Intrusion of recent air in midlatitude stratosphere revealed by in situ tracer measurements and trajectory calculations

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[1] In the midlatitude stratosphere the mean circulation is downward leading to relatively old air. Mixing between this old air and more recent air from the troposphere or the tropical stratosphere can nevertheless occur. In particular, mixing between the lower part of the tropical stratospheric reservoir and the midlatitude stratosphere was reported in the so-called ''tropically controlled transition region'' which extends from the tropics to midlatitudes between the isentropic surfaces 380 K and 450 K (15–18 km altitude). O_3/CO and HCl/O_3 relations derived from the measurements of the in situ tunable lasers (TDLAS) balloon-borne instrument Spectroscopie Infrarouge par Absorption de Lasers Embarqués (SPIRALE), and 3D air parcels trajectories are used to analyze the characteristics of this region at the time and location of SPIRALE. These measurements took place above Aire sur l'Adour (43.6°N, 0°E) in October 2002. Two-month 3D backward trajectories ending around each individual measurement recorded with a time step of 1 s are computed on the basis of European Centre for Medium-Range Weather Forecasts (ECMWF) wind fields. The region is found to extend from 380 K to 465 K, and mixing is found to take place 1 to 2 months before the SPIRALE measurements. The region is composed of two layers with different characteristics. In the 380–405 K layer the recent air is uplifted from below the 380 K isentropic surface, and the percentage of recent air is high (minimum estimate 32%). In the 405–465 K layer the origin of recent air is the tropical stratosphere, and the percentage of the recent air is lower (minimum estimate 12%).

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1. Introduction

[2] According to *Hoskins* [1991], the midlatitude lower stratosphere can be divided into the overworld and the lowermost stratosphere. The overworld is the region above the 380 K isentropic surface. This surface corresponds approximately to the tropical tropopause. The lowermost stratosphere is located below the 380 K isentropic surface and above the midlatitude tropopause. In the whole midlatitude stratosphere the mean circulation is downward [*Holton et al.*, 1995] leading to the presence of relatively old air (a few years) which has been lifted in the tropics and then pushed poleward and downward at mid and high latitudes. This simple view is nevertheless complicated by mixing of midlatitude stratospheric air with more recent air coming from the tropical stratosphere or from the troposphere.

[3] In the lowermost stratosphere, recent air is coming either from the midlatitude troposphere [*Poulida et al.*, 1996; *Lelieveld et al.*, 1997; *Jost et al.*, 2004] because of meteorological perturbations or forest fire plumes, or from the tropical troposphere along the isentropic surfaces [*Ray et al.*, 1999; *Stohl et al.*, 2003] across the tropopause.

[4] In the midlatitude overworld a large number of observations [*Durry et al.* 2002; *Portafaix et al.*, 2003; *Grant et al.*, 1998] and modeling studies [*Orsolini and Grant*, 2000] report on laminae from the tropical stratospheric reservoir (TSR) into the midlatitude stratosphere. These laminae lead to irreversible mixing between old midlatitude stratospheric air and more recent air from the tropics across the subtropical barrier. This subtropical barrier seems to vanish in the lower part of the overworld leading to more mixing between the tropical and the midlatitude air. Indeed, *Grant et al.* [1998] indicate that the exchanges between the midlatitudes and the tropics are mainly located below 21 km. Aerosol [*Grant et al.*, 1994; *Hitchman et al.*, 1994] and water vapor measurements [*Rosenlof et al.*, 1997; *Randel et al.*, 2001] also suggest a large two-way transport between the lower part of the TSR and the midlatitude stratosphere. Model calculations [*Douglass et al.*, 1996] confirm that transport between the tropics and the midlatitudes decreases above 20 km. *Rosenlof et al.* [1997] point out a tropically controlled transition region which extends from the tropics to

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midlatitude above the top of the lowermost stratosphere (380 K) up to the 450 K potential surface (approximately 18 km).

[5] Mixing processes can be studied in using tracer correlations [*Plumb and Ko*, 1992; *Plumb et al.*, 2000; *Morgenstern et al.*, 2002; *Jost et al.*, 2002; *Ray et al.*, 2002]. Since dynamical barriers limit the exchange between the stratospheric regions, tracer correlations in the tropical stratospheric reservoir, in the midlatitude stratosphere and in the polar vortex are different. Typical tracer/tracer relations for each region have been established [see, e.g., *Michelsen et al.*, 1998b]. Such relations are available for background conditions for which exchanges between the regions are small. If significant exchange occurs, "anomalous" tracer/tracer relations are expected.

[6] *Huret et al.* [2006] presented in situ measurements of N_2O and CH_4 mixing ratios by the tunable lasers (TDLAS) balloon-borne instrument Spectroscopie Infrarouge par Absorption de Lasers Embarqués (SPIRALE) at midlatitude above Aire sur l'Adour (43.6°N, 0°E) on 2 October 2002 and at high latitude above Kiruna (68N, 20E) on 21 January 2003. They showed that the correlation of the abundance of these two species can be used to study the large exchanges of air masses across the subtropical barrier and across the Arctic polar vortex edge. The CH_4/N_2O relation derived from the SPIRALE midlatitude flight shows some anomalous behavior relatively to the typical relations derived from the measurements of ATMOS [*Michelsen et al.*, 1998a] corrected for the year 2002. The main differences are observed in the region between the tropopause and the potential temperature surface 480 K (20 km). Taking as the reference, the CH_4/N_2O relation derived from the outside vortex measurements of the high-latitude flight of SPIRALE, *Huret et al.* [2006] concluded that the air masses sampled by SPIRALE at midlatitude from the tropopause up to 480 K are due to irreversible mixing following the isentropic intrusion of recent air from the tropics into the midlatitudes. Such observations are similar to those reported by *Ray et al.* [1999] and *Stohl et al.* [2003] in the lowermost stratosphere and by *Grant et al.* [1994], *Rosenlof et al.* [1997] and *Randel et al.* [2001] in the tropically controlled transition region.

[7] CH_4/N_2O relations close to the SPIRALE anomalous relation have been reported by other groups [*Engel et al.*, 1996; *Herman et al.*, 1998; *Michelsen et al.*, 1999; *Kanzawa et al.*, 2003] in the tropically controlled transition region. No attempts have been made in these papers to document this region more. The aim of the present paper is to take advantage of the very high spatial resolution of the TDLAS balloon-borne SPIRALE instrument to analyze the characteristics of this region at the time and at the location of the SPIRALE measurements.

[8] The O_3/CO and HCl/O_3 relations derived from the SPIRALE flight on 2 October 2002 and a trajectory analysis are used to this goal. The O_3/CO relation was used by *Hoor et al.* [2002] to study mixing in the extratropical tropopause layer (EXTL) which extends from the local tropopause to 330–360 K. This relation is used here to study mixing in the tropically controlled transition region. The HCl/O_3 relation was used by *Marcy et al.* [2004] to quantify the stratospheric ozone intrusions in the troposphere. It is used in the present paper to show evidences of mixing between recent air and midlatitude stratospheric air. The respective 3 months and 1 month photochemical lifetimes of CO and HCl in the lower stratosphere enable us to estimate the approximate time it takes to the mixed air parcels to reach the location of SPIRALE in the midlatitude lower stratosphere. Trajectory calculations are also used to confirm these time values and to find the origin of the recent air.

2. Instrument and Trajectory Model

2.1. SPIRALE Instrument

[9] Spectroscopie Infrarouge par Absorption de Lasers Embarqués (SPIRALE) is an Infrared spectrometer with six tunable lasers (TDLAS) devoted to in situ measurements of atmospheric tracers and active species from 10 km to 35 km altitude. A detailed description and the performances of this balloon-borne instrument are given by *Moreau et al.* [2005]. Vertical profiles of the species are obtained at the ascent and at the descent. SPIRALE has a very fine time resolution (1 s), a very good accuracy (10% for CO, 6% for O_3 and 13% for HCl in the range 18–23 km) and a low detection limit (20 pptv for HCl in general and 1 ppbv for N_2O for example).

2.2. Measurements of SPIRALE in the Lower Part of the Midlatitude Overworld on 2 October 2002

[10] SPIRALE flew in the morning of 2 October 2002 above Aire sur l'Adour (43.6°N, 0°E) in the frame of the ENVISAT validation program. Vertical profiles of CH_4 and N_2O were measured from 12 to 33 km at the ascent of the balloon. These data were used by *Huret et al.* [2006]. Vertical profiles of CO, O_3 and HCl were measured at the descent of the balloon down to 14.5 km (375 K). The latter profiles are used in the present paper to study the lower part of the overworld (380–480 K) where the tropically controlled transition region is located. As shown by *Huret et al.* [2006] and in the supplementary material (Figure S1), potential vorticity (PV) maps indicate that in this potential temperature range the SPIRALE profile is not include in any laminae from the tropical reservoir.[1]

2.3. Trajectory Model

[11] Reverse integrations of trajectories initialized along each transect have been performed with TRACZILLA [*Legras et al.*, 2003; *Pisso and Legras*, 2007], a modified version of FLEXPART [*Stohl et al.*, 2005] which uses European Centre for Medium-Range Weather Forecasts (ECMWF) winds at 1° horizontal resolution and on 60 hybrid levels with 3 h resolution obtained by combining analysis available every 6 h with short time forecasts at intermediate times (3 h resolution). The modifications from FLEXPART advection scheme consists mainly in discarding the intermediate terrain following coordinate system and in performing a direct vertical interpolation of winds, linear in log pressure, from hybrid levels. The vertical velocities used in this work are computed by the FLEXPART preprocessor using a mass conserving scheme in the hybrid ECMWF coordinates as in the ECMWF model. Other schemes are possible, like diabatic vertical velocities currently under

[1] Auxiliary materials are available in the HTML. doi:10.1029/2007JD009188.

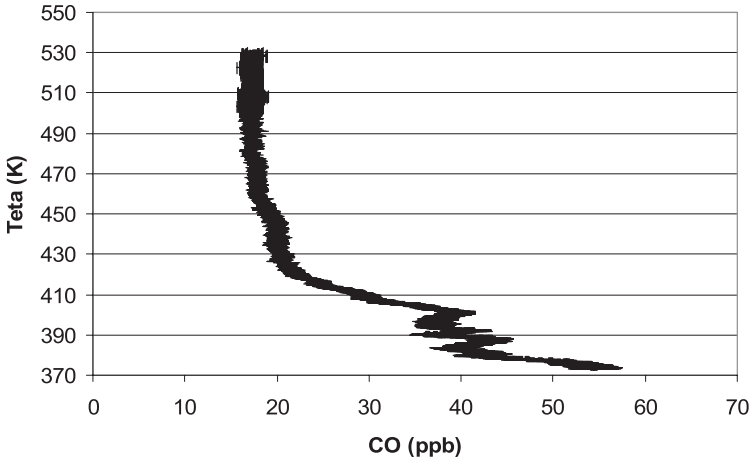

Figure 1. CO mixing ratio profile obtained during the descending phase of the Spectroscopie Infrarouge par Absorption de Lasers Embarqués (SPIRALE) flight on 2 October 2002 at Aire sur l'Adour as a function of potential temperature in the range 375–530 K (14.5–21 km). Error bars on CO mixing ratio are shown on each data point.

development. The model uses a fixed time step of dt = 900 s. Time lag between particle releases is 1 s along the balloon track. We use the balloon GPS data and the onboard pressure sensor to locate the launching point of each parcel horizontally and vertically, respectively.

3. Results

3.1. CO Profile in the Lower Stratosphere

[12] In the midlatitude stratosphere, the Brewer-Dobson circulation is downward. CO is produced by oxidation of methane. CO mixing ratio decreases when altitude decreases since the sink of CO by reaction with OH is more efficient than the temperature-dependent CO production at the low temperatures prevailing in the lower stratosphere [*Hoor et al.*, 2002]. According to *Hoor et al.* [2002] the CO mixing ratio reaches an almost constant value (typically 10–15 ppb) within the lowermost stratosphere. In the troposphere CO is highly variable with mixing ratios ranging from 60 ppbv to 200 ppbv [*Novelli et al.*, 1992], with peaks reaching 800 ppbv [*Nedelec et al.*, 2005]. CO mixing ratio values greater than approximately 10–15 ppbv in the midlatitude stratosphere may be therefore interpreted as mixing between recent air and midlatitude air [*Hoor et al.*, 2002].

[13] Figure 1 shows the profile of CO measured by SPIRALE on 2 October 2002 at midlatitude as a function of potential temperature ranging from 375 K to 530 K. CO mixing ratios are very close to the midlatitude stratosphere typical values (10–15 ppbv) in the upper part of the profile (17.4 ppbv ± 1.7 ppbv). CO mixing ratios are greater than these typical values in the lower part of the profile meaning that mixing occurs.

3.2. Correlation of the Abundance of CO and O_3

[14] Unlike CO, O_3 is mainly produced in the stratosphere. Its mixing ratio is therefore strongly decreasing with altitude. The idealized relation O_3/CO assuming no mixing have therefore an "L-shape" as shown in Figure 2 [see also *Hoor et al.*, 2002, Figure 2]. The vertical line of the L-shape curve is the stratospheric branch. It intercepts the CO axis at the stratospheric CO low value (see Figure 2). In this ideal case the CO mixing ratio increases sharply at the tropopause to the tropospheric high values. This steep increase is the horizontal line of the L shape curve assuming a constant low O_3 mixing ratio at the tropopause. According to *Fischer et al.* [2000] irreversible mixing between "old" midlatitude air and recent tropospheric air is represented by mixing lines [*Waugh et al.*, 1997] which connect a tropospheric "end member" on the horizontal line of the L shape curve to a stratospheric end member on the vertical line (Figure 2). The tropospheric end member depends on the ozone and CO mixing ratio of tropospheric air which is mixed with stratospheric air. This end member may be highly variable depending on the time and location of the entry point of tropospheric air. A set of data measured in a mixing region may therefore exhibit several mixing lines. Two possible mixing lines are shown in Figure 2. Such a mixing is generally assumed to take place on isentropic surfaces across the subtropical tropopause between the tropical troposphere and the midlatitude lowermost stratosphere in the potential temperature interval 330–360 K [*Fischer et al.*, 2000, *Hoor et al.*, 2002].

[15] Figure 3a shows the O_3/CO relation derived from the measurements of SPIRALE. For ozone mixing ratios between 1.62 ppmv and 3 ppmv the CO mixing ratio has a very constant value of 17.4 pppv ±1 ppbv. This corresponds to potential temperatures from 465 K to 530 K (Figure 1). According to the previous discussion (section 3.1), air sampled by SPIRALE in this altitude range is pure midlatitude stratospheric air. It represents the stratospheric branch of the L shape idealized curve. Below 465 K, CO mixing ratio increases up to 55 ppbv at 375 K while altitude decreases. The shape of this curve is consistent with the relation shown by *Hoor et al.* [2002] but the increase of the CO mixing ratio is observed in the lower stratosphere (below 465 K) instead of in the lowermost stratosphere (below 360 K) by *Hoor et al.* [2002]. Thus, it is likely that the O_3/CO relation derived from SPIRALE results from the

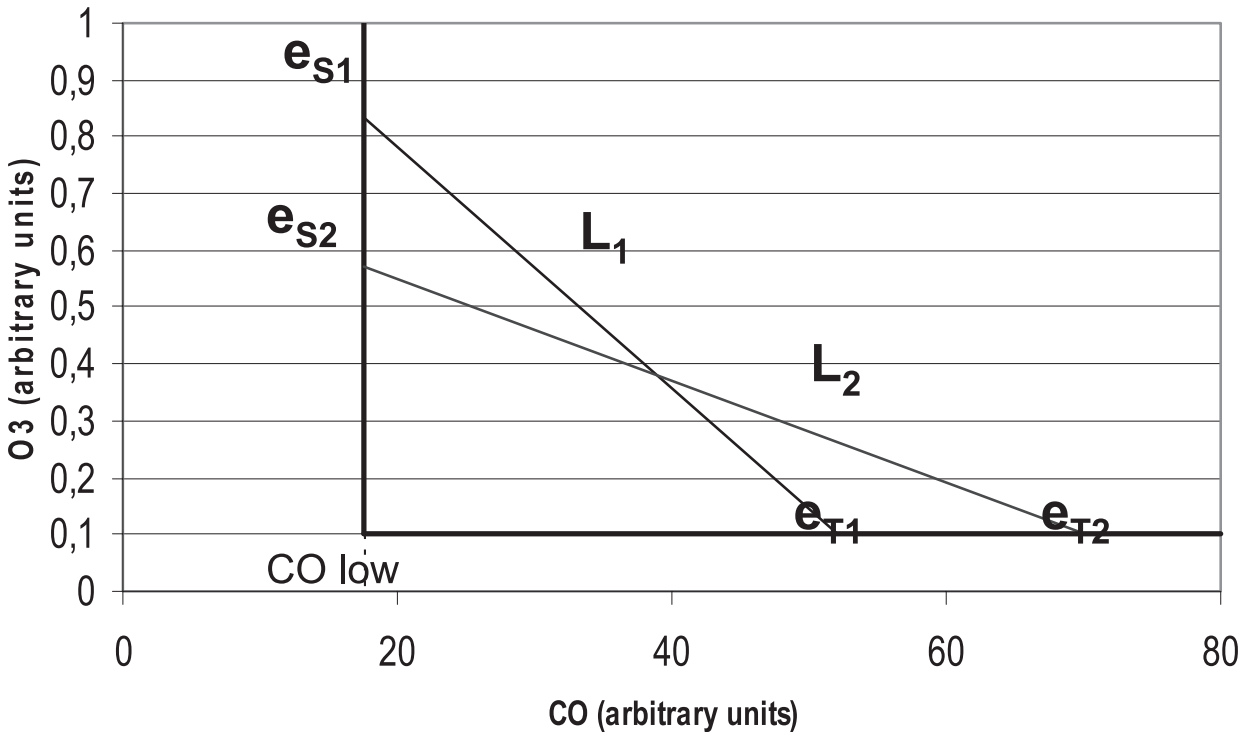

Figure 2. L shape idealized O_3/CO relation (thick solid line). The vertical line of the "L" represents the O_3 mixing ratio as a function of CO in the stratosphere. The horizontal line of the L represents the steep increase of CO at the tropopause. Thin solid lines L_1 and L_2 are examples of mixing lines; e_{S1} and e_{T1} are the stratospheric and the tropospheric end members, respectively, of the mixing line L_1; e_{S2} and e_{T2} are the stratospheric and the tropospheric end members, respectively, of the mixing line L_2.

isentropic mixing between tropical lower-stratospheric air (instead of tropical tropospheric air by *Hoor et al.* [2002]) and midlatitude lower-stratospheric air (instead of lowermost-stratospheric air by *Hoor et al.* [2002]) across the subtropical PV barrier in the lower stratosphere (instead of the subtropical tropopause by *Hoor et al.* [2002]).

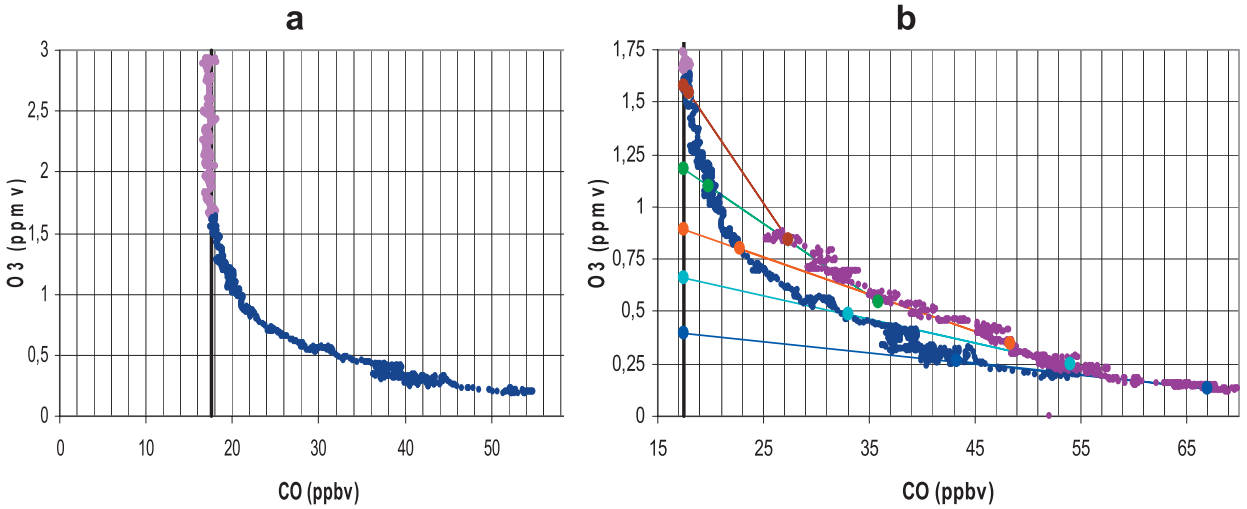

Figure 3. (a and b) Vertical line of the L shape curve (black solid line); O_3/CO relation derived from measurements of SPIRALE on 2 October 2002. Light purple data points are in the range 465–530 K; dark blue data points are in the range 375–465 K. (Figure 3b) O_3/CO relation derived from measurements of SPIRALE on 22 June 2005 above Teresina, Brazil (5°S) (purple full circles); mixing lines on the isentropic surfaces 460 K (brown solid line), 440 K (green solid line), 420 K (red solid line), 405 K (light blue solid line), and 380 K (dark blue solid line). For each mixing line, the midlatitude end member (on the vertical line of the L), the tropical end member (on the tropical O_3/CO relation), and the resulting mixing point (on the midlatitude O_3/CO relation) are shown (full large circles).

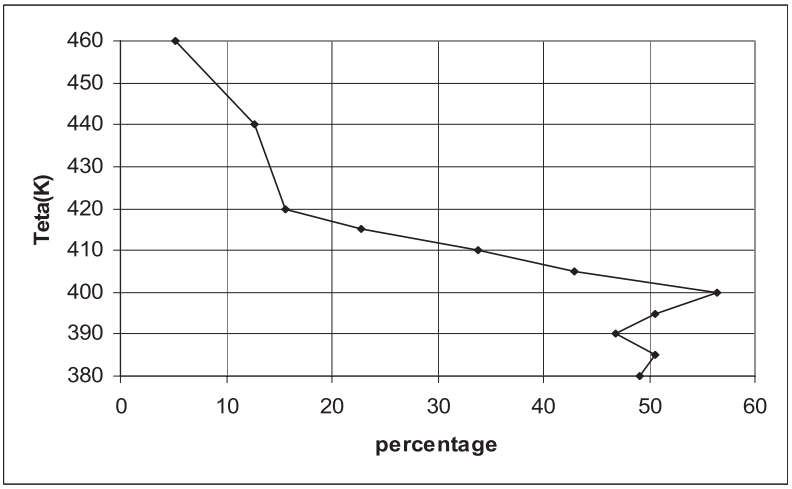

Figure 4. Percentage of recent air in the mixed parcels sampled by SPIRALE on 2 October 2002 as a function of potential temperature.

[16] In the SPIRALE case, the irreversible mixing between the old midlatitude air and the recent air must be therefore represented by mixing lines that connect midlatitude stratospheric end members located on the vertical line of the L shape curve as on Figure 2 to tropical end members. The location of these tropical end members is no more the horizontal line of the L shape curve. It must be an O_3/CO relation curve typical of the tropical stratosphere. This relation is not available from measurements for our case study. Instead, the measurements in the tropical stratosphere of the SPIRALE instrument in June 2005 above Teresina in Brazil (5°S) have been used for the present analysis. This can be justified by the fact that the SPIRALE instrument was flown well in the tropical latitude band far away from the midlatitudes. These measurements are shown in Figure 3b (purple circles) and exhibit a different O_3/CO relation with higher values of CO for any O_3 values compared to the midlatitude stratospheric SPIRALE measurements. This result is consistent with the fact that, in the tropics, CO mixing ratio decreases more smoothly as a function of altitude than in the midlatitude stratosphere [*Flocke et al.*, 1999].

[17] In Figure 3b, each point of the O_3/CO relation derived from measurements of SPIRALE at midlatitude below 465 K (O_3 mixing ratio lower than 1.62 ppmv) is located on a mixing line between a midlatitude stratospheric end member and a tropical stratospheric end member. Mixing lines on isentropic surfaces 460 K, 440 K, 420 K, 405 K and 380 K are shown in Figure 3b. The percentage of recent air in the mixed air parcels as a function of potential temperature can be calculated for each mixing line from the relative distance of the midlatitude SPIRALE measurements to the end members. Figure 4 shows the result of these calculations. Above 415 K, the percentage of recent air in the mixed air parcels is between 0 and 20%. Below 405 K it is between 40 and 60% showing a sharp increase from 420 K to 400 K.

[18] The O_3/CO relation confirms that SPIRALE has sampled midlatitude stratospheric air mixed with more recent air on 2 October 2002 [*Huret et al.*, 2006]. Nevertheless, the top of the mixing layer (465 K, 19.3 km) is below the top of the mixing layer (480 K, 20 km) derived from the relation CH_4/N_2O [*Huret et al.*, 2006]. It is likely that between 465 K and 480 K the air is recent but older than the photochemical lifetime of CO in midlatitude lower stratosphere. This could explain the decrease of the CO abundance in the mixed layer down to the typical stratospheric values. Loss of CO in the lower stratosphere is mainly due to its reaction with OH. Assuming an OH concentration of 8×10^5 cm^{-3} [*Flocke et al.*, 1999] the photochemical lifetime of CO in the midlatitude stratosphere is 3 months [*Sander et al.*, 2002]. It is likely therefore that the mixing of recent air with midlatitude stratospheric air in the layer 465–480 K takes place 3 months or more before the mixed air parcels reach the SPIRALE location. In the layers below the mixing is probably younger ranging from 0 to 3 months. The percentage of midlatitude stratospheric air in the mixed air parcels sampled by SPIRALE from 380 K to 465 K is found to be between 0 and 20% above 415 K and between 40 and 60% below 405 K. One has to note nevertheless that these values are calculated assuming a realistic but not the actual O_3/CO relation in the tropics for the considered case study. The air parcels sampled by SPIRALE are expected to be a mixing between air parcels which have left the tropical stratosphere between 0 and approximately 3 months. According to *Schoeberl et al.* [2006], the CO zonal mean mixing ratio measured by Aura-MLS in the latitude interval ±12° varies by about 8 ppbv over 3 months from the end of June to the beginning of October in the potential temperature range of interest (380–465 K). The SPIRALE profile used as the tropical end member for the calculations of the percentage of recent air in the mixed air parcels was measured at the end of June. Depending on the time of entry of the tropical air in the midlatitude stratosphere the actual tropical end member could be therefore between 0 and 8 ppbv smaller than the one used in this study. This would result in a negative bias of the computed percentage of recent air assuming the tropical SPIRALE profile as the tropical end member. Superimposed to the seasonal variation of the stratospheric tropical CO profile, local or/and day-to-day variations of the actual CO tropical profile are

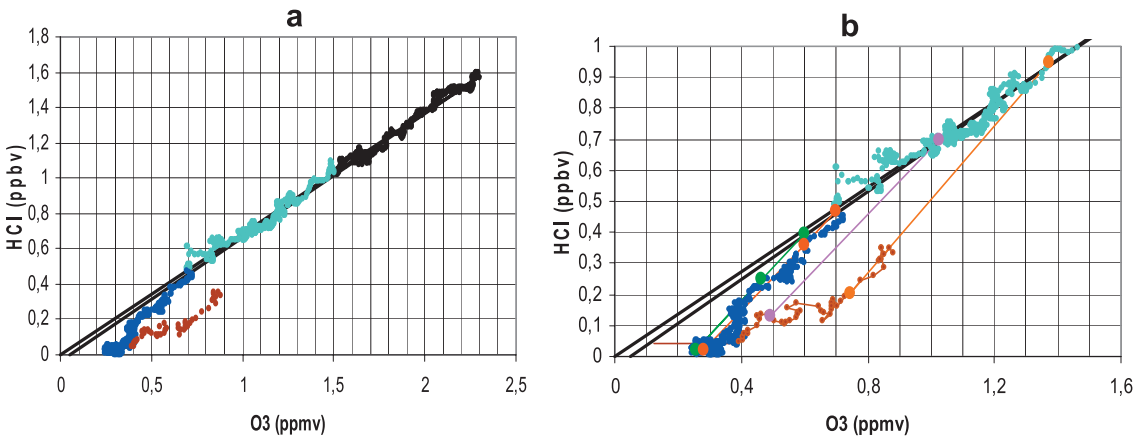

Figure 5. (a and b) HCl mixing ratio (ppbv) as a function of O_3 mixing ratio (ppmv) measured by SPIRALE on 2 October 2002 at Aire sur l'Adour during the descending phase of the balloon flight, in the potential temperature ranges 380–415 K (dark blue full circles), 415–465 K (light blue full circles), and 465–500 K (black full circles); linear fits of the data points above 465 K assuming 0 intercept and 50 ppbv intercept of the O_3 axis, respectively (black solid lines). They are assumed to be the location of the midlatitude stratospheric end members; HCl mixing ratio (ppbv) as a function of O_3 mixing ratio measured by SPIRALE on 22 June 2005 above Teresina in the potential temperature interval 420–465 K (brown full circles). Below 0.4 ppmv ozone mixing ratio, the HCl mixing ratio is an upper limit (40 pptv). This curve is assumed to be the location of the tropical end members. (Figure 5b) Mixing lines on the isentropic surfaces 405 K (green thin solid line), 410 K (red thin solid line), 430 K (pink thin solid line), and 455 K (orange thin solid line). The large green, red, pink, and orange full circles show the location of the tropical end member, the midlatitude stratospheric end member, and the resulting mixing point for each mixing line.

also expected. The uncertainties on the percentage of recent air in the mixed air parcels computed above could therefore be relatively large and cannot be estimated.

3.3. Correlation of the Abundance of HCl and O_3

[19] *Marcy et al.* [2004] studied the relation between O_3 and HCl from both measurements and 3D model outputs. The measurements were made aboard the NASA WB-57F aircraft during two flights of the CRYSTAL-FACE campaign in Florida on July 2002, between 24°N and 39°N, from 11 to 18 km. HCl was measured with a high precision and a very low detection limit of 5 pptv by a Chemical Ionization Mass Spectrometry (CIMS) instrument. The model is the 3D CTM IMPACT with a full treatment of chemistry and transport processes [*Rotman et al.*, 2004]. Model results at latitudes of 26°N and 46°N are shown by *Marcy et al.* [2004]. Both observations and model results showed that the HCl/O_3 relation curve typical of the midlatitude lower stratosphere is linear and intercepts the O_3 axis at low values (<50 ppbv).

[20] Figure 5a shows the HCl/O_3 relation derived from measurements of SPIRALE between the 380 K and 500 K isentropic surfaces. As discussed in section 3.2, pure midlatitude stratospheric air was sampled by SPIRALE above 465 K. Two linear fits of the data points above 465 K are shown on Figure 5a. One of these fits assumes the intercept to be 0 and the second one assumes the intercept to be 50 ppbv. For the first one the slope of the linear fit is $6.84\ 10^{-4}$ with a very good correlation factor $R^2 = 0.9679$. For the second one the slope is $7.02\ 10^{-4}$ also with a very good correlation factor $R^2 = 0.9698$. The correlation of the abundance of O_3 and HCl in the midlatitude stratosphere is therefore linear and the linear fit intercepts the O_3 axis at a low O_3 mixing ratio value. This is in agreement with *Marcy et al.* [2004].

[21] In the potential temperature range 423–465 K (O_3 mixing ratio in between 0.9 ppmv and 1.5 ppmv) the midlatitude data points are onto the linear fits of the data points above 465 K (Figure 5a). On average, the data points are slightly above the linear fits in the altitude range 415–423 K (Figures 5a and 5b; O_3 mixing ratio in between 0.7 and 0.9 ppmv). In Figures 5a and 5b, we can notice a sharp decrease of the HCl mixing ratio for the O_3 mixing ratio below 0.7 ppmv. This sharp decrease takes place at the level 415 K. As by *Marcy et al.* [2004] we attribute this sharp decrease to dynamical mixing between stratospheric midlatitude air containing high HCl and more recent air containing low HCl. The relation HCl/O_3 derived from the measurements of SPIRALE instrument in June 2005 above Teresina in Brazil (5°S) is shown on Figure 5a. As in the troposphere [*Marcy et al.*, 2004], we observe that the HCl mixing ratio is very low in the tropical stratosphere relatively to midlatitude stratosphere. The more recent air which is mixed with the midlatitude stratospheric air may therefore originate from the troposphere or from the tropical stratosphere.

[22] The analysis made for O_3/CO relation has been repeated for the relation HCl/O_3 in the 380–465K layer to derive another evaluation of the percentage of recent air in the mixed air parcels sampled by SPIRALE. In this analysis the measurements of the SPIRALE instrument in June 2005 above Teresina in Brazil (5°S) are assumed as in section 3.2 to be the stratospheric tropical end members. Figure 5b shows

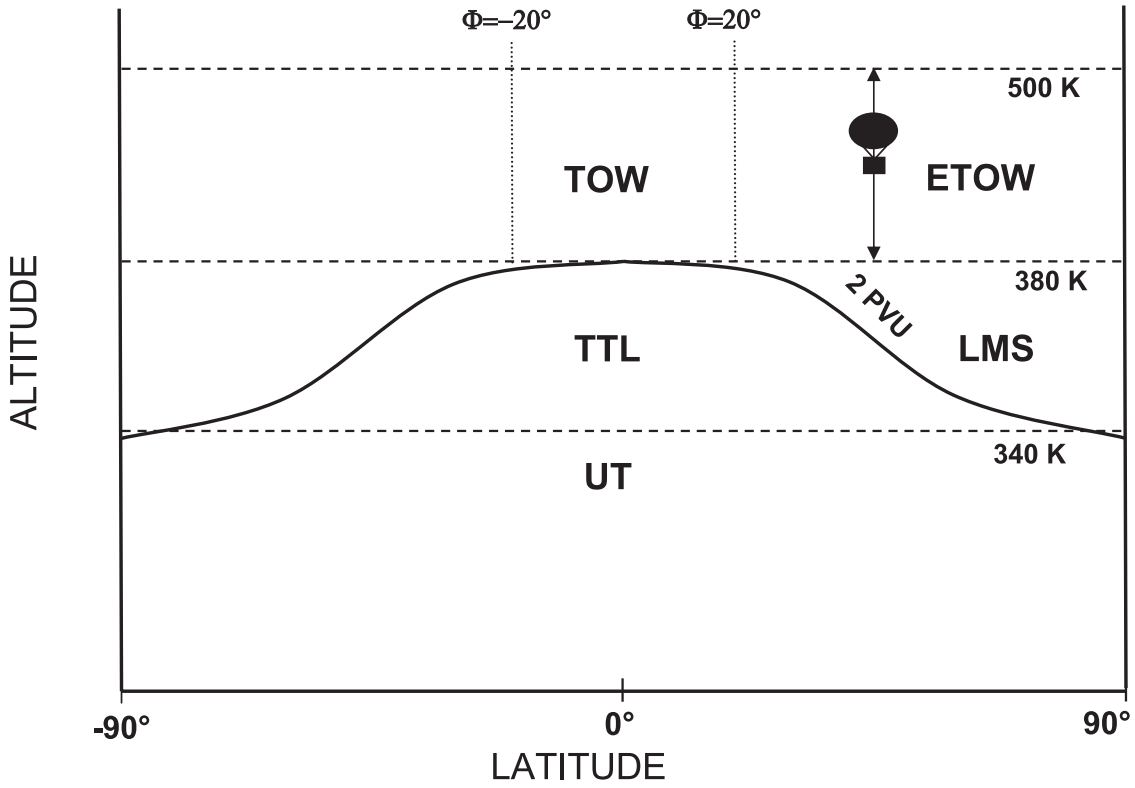

Figure 6. Schematic altitude-latitude cross section of the atmosphere. The Upper Troposphere (UT) region is assumed to be below the 340 K isentropic surface. The 2 PVU line is the tropopause. The lowermost stratosphere (LMS) and the tropical transition layer (TTL) are between the 340 K and 380 K isentropic surfaces. The PV are larger than 2 PVU in the LMS region and lower than 2 PVU in the TTL region. The tropical overworld (TOW) region and the extratropical overworld (ETOW) region are above the 380 K isentropic surface. The TOW region is defined by the (−20°; +20°) latitude band, and the ETOW region is for latitudes > +20°. The SPIRALE measurements used in the present paper are located in the lower part of the ETOW region (balloon symbol) on the figure.

the mixing lines on the isentropic surfaces 405 K, 410 K, 430 K and 455 K between the midlatitude stratospheric end members and the tropical stratospheric end members intercepting the SPIRALE measurements at midlatitude. Between 380 K and 400 K it is not possible to define such mixing lines. This shows that in this potential temperature interval mixing cannot be isentropic. From Figure 5b, the percentages of recent air in the mixed air parcels are 23% at 410 K and 37% at 405 K which are consistent with those derived from the O_3/CO correlation (34% and 43%, respectively, as shown in Figure 4). Above 415 K, the percentage of recent air in the air sampled by SPIRALE is nearly zero. This is illustrated in Figure 5b for isentropic surfaces 430 K and 455 K showing the SPIRALE measurements at midlatitude being equal to the midlatitude stratospheric end member. This result is not in agreement with the result obtained in section 3.2. This can be explained by the fact that only dynamical mixing processes are taken into account in the present analysis and no HCl chemistry. The lifetime of HCl is of the order of 1 month in the lower stratosphere assuming as before an OH concentration of 8×10^5 cm^{-3}. As discussed in section 3.2, the mixing between recent air and midlatitude stratospheric air occurs within 3 months. Thus the mixing of midlatitude air with more recent air is of the same order or larger than the characteristic time of the HCl chemistry. This argument on chemistry does not hold for the relation O_3/CO since the lifetime of CO is larger than the lifetime of HCl. We assume therefore that the percentages of recent air computed in section 3.2 are more reliable that those derived from the HCl/O_3 relation.

[23] In summary, the correlation of the abundance of HCl and O_3 also confirms that SPIRALE sampled midlatitude stratospheric air mixed with recent air on 2 October 2002 between 380 K and 415 K. The isentropic mixing takes place between 400 K and 415 K leading to percentages of midlatitude stratospheric air in the mixed air parcels consistent with those derived from the O_3/CO relation (see section 3.2). Mixing could be not isentropic from 380 K to 400 K. The mixing in the range 415–465 K revealed by the relation O_3/CO is not confirmed by the HCl/O_3 relation. This can be due to chemical processes not taken into account in the analysis of the HCl/O_3 relation if mixing of recent air with midlatitude stratospheric air in the range 380–465 K took place 1 month or more before the mixed air parcels reach the SPIRALE location.

3.4. Conclusion

[24] There are therefore experimental evidences that air sampled by SPIRALE on 2 October 2002 at midlatitude in the altitude range 15–19.3 km (380–465 K) is composed of old stratospheric midlatitude air mixed with more recent air. The mixing could have taken 1 month or more before the mixed air parcels reach the SPIRALE location. The

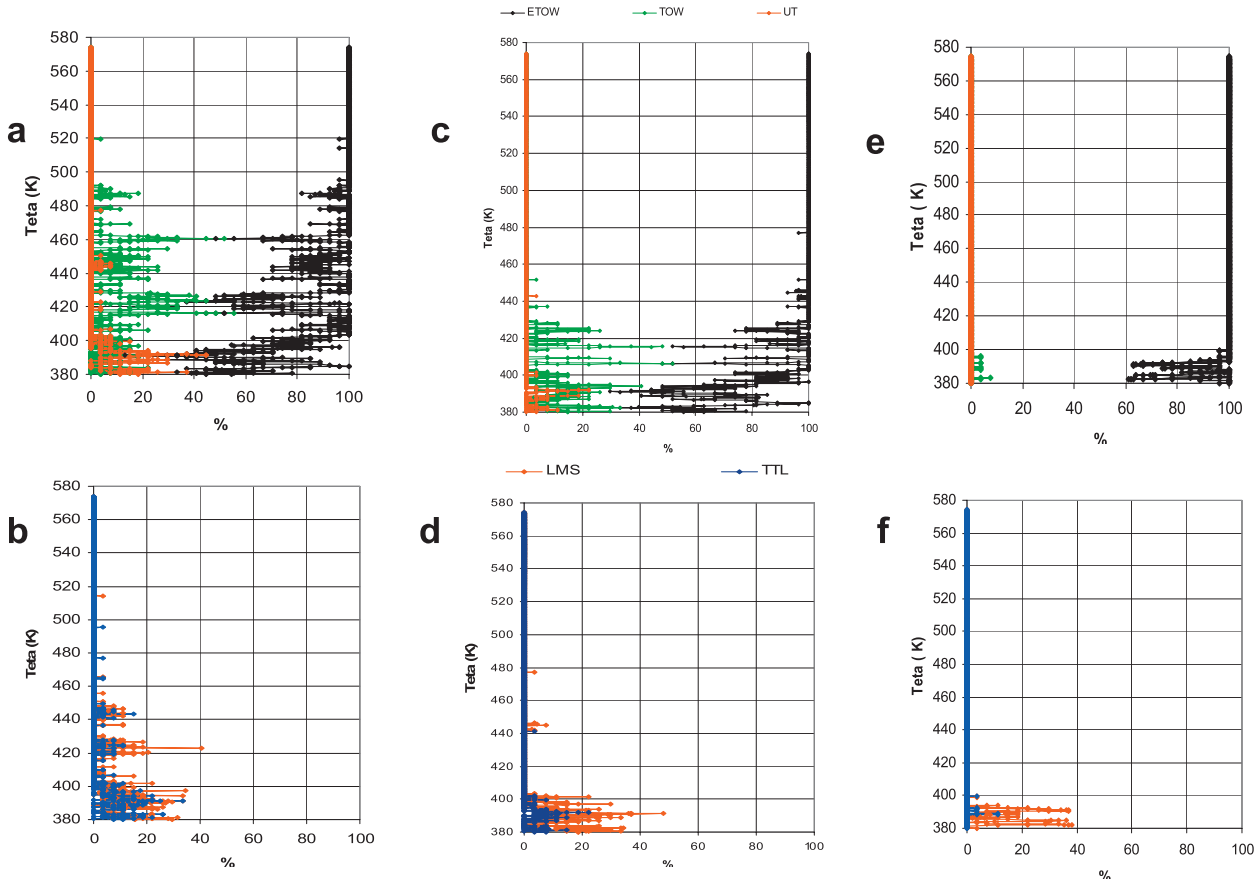

Figure 7. Percentage of the 27 particles in each parcel associated to an individual SPIRALE measurement as a function of the potential temperature in the regions ETOW, TOW, and UT defined in Figure 6 and in section 4.1, 60 days before their arrival at the SPIRALE location (a), 40 days before their arrival (c), and 20 days before their arrival (e). The same for regions LMS and TTL, 60 days before (b), 40 days before (d), and 20 days before (f).

percentage of recent air in the mixed air parcels is found to be 40 to 60% between 380 K and 405 K and between 0 and 20% above 415 K. Correlation of the abundance of CO and ozone shows that the origin of the recent air is probably the tropical stratosphere. Correlation of the abundance of HCl and ozone shows the same above 400 K. The origin of recent air could be different between 380 K and 400 K. Since the tropical profiles of HCl, CO and O_3 used here were obtained at a different date from the midlatitude profiles, there is obviously an uncertainty on the actual origin of the recent air and its percentage in the mixed air parcels. Trajectory calculations are therefore used in section 4: (1) to look for other evidences of the origin of the recent air; (2) to compute differently the percentage of the recent air in the mixed air parcels; and (3) to evaluate their "mixing age"; that is, the time between the mixed air parcel is formed when the recent air enters the midlatitude stratosphere and the SPIRALE location is reached.

4. Trajectory Calculations

4.1. Lower Limit of the Percentage of the Recent Air in the Mixed Air Parcels and Mixing Age

[25] Backward trajectories ending at the location of the SPIRALE measurements in the range 15–24 km (380–575 K) were computed during the descending phase of the balloon flight. In the model a parcel is associated to an individual measurement which is recorded every 1s by the instrument. This corresponds to 1500 parcels in the altitude range of interest. Each parcel is composed of 27 particles ending around each individual measurement location. Particles are distributed in a cubic array ($3 \times 3 \times 3$) with the measurement in the center of the cube. The vertical separation is 10 m, the scale of the vertical separation between measurements and the horizontal scale is 2.5 km according to the typical aspect ratio (250) of tracer structures in the lower stratosphere [*Haynes and Anglade*, 1997]. Sixty days backward trajectories for each of the 40500 (1500×27) particles are computed. The locations of these particles are recorded 60, 40, and 20 days before their arrival at the SPIRALE measurement location and dispatched in five schematic geographical regions (Figure 6): (1) The region called ETOW is the extratropical overworld: potential temperature $\theta > 380$ K, latitude $\phi > +20°$; SPIRALE is located in this region. (2) The region called TOW is the tropical overworld: $\theta > 380$ K, $-20° < \phi < +20°$. (3) The region called LMS is lowermost stratosphere: $\theta < 380$ K and potential vorticity PV > 2 PVU. (4) The region called TTL is the tropical transition layer: $\theta < 380$ K, PV < 2 PVU. (5) The region called UT is the Upper

Table 1. Percentage of Particles Which are Outside the ETOW Region 60, 40, and 20 Days Before Their Arrival at the SPIRALE Location, in the Potential Temperature Intervals 380–405 K, 405–430 K, 430–465 K, and 465–500 K[a]

Days Before Arrival	380–405 K	405–430 K	430–465 K	465–500 K
60	32.3	16.8	8.7	1.4
40	25.1	6.6	0.2	0.0
20	7.0	0.0	0.0	0.0

[a]ETOW: extratropical overworld; SPIRALE: Spectroscopie Infrarouge par Absorption de Lasers Embarqués.

Troposphere, $\theta < 340$ K. The percentages of the 27 particles of one parcel in each region are shown in Figure 7 as a function of the potential temperature of the parcel at the end of the trajectories (SPIRALE location).

[26] According to Figure 7, the percentages of particles which are not in the ETOW region 60 days before their arrival at the SPIRALE location vanish above 500 K. These percentages vanish above 430 and 400 K, 40 and 20 days, respectively, before their arrival at the SPIRALE location. The particles which are not in the ETOW region 60 and 40 days before their arrival are mainly in the tropical overworld region (TOW) for potential temperature larger than 405 K and mainly in the regions located below the 380 K isentropic surface (LMS, TTL and UT) for potential temperature lower than 405 K. Twenty days before they are mainly below the 380 K isentropic surface.

[27] Table 1 shows the percentage of particles which are not yet in the ETOW region 60, 40, and 20 days before their arrival at the SPIRALE location in four potential temperature intervals: 380–405 K, 405–430 K, 430–465 K and 465–500 K. The first row of Table 1 represents the lower limit of the percentage of recent air in the mixed air parcels sampled by SPIRALE: 32.3% in the 380–405 K layer, 16.8% in the 405–430 K layer, 8.7% in the 430–465 K layer and 1.4% in the 465–500 K layer. These values are lower limits since other particles could have entered the ETOW region more than 60 days before their arrival at the SPIRALE location according to ECMWF fields. These lower values are consistent with the percentage of recent air calculated in using the CO/O_3 correlation (Figure 4): 50% in the layer 380–405 K, 23% in the layer 405–430 K and 10% in the layer 430–465 K. The small but not negligible percentage of recent air (1.4%) in the layer 465–500 K shows that mixing could have occurred above 465 K as shown by *Huret et al.* [2006]. From the results in Table 1, it is also possible to derive a mean mixing age of the mixed air parcels.

Table 2. Percentage of Particles in the TOW, LMS, TTL, and UT 60, 40, and 20 Days Before Their Arrival at the SPIRALE Location in the 430–465 K Layer[a]

Days Before Arrival	TOW	LMS	TTL	UT
60	6.9	0.9	0.4	0.4
40	0.1	0.1	0.0	0.0
20	0.0	0.0	0.0	0.0

[a]Tropical overworld: TOW; lowermost stratosphere LMS; tropical transition layer: TTL; and upper troposphere: UT.

Table 3. Percentage of Particles in the TOW, LMS, TTL, and UT 60, 40, and 20 Days Before Their Arrival at the SPIRALE Location in the 405–430 K Layer[a]

Days Before Arrival	TOW	LMS	TTL	UT
60	12.4	3.2	1.0	0.1
40	6.6	0.0	0.0	0.0
20	0.0	0.0	0.0	0.0

[a]See definitions in footnote for Table 2.

[28] 1. In the layer 380–405 K, 7% of the particles need 0 to 20 days to reach the location of SPIRALE, 18.1% need 20 to 40 days and 7.2% need 40 to 60 days. From these values we can derive an approximate mixing age of 30 days.

[29] 2. In the layer 405–430 K, 6.6% of the particles need 20 to 40 days and 10.2% need 40 to 60 days. Taking into account that other particles can reach the SPIRALE location more than 60 days after they enter the ETOW region, we estimate an approximate mean mixing age of 60 days.

[30] 3. In the layer 430–465 K, 0.2% of the particles need 20 to 40 days and 8.5% of the particles need 40 to 60 days. In this layer, we estimate an approximate mean mixing age of 60 days or more.

[31] 4. In the layer 465–500 K, 1.4% of the particles need 40 to 60 days. In this layer we estimate a mixing age larger than 60 days.

[32] These mixing ages are very consistent with those derived from the analysis of the CO/O_3 and the HCl/O_3 correlations.

4.2. Origin of the Recent Air

[33] In the layer 430–465 K (see Table 2) and in the layer 465–500 K (Figure 7), almost all the particles that were outside the extratropical overworld region (ETOW) 60 days before their arrival at the SPIRALE location were in the tropical overworld region (TOW). This demonstrates that the mixing takes place on the isentropic surfaces in this layer. This is in agreement with the conclusions derived from the analysis of the CO/O_3 correlation (see section 3.2).

[34] In the layer 405–430 K layer (see Table 3), all the particles that were outside the ETOW region 40 days before their arrival at the SPIRALE location (6.6%) were in the TOW region. They were also mainly in the TOW region (12.4%) 60 days before their arrival but some of them (3.2%) were in the lowermost stratosphere (LMS) and in the TTL (1%). As for the layer 430–465 K, this confirms that the mixing takes place preferentially on the isentropic surfaces. It appears nevertheless that some particles were

Table 4. Percentage of Particles in the TOW, LMS, TTL, and UT 60, 40, and 20 Days Before Their Arrival at the SPIRALE Location in the 380–405 K Layer[a]

Days Before Arrival	TOW	LMS	TTL	UT
60	5.3	10.7	7.3	9.0
40	9.1	11.3	2.7	2.0
20	0.2	6.5	0.3	0.0

[a]See definitions in footnote for Table 2.

lifted diabatically from the LMS and the TTL, i.e., from below the 380 K isentropic surface up to the layer 405–430 K.

[35] In the layer 380–405 K, the feature observed in the layer above is more pronounced (see Table 4). A large number of particles were below the 380 K isentropic surface: 27% (60 days before arrival), 16% (40 days before) and 6.8% (20 days before) while only a small fraction were in the tropical overworld, 5.3% (60 days before), 9.1% (40 days before) and 0.2% (20 days before). Some of these particles (9%) were even below the 340 K isentropic surface 60 days before their arrival at the SPIRALE location. In the 380–405 K layer, the recent air mixed with the midlatitude stratospheric air is therefore mainly diabatically lifted from below the 380 K level. This is consistent with one of the hypotheses proposed to explain the HCl/O_3 correlation below 400 K. A small but significant fraction is nevertheless also isentropically mixed.

5. Conclusions

[36] The tracer correlations derived from the in situ measurements of O_3, CO and HCl made by the high-resolution TDLAS balloon instrument SPIRALE on 2 October 2002 above Aire sur l'Adour (43.6°N, 0°) and trajectory calculations have revealed that some mixing has occurred in the lower part of the overworld (380–465 K) between old midlatitude stratospheric air and more recent air. This is in agreement with the tropically controlled transition region concept developed by *Rosenlof et al.* [1997]. The characteristics of this region at the time and at the location of the SPIRALE flight were analyzed in the present paper.

[37] Using aircraft and satellite water vapor and methane measurements *Rosenlof et al.* [1997] found that this region extends up to 450 K. In the present paper the top of the layer is found to be 465 K (19.3 km) and the mixing age of the mixed air parcels sampled by SPIRALE is estimated to be more than 1 month in the potential temperature layer 380–465 K. The mixed air parcels are found to be composed of 50% of recent air in the layer 380–405 K. This percentage decreases sharply above this layer. It is of about 10% above 430 K.

[38] Trajectory calculations confirm both the mixing age of the mixed air parcels and the sharp decrease of the percentage of recent air in these air parcels above 405 K. Trajectory calculations also enable us to derive lower limits for the percentage of recent air in the mixed air parcels which are consistent with the tracer correlations studies: 32.3% in the 380–405 K layer and 8.7% in the 430–465 K layer. Above 405 K, the origin of the recent air is found to be the tropical overworld by both the tracer correlation studies and the trajectory calculations. The mixing between this recent air and the old midlatitude stratospheric air is isentropic. In the layer 380–405 K recent air is shown by the trajectory calculations to be uplifted from below the 380 K isentropic surface.

[39] At the location and at the time of the SPIRALE flight (2 October), the tropically controlled transition region is therefore composed of two very different layers. In the layer 380–405 K, the recent air is uplifted and its percentage in the mixed air parcels is high. In the layer 405–465 K, the origin of the recent air is the tropical overworld and its percentage in the mixed air parcel is lower. In both layers the mixing age is between 1 and 2 months. This mixing age suggests that the recent air in the layer 380–405 K could have been uplifted by the Asian monsoon whose maximum is in July (2 months before the flight of SPIRALE) decreasing slowly until September.

[40] HCl/O_3 and O_3/CO correlations derived from the SPIRALE in situ measurements with a high resolution and a very good accuracy have shown to be a powerful tool to document the characteristics of the tropically controlled transition region. The characteristics of this region could be seasonally dependent. Other measurements in different seasons are therefore necessary to study this possible seasonal dependence.

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