospheric air composition in the tropics since it can lift rapidly boundary layer tropospheric emissions (e.g. Wang et al. 1996, Marécal et al., 2006). Although the main outflow of tropical convection generally reaches 12–13 km altitude with subsequent radiative cooling driving air downwards, tropical deep convection is also observed above, i.e. in the TTL. If sources reach levels above the zero net radiative heating level (typically 15.5 km or 360 K) they can then slowly rise radiatively in the lower stratosphere.

In this context, several field campaigns aiming at documenting the TTL were conducted at different locations mainly during the convective season: APE-THESEO in western Indian Ocean, NASA Pre-AVE, CR-AVE and TC-4 aircraft campaigns in eastern tropical Pacific, HIBISCUS and TROCCINOX balloon and aircraft campaigns in South Brazil, STEP and SCOUT-O3/ACTIVE aircraft campaigns in the Maritime Continent and AMMA/SCOUT-AMMA aircraft and balloon campaign in West Africa. The analysis of these campaign measurements and the associated modelling studies provided an improved knowledge on the processes affecting the TTL gas composition (e.g. Marcy et al., 2007; Konopka et al., 2007; Heyes et al., 2009; Palazzi et al., 2009; Homan et al., 2010).

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It was shown that the TTL characteristics vary geographically (Viciani et al., 2008; Homan et al., 2010) and with the season (Heyes et al., 2009). This variability can be related to the origin of the sampled air masses (clean or polluted) and to the processes undergone by these air masses until they reach the TTL, mainly deep convection, large scale isentropic or radiative transports, chemical processing for reactive species and scavenging for soluble species and aqueous chemistry. The signature of the detrainment by deep convection at the cloud top can be identified locally on measurements by an enhancement of tropospheric tracer mixing ratios compared to background levels and by lower ozone (Marcy et al., 2007; Viciani et al., 2008; Law et al., 2010). In conjunction with the convective transport, the TTL composition is linked to the local and regional sources of chemical species (Schoeberl et al., 2006; Heyes et al., 2009). In the tropics, biomass burning is a major source of pollutants in the boundary layer taking place over continental areas, preferentially during the dry season (Crutzen and Andreae 1990, Duncan et al., 2003). Megacities are additional important local sources of pollutants. In this respect the South American continent is an interesting region because of its large biomass burning activity, its megacities and its high reaching convection (Liu and Zipser, 2005). The transition from wet to dry season is favourable for transport of significant amounts of chemical species from the low troposphere into the TTL since it is characterised at the regional scale by an increasing biomass burning activity with time and by a decreasing but still significant convective activity. This is one of the reasons why this transition period was chosen for the two large balloon campaigns conducted from Teresina (5.1° S, 42.9° W) in North-east Brazil in June 2005 in the frame of the ENVISAT satellite validation and in June 2008 in the frame of the SCOUT-O3 FP7 European Commission integrated project. Here we analyse the measurements of temperature, CO and ozone gathered in the TTL by the SPIRALE (Spectroscopie Infa-Rouge par Absorption de Lasers Embarqués) in situ instrument (Moreau et al., 2005) along the balloon trajectory during these two campaigns. The general objective of this study is to analyse from these measurement the combined effect of deep convection and biomass burning emissions on the TTL composition during the transition from the

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wet to the dry season. More specifically, we analyse here the characteristics of the TTL from temperature, CO and O_3 SPIRALE profiles and we determine the processes responsible for the different features observed in the TTL in relation with deep convection activity and with biomass burning emissions.

The paper is organised as follows. The SPIRALE instruments and the Teresina flights are described in Sect. 2. The environmental conditions (convective activity and sources of CO) of the flights are discussed in Sect. 3. The SPIRALE balloon-borne measurements are shown and analysed in Sect. 4. Conclusions are given in Sect. 5.

2 Description of the SPIRALE instrument and flights

SPIRALE is a tunable diode laser spectrometer devoted to in situ measurements of trace gas species from the upper troposphere (~12 km height) to the middle stratosphere (~35 km height). To date, SPIRALE has performed eight successful flights at different latitudes from Kiruna (Sweden, 67.9° N), Gap (France, 44.6° N), Aire sur l'Adour (France, 43.7° N) and Teresina (Brazil, 5.1° S).

A detailed description of the instrument can be found in Moreau et al. (2005). To summarise, six laser diodes connected to a multipass Herriott cell (~3.5 m length) fully open to the atmosphere are used as light sources in the mid-infrared region (3-8 μm). Each tunable diode laser emits in a specific micro-window including the ro-vibrational line of the species of interest. The absorption spectra micro-domains for ozone and carbon monoxide were 2085.93–2086.63 and 2123.40–2123.95 cm^{-1} for the 2005 and 2008 flights, respectively.

Laser absorption takes place between two mirrors of the optical cell located at the extremities of a deployable mast below the gondola, enabling simultaneous measurements of several chemical species. The very high spectral resolution (~0.0005–0.001 cm^{-1}) and the long optical path (300.31 m in June 2005 and 430.78 m in June 2008) allow measurements of trace gas concentration with a good sensitivity. The very high vertical resolution (~5 m) was achieved thanks to the low vertical velocity of the

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gondola and the rapid measurement acquisition system (within 1.1 s).

Species concentrations are retrieved from direct infrared absorption, by fitting experimental spectra with spectra calculated using the HITRAN 2008 database (Rothman et al., 2009) and the temperature and pressure measured onboard the gondola. Measurements of pressure (by two calibrated and temperature-regulated capacitance manometers) and temperature (by two probes made of resistive platinum wire) also allow for converting the species concentrations in volume mixing ratios (vmr).

An assessment of the error sources on the retrieved vmr has been reported in Moreau et al. (2005). In brief, uncertainties in the pressure and temperature parameters have been evaluated to be negligible relative to other uncertainties discussed below. The overall uncertainties take into account the random and systematic errors, combined as the square root of their quadratic sum. The two important sources of random errors are the fluctuations of the laser background emission signal and the signal-to-noise ratio. At lower altitudes, these are the main contributions to overall uncertainties. Systematic errors originate essentially from the laser linewidth (an intrinsic characteristic of the laser diode), which contributes more at lower pressure (higher altitudes) than at higher pressures. For CO, the overall uncertainties are 6±1% (1σ) below 17 km and increase up to around 17±6% (1σ) above 17 km. For O_3, the overall uncertainties decrease significantly from 50% to 9% between 13 and 17 km, and down to 6±2% above 17 km.

The SPIRALE balloon-borne instrument was flown on 22 June 2005 and during the night of 9 to 10 June 2008. During the first flight, the measurements started at 09:32 UTC (i.e. 06:32 local time). The balloon reached the maximum altitude of 33.2 km (7.4 hPa) at 11:01 UTC. The descent phase started at 11:47 UTC (33.2 km) and the measurements ended at 14:28 UTC (i.e. 11:28 local time) at 15.1 km (122 hPa). For the second flight, the measurements started at 23:30 UTC (i.e. 20:30 local time) and reached the maximum altitude of 33.6 km (6.9 hPa) at 01:14 UTC (i.e. 22:14 local time). The descent phase started at 01:52 UTC (i.e. 22:52 local time) at 33.7 km and the measurements ended at 05:35 UTC (i.e. 02:35 local time), at 16.9 km (95 hPa). For both

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flights, retrievals of mixing ratios have been performed using ascent and descent. Only the ascent profiles are presented here since they cover the whole TTL. The descent profiles have been used as a check of the consistency of the measurements.

For both flights, an ozone sonde (Vaisala Radiosonde RS92 type, www.vaisala.com) and an aerosol particle counter (Renard et al., 2008) were flown as piggy backs on the SPIRALE instrument. The ozone sonde data are usually used to check the consistency of the SPIRALE ozone retrieval. For 2005 comparisons between the SPIRALE and ozone sonde measurements showed an excellent agreement (<5%). Unfortunately the ozone sonde did not work properly during the 2008 flight. The analysis of the measurements of the aerosol particle counter does not show the presence of ice particles along the balloon track for both flights. This means that the SPIRALE measurements were performed in cloud-free conditions.

3 Environmental conditions of the flights

3.1 Convective activity

Both flights were performed in June, a time period that corresponds climatologically to the end of the wet season. In June, convective activity still occurs in the Teresina region and therefore the measurements performed in the TTL by SPIRALE might be affected by local convection. The surface rainfall rate from TRMM (Tropical Rainfall Measuring Mission, http://trmm.gsfc.nasa.gov/) accumulated over the 24 h preceding approximately each flight is displayed in Fig. 1a and c. The dataset used is a 3-hourly average with a 0.25° by 0.25° resolution and was produced by the 3B42 algorithm (Huffman et al., 2007). For the 2005 flight, surface precipitation is sparse in North-east Brazil. The surface rainrate accumulated 24 h prior to 2008 flight shows a different picture with intense convection mainly North of Teresina.

SPIRALE measurements in the TTL might have been affected not only by local convection but also by remote convection occurring several days before the flights.

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Figure 1b and d show the surface rainrates accumulated over the 10 days preceding each flight. The signature of the ITCZ (Inter Tropical Convergence Zone) can be identified as the main band of intense precipitation. It is wider and located more to the south in 2008 than in 2005. This can be partly explained by the fact that the 2008 flight was on 9–10 June while the 2005 flight was on 22 June. Earlier in the month of June the ITCZ is expected to have a more southerly location. This is also explained by the fact that the 2005 and 2008 years show a clear shift of the ITCZ in June. This is illustrated by Fig. 2 showing the 2.5° by 2.5° surface precipitation anomaly for June compared to climatology (1979–2008) from GPCP (Global Precipitation Climatology Project) Version 2.1 Combination product (Adler et al., 2003). June 2005 is characterised by a positive anomaly around 10° N and a negative anomaly around 0°. This means that the ITCZ was more northerly than climatology. For June 2008 a positive anomaly is found over the Atlantic Ocean between 5° N and 7° S showing that the ITCZ was more southerly than climatology. Although the two flights were conducted during the same month (about 12 day difference), measurements in 2005 were obtained in a less convective environment than in 2008.

3.2 Biomass burning and anthropogenic sources

The TTL chemical composition can depend on local and remote sources from biomass burning and anthropogenic emissions. In Fig. 3 are represented the number of biomass burning fires from the Moderate Resolution Imaging Spectroradiometer (MODIS) fire product (http://modis-fire.umd.edu; Giglio et al., 2003) during the 10 days and the 20 to 11 days prior to each flight. For both 10-day periods, there are significantly more fires occurring in 2005 than in 2008. In 2005 the period preceding the flight was less convective (see Sect. 3.1) and consequently characterised by more frequent fires, particularly in Central Brazil. Note that there is already a large number of fires in the 20–11 days period before the flight in 2005, meaning that the biomass burning season started earlier in 2005 compared to 2008.

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Another possible source of CO that could affect the TTL composition is urban anthropogenic emissions from large cities. SPIRALE flights were performed very far from the main sources that are located near the Brazilian Atlantic coast and in the northwestern part of the South American continent (not shown) and thus likely do not affect SPIRALE measurements.

4 Analysis of SPIRALE measurements in the TTL

4.1 SPIRALE profiles

The vertical profiles of temperature, carbon monoxide (CO) and ozone (O_3) derived from SPIRALE measurements for both flights are displayed in Fig. 4. The temperature profile for 2008 in Fig. 4a exhibits large oscillations above 16 km altitude, including a well defined cold point (−85 °C) located at 16.9 km, followed by a strong increase up to 17.2 km and then a decay up to 18 km. The 2005 profile is smoother and exhibits two temperature minima close to −80 °C around 16.3 and 17.2 km altitude. In the 16–18.7 km altitude range, colder temperatures are found in 2008 on average. This is consistent with previous studies based on measurements in the tropics showing a cooling of the TTL where intense convection is observed (Johnson and Kriete, 1982; Kim and Dessler, 2004; Pommereau and Held, 2007). Note that the temperature maximum located around 19 km altitude in the June 2008 flight, correlated with an ozone anomaly, is out of the scope of the present paper since it is located above the TTL. It will be discussed in a separate paper.

Carbon monoxide mixing ratios in Fig. 4b are larger by ∼20 ppbv in 2005 than in 2008 except in the 17–18 km altitude range where they are almost equal. Below 17 km, where tropospheric influence is expected, the large mixing ratios of CO in 2005 can be related to the enhanced biomass burning activity compared to 2008 (see Fig. 3). The 2005 profile exhibits a series of minima and maxima below 17 km altitude with two main peaks around 14.2 km (maximum = 101 ± 5 ppbv) and 16.3 km altitude (maximum

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= 85±5 ppbv). The 2008 profile is characterised mainly by two layers having nearly constant mixing ratios of about 80 ppbv at 14–15.2 km and 60 ppbv at 16.0–17.3 km. Above CO decreases at about a similar rate as in 2005.

The ozone profiles are displayed in Fig. 4c and d with two different scales. Similarly to the temperature measurements, the ozone profile is much smoother in 2005 than in 2008. Mixing ratio values are close to each other up to ∼17.3 km altitude and the 2005 ones are larger above, up to 18.8 km. The 2008 ozone profile exhibits a step-like shape with, in particular, a large positive gradient in the 16.9–17.0 km layer, fairly constant values above up to 18.0 km and again a large positive gradient above. This ozone feature is also correlated with a steep increase of temperature in the 16.9-17.0 km layer followed by a negative gradient in the 17.0–18.0 km layer and a steep increase again above.

4.2 TTL characteristics

Table 1 gives the height of the TTL top and bottom. Several definitions have been proposed in the literature for the TTL boundaries based on convection (Sherwood and Dessler, 2001; Fueglistaler et al., 2009), chemical characteristics (Tuck et al., 1997; Folkins et al., 1999) or thermal structure (Gettelman and Forster, 2002). Here, the height of the TTL top and bottom is estimated from chemical composition criteria. Above the TTL top, pure stratospheric conditions are expected with ozone increasing steadily. This leads us to locate the TTL top at ∼17.8 km in 2005. Note that in 2008 we disregard in Fig. 4c the feature at 19 km in the ozone profile and we use the methane profiles giving the start of the stratospheric monotonic decrease above 18.0 km (not shown). The bottom of the TTL in 2005 can be identified in the ozone profile showing a transition at ∼14 km from typical tropospheric ozone values to a distinct increase of ozone in the TTL. This level is consistent with the level of change in the methane slope from constant to slightly negative (not shown). It is not possible to clearly identify the bottom of the TTL in 2008 in the ozone profile because of its step-shape structure. We use instead the methane profile (not shown) indicating the bottom of the TTL is at

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~14.4 km in 2008.

Table 1 also presents the cold point tropopause (CPT). One difficulty in 2005 is that the temperature profile has two minima at 16.3 and 17.2 km altitude with very close temperature values (Fig. 4a). To discriminate the CPT between these two minima we have considered the radiosounding launched on 22 June 2005 at 04:50 UTC, before the SPIRALE flight (not shown) and also the temperature profile measured during SPIRALE descent (from 11:47 UTC to 14:28 UTC). They both show a clear absolute minimum corresponding respectively to ~16.4 km altitude and ~16.2 km and a warmer (~1°C) minimum around 17.2 km (not shown). We conclude that the cold point tropopause for 2005 is at 16.3 km.

The characteristic levels for the two flights are summarised in Table 1. In general the two sets of measurements show very close TTL characteristic levels. Therefore these levels can be regarded as representative for North-east Brazil during the transition period from wet to dry season. The 2005 levels are slightly lower than 2008 ones likely because of the more intense convective activity in 2008 in the vicinity of Teresina leading to higher convective cloud tops on average and to colder temperatures.

TTL tops and bottoms are defined here from the characteristics of the chemical species profiles. Nevertheless the values found fit the altitudes of the top and bottom of the TTL defined from the synthesis definition recently proposed by Fueglistaler et al. (2009) and based upon dynamic characteristics. They set the lower boundary of the TTL above the level of main convective outflow, i.e., at ~150 hPa (355 K, 14 km) and the upper boundary at stratospheric levels not affected by convection, i.e. at ~70 hPa (425 K, 18.5 km).

To go a step further in the TTL characterisation we use correlations between CO and O_3. The approach based on the correlation of a tropospheric tracer and a stratospheric tracer has already been used at different latitudes to study the transition between the troposphere and the stratosphere (e.g. Pan et al., 2004; Pirre et al., 2008; Palazzi et al., 2009). The CO-O_3 correlation from the 2005 and 2008 flight measurements are displayed in Fig. 5a and b. If there were no mixing between tropospheric and

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stratospheric air, the CO-O_3 plot would have a "L-Shape", the vertical branch of the "L" corresponding to stratospheric air and the horizontal branch to tropospheric air. Both 2005 and 2008 data do not follow the "L"-shape, as expected in the tropics where the transition between the troposphere and the stratosphere is not sharp.

The CO-O_3 relationship for 2005 is characterised by a negative exponential shape with a rapid CO decrease with O_3 increase with altitude in the TTL. This indicates that the effect of vertical transport of tropospheric air masses in the TTL decreases rapidly with altitude. This behaviour is consistent with previous CO-O_3 correlations obtained from measurements in the tropics (Marcy et al., 2007, Palazzi et al., 2009, James and Legras 2009). To analyse in more detail the CO-O_3 correlation in the TTL we plotted the joint Probability Distribution Function (PDF) of CO-O_3 pairs only in the TTL altitude range (14–17.8 km for 2005 and 14.4–18 km for 2008, see Table 1). Figure 5c shows a general exponential decrease similarly to Fig. 5a. There is a maximum departing from this exponential shape around CO = 85 ppbv and O_3 = 100 ppbv. It corresponds to the peak of CO observed around 16.3 km altitude (16.0–16.5 km layer, see Fig. 4b) and may be related to air masses having a tropospheric signature (larger CO than background). Apart from this peak, the PDF exhibits very few discontinuities indicating that the air composition changes very progressively from tropospheric to stratospheric characteristics in the TTL. Note that the peak of CO around 14.2 km altitude cannot be seen on the PDF plot since part of it is located below the TTL. The 2008 CO-O_3 correlation (Fig. 5b) is very different from 2005. It exhibits a step-like shape within and above the TTL. The PDF figure (Fig. 5d) is very interesting since it is mainly composed of 3 sets of points. They correspond to layers where both CO and O_3 remain nearly constant. This means that in these layers tropospheric and stratospheric air is well mixed.

4.3 Analysis

In Sects. 4.1 and 4.2 we showed that although SPIRALE measurements were sampled at the same time of the year and in the same region and have similar TTL characteristic

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levels, the profiles and correlations for the two years show large differences. We analyse in the present section the origin of the specific features for each flight. For this purpose we use backward trajectories (Freitas et al., 2000) calculated from 3-D-wind fields provided by a simulation from the BRAMS (Brazilian Regional Atmospheric Modeling System, Freitas et al., 2009) regional meteorological model. Details on the model, simulations and trajectory code used in this section are given in the Appendix A. All backward trajectories are calculated at SPIRALE measurement times and altitudes. To take into account the model uncertainty on the 3-D-wind linked to its relatively coarse horizontal resolution (50 km), we have calculated the trajectories not only from the exact latitude/longitude location of SPIRALE data but also for each altitude at eight points around located at SPIRALE measurement point, displaced ±0.25° in latitude and longitude. 0.5° corresponds approximately to the distance between two model horizontal grid points.

The general shape of the 2005 CO profile in the TTL is characterised by a decrease from tropospheric to stratospheric values with two significant features: the layers around 14.2 km and 16.3 km altitude showing a large enhancement of CO with values reaching 101 ppbv and 85 ppbv, respectively. Figure 6 shows the accumulated surface precipitation rates from the BRAMS simulation and from TRMM together with the backward trajectories from the SPIRALE geographical location at 14.2 km altitude (altitude of the CO maximum). The back-trajectories are displayed until 16:50 UTC on 20 June 2005, ~40 h before SPIRALE measurements. This corresponds to the time when a rapid uplift related to an intense convective system (~62.2° W, ~7.3° S) is found for two trajectories (red trajectories), as illustrated by the model accumulated surface precipitation from 16:30 to 17:00 UTC. We have confidence in the model simulation for this convective system since TRMM surface precipitation accumulated between 16:30 and 19:30 UTC also shows a small but intense convective system at the same place. Note that TRMM does not provide accumulated rainfall rates over less than 3 h. A similar analysis is done to determine the origin of the peak on CO at 16.3 km altitude measured by SPIRALE. Results of 65-h back-trajectory calculations (back to 19 June

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16:40 UTC) are shown in Fig. 7. This corresponds to the time when the trajectories encounter a convective system in the model. This system is also observed by TRMM. In this figure we discarded four of the trajectories since they encountered convective uplift before 19 June 2005 at 16:40 UTC in areas where the model predicts unrealistic convection compared to TRMM observations. The peak at 14.2 km altitude exhibits larger CO mixing ratios than the peak at 16.2 km altitude. The convective system leading to the peak at 14.2 km is located closer to the main biomass burning area (see Fig. 3) than the convective system leading to the peak at 16.3 km. Therefore air lifted to 14.2 km is possibly more polluted than the air lifted to 16.3 km.

To confirm this hypothesis we used the Measurement of Pollution In The Troposphere (MOPITT) CO (Deeter et al., 2003; Emmons et al., 2004, 2007). We analysed the MOPITT version 5 retrievals which utilise the information from the NIR (Near Infra-Red) channel sensitive to the surface (Deeter et al., 2011). The data were averaged in boxes of $1^{\circ}\times1^{\circ}$ between 13 and 22 June 2005 to allow the reduction of the noise variability, in particular at the surface. Although the MOPITT V5 retrievals are not validated, it shows more sensitivity at the surface. This is shown by the surface averaging kernels greater than zero in Fig. 8 (bottom left panel) compared to MOPITT V4 (not shown). In addition the sensitivity at 200 hPa, as represented by their averaging kernels (Fig. 8 bottom right panel) is also improved. Figure 8 (top left panel) shows the CO distribution at the surface with a large area around 100 ppbv values in the north-west part of South America. In the south-west part, a maximum of CO is measured which corresponds to the fire emissions. One of the convectively lifted trajectories from Fig. 6 starting at location P1 and one from Fig. 7 starting at location P2 are also plotted in Fig. 8. P1 and P2 are located respectively at the outermost borders of the CO maximum and in a region representative of the background. This figure confirms that the P1 trajectory, which corresponds to the peak at 14.2 km altitude in SPIRALE data, starts where MOPITT measures significantly larger CO surface mixing ratios than the P2 trajectory which correspond to the peak at 16.3 km altitude.

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Nevertheless, the values of the peaks observed by SPIRALE at 14.2 and 16.3 km are lower than the CO measured by MOPITT at the surface but higher than at 900 hPa (not shown). This indicates that the air was likely uplifted by convection from levels between the surface and 900 hPa. Note also that the layers of enhanced CO measured by SPIRALE several days after they were convectively lifted could also be affected by diffusion leading to a decrease of their mixing ratios with time. To complement our analysis we also present in Fig. 8 (top right panel) the CO distribution at 200 hPa. The north-western area exhibits large values of CO (>100 ppbv) and corresponds to the maximum of TRMM accumulated surface rainrate area (Fig. 1), and thus to a convective area. This indicates that convection transports efficiently surface emissions in the upper troposphere in this region. The area over Teresina shows also concentrations close to 100 pbbv indicating transport from the north-western area as suggested by the back-trajectories. The new product MOPITT V5 presents CO in good agreement with our transport scheme and in terms of concentration compared to the Teresina profile.

In summary, the two peaks observed in the CO profile very likely come from the vertical transport by remote convection of lower tropospheric CO. The height and the intensity of the peaks are related to the intensity of convective updraft and on the sources of CO in the troposphere. The main emissions that are from biomass burning in Central Brazil encounter convection in the Amazonian region and are then driven by upper tropospheric winds towards the Teresina region. The layer of enhanced CO around 14.2 km altitude is below the level of zero radiative heating (LZRH ~15.5 km). Thus CO in this layer, if unperturbed by convection, will be radiatively driven downwards and will not reach the stratosphere. Contrarily the CO layer of enhanced CO around 16.3 km altitude will ascend radiatively to the stratosphere. This shows that during the transition from wet to dry season large amounts of biomass burning CO can be transported to the TTL and ultimately to the lower stratosphere.

In 2008 the ozone profile measured and the CO-O_3 correlation are characterised by a fairly unusual step-like shape as mentioned in Sects. 4.1 and 4.3. We have identified the presence of a layer between 17.0 and 18.0 km altitude characterised by a nearly

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constant ozone mixing ratio and by a decrease of temperature just above a strong increase. To our knowledge, similar measurements of ozone and temperature variations have only been reported by Danielsen (1993) and by Teitelbaum et al. (1999). Danielsen (1993) analysed airborne coincident measurements of ozone, temperature, water vapour and total water gathered near Darwin (Australia) in the frame of the tropical phase of the STEP project. He interpreted the constant ozone layer as a mixing layer formed by overshooting cloud turrets penetrating the tropopause and mixing air within the anvil cloud. This mechanism cannot be used to interpret the 2008 SPIRALE profiles since no ice particles were detected by the aerosol counter during the flight.

Teitelbaum et al. (1999) and Moustaoui et al. (2004) interpreted the measurements of temperature and ozone from a sounding launched in central Equatorial Pacific during the CEPEX experiment. They showed that some of the mixing layers observed near the tropopause were likely produced by the interaction between gravity waves and their critical levels. Critical levels arise when a wave encounters a mean horizontal wind whose horizontal velocity matches its horizontal phase speed. Here the gravity waves are likely to be due to convection, and are therefore expected to have a moderate phase speed (typically that of the convective clouds that produce them). So they certainly have critical levels when the wind becomes easterly and because the Quasi-Biennial Oscillation (QBO) is in its easterly phase. This interpretation does not require overshooting convection to occur. The mixing layer is identified by a step-shape of the ozone and potential temperature profiles coincident with a wind shear at the basis of the mixing layer. This feature can persist for some time and was identified by Teitelbaum et al. (1999) on soundings launched 3 h later.

The potential temperature measured by SPIRALE is displayed together with the ozone profile in Fig. 9a. It shows a nearly constant value in the mixing layer (17.0–18.0 km) which is similar to the CEPEX case study. To go further in the analysis we have used wind measurements from the radiosounding launched from Teresina on 10 June 2008 at 12:00 UTC. In Fig. 9b is displayed the potential temperature and zonal wind from this radiosounding. The SPIRALE potential temperature profile is also plot-

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ted in this figure. The radiosounding provides a potential temperature profile similar to SPIRALE in the mixing layer showing that it likely persisted several hours after the flight. The basis of the mixing layer is at the altitude of a strong wind shear (with a change from $u = 3\,\mathrm{m\,s^{-1}}$ to $u = -13\,\mathrm{m\,s^{-1}}$ in the layer 16.6–16.9 km) linked to the reversal of upper tropospheric westerlies to easterlies corresponding to the very end of the easterly phase of the QBO in the TTL and lower stratosphere. This evidence indicates that the 17.0–18.0 km mixing layer observed in the SPIRALE flight could be generated by the same processes as in the CEPEX measurements, i.e., the interaction of quasi-stationary convective gravity waves with their critical levels. This requires convection to have occurred close in time and space of the SPIRALE measurements. TRMM accumulated rainfall rates between 9 June 22:30 UTC and 10 June 01:30 UTC show the presence of a convective system about 50 km North-west of SPIRALE flight. To find a possible link between this convective zone and the SPIRALE measurements we have calculated backward trajectories from the 2008 model simulation. The trajectories below 16.2 km originate from the intense convective system located at 4° S–43° W (see Fig. 1c). Therefore the mixing could have been produced there and the characteristics of the mixed air transported from this mixing region to Teresina where SPIRALE measured it.

5 Conclusions

In this paper we present and analyse for the first time measurements of CO, ozone and temperature in the tropical tropopause layer in Equatorial Brazil. They were sampled by the SPIRALE balloon-borne instrument during two field campaigns that took place in June 2005 and June 2008 in Teresina (Brazil) at the transition period between the wet and dry season.

The characteristic levels in the TTL (top, bottom, cold point, 380 K) have been determined for the two flights. As expected, they are similar since the flights were done at the same time of the year. Nevertheless, the TTL is slightly higher and colder in

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2008 than in 2005. This can be related to a more convective environment in 2008. In 2008 the ITCZ is more southerly than climatology while it is the contrary for 2005. As a consequence there were significantly more biomass fires in Brazil during the 20 days preceding the flight in 2005.

The profiles of CO and ozone measured in 2005 and in 2008 have different shapes and mixing ratio values. The most important features are the larger values of CO mixing ratios in 2005 than in 2008 with peaks of CO at different altitudes and a step-shape for the ozone profile in 2008. We used the Probability Distribution Function (PDF) of CO-O_3 as a complementary tool to identify specific features. The analysis of the PDF of CO-O_3 in the TTL shows that in the 2005 profile the layer with high CO around 16.3 km altitude departs from the general smooth transition from tropospheric to stratospheric values, meaning that this layer has a different origin. The PDF for 2008 is mainly composed of three distinct sets of points which correspond to layers with fairly low variations in CO and O_3, i.e. well mixed layers.

The layers of enhanced CO from the 2005 flight were analysed using backward trajectories calculated from 3-D winds simulated by the BRAMS regional atmospheric model. The measurements in 2005 were done in dryer and more polluted conditions because of more frequent biomass burning than in 2008. This is why the CO mixing ratios are generally larger by about 20 ppbv in 2005 in the upper troposphere and in the TTL. We showed that the two layers of enhanced CO in the upper troposphere and in the TTL come from the vertical transport of low troposphere CO-rich air by remote convection occurring 2 and 3 days before the flight. The layer peaking at 83 ppbv around 16.3 km altitude is located above the LZRH (Level of zero radiative heating, ~15.5 km). At this season, continental deep convection is thus strong enough to drive air from the low troposphere up to altitudes where it can be then transported into the lower stratosphere by radiative ascent. The main source of pollutants here is biomass burning which mainly occurs in Central Brazil. It is transported vertically by deep convection in the Amazonian region and then towards the Teresina region by upper tropospheric winds. Therefore our results show that the transition between the wet and the dry

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season tends to favour the troposphere to stratosphere transport of CO in Equatorial Brazil thanks to the combination of convective activity and relatively polluted conditions. This also shows that Teresina is well located to capture the effects of convective and large scale transport of Central Brazil emissions.

In 2008 SPIRALE sampled mixed layers, in particular a layer identified on O_3 and potential temperature in the TTL by a step-shape structure with a very strong gradient between 16.9 and 17 km altitude and fairly constant values from 17.0 to 18.0 km. This feature is of interest since very few measurements of this type are reported in the literature (Danielsen, 1993; Teitelbaum et al., 1999). Our case show large similarities with the CEPEX observations discussed in Teitelbaum et al. (1999) and Mastaoui et al. (2004). We showed that this layer is possibly linked to the interaction of quasi-stationary convection gravity waves, provided by the presence of an upper tropospheric wind shear, with their critical levels. TRMM surface rainrates indicate the presence of a strong convective system in the vicinity of the flight. Backward trajectories indicate that the air sampled by SPIRALE comes from this convective system. Moreover there is a strong wind shear that is observed by the radiosounding two hours after the flight. This shear is linked to the reversal of upper tropospheric westerlies to easterlies in the TTL and lower stratosphere. These easterlies correspond to the very end of the easterly phase of the QBO. To go a step further in the analysis of this case study, very high model resolutions could be done to resolve the convective waves and their interaction with the wind shear as in Mastaoui et al. (2004).

This study confirms that, similarly to what was found in other tropical regions, deep convection is able to transport tropospheric species into the TTL above LZRH. In our analysis we showed that the TTL composition can be significantly affected by remote convection occurring thousands of kilometres away. The impact of deep convection on troposphere to stratosphere transport is only important if deep convection occurs where tropospheric air is polluted. This happens during the period of transition between the wet to dry season. It is expected to be even more favourable during the transition from the dry to wet season because tropospheric air can be strongly loaded with pollutants at

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the end of the biomass burning season. Therefore, it would be interesting to make new balloon-borne measurements from Teresina at the transition from dry to wet season.

Appendix A

Description of the model and simulations used for backward trajectories

Backward trajectories are used in Sect. 4 to diagnose the origin of the air masses sampled by SPIRALE. The wind fields used to calculate the backward trajectories are from simulations of the BRAMS (Brazilian Regional Atmospheric Modeling System, http://brams.cptec.inpe.br) model (Freitas et al., 2009). BRAMS is based on the limited-area meteorological model RAMS (Cotton et al., 2003). BRAMS version is tailored to the tropics with several improvements such as the cumulus convection parameterization, soil moisture initialization and surface scheme. For the simulations used here, the radiative calculations are done with the Harrington (1997) scheme. It is a two-stream scheme which is sensitive to water vapour and hydrometeor spatial distributions. For the microphysics parameterization we use a one-moment bulk scheme which includes cloud water, rain, pristine ice, snow, aggregates, graupel and hail (Walko et al. 1995). Shallow and deep convection are parameterized as described in Grell (1993) and Grell and Devenyi (2002). Trajectories are calculated from the simulation outputs using the trajectory model developed by Freitas et al. (2000) which takes into account the sub-grid effects of convective processes. The trajectories determined by this method are reliable providing that the modelled convective precipitation is well located.

The simulations run for 2005 and 2008 flights have the same set-up. The domain ranges from 90° W to 30° E and from 20° S to 14° N. Horizontal grid spacing is 50 km. The simulations are set with 58 vertical levels from surface to 39 km altitude with a vertical grid-spacing of 400m in the upper troposphere and lower stratosphere. The simulations are started 24 days prior to each flight. The large size of the domain together with the length of the simulations are chosen in order to allow long backward trajectories,

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possibly originating in Africa. 3-D fields of pressure, temperature, water vapour and horizontal wind from ECMWF (European Centre for Medium-range Weather Forecasts) operational analysis are used to initialize and to constraint the model at the lateral boundaries. At the top of domain, we use a rigid lid with a high viscosity layer above 22 km altitude to damp gravity waves. Soil moisture initialisation is obtained by providing satellite TRMM precipitation estimates to a hydrological model (Gevaerd and Freitas, 2006). Sea surface temperatures (SSTs) are constrained using weekly SST analyses derived from satellite data on a $1^{\circ}\times1^{\circ}$ grid. SSTs are produced by blending in situ observations and satellite data (Reynolds et al., 2002) from the Advanced Very High Resolution Radiometer (AVHRR) sensor aboard NOAA (National Oceanic and Atmospheric Administration) satellites.

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Table 1. Characteristic levels of the TTL from SPIRALE 2005 and 2008 flights.

Year of flight	TTL top	TTL bottom	Cold point tropopause (temperature minimum)	380 K
2005	17.8 km	14 km	16.3 km (−80 °C)	16.7 km
2008	18.0 km	14.4 km	16.9 km (−85 °C)	16.9 km

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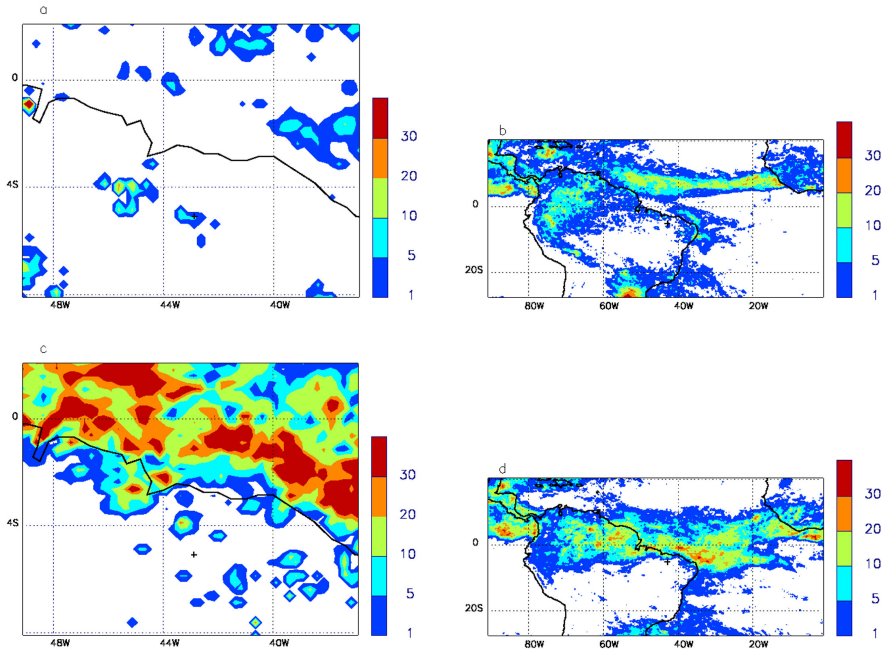

Fig. 1. TRMM accumulated surface rainrate in mm day^{-1} (**a**: top left panel) over 24 h from 21 June 2005 10:30 UTC, (**b**: top right panel) over 10 days from 12 June 2005 10:30 UTC (**c**: bottom left panel) over 24 h from 9 June 2008 01:30 and (**d**: bottom right panel) over 10 days from 31 May 2008 01:30 UTC. The black cross corresponds to Teresina location. Blue and red colours indicate weak (<5 mm day^{-1}) and heavy (>30 mm day^{-1}) surface rainrates, respectively.

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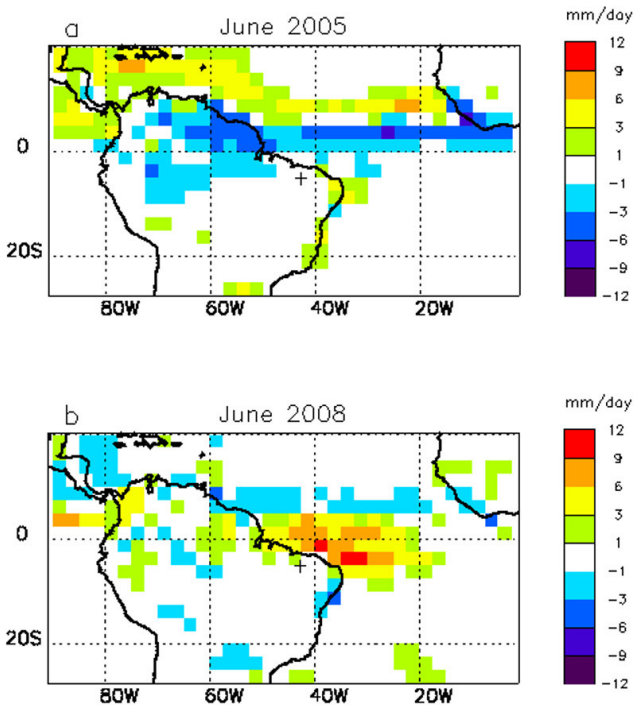

Fig. 2. Surface precipitation anomaly in mm day^{-1} **(a)** for June 2005 and **(b)** for June 2008 calculated from 2.5° latitude×2.5° longitude GPCP monthly mean Version 2.1 combination product and Version 2.1 1979-2008 climatology. The black cross corresponds to Teresina location. Black (resp. red) colour corresponds to a −12 K (resp 12 K) difference with climatology.

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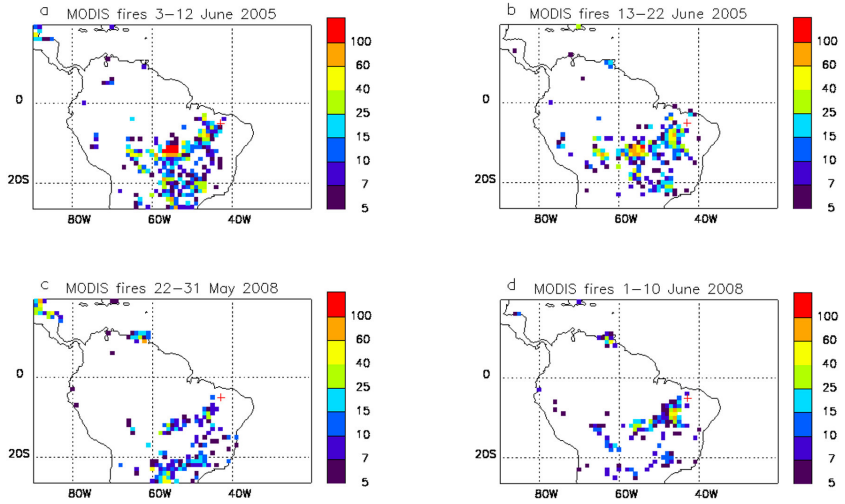

Fig. 3. Number of MODIS fires in 1° longitude×1° latitude boxes detected **(a)** between 3 June 2005 and 12 June 2005, **(b)** between 13 June 2005 and 22 June 2005, **(c)** between 22 May 2008 and 31 May 2008 and **(d)** between 1 June 2008 and 10 June 2008. The red cross corresponds to Teresina location. The black (resp. red) colour corresponds to 5 to 7 (resp. >100) fires per 1°×1° box over the each 10 days period.

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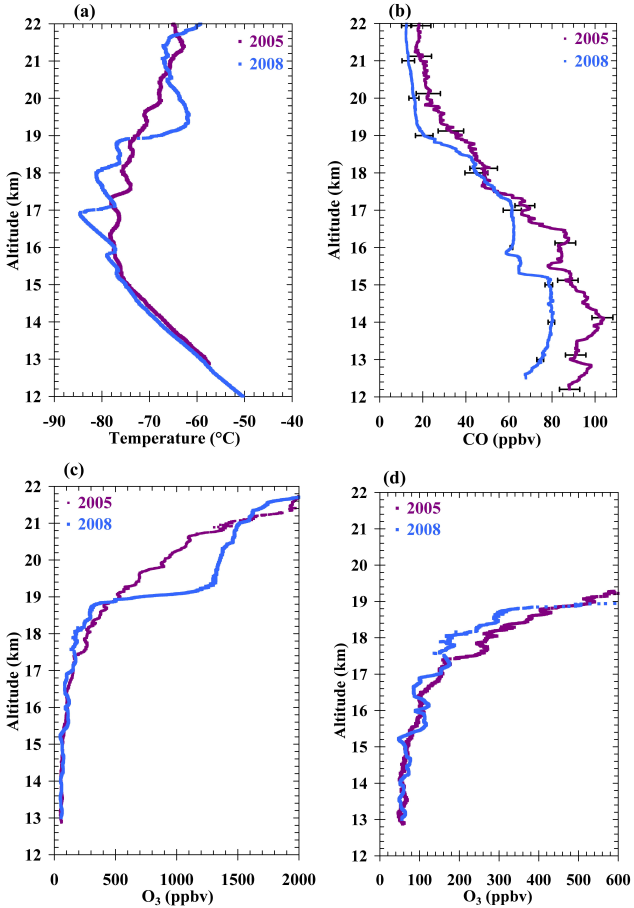

Fig. 4. SPIRALE 2005 and 2008 ascent measurements of **(a)** temperature (°C), **(b)** CO mixing ratio (ppbv) with error bars, **(c)** ozone mixing ratio (ppbv) up to 2000 ppbv and **(d)** ozone mixing ratio (ppbv) up to 600 ppbv. Purple and blue points correspond respectively to 2005 and 2008 data.

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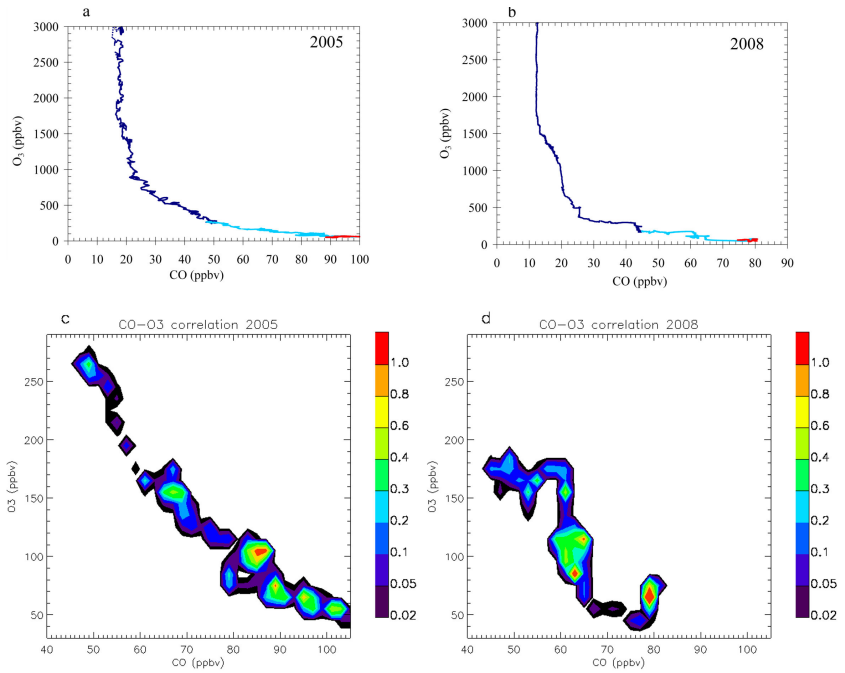

Fig. 5. CO-O_3 correlations (**a**: top left panel) from 2005 and (**b**: top right panel) from 2008 SPIRALE measurements. CO and O_3 are is in ppbv. Red, light blue and dark blue points correspond respectively to tropospheric, TTL and stratospheric points with TTL boundaries defined in Sect. 4.2. Joint probability distribution functions (PDFs) in % of the total number of CO-O_3 pairs measured within the TTL (**c**: bottom left panel) for 2005 and (**d**: bottom right panel) for 2008.

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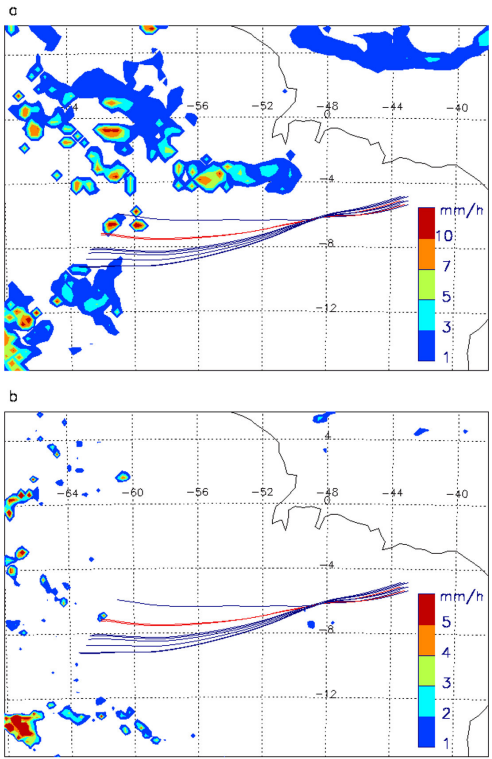

Fig. 6. Backward trajectories from SPIRALE measurements at 14.2 km altitude back to 16:50 UTC on 20 June 2005 and surface rainfall **(a)** from the model simulation accumulated on 20 June 2005 from 16:30 UTC to 17:00 UTC and **(b)** from 3B42 TRMM product accumulated on 20 June 2005 from 16:30 UTC to 19:30 UTC. The trajectories in red (resp. in blue) correspond to trajectories affected (resp. not affected) by rapid convective uplift in the model.

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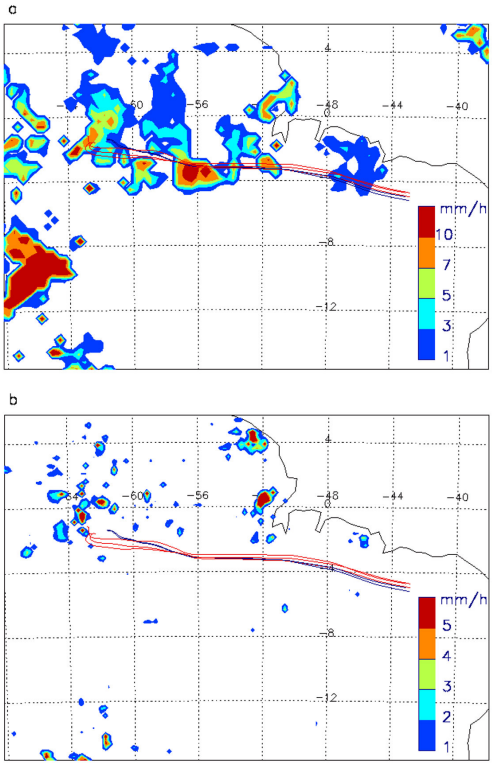

Fig. 7. Backward trajectories from SPIRALE measurements at 16.3 km altitude back to 16:40 UTC on 19 June 2005 and surface rainfall **(a)** from the model simulation accumulated on 20 June 2005 from 16:30 UTC to 17:00 UTC and **(b)** from 3B42 TRMM product accumulated on 19 June 2005 from 16:30 UTC to 19:30 UTC. The trajectories in red (resp. in blue) correspond to trajectories affected (resp. not affected) by rapid convective uplift in the model.

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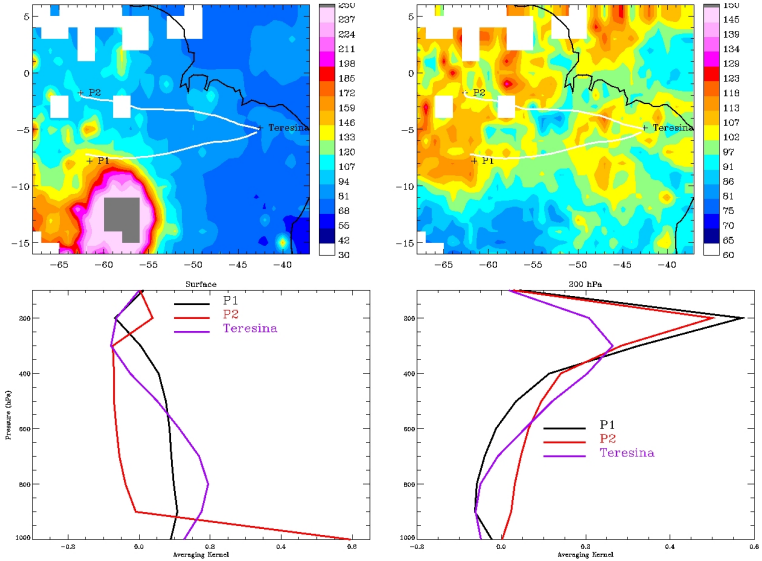

Fig. 8. Top left: Distribution of carbon monoxide (CO) at surface from MOPITT in ppbv. Top right: same but at 200 hPa. The data were averaged in boxes of 1°×1° and from 13 June to 22 June 2005. The red curves represent convectively uplifted back-trajectories from Teresina, one from Fig. 6 and one from Fig. 7. Note that the colour code is different for the two figures. Bottom left: Surface averaging kernel taken at the locations P1, P2 and Teresina as represented in the top figures The blue and pink colours correspond respectively to low (<100 ppbv) and high (>200 ppbv) CO mixing ratios. Bottom right: as bottom left but for the 200 hPa level. The blue and pink colours correspond respectively to low (<90 ppbv) and high (>130 ppbv) CO mixing ratios.

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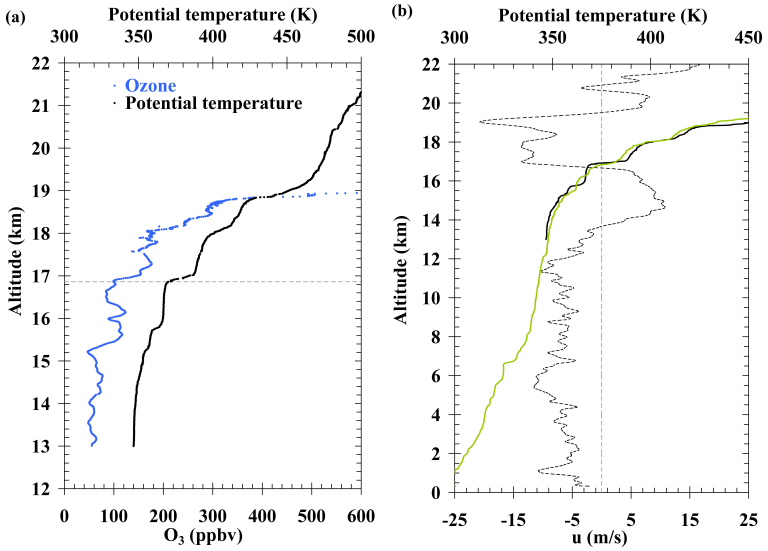

Fig. 9. **(a)** 2008 SPIRALE measurements of ozone (blue) and potential temperature (black). **(b)** SPIRALE 2008 measurements of potential temperature (black) and Teresina radiosounding data gathered on 10 June 2008 at 12:00 UTC (about 12 h after SPIRALE measurements) of potential temperature (light green) and zonal wind (dashed black line).

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