Isentropic advection and convective lifting of water vapor in the UT – LS as observed over Brazil (22° S) in February 2004 by in situ high-resolution measurements of H2O, CH4, O3 and temperature

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Abstract

The micro-SDLA balloonborne diode laser spectrometer was flown twice from Bauru (22° S, Brazil) in February 2004 during HIBISCUS to yield in situ H_2O measurements in the Upper Troposphere (UT) and Lower Stratosphere (LS) and in particular in the Tropical Tropopause Layer (TTL). The overall TTL was found warmer (with a subsaturated cold point near –79°C) and the LS moister compared to former measurements obtained in tropical oceanic conditions. The use of specific balloons with a slow descent, combined with the high-resolution of the laser sensor, allowed us to observe in situ in the UT, the TTL and the LS several thin layers correlated on H_2O, CH_4, O_3, temperature and PV. A component of these layers is associated with the isentropic transport into the UT- LS of extratropical stratospheric air masses. Moreover, the examination of temperature and tracer (CH_4, O_3) profiles gives insights on the potential contribution of convective transport of H_2O in the TTL.

1 Introduction

It is widely accepted that there is a subtropical dynamical barrier, which prevents transport into the tropical tropopause, identified by strong gradients of potential vorticity (PV) and chemical species (Polvani et al., 1995; Holton et al., 1995; Hall and Waugh, 1997; Haynes and Shuckburgh, 2000). Nevertheless, the subtropical Rossby wave-breaking processes can cause the transport of tongues of extratropical air through this dynamical barrier deep into the tropics (Plumb, 1996 ; Horinouchi, 2000). The observation in the tropical upper troposphere of such intrusion events has previously been discussed in the literature. For instance, Scott et al. (2001) have investigated, over the southern tropical Atlantic, subtropical intrusions in the tropical upper troposphere and their impact on H_2O and O_3, by examining ozone from the Measurements of Ozone by Airbus In-Service Aircraft (MOZAIC) program in conjunction with PV-advection and meso-scale models. They have suggested that these intrusions due to Rossby wave

breakings may have an impact on the distribution of tracers in the tropical upper tro-
posphere and a significant contribution to the exchange of air between tropical and
extratropical regions. The investigation of the vertical extension of such subtropical in-
trusions was made by Waugh and Funatsu (2003) by using ozone from TOMS and 20-
years of NCEP-NCAR PV-data over the northern tropical Pacific. They have suggested
that intrusions events could potentially affect the tropical tropopause layer (TTL) and
the lower stratosphere, as well as the upper troposphere. In particular, they indicated
that the impact on H_2O in the TTL and the tropical lower stratosphere (LS) needed to
be further investigated. Note that in the present study, UT will refer to the part of the
upper troposphere located below the TTL. An other open issue is the effect of such sub-
tropical intrusions on deep convection (Kiladis and Weickmann, 1992; Kiladis, 1998;
Matthews and Kiladis, 1999). Indeed, the anomalous PV in the intruding tongue of ex-
tratropical air could produce a region of ascent and of reduced static stability, initiating
thereby convection. Kiladis (1998) suggested that the diabatic heating associated with
convection may play a role in the weakening of the PV-anomaly. Hence the intrusion
would generate convection which could conversely cause its erosion and the mixing of
tropical and extra-tropical air. Therefore, Waugh and Funatsu (2003) recommended in
their study that further investigations on transport and on convection associated with
intrusion events be combined.

The micro-SDLA balloonborne diode laser spectrometer was flown twice during the
HIBISCUS field campaign (Pommereau et al., 2006[1]) to provide in situ high-resolution
H_2O measurements in the UT-LS. The data were recorded near Bauru (22.4° S, 49° W)
in Brazil during the wet season, in February 2004. In contrast to most previous studies
on tropical H_2O in the UT and TTL (Jensen et al., 2005; Tuck et al., 2004; Vömel et
al., 2002; Weinstock et al., 1995; Richard et al., 2006), the HIBISCUS measurements
were performed over a continental region instead of a maritime one. Continental re-
gions and in particular South America are favorable to the development of very deep

[1]Pommereau, J.-P., Garnier, A., Held, G., and the Hibiscus team: An overview of the HIBIS-
CUS campaign, Atmos. Chem. Phys. Discuss., in preparation, 2006.

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convective events as shown by Liu and Zipser (2005) from TRMM (Tropical Rainfall Measuring Mission) observations. The micro-SDLA was flown over a continental convective region located within or in the vicinity of the South Atlantic Convergence Zone (SACZ) in the summer. In the present paper, we examine these high-resolution H$_2$O measurements using temperature profiles and tracer data (CH$_4$ and O$_3$), as well as PV from a high-resolution PV-advection model. This is done with the purpose of identifying the respective signatures of isentropic and convective processes affecting H$_2$O in the UT-LS. In Huret et al. (2006)[2] we use a meso-scale modeling approach with a sufficient vertical resolution and based on local meteorology to further investigate the impact of convection on H$_2$O in the TTL.

In Sect. 2, we describe the micro-SDLA instrument. In Sect. 3, we define the TTL extension for both flights and report the H$_2$O, CH$_4$ and O$_3$ data in the UT, TTL and LS. Finally, we will discuss in Sect. 4 the data in the UT-LS by examining the tracer measurements and the PV-maps from the PV-advection model.

2 Instrumental

The micro-SDLA sensor is a balloon borne diode laser spectrometer devoted to the in situ monitoring of H$_2$O, CH$_4$ and CO$_2$ in the UT-LS by infrared absorption spectroscopy (Durry et al., 2004). Figure 1 is a picture of the sensor during the HIBISCUS campaign in Brazil in February 2004. With the micro-SDLA, three near-infrared telecommunication-type InGaAs laser diodes are connected by means of optical fibers to a two-mirrors multipass optical cell operated open to the atmosphere that provides a 28 m absorption path length (Durry et al., 2002a). The laser beams are absorbed in situ by the ambient CH$_4$, CO$_2$ and H$_2$O molecules as it is reflected back and forth between

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[2]Huret, N., Durry G., Freitas S., et al.: In situ laser diode measurements of H$_2$O during HIBISCUS: investigation of convective event impact on H$_2$O distribution in the TTL by a meso-scale model, Atmos. Chem. Phys. Discuss., in preparation, 2006b.

the mirrors of the optical cell. In situ absorption spectra of CH_4 in the 1.65-micron region, CO_2 in the 1.60-micron and H_2O in the 1.39-micron region are recorded at the cell output using a differential detection technique (Durry et al., 2000). By combining the direct-differential detection with an adequate selection of the H_2O rotation-vibration transitions scanned over by the laser, the achieved dynamical range of the measurements is of four orders of magnitude, which permits a continuous probing of H_2O in both the troposphere and the LS (Durry and Megie, 2000). The H_2O used transitions and their associated spectroscopy are described in Durry et al. (2005). The Beer Lambert law is then applied to relate the absorption of laser energy to the molecular concentration. In practice, the molecular mixing ratio is retrieved from the absorption spectra using a non-linear least-squares fit to the full molecular line shape in conjunction with onboard in situ atmospheric pressure and temperature measurements and by use of a molecular model (Durry and Megie, 1999). The achieved inaccuracy in the H_2O and CH_4 concentration measurements is of 5% in the UT-LS, altitude region, which is studied in this paper, for an average measurement time of 160 ms (co-addition of twenty 8-ms laser scans). More details on the used balloon borne laser probing technique used for H_2O and CH_4, the concentration retrieval and the various sources of errors are given in Durry and Megie (1999) and Durry et al. (2002b).

The micro-SDLA sensor was flown twice from Bauru (22° S, 49° W). The objective was to probe in situ the UT-LS and in particular the TTL at high-resolution. Small open stratospheric balloons inflated with 3000 m^3 of helium were used, which provided a float altitude near 21 km. The H_2O data in the stratosphere and the tropopause were taken during the descent of the instrument to prevent the contamination of the measurements by water vapor outgassing from the balloon envelope (Durry and Megie, 2000). As no valve was available with the small balloons, the slow descent in the TTL was achieved using a transition from day to night (i.e., the balloon temperature decreases at sunset and the balloons starts a slow descent). The UT-LS was hence probed at a mean descent speed near 1.5 m s^{-1} during nighttime. The measurements in the troposphere were taken during the final descent after cut-down under parachutes. For the first flight,

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additional H_2O measurements were also taken during the ascent in the troposphere. The micro-SDLA was flown without telemetry the data being recorded on-board and processed after the recovery of the gondola. For this study focused on the TTL transition layer and the UT, we will make use of the micro-SDLA data in the altitude range from 9 km to 18.5 km. A further discussion of the H_2O data in the LS, near the flight ceiling, is made in the paper by Nielsen et al. (2006).

3 Observations

The micro-SDLA was flown twice during the 2004 HIBISCUS field campaign. An overview of the overall HIBISCUS campaign can be found in the paper by Pommereau et al. (2006)[1]. The first flight, called "SF2", which stands for "Short duration Flight 2", took place on 13 February 2004 with a launch at 20:18 UT. The second flight, called "SF4" took place on 24 February 2004, with a launch time at 20:03 UT. At the balloon launch site, a meteorological S-band Doppler radar, operated by the IPMet (Instituto de Pesquisas Meteorológicas), was providing real-time observations of precipitation within a 450 km range.

3.1 Brief overview of the meteorological situations

During the flight period, in February 2004, the rainfall anomaly was positive in most part of Brazil. The eastward propagation of intra-seasonal atmospheric oscillations, emanating from the Indian and Pacific Oceans, continued to induce convective activity over Brazil. The frontal systems rapidly advanced over southern Brazil and became stationary over the Southeast Brazil region, establishing two episodes of convection in the Bauru region associated to the South Atlantic Convergence Zone (SACZ). The SACZ produces an elongated band of high convective activity emanating from the Amazon basin and running southeastwards into the South Atlantic Ocean, typical in summer over the South American continent.

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The two flights took place in different meteorological conditions. On the synoptic scale, the flight of SF2 took place under diminishing influence of a high pressure system at the surface and the 200 hPa divergence slowly advancing southwards from Northeast Brazil, resulting in unstable conditions over most of the State of São Paulo. During the afternoon of 13 February 2004, the Bauru radar observed moderate convective activity within the 240 km radar range. At the time of the launch of SF2, the nearest storm was about 60–70 km south-east of the Bauru radar, but remained more or less stationary and at no stage did it impact on the flight trajectory (Fig. 2b, left). However, at the same time, the radar in Presidente Prudente, 240 km west-north-west of Bauru, showed a large area of instability (echo tops up to 16 km–17 km) moving in from north-west and west, but it never came closer than 300 km from the balloon track, as illustrated in Fig. 2a (left), showing the TRMM surface rainfall accumulated approximately during the duration of the flight for the region of interest.

From 20 to 24 February, the Bauru area was under the SACZ influence leading to convective conditions during the SF4 flight. This is illustrated by the TRMM accumulated rainfall depicted in Fig. 2a (right). A large area of instability within the SAZC, with more or less randomly developing intense cells was first observed by IPMet's radars in the far north-west of the State of São Paulo, between 07:00 and 08:00 LT (10:00–11:00 UT), expanding eastwards while moving towards south-east as a whole. Significant intensification occurred from noon onwards, resulting in a extensive multi-cellular complex, with echo tops of up to 13–15 km (Pommereau et al., 2006[1]), heading towards Bauru during the late afternoon. At the time of the launch, the storm complex was about 120 km to the north-west, moving at 40 km h^{-1} south-eastwards and reaching Bauru shortly after 23:00 UT. The radar accumulated rainfall for a 6-h period, commencing one hour before the launch, is shown in Fig. 2b (right). The convective activity during SF4 flight was significantly higher than for the SF2 case, but both flights were performed in the vicinity of cloudy areas (Huret et al., 2006[2]). The wind intensity is similar on both days, with a maximum of 25–30 m s^{-1} between 11 km and 15 km altitude, but from south-west for SF2 and from the north-west for SF4, and light winds

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(5–10 m s^{-1}) in the 15 km to 18 km altitude range, turning to the east-south-east in the stratosphere (Pommereau et al., 2006[1]).

3.2 Location of the TTL for both flights

Measured ozone and temperature profiles were used to determine the extent of the TTL for both flights. The TTL is the transition layer between air with tropospheric properties and air with stratospheric properties (Highwood and Hoskins 1998, Folkins et al., 1999). In practice, there are several definitions relevant for the TTL, based either on thermodynamic properties, or on ozone mixing ratio. Following Sherwood and Dessler (2000), the TTL ranges from the zero net radiative heating level to the highest level that convection reaches. However this definition is difficult to use in the present study, which is based on profile measurements of temperature, H_2O, and chemical species. Sherwood and Dessler (2000) report that the TTL lies in the altitude range from approximately 13 km to 18 km, which corresponds to an isentropic surface range from roughly 345 K to 410 K. According to Folkins et al. (1999), the TTL is a mixing layer and its bottom is at the chemopause, i.e. the altitude from which there is an increase in the ozone mixing ratio. In the present study, we base our definition of the TTL on temperature profiles, lapse rates and ozone profiles (Fig. 3).

To locate the top of the TTL, we use the definition from the SPARC tropopause workshop (Haynes and Shepherd, 2001), which suggested the use of either the lapse rate tropopause (LRT) or the cold point tropopause (CPT). In the tropics the LRT and the CPT are usually less than 0.5 km apart. Here we locate the top of the TTL at the lapse rate point tropopause (using WMO definition: the base of a layer at least 2 km thick, in which the vertical temperature gradient, $\partial T/\partial z,$ exceeds the value -2 K km^{-1}) which corresponds to the dynamic barrier level. This gives a top TTL level at 15.5 km for SF2 and at 17.8 km for SF4. Note that in both cases the LRT is close in altitude to the CPT: the cold point tropopause is at an altitude of 15.6 km for SF2 and 18.0 km for SF4. The temperature minimum value is $-79°$C for both flights. We can deduce the altitude of

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the bottom of the TTL using the chemopause determined from the ozone soundings performed during HIBISCUS by the electrochemical concentration cell (ECC) ozone sensor from the Danish Meteorological Institute (DMI). The accuracy of the ozone sensor is 5% (Komhyr et al., 1995). The two sondes used were launched on 13 February at 22:35 UT and 24 February at 22:30 UT. Since they are very close in time respectively to the SF2 and to the SF4 flights, it is possible to use them to determine the chemopause level. Figure 3 shows that for SF2 the chemopause is observed around 13.5 km altitude and for SF4, around 13.3 km, which is the altitude of the maximum lapse rate of about 8 K/km. Thus, the TTL extension is 13.5–15.5 km for SF2 and 13.3–17.8 km for SF4. In SF2, the TTL is significantly thinner than in SF4. In SF2, there is a sharp transition between the TTL and the LS as illustrated by the lapse rate gradient in Fig. 3. This indicates that the dynamical barrier between the troposphere and the stratosphere is well established in SF2. For SF4, the transition is smoother with an alternate of positive and negative lapse rate values between 15.7 and 17.8 km. This and the large TTL extent (13.3–17.8 km) indicates, that there is likely to have been more mixing of tropospheric air with stratospheric air in SF4 than in SF2.

For both flights the temperature range encountered in the TTL varies from –65°C at the bottom to –79°C at the cold point tropopause. The TTL appears rather warm compared to previous observations. For instance, Jensen et al. (2005), have reported a cold point at –86°C, over the Pacific south of Costa-Rica (pre-Aura validation experiment) during January 2004, close in time to the SF2 and SF4 dates. This temperature difference in the TTL may be attributed to the fact that HIBISCUS measurements were obtained above a continental surface, while previous studies were done essentially in oceanic conditions.

3.3 H_2O and CH_4 in the UT-LS

Figure 4 displays the retrieved H_2O and CH_4 data from micro-SDLA for the SF2 and SF4 flights. The temporal resolution was of one H_2O (and CH_4) concentration sample per second for the first flight (SF2) and of four per second for the second flight (SF4).

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The vertical concentration profiles are therefore composed of a few thousands of individual measurements. Additional meteorological balloonborne soundings obtained close in time to the flights (13 February 2004 at 22:30 UT and 24 February 2004 at 20:15 UT) with Vaisala RS-90 and RS-80 sondes by IPMET/UNESP (Institute for Meteorological Research of the São Paulo State University) and by DMI (Danish Meteorological Institute) are added to the figure for comparison with the measurements made by the micro-SDLA in the troposphere. Another sounding performed on 23 February at 22:30 UT (green line in Figs. 4a and b) is also displayed. This sounding will be used later in the discussion as a reference for H_2O and O_3 since this sounding took place in tropical air masses far from any extratropical intrusions.

For the SF2 flight in the UT (Fig. 4a), a water vapor minimum of 100 ppmv is observed in the troposphere around 6.5 km altitude. At 8–10 km, the instrument measures approximately 800 ppmv. The sounding close to SF2 gives the same water vapor profile shape, and the achieved water vapor contents are in between the micro-SDLA ascent and descent data. CH_4 measurements in the UT for SF2 flight remain close to the expected surface value (1.8 ppmv) with no special features. For the SF4 flight in the UT (Fig. 4), a very dry layer is observed around 9.5 km (340 K potential temperature surface) in a $\sim$2 km thick layer with a water vapor mixing ratio lower than 100 ppmv. One can even see a very thin substructure (less than 100 m) at 9 km altitude with an increase of water vapor amount up to 900 ppmv. The corresponding CH_4 data provided by micro-SDLA in this altitude range are also displayed in Fig. 4 and show a good correlation with H_2O dry layer with CH_4 values 0.2 ppmv lower than the expected surface value and than the CH_4 observed in the whole troposphere. The water vapour sounding launched two hours before the SF4-flight also shows this type of shape with an alternation of very dry thin layers. The vertical extent and the intensity of this dry layer is larger but in the same altitude range. The sounding launched one day before the SF4 flight (in green) does not show this dry layer with a water vapor content more than ten times greater than SF4 at 9.5 km, i.e. 1500 ppmv. A specific process has happened between, 23 and 24 February in the UT leading to the drying of the layer around 9.5 km.

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For both flights, a CH_4 decrease of roughly 0.2 ppmv is detected around 15 km for SF2 and 14.5 km for SF4. Nearly at the same altitude levels as these methane structures, a sharp decrease in the water vapor profiles can be observed: within roughly five hundred meters, the H_2O mixing ratio changes from 20 ppmv down to 6 ppmv for SF4 and from 10 ppmv down to 6 ppmv for SF2 (descent data).

In the TTL and LS, the measured H_2O mixing ratios for both flights range roughly from 20 ppmv at 14 km down to 5 ppmv at 18 km with values between 3.5 and 6 ppmv in the 16–18 km range. Also displayed in Fig. 4 is the saturation mixing ratio over ice above 14 km altitude. For flight SF2, the air is largely supersaturated (over 150% in relative humidity with respect to ice saturation) up to 15.5 km and largely undersaturated above (down to ∼20% in relative humidity). Dehydration by ice nucleation and subsequent sedimentation is likely to take place in the upper troposphere and the TTL on this occasion. Additionally, for this flight, above the top of the TTL, the undersaturated conditions indicate that the vertical dynamical barrier is strong enough to prevent troposphere to stratosphere exchange at the lapse rate tropopause level. For SF4, the air is also supersaturated (in the order of 120%) up to 15 km and undersaturated above (down to ∼30%). In this case, the upper part of the TTL is dry, leading to no possible dehydration by ice condensation and sedimentation. In the LS, the air is significantly undersaturated for both flights. A detailed calculation of the saturation ratio for SF2 and SF4 can be found in Marécal et al. (2006). Nevertheless, compared to the measurements presented by Vömel et al. (2002) and Jensen et al. (2005), both SF2 and SF4 H_2O mixing ratios are significantly larger above 17–17.5 km with values generally greater than 4 ppmv. They are also larger than those reported by Richard et al. (2006) during ER-2 flights near Costa-Rica above the 380 K isentropic level. Air in the LS over Brazil is thus moister than in tropical oceanic conditions. This is consistent with the warmer conditions recorded in the LS over Brazil leading to less dehydration than in colder conditions recorded in the LS over the tropical ocean.

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3.4 Ozone in the UT-LS

The examination of ozone measurements in the UT-LS provides complementary information on the dynamical processes affecting the probed air masses. During HIBISCUS, ozone soundings were performed by the DMI ECC ozone sensors but also by the O_3-SSS sensor, a solid-state semiconductor sensor developed at UCAM Cambridge (Hansford at al., 2005) that was operated as a piggy-back on the flight chain of the micro-SDLA flights. Figure 5 shows the ozone measurements achieved in the UT-LS at the time of the flights above the 330 K isentropic level. Two soundings performed on the 10 and 23 February have been added to the figure. The measurements on 23 February were used in Fig. 4 (green line) as a reference for water vapour. The corresponding ozone data will be used here as a reference for ozone in the UT, since the sonde probed tropical air masses free from extratropical intrusions. The measurements on 10 February will be used as a reference profile for O_3 in the TTL and LS. Figure 5 shows that the ECC and O3-SSS measurements generally agree well. The differences found below 10 km on 24 February can be attributed to the difference in time between the two soundings.

In the UT, near 340 K ($\sim$9.5 km), the SF4 flight appears rather rich in ozone compared to the reference sonde on 23 February. This ozone-rich layer is at the same isentropic level where a very dry and CH_4-poor layer was observed (335–340 K). An analysis of the O_3 profiles shows that for both flights, there is a local ozone maximum just below the chemopause ($\sim$350K). In their study over the equatorial Pacific and equatorial Eastern Brazil, Vömel et al. (2002) have classified O_3 and H_2O profiles in the upper troposphere. Profiles associated with deep convection have a strong decrease in ozone concentration in the UT from 10 ppbv at 10 km to 1 ppbv at the chemopause. In non-convective cases or when convection did not reach a sufficient altitude in the UT, the O_3 mixing ratios are higher (40 ppb) and the profiles are constant up to the chemopause as observed on 23 February 2004 (green line in Fig. 5b). Ozone and water vapor profiles from both micro-SDLA flights are similar to those reported by

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these authors over South America on February 1997, with a local maximum of ozone below the chemopause (around 350 K). Therefore, if referring to Vömel et al. (2002), none of our ozone measurements seem to be associated with cases affected locally by convection in the UT.

In the TTL and LS, there is a clear increase in the ozone content for SF2 compared to SF4 in the 17 km to 18 km range, with a maximum difference of up to 80 ppbv at 18 km. We also observe that the O_3 content for both flights is significantly greater (by up to 100 ppbv) than the 10 February reference ECC sonde. A similar order of magnitude in the O_3 excess was also reported by Waugh and Funatsu (2003) from the ozone sounding over Hilo, Hawaii, in a case of extratropical intrusion in January 1987.

From the comparison with all the ozone soundings achieved during HIBISCUS (not shown), it appears that in both flights the micro-SDLA has probed air-masses enriched in ozone.

4 Discussion of H_2O in the UT, TTL and LS

To summarize the observations, for both flights, the overall TTL was found warmer (with a cold point near −79°C) and the LS moister compared to former measurements obtained in tropical oceanic conditions. The vertical extent of the TTL transition layer for SF4 is nearly twice as much as for SF2, and the lapse rate features a sharp transition between TTL and LS for SF2 and smoother for SF4. The LS for SF2, and the upper part of the TTL and the LS for SF4, which are discussed in next section, are found largely subsaturated. The ozone measurements give insights of potential occurrence of extratroptical intrusions, particularly in the UT for SF4 and in the LS for SF2.

4.1 Intrusions of extratropical air into the UT-LS

In the UT, there is, for SF4, a very dry layer with low CH_4 and high O_3. To identify the occurrence of an extratropical intrusion in the UT, we have run the MIMOSA contour ad-

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vection model (Hauchecorne et al., 2002) of the potential vorticity (PV). The MIMOSA model was used previously in several studies, for instance, to follow the evolution of polar and tropical intrusions at mid-latitudes (Heese et al., 2001; Durry et al., 2002b; Durry and Hauchecorne, 2005; Huret et al., 2006). During HIBISCUS, PV maps have been calculated with MIMOSA every six hours on isentropic surfaces in the TTL with a 5 K resolution using the ECMWF fields. MIMOSA has to be used with caution in the UT and the TTL because it is based on isentropic advection and does not take into account a possible vertical transport of air masses. The MIMOSA predictions are nonetheless for each case reinforced by the exmaination of the corresponding tracer measurements. Figures 6 and 7 show the PV-maps in the 340 K to 415 K range with a 5 K-resolution for SF2 and SF4, respectively. The micro-SDLA instrument is located on the PV-maps using its onboard GPS.

In the upper troposphere, MIMOSA confirms for SF4 the presence of a filament of extratropical stratospheric air at 340 K (see Fig. 7, PV map at 340 K). This filament explains the presence of the very dry layer observed in SF4 at 9.5 km with low CH_4 and high O_3, typical of stratospheric air. The process is similar to that observed in the UT by Scott et al. (2001) over the southern tropical Atlantic when the aircraft in the UT crossed a potential vorticity filament coming from mid-latitude showing a strong discontinuity in potential temperature, a low relative humidity and an ozone mixing ratio peak greater than 70 ppbv at 10 660 m. It is remarkable that MIMOSA is able to simulate the filament, since in the tropical UT diabatic processes, and in particular convection, possibly degrade the prediction of PV advection. Nevertheless, according to the examination of the ozone profile described in Sect. 3, the situation is non-convective in the UT at the time and place of the flight. Within one day, the filament is formed and advected in a convection-free atmosphere until it reaches the convection zone located near Bauru as illustrated by Fig. 2a (left) showing TRMM surface rain-rates during the flight period. The BRAMS meso-scale model, which is used in the companion paper associated to this study (Huret et al., 2006[2]), also predicts the presence of this filament in the UT.

We see in Figs. 6 and 7 that extratropical intrusions were also present in the TTL and LS for both flights. One remarkable feature provided by the PV maps is the tongue of high PV ($|PV|>2$) stratospheric mid-latitude air in the 385 K to 405 K range for SF2 in the LS (Fig. 6). To better examine the impact of isentropic transport in this altitude range,

5 vertical PV profiles were extracted from the MIMOSA simulations for both flights and plotted in Fig. 8, in conjunction with the H_2O, CH_4, O_3 and temperature. For SF2 (in blue in Fig. 8), the signature of the intruding tongue can be clearly seen in the 385 K to 405 K range. In this layer, there is a correlation between PV, CH_4, O_3 and temperature. Low CH_4 and high O_3 are observed in air masses of high-PV suggesting an extratropical

10 stratospheric origin. Furthermore, from 370 K to 390 K the temperature is nearly constant but above 390 K, a positive gradient of temperature, typical of the stratosphere, is observed in the region of interest. The hydrogen budget $[H_2O]+2[CH_4]$ yields values within 7.2 to 7.5 ppmv, which is a further indication of mid-latitude stratospheric origin (Durry et al., 2002b, Zöger et al., 1999). Hence, there are several lines of evidence

15 (low CH_4, high O_3, high PV, hydrogen budget, temperature measurements) showing that the micro-SDLA has probed air masses originating from the southern mid-latitude stratosphere during SF2. This also explains the ~4 ppmv H_2O content recorded by the micro-SDLA, which is typical of mid-latitude H_2O values. Also in SF2, a thin layer of low H_2O (3.3 ppmv), anticorrelated with CH_4 can be seen at 400 K. When taking into

20 account the systematic 300 m shift upwards of ozone resulting from the 50 s time constant of the ECC sensor, it would also correspond to an ozone dip. Indeed, the PV profile and the maps of Fig. 6 show, that at this altitude, the balloon is located within a low $|PV|$ tropical air parcel. For the isentropic levels lower than 385 K, it is difficult to conclude about potential impact of isentropic transport, as the instrument is found at

25 the edge of the intrusion (Fig. 6).

4.2   Vertical transport by convection into the TTL for SF4

For the SF4 data (in red in Fig. 8), the isentropic transport is a less convincing explanation for the large amount of 5.5 ppmv of H_2O observed between 390 K and 405 K.

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The MIMOSA predictions, as well as the CH_4 or O_3 profiles, do not show any particular structures. At 385 K (∼16.5 km), the micro-SDLA measures 3.5 ppmv of H_2O while at 395K (∼17.5 km), it gives 5.5 ppmv. The examination of the corresponding PV-maps in Fig. 7 shows that the micro-SDLA was probing tropical air masses with the same low

5 PV at both isentropic levels. From MIMOSA results, the isentropic transport can not explain for the difference of 2 ppmv observed between both isentropic levels. The calculation of the hydrogen budget $[H_2O]+2[CH_4]$ between 390 K and 405 K gives a value near 8.5–9 ppmv, which is too high to be of mid-latitude stratospheric origin. For SF4, the O_3 profile is unstructured in the whole TTL. While an increase in H_2O with altitude

10 is observed in SF4, the methane mixing ratio remains fairly close to the surface value at 1.75 ppmv. This could be due to a convective transport of tropospheric moist air in this region which is far from the saturation with respect to ice over 370 K. For instance, at 395 K, the water vapor saturation mixing ratio over ice is 11 ppmv. The vertical transport by convection could also explain why, for SF4, methane concentration remains

15 close to the surface values in the TTL. A more precise examination of the CH_4 profile (see dashed black line in Fig. 8 which gives average values for CH_4 in SF4), shows that methane decreases between 370 K–380 K. Above 380 K, it increases again to converge towards surface values near 1.75 ppmv in the region of interest. Above 400 K, the methane starts to decrease again to 1.6 ppmv as the instrument enters the lower

20 tropical stratosphere. From 370 K to 390 K, the temperature is nearly constant like for SF2; nevertheless, above 390 K, we observe a cooling up to the cold point tropopause. This cooling could be associated with the nearly adiabatic vertical transport by convection. But with the assumption of a convective transport close in time and location of the SF4 flight, the ozone mixing ratio would be expected to decrease because the air

25 masses originating from the lower troposphere have lower ozone mixing ratios. The absence of a clear feature in ozone could suggest that the transport by convection of moist tropospheric air had occurred significantly before the flight, leading to a partial mixing of this air with its environment. Another potential explanation for this insignificant signature is a local photochemical production of ozone related to the transport of ozone

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precursors by convection and the production of NO$_x$ by lightning (Rivière et al., 2006). Whatever the explanation is, mixing or photochemical production, it is likely that there was some delay between the time of the in situ measurements and the occurrence of the convective event causing the vertical elevation of the moist tropospheric air mass. This delay must be long enough for these processes, mixing or ozone photochemical production, to be effective. Hence, to support this hypothesis, it is required to calculate backwards trajectories in the TTL over a few days corresponding to the period when the SACZ was over the Bauru region. For this purpose, a meso-scale model with sufficient vertical resolution and based on local meteorology is used in Huret et al. (2006)[2].

4.3 Comparison with mid-latitude H$_2$O data

A further insight for the occurrence of a convective transport of H$_2$O into the TTL for SF4 can be found in the comparison between HIBISCUS data and previous observations at mid-latitudes H$_2$O measurements. Indeed, the H$_2$O mixing ratios reported by micro SDLA in the TTL in this flight are comparable to that observed at mid-latitudes at identical potential temperature levels. To illustrate this point, Fig. 9 displays the measured H$_2$O data in the UT-LS, superimposed to previous northern mid-latitudes H$_2$O measurements from the balloonborne SDLA laser hygrometer (Durry et Megie, 1999) and from the ELHYSA frost-point hygrometer (Ovarlez, 1991) at Aire sur l'Adour (44˚ N, France). Above the tropopause, at about 360 K, the mid-latitude observations display the seasonal cycle of water vapor (Randel et al., 2001), the lower stratosphere being more moist in autumn compared to spring. Although the HIBISCUS measurements were performed in the Southern Hemisphere, the small difference between both hemispheres makes the comparison valid. Up to 380 K, the two tropical profiles, recorded in summer, are close to the mid-latitude autumn data. Above this level, they shift between spring and autumn mid-latitude concentrations. The SF2 layers and the driest layer in SF4 (3.5–4 ppmv) are close to spring mid-latitude values while the humidity of the moist layers in SF4 (∼5 ppmv, between 390 K to 400 K) is close to that of the autumn mid-latitude profiles. If the moist layers in SF4, between 390 K and 400 K,

were due to convection as discussed above, this comparison would suggest that the increased humidity in the autumn could be the result of re-hydration of the lower stratosphere in the tropics during the convective summer, followed by isentropic exchange with the mid-latitudes.

5   Conclusions

High resolution H_2O measurements have been performed by the microSDLA balloon-borne laser hygrometer in the UT, the TTL and the LS over Brazil (22° S) during the HIBISCUS field campaign in February 2004. The combination of balloon flights descending slowly in the TTL and the high resolution of microSDLA has allowed observing thin layering in the H_2O profiles in the UT, TTL and LS. Simultaneous or quasi-simultaneous CH_4, O_3 and temperature measurements and PV-profiles provides insight on the origin of the strong layering.

In the UT for SF4, the unambiguous signature of a filament of extratropical stratospheric air, crossed by the sensor was identified near 10 km (340 K), corresponding to a very dry layer, with low CH_4 and high O_3. In the LS, for SF2, in the 17 km to 18 km range (385 K to 405 K) the instrument crossed a tongue of high-PV stratospheric mid-latitude air intruding the tropical LS. A very good agreement was found between the predictions of the MIMOSA PV-advection model in the TTL and LS and the in situ tracer measurements: the MIMOSA model predicts a strong increase in the PV when low CH_4 and high O_3 are detected. The corresponding H_2O data are typical of mid-latitude stratospheric values and range from 3.5 ppmv to 5 ppmv. No significant dehydration by ice nucleation and subsequent sedimentation was detected in the TTL and LS, likely because significant undersaturated conditions occur. For the second flight (SF4), in the same altitude range, a high H_2O mixing ratio of 5.5 ppmv is observed, as well as a cooling and an enrichment of methane. It could be related to a convective transport of tropospheric air moistening the subsaturated TTL in the 385 K–405 K range. Compared to previous measurements of water vapour in the UTLS in oceanic conditions (Vömel et al, 2002;

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Jensen et al., 2005; Richard et al., 2006), the micro-SDLA observations exhibit larger H_2O mixing ratios in the LS. This is consistent with the warmer temperatures measured at and around the cold point tropopause by micro-SDLA, leading to less dehydration. This seems to indicate that the tropical LS is moister over continent than over ocean. This suggests further that the land convective systems could be a source of stratospheric moisture in the summer season. Nevertheless, more measurements in the LS in continental conditions are needed to confirm this result.

The examination of the TTL based on the two flights of the HIBISCUS campaign underlines the complexity of the processes controlling H_2O in the UT-LS. Within the 10 days separating both flights of the micro-SDLA, at the same isentropic levels, between 385 K and 405 K, tongues of stratospheric mid-latitude air, followed by convective transport, have largely modified the H_2O profiles. The relatively warm TTL, over the land convective region of the South Atlantic Convergence Zone, appears as a highly structured layer of mixing between air parcels advected isentropically from the mid-latitudes lower stratosphere, the overworld of Holton et al. (1995), and convectively lifted tropospheric air.

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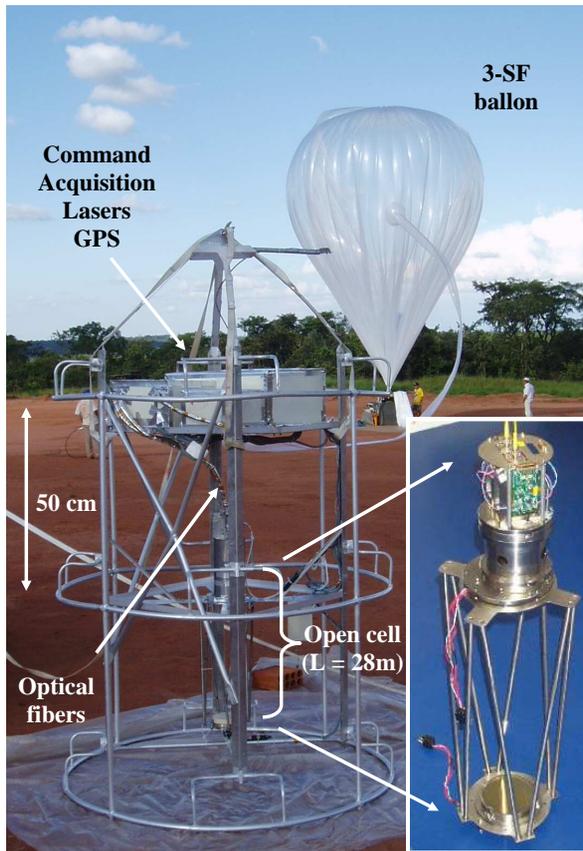
Fig. 1. The micro-SDLA diode laser spectrometer during HIBISCUS in Bauru, Brazil in February 2004. With an overall weight of less than 40 kg, the micro-SDLA sensor is flown from small-size 3000 m^3 balloons to reach a float altitude near 22 km.

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(a)

(b)
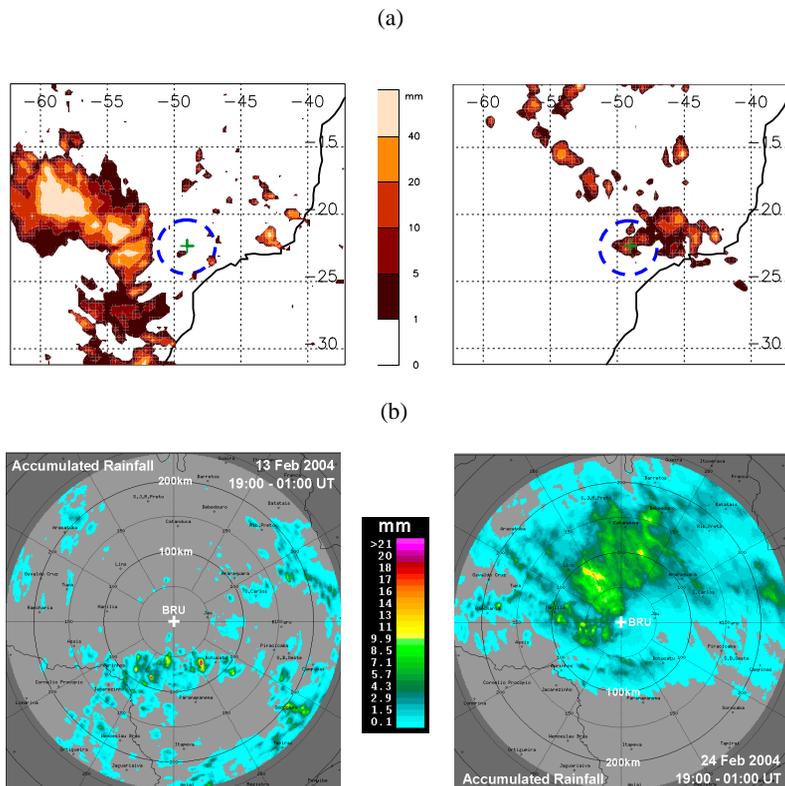
Fig. 2. (a) TRMM surface rainfall in mm accumulated from 19:30 UT on 13 February 2004 to 01:30 UT on 14 February 2004 (left panel, SF2 flight) and from 22:30 UT on 24 February 2004 to 01:30 UT on 25 February 2004 (right panel, SF4 flight). The green cross corresponds to the location of the Bauru launch site. The blue circle indicates approximately the 200 km range of the Bauru Radar (BRU). **(b)** 6-h accumulated radar rainfall within a 240 km range from Bauru from 19:00 UT (about one hour before launch) until the end of the balloon flights at 01:00 UT on the following day. Left: SF2 on 13 February 2004. Right: SF4 on 24 February 2004.

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Fig. 3. Temperature (micro-SDLA raw and smoothed data), temperature gradients and ozone profiles for both flights, SF2 (in blue) on 13 February and SF4 (in red) on 24 February 2004.

Fig. 4. In situ vertical concentration profiles of H_2O and CH_4 yielded by the micro-SDLA instrument during HIBISCUS, on 13 February (SF2, **(a)**) and on 24 February (SF4, **(b)**) 2004. Additional tropospheric soundings with RS-90 and RS-80 sondes are also displayed.

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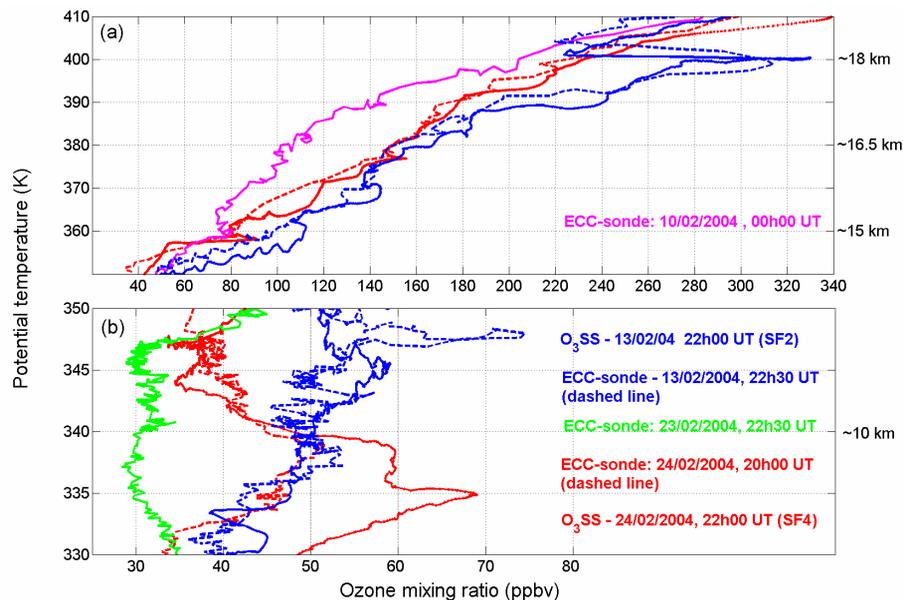
Fig. 5. Ozone measurements in the TTL and LS **(a)** and the UT **(b)** obtained with ECC balloonborne sondes and with the OSSS-sensor (piggy-back in the micro-SDLA flight chain) at the time and place of the micro-SF2 and SF4 flights. Two soundings, on 10 and 23 February 2004 are displayed and used as references for O$_3$ in the LS, the TTL and the UT.

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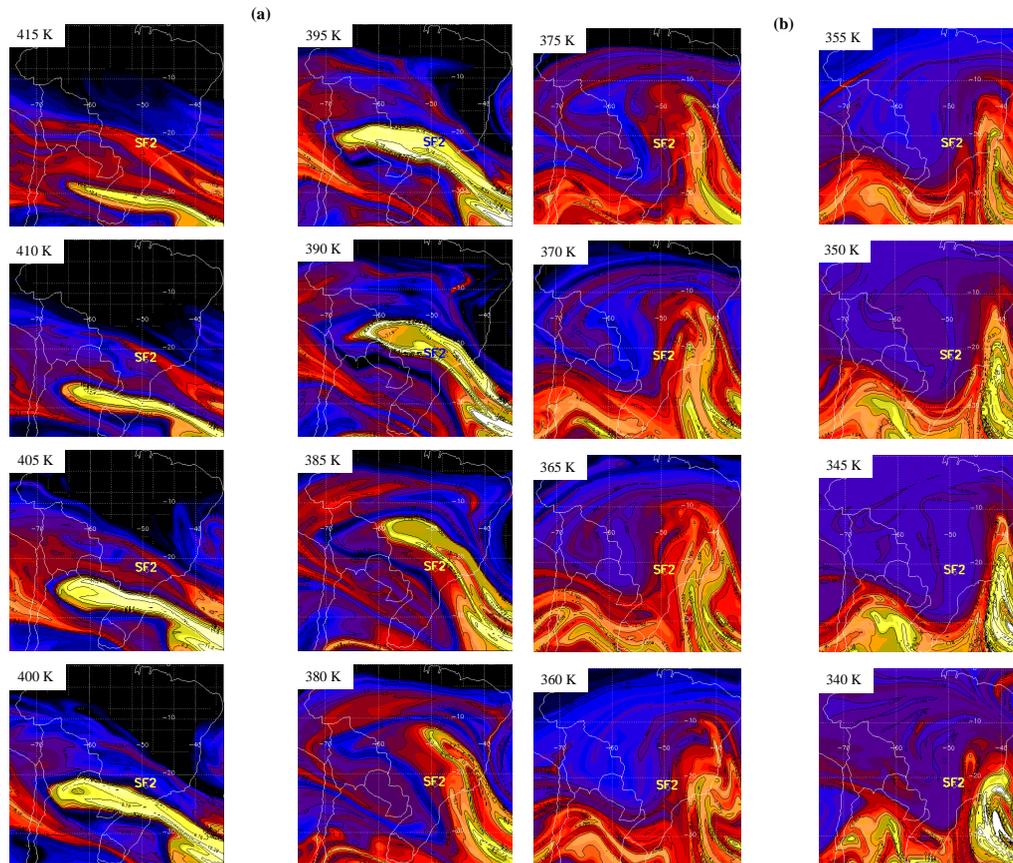
Fig. 6. Distribution of the Potential Vorticity predicted by the MIMOSA with a 5 K resolution in the UT, the TTL and the LS for the SF2 flight on 13 February 2004. The micro-SDLA location in each isentropic level is indicated with the mark SF2. Contours are of potential vorticity (10^{-8} K m^2 s^{-1} kg^{-1}).

(a)　　　　　**(b)**
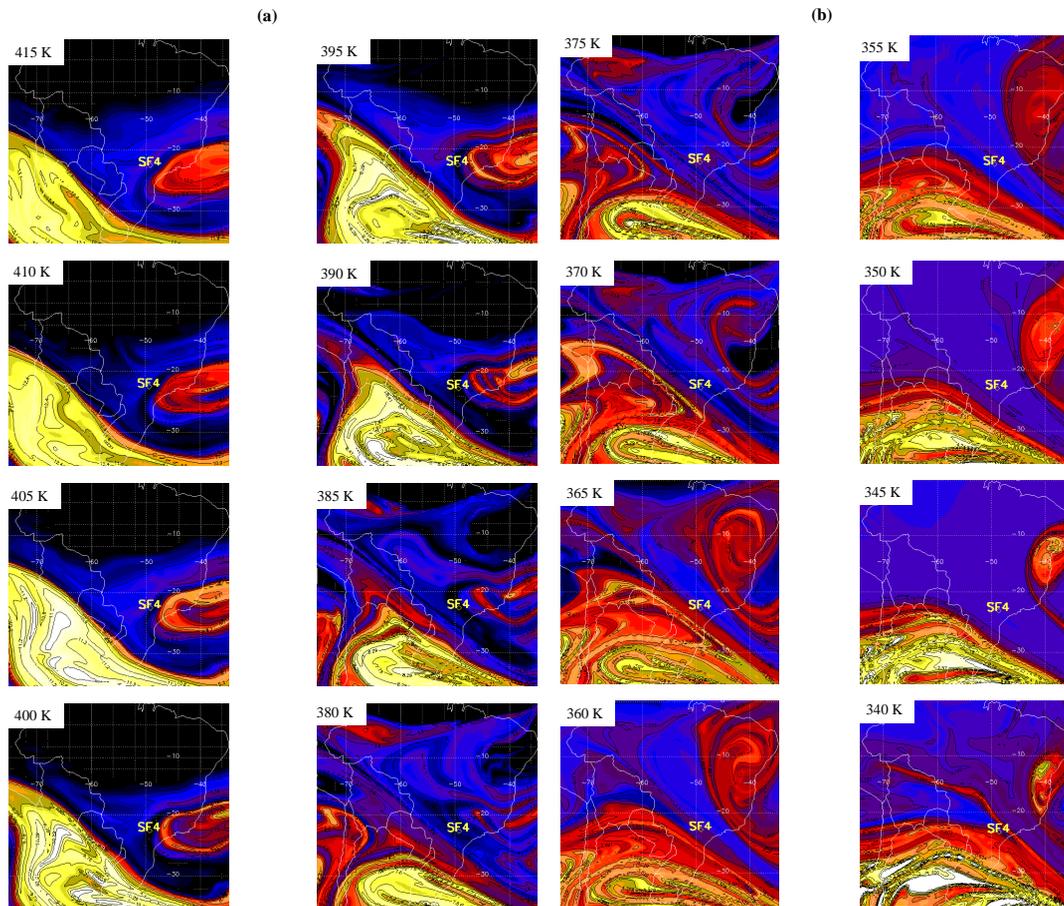
Fig. 7. Same as in Fig. 6 but for the SF4 flight on 24 February 2004.

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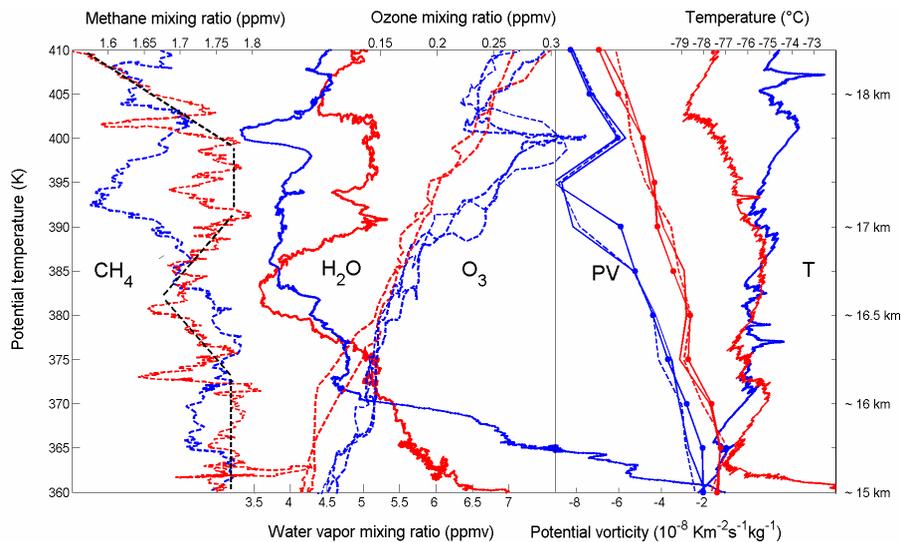
Fig. 8. A zoom on the CH$_4$, H$_2$O, O$_3$ and temperature measurements achieved in the TTL for the SF4 (in red) and SF2 (in blue) flights. The PV predicted by MIMOSA is also added on the figure. Both the potential temperature and altitude vertical scales are indicated.
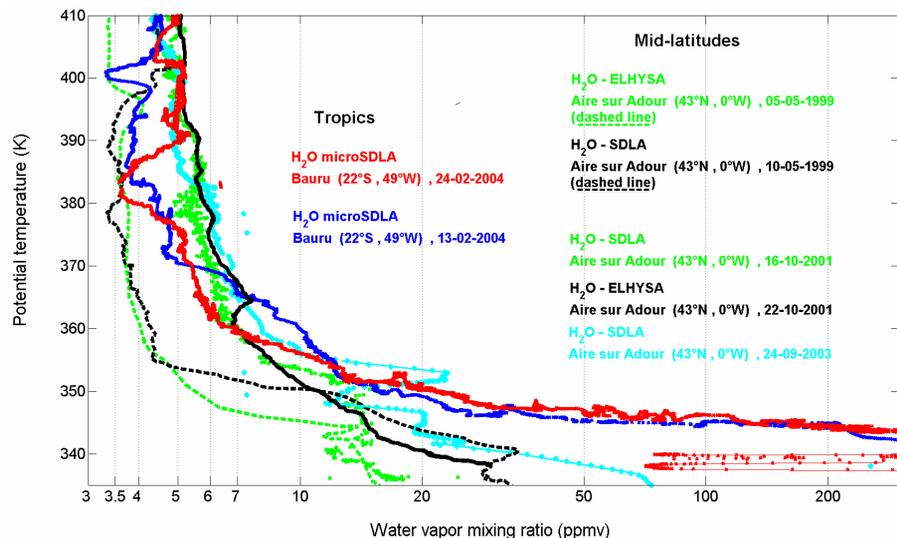
Fig. 9. The H$_2$O profiles obtained during HIBISCUS (in red and blue) superimposed to previous mid-latitudes H$_2$O determinations from the SDLA and the ELHYSA hygrometers. See text for more details.