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On the vertical structure of the stratosphere at midlatitudes during the first stage of the polar vortex formation and in the polar region in the presence of a large mesospheric descent

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[1] CH₄ and N₂O measured by the balloon-borne Spectromètre Infra Rouge pour l'Étude de l'Atmosphère par Diodes Laser Embarquées (SPIRALE) instrument at midlatitude on 2 October 2002 and in the polar region on 21 January 2003 are used to study the vertical structure of the stratosphere. SPIRALE utilizes a direct absorption technique using tunable diodes laser in the midinfrared. For the two flights, detailed structures with vertical extents less than 1 km and nonmonotonic profiles are observed. Measured N₂O and CH₄ have been compared with correlation curves deduced from ATMOS space shuttle measurements. The high vertical resolution (5 m) and high precision of the measurements allow us to discuss the air mass origin in detail. To help with the interpretation, the potential vorticity maps have been used. At midlatitude we have identified a tropospheric tropical air mass intrusion. The layer located just above has been identified as the "tropically controlled transition region." For this layer a specific correlation line has been deduced from our measurements. The nonmonotonic part of the profile in the 23–29 km altitude range corresponds to a transition layer where mixing is still occurring. For the polar flight the instrument has sampled air masses outside the polar vortex at the lower levels and inside the polar vortex above. An intense mesospheric descent is observed. We provide new CH₄:N₂O correlation values that can be encountered in cases of large mesospheric descent. Our measurements show also a nontypical CH₄:N₂O correlation line in a thin layer corresponding to a midlatitude filament crossing the balloon trajectory.

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1. Introduction

[2] The distribution of stratospheric N₂O and CH₄ is controlled by large-scale transport, chemical reactions and mixing processes. These species are injected into the stratosphere through the tropical tropopause, then transported poleward and downward in extratropical regions. During their transport, chemical reactions occur with OH, Cl and O(¹D) for CH₄ and with O(¹D) and via UV radiation for N₂O. The timescale of the chemical reactions is shorter than the timescale of the large-scale transport and more effective at high altitude leading to vertical profiles decreasing with altitude and to lower species abundance poleward at a given altitude.

[3] Using ATMOS measurements, *Michelsen et al.* [1998a] (hereafter referred to as M1998a) have shown

that correlations between these species are compact and representative of different regions (tropics, midlatitudes/protovortex and well-formed vortex). Indeed the stratospheric dynamical structure consists of the subtropical dynamical barrier and the polar vortex boundary during winter which isolates the regions in which mixing processes are more effective in comparison with chemical processes [*Plumb and Ko*, 1992; *Waugh et al.*, 1997]. The compact relations are very robust regarding the seasonal, yearly and hemispherical variations. Nevertheless episodic fast events affect the vertical profiles of these species leading to anomalous correlations, as for example during the formation/breakdown of the polar vortex associated with inversion in the vertical profiles [*Waugh*, 1993; *Waugh et al.*, 1997]. In this case incompletely mixed air masses are observed.

[4] Correlations between tracers have been used to study mixing processes across the boundary of the polar vortex [*Morgenstein et al.*, 2002; *Jost et al.* 2002; *Ray et al.*, 2002; *Plumb et al.*, 2000], anomalous chemistry such as denitrification mechanisms in the polar vortex [*Esler and Waugh*, 2002], entrainment of air masses from midlatitudes to tropical regions [*Herman et al.*, 1998] or from extravortex region to vortex region *Michelsen et al.* [1999a], laminae

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Manney et al. [2000] and mesospheric descent [*Plumb et al.*, 2002]. On the basis of CH₄ and H₂O satellite measurements, *Rosenlof et al.* [1997] define three vertical layers in the midlatitude stratosphere: (1) the overworld above the 450 K isentropic surface controlled by chemistry and mixing; (2) the tropically controlled transition region between the 380 and 450 K isentropic surfaces where dynamical barriers are weak [*Grant et al.*, 1998] leading to large isentropic mixing between recent tropical stratospheric air and older midlatitude stratospheric air; and (3) the lowermost stratosphere below the 380 K isentropic surface where tropical tropospheric air can be mixed isentropically with the midlatitude stratospheric air across the subtropical tropopause.

[5] Correlations between tracers allow the characterization of the air masses origin and mixing timescales. They have been used to test and adjust the dynamical and mixing codes in three-dimensional (3-D) models [*Avallone and Prather*, 1997; *Hall and Prather*, 1995]. Associated with a tropical tracer model *Volk et al.* [1996] evaluate the rate of transport between the tropics and the midlatitudes. Ozone loss due to chemistry and due to vortex and midlatitude air mass mixing can be separated using these tracer correlations [*Konopka et al.*, 2004]. Most of the measurements available come from the Space Shuttle (ATMOS) or satellites (ILAS (N₂O and CH₄), HALOE (CH₄), ODIN (N₂O)) with high spatial coverage but low vertical resolution, or in situ measurements from aircraft with high spatial resolution but only observing the lower stratosphere. The measurements from ATMOS and ALIAS in situ instrument onboard the ER2 aircraft have been intercompared and a good agreement has been observed [*Michelsen et al.* [1999b]. They have studied the air enters in the vortex. In addition, a set of various balloonborne instruments using different techniques measure these tracers together with short-lived species. In this last set, studies focus on the impact of the dynamical state on the detailed chemical composition of the stratosphere and in particular on the partitioning inside the NO_y or Cl_y families [*Wetzel et al.*, 2002; *Stowasser et al.*, 2002; *Payan et al.*, 1999]. However, as for satellite instruments, the vertical resolution of up to 1 km of these instruments is not enough to investigate detailed mixing processes. *Orsolini et al.* [1998] have studied the vertical structure of the stratosphere and shown the relevance of N₂O, halocarbons and H₂O in situ balloonborne measurements at midlatitudes in detecting laminae. *Manney et al.* [2000] highlight also the detection of laminae using N₂O:CH₄ correlation from ATMOS data. Using a reverse trajectory model they reproduce the majority of the laminae observed and conclude that advection by large-scale winds is the major process responsible of such laminae.

[6] In this study we combine in situ measurements of CH₄ and N₂O mixing ratios and high vertical resolution (a few meters) to investigate the small-scale structure (smaller than 1 km) of the stratosphere from 12 km to 34 km.

[7] Measurements were obtained by the balloonborne instrument SPIRALE [*Moreau et al.*, 2005] at midlatitudes on October 2002 during the first stage of the polar vortex formation and in the polar region on January 2003 at the edge of the vortex and inside the vortex during a large mesospheric descent. For each flight the detailed interpre-

tation of the measurements is aided by the potential vorticity maps calculated by the MIMOSA contour advection model [*Hauchecorne et al.*, 2002].

[8] In the first part of this paper we present a short description of the SPIRALE instrument, the CH₄:N₂O correlation curves derived from the measurements of ATMOS version 2 (M1998a) and slightly updated, and the MIMOSA model. Slight modifications of the ATMOS correlations have been made to take into account the CH₄ and N₂O variation from 1992 to 1994. Then we present the SPIRALE N₂O and CH₄ mixing ratio measurements and we compare the derived CH₄:N₂O correlation curves with those derived from ATMOS. In the last part we discuss, for the two flights, the detailed vertical structure of the stratosphere from the lowermost stratosphere up to 34 km.

[9] At midlatitudes the profiles obtained are nonmonotonic and a highlight is the occurrence of thin layers where mixing processes have probably occurred. The mixing processes are investigated by discussing the air masses origin using the potential vorticity map (PV) and the correlation points obtained. In the polar flight a large mesospheric descent has been observed by SPIRALE inside the vortex. In a well-defined layer this mesospheric air has probably been mixed with stratospheric air coming from midlatitudes. The PV map shows an intrusion of a midlatitude filament into the polar vortex which was crossed during the ascent of the balloon.

2. Observations and Model Description

[10] This part is devoted to the description of the SPIRALE instrument and the tools used to interpret its measurements. Updated N₂O:CH₄ correlations based on the M1998a study, and the MIMOSA contour advection model [*Hauchecorne et al.*, 2002] are presented.

2.1. SPIRALE Measurements

[11] SPIRALE (French acronym for Infrared Spectroscopy by Tunable Diode Lasers) is a balloonborne instrument devoted to in situ measurements of atmospheric trace and active species from the tropopause up to 40 km. Infrared absorption spectroscopy using lead salt diode lasers provides coverage of the mid-infrared region (3 μm to 10 μm) where molecular line strengths are greatest. Six laser beams are directed to a 3.5 m multipass Herriott cell located under the gondola, largely exposed to ambient air, and deployed above the tropopause. Eighty-six reflections occur in the cell giving a 300 m optical path. Species concentrations are retrieved from direct absorption, by fitting experimental spectra and spectra calculated using spectroscopic parameters of absorption lines from the HITRAN 2001 database. The instrument performance is as follows: 1) a vertical resolution of a few meters, 2) an absorption limit close to 10⁻⁵, 3) ability to detect several species with the same diode laser by judicious choice of the micro spectra in specific modes. By this technique the instrument measures simultaneously CH₄, N₂O, NO₂, HNO₃, HCl, CO and O₃. Accuracies of the measured concentrations have been evaluated to be 3% for CH₄, N₂O, 3–5% for O₃ and CO, 3–30% for NO₂, HCl, 5–20% for HNO₃. A detailed description of the instrument and of its operating modes can be found in the work of *Moreau et al.* [2005].

[12] This study focuses on N_2O and CH_4 measurements made at midlatitudes on 2 October 2002 above Aire sur l'Adour (43.6° N, 0°E) in the morning and in the polar region on 21 January 2003 during the first part of the night above Kiruna (68°N, 20°E).

2.2. N_2O and CH_4 Earth Surface Mixing Ratios and Correlations in 2002–2003

[13] The CH_4 and N_2O mixing ratios at Earth's surface in 2002 can be evaluated from the mixing ratio and the tendency measured in 2000 [*World Meteorological Organization (WMO)*, 2002]. Globally averaged Earth surface CH_4 was measured to be between 1751.6 ppbv with a tendency of +0.7 ppbv per year and 1784.4 ppbv with a tendency of +1.3 ppbv per year. Globally averaged Earth surface N_2O was measured in 2000 to be between 315.2 ppbv with a tendency of +1 ppbv per year and 317.2 ppbv with a tendency of 1.1 ppbv per year. From these measurements, the following 2002 globally averaged Earth surface mixing ratios assumed in this paper are: 1770 ppbv \pm 17 ppbv for CH_4 and 318.3 ppbv \pm 1.1 ppbv for N_2O .

[14] We have used correlation curves between the CH_4 and N_2O mixing ratios derived from the Version 2 of the Atmospheric Trace Molecular Spectroscopy (ATMOS) measurements made in November 1994 and April 1993 and shown in M1998a. Comparing the ATMOS Version 2 measurements with ER-2 measurements, *Chang et al.* [1996] have demonstrated an agreement better than 3% for CH_4 , as a function of N_2O , for all values of N_2O . The ATMOS version 2 measurements have been compared with the version 3 of ATMOS measurements retrieval presented by *Irion et al.* [2002] and do not show for CH_4 large differences for stratospheric levels in comparison with Version 2 used in the work of M1998a. Finally, $\text{N}_2\text{O}:\text{CH}_4$ correlations derived from ATMOS version 3 have been shown to be consistent with those derived from version 2 [*Michelsen et al.*, 1999a].

[15] In M1998a a very detailed study has been conducted to classify the ATMOS profiles. The three main groups are: vortex, tropics and extratropical/extravortex. The extratropical/extravortex data have been subdivided into proto-vortex, midlatitudes and mixed groups. The mixed group refers to semitropical, subtropical or vortex edge. Polynomial fits to $[\text{CH}_4]$ versus $[\text{N}_2\text{O}]$ for the vortex, midlatitudes and tropics groups are given in the work of M1998a and used as a reference in this paper. Mixed group is not shown in the work of M1998a for simplicity and the protovortex group has found to be very similar to the midlatitude group. The same polynomial fits are given to midlatitude and tropics data for N_2O mixing ratio larger than 293 ppbv. Midlatitude and tropics correlations are shown to be very compact and independent of season, year and hemisphere. In contrast polar vortex correlations could vary from year to year and depend on the extent of the descent within the vortex and on the permeability of the vortex boundary.

[16] The M1998a correlation curves have been slightly modified before comparison with SPIRALE measurements. These modifications are needed because SPIRALE measurements took place 8 and 9 years after ATMOS measurements and during this period N_2O and CH_4 concentrations have varied, principally due to surface emission.

[17] N_2O at Earth's surface has increased steadily since 1980 at an average rate of 0.75 ppbv per year [*WMO*, 2002]. The corresponding increase from 1994 to October 2002 is 2%. This increase can be assumed to be the same in all the air parcels sampled by SPIRALE in the altitude range 10–35 km. To take into account such an increase the M1998a curves have been translated by +2% along the horizontal axis (N_2O axis).

[18] The growth rate of the CH_4 Earth surface mixing ratio has been decreasing since 1980. It was 14 ppbv per year in the early 1980s, 5 ppbv per year in the late 1990s and only 1 ppbv per year in 2000 [*WMO*, 2002]. As a consequence the increase of the CH_4 mixing ratio in the air parcels depends on their mean age. In the regions sampled by SPIRALE the mean age of the air parcels can be derived from the model calculations shown in the work of *Plumb et al.* [2002] to be 2 ± 2 years. We can then assume an increase of $2.5\% \pm 0.7\%$ from 1994 to 2002. To take into account such an increase the M1998a curves have been translated by +2.5% along the vertical axis (CH_4 axis).

2.3. MIMOSA

[19] MIMOSA is a contour advection model, in which the potential vorticity (PV) is advected on several isentropic levels by the horizontal wind components on an x-y grid centered on the North Pole and with a resolution of 3 points per degree. To preserve the homogeneity of the field, a regridding of the PV field on the original grid is made every 6 hours. The diabatic evolution of the PV field at large scales can be extracted from ECMWF fields. In MIMOSA this is made by applying a relaxation toward the ECMWF PV field with a time constant of 10 days. Using this procedure, it is possible to run continuously the model during long periods and to follow the evolution of polar and tropical intrusions at midlatitudes [*Heese et al.*, 2001]. It has been used in several studies to investigate ozone filaments in the polar region [*Hauchecorne et al.*, 2002] and tropical air mass intrusion at midlatitudes [*Durry et al.*, 2002].

[20] This model allows characterizing the air masses origin in identifying dynamical barriers characterized by the maximum of the PV gradient. For measurements made on October 2002 the dynamical barrier of interest is between the midlatitudes and tropical air. For polar measurements the PV barrier at the polar vortex boundary separates midlatitudes and polar vortex air. PV maps have been calculated on isentropic surfaces between 340 K and 810 K on 2 October 2002 and between 417 K and 885 K on 21 January 2003 at the time of the SPIRALE measurements ± 0 to 30 min.

3. Results

[21] In this section we present and describe the vertical profiles obtained and we compare the derived $\text{CH}_4:\text{N}_2\text{O}$ correlation curves with the slightly modified M1998a correlation curves.

3.1. SPIRALE Measurements

[22] Vertical profiles of N_2O and CH_4 mixing ratios measured by SPIRALE at mid and high latitudes are shown on Figures 1a and 1b. For both flights N_2O and CH_4 are

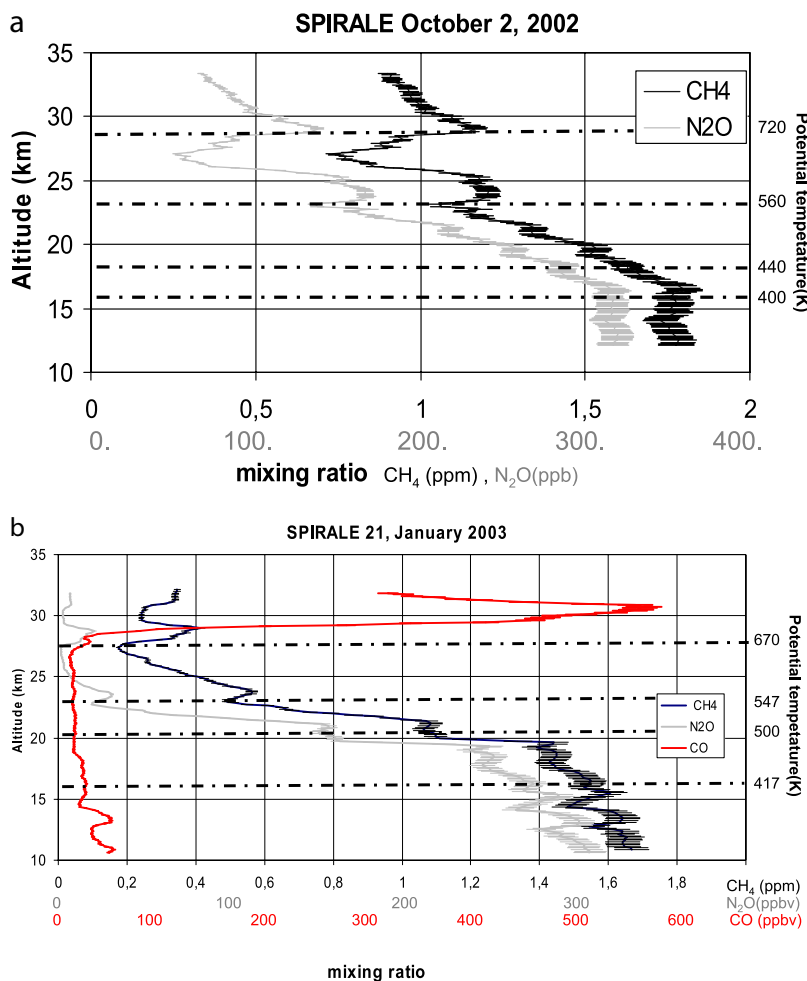


Figure 1. Vertical profiles of mixing ratios measured by the SPIRALE instrument. (a) CH₄ and N₂O on 2 October 2002 above Aire sur l'Adour (43.6° N, 0°E) in the morning (0600–0730 UT) and (b) CH₄, N₂O, and CO on 21 January 2003 during the first part of the night (1930–2100 UT) above Kiruna (68°N, 20°E).

very well correlated. The high vertical resolution of the SPIRALE instruments allows us to identify small structures in the profiles on a scale of less than 1 km.

[23] In Figure 1a (midlatitude measurements) four layers can be seen. Between 12 and 16 km mixing ratios are respectively 1765 ± 40 ppbv for CH₄ and 316.0 ± 5.0 ppbv for N₂O. The 3% SPIRALE error bars of these low altitude mixing ratios overlap the error bars of the globally averaged Earth surface mixing ratios evaluated for 2002 in part 2.2. From 16 km to 23 km, both profiles are decreasing and small vertical structures can be seen. Then, in the layer between 23 km and 29 km, a succession of increases and decreases occur. In the upper part of the profiles above 29 km the mixing ratios are decreasing as is typically observed in the stratosphere.

[24] In Figure 1b (high-latitude measurements) the N₂O and CH₄ mixing ratios decrease slightly as a function of altitude at lower heights (below 20 km) and small structures with a vertical extent less than 1 km can be seen. Strong vertical gradient in the N₂O and CH₄ mixing ratios is observed at 20 km. Very low mixing ratios (1.5 ppbv for N₂O and 0.17 ppmv for CH₄) occur at 27.5 km. Between 27.8 km and 29.5 km larger amounts of N₂O and CH₄ are

observed with maximum values of 20 ppbv for N₂O and 0.4 ppm for CH₄.

[25] The CO vertical profile measured by SPIRALE is also presented in Figure 1b. Small values (10–20 ppbv) typical of the stratosphere are observed below 27 km. Above this altitude a large vertical gradient leads to very high values of the CO mixing ratio above 29 km (with a maximum close to 600 ppbv at 31 km). ATMOS measurements shown in the work of *Plumb et al.* [2002] indicate that such a large mixing ratio is typical of altitudes between 60 and 70 km at midlatitudes. This means that a very large mesospheric descent of air was observed above 29 km at the location of the SPIRALE measurements. Careful examination of CO mixing ratio increasing between 27 km and 31 km, shows that the increase is not strictly monotonic and a secondary minimum is observed between 28 and 28.5 km corresponding to the relative large amounts of CH₄ and N₂O in the layer 27.8–29.5 km.

[26] For SPIRALE flights the structure of the vertical profiles of the CH₄ and N₂O mixing ratios are very structured. Different layers are clearly identified at different altitudes. Before discussing the characteristics of these layers, correlations of the abundance of these two species

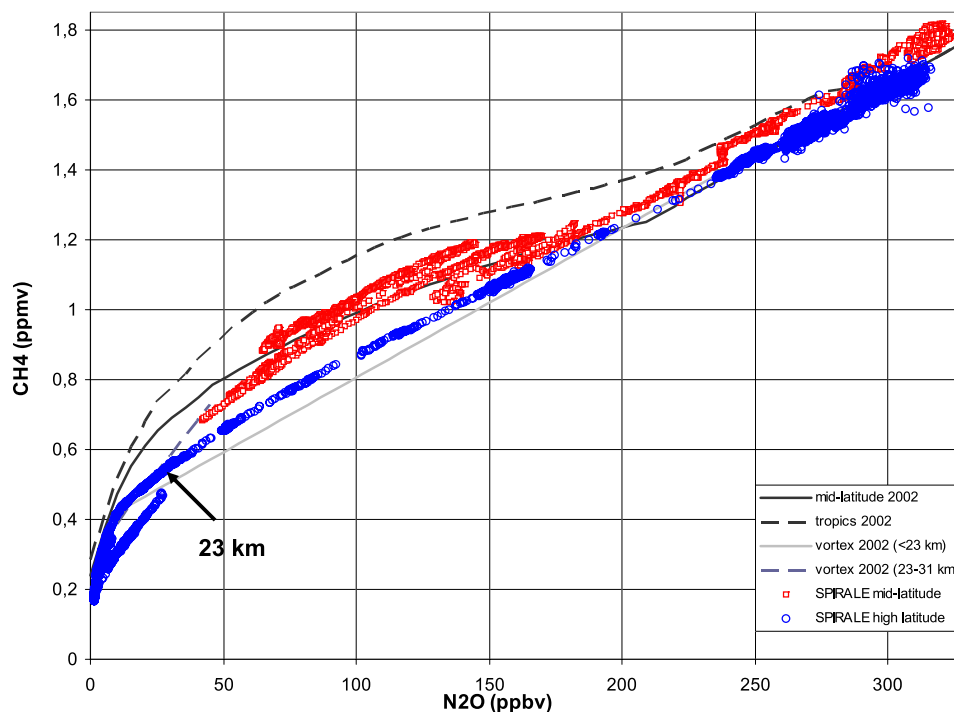


Figure 2. $\text{CH}_4:\text{N}_2\text{O}$ correlation values derived from SPIRALE for the midlatitude flight (red squares) and the high-latitude flight (blue circles); $\text{CH}_4:\text{N}_2\text{O}$ correlation curves from M1998a updated for the year 2002 for midlatitudes (black solid line), the vortex below 23 km (gray solid line), and the tropics (black dashed line).

are compared with correlations derived from ATMOS measurements.

3.2. $\text{CH}_4:\text{N}_2\text{O}$ Correlations

[27] Correlation between tracers has been recognized to be a good tool for comparing measurements by different instruments [Michelsen *et al.*, 1998b]. Figure 2 presents the correlations for the midlatitudes and the polar SPIRALE flights as well as the M1998a curves, slightly modified as discussed before, for tropical, midlatitudes/protovortex and well-formed vortex conditions below 23 km. There is a roughly good agreement between the measurements from the midlatitude SPIRALE flight and the midlatitude ATMOS measurements, and also between the SPIRALE high-latitude measurements and ATMOS measurements of the well-formed vortex.

[28] However by analyzing this figure in detail, some differences between the SPIRALE and ATMOS correlation curves can be pointed out:

[29] 1. Large values of midlatitude N_2O and CH_4 mixing ratios measured by SPIRALE at low altitude (below 16 km) are greater than the values given by M1998a. For the N_2O range greater than 285 ppbv the correlation data points from the SPIRALE midlatitude flight are above the M1998a midlatitude curve. Between 180 ppbv and 285 ppbv correlation data points fall between the midlatitude and tropical curves. Then for N_2O lower than 180 ppbv SPIRALE correlation data points are in agreement with the M1998a midlatitude curve but distributed around it.

[30] 2. The correlation curve derived from the measurements of the SPIRALE polar flight is in full agreement

with the common midlatitudes/protovortex and well-formed vortex M1998a curve above 220 ppbv and a little above the well-formed vortex curve from 10 to 200 ppbv. In the work of M1998a, correlation curves are computed for different altitude ranges: lower than 23 km, between 23 and 31 km, and above 31 km. Below 31 km, the minimum value for which the M1998a curves are computed is 10 ppbv while the N_2O SPIRALE mixing ratio values are often less than this (the minimum value at 27.5 km is 1.5 ppbv). Below 30 ppbv the correlation data points are well ordered but below the vortex line reported by M1998a.

[31] The differences highlighted between SPIRALE measurements and M1998a correlations curves need to be investigated. The very good precision and the high spatial resolution of the SPIRALE instrument can be very useful in identifying the sampled air masses origin.

4. Discussion

[32] Potential vorticity maps have been calculated using the MIMOSA model for the time of the measurements by the SPIRALE flights. These PV maps provide information about the location of the measurements relative to different dynamical barriers and therefore, in a first approach, the region of which air masses are representative: troposphere, tropical stratosphere, midlatitude stratosphere or polar vortex. Owing to mixing across the dynamical barriers, the chemical composition of the air parcels is not necessarily fully typical of these regions. The combination of the PV maps and a careful examination of the correlation curves can help us to know more about this mixing. We discuss

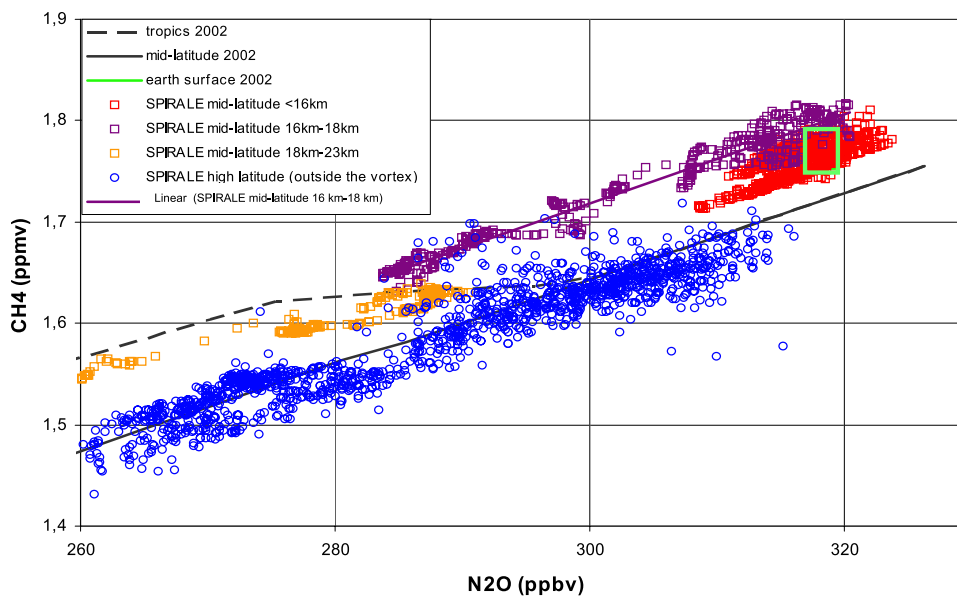


Figure 3. Detailed CH_4 : N_2O correlation values derived from SPIRALE in the range 260–330 ppbv for N_2O for the midlatitude flight: below 16 km (red squares), 16–18 km range (purple squares), 18–23 km range (orange squares), for the high-latitude flight outside the vortex (blue circles); CH_4 : N_2O correlation curves from M1998a updated for the year 2002 for midlatitudes (black solid line), and the tropics (black dashed line); 2002 Earth surface correlation from *WMO* [2002] (green square); and linear fit to CH_4 versus N_2O in the 16–18 km range (purple solid line).

first the results obtained during the midlatitude flight and then during the high-latitude flight.

4.1. Midlatitude Flight

4.1.1. CH_4 : N_2O Between 12 and 16 km

[33] Between 12 to 16 km, the N_2O and CH_4 mixing ratios measured by SPIRALE at midlatitudes are very close to the globally averaged Earth surface values [*WMO*, 2002] shown in Figure 3 in green, and the corresponding points on the CH_4 : N_2O correlation diagram are above the common midlatitudes/protovortex and tropical curves derived from *ATMOS*. This indicates that air masses sampled by SPIRALE have formed very recently and are likely to have a tropospheric origin. In this layer the potential vorticity map computed by *MIMOSA* below 16 km (400 K) shows a large intrusion of air coming from the tropical upper troposphere (Figure 4a on the isentropic surface 340 K). The SPIRALE location (cross) is in the lowermost stratosphere but very close to the maximum gradient of PV indicating the location of the local tropopause. This suggests that below 16 km air sampled by SPIRALE on 2 October 2002 is in fact typical of the tropical troposphere. This is in agreement with the results of *Ray et al.* [1999] showing that exchanges between the lowermost stratosphere and the tropical troposphere are seasonally dependant and reach a maximum in September.

4.1.2. CH_4 : N_2O Between 16 and 18 km

[34] For N_2O greater than 285 ppbv, SPIRALE N_2O versus CH_4 correlation curves obtained from the mid and high-latitude flights are different as shown on Figure 3, whereas M1998a recommends the same curve for tropical and midlatitudes/protovortex regions for the largest values of the N_2O mixing ratio. The PV maps for the high-latitude flight (Figure 4b) show that at the level 452 K (18 km)

and below, SPIRALE has sampled air masses outside the vortex which are more typical of midlatitudes. The correlations derived from these SPIRALE high-latitude data are in agreement (Figure 3) with the M1998a midlatitudes/protovortex polynomial fit.

[35] In contrast, the SPIRALE midlatitude correlation points are above the common M1998a tropics and midlatitude curve. They are on the straight line continuation of the tropics M1998a curve below 275 ppbv. This group of the correlation points corresponds to the altitude range 16–18 km shown on Figure 1a.

[36] For each point of the measurements, we have calculated the relative difference between the M1998a curve and the SPIRALE measurements for the midlatitudes and the high-latitude flights. We have checked for statistical significance of this difference between the two sets of data in comparison with the M1998a curve by applying the Student's *t* test [*Press et al.*, 1992]. The result obtained shows that the relative difference between the M1998a curve and the SPIRALE measurements at middle and high latitudes is significant at the 5% level. We can conclude that the air mass sampled between 16 km and 18 km at midlatitudes differs significantly from the air masses sampled at high latitudes outside the vortex. This air mass is composed by recent air with high mixing ratios of CH_4 and N_2O compared to the typical midlatitude air given by M1998a. This air mass appears to be typical of the tropical region because the CH_4 : N_2O points are linked with the tropical M1998a curve and with the N_2O and CH_4 Earth surface mixing ratios.

[37] PV maps between 16 km and 18 km (Figure 4c isentropic surface 425 K) do not indicate the presence of tropical air close to the location of SPIRALE. Nevertheless we can see that the PV barrier is very weak and mixing

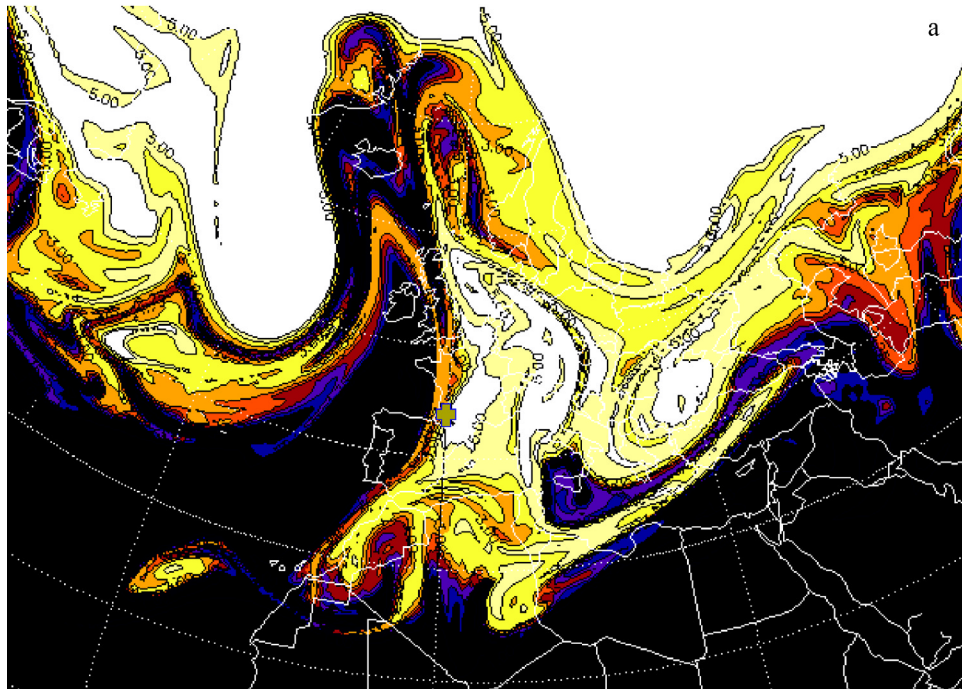


Figure 4. PV isocontours calculated by the MIMOSA model at the time (± 30 min) of the SPIRALE measurements. The cross denotes the instrument location (a) at midlatitudes at 340 K (12 km). The black region denotes PV < 2 PVU (1 PVU = 10^{-6} m² s⁻¹ kg⁻¹ K) (b) at high latitudes at 452 K (18 km), and the black region denotes air mass outside the vortex (c) at midlatitudes at 425 K (17.6 km).

could occur with the tropical stratosphere. This layer probably corresponds to the “tropically controlled transition region” pointed out by *Rosenlof et al.* [1997] where relatively young air is present. The correlation coefficient between CH₄ and N₂O in this layer is 0.9794 showing a very well correlation of these two species. To characterize this layer we have derived the following correlation curve for N₂O > 285 ppbv in the “tropically controlled transition region” from SPIRALE measurements:

$$y = 4.87 \cdot 10^{-1} + 4.10 \cdot 10^{-3}x$$

where y represents the CH₄ mixing ratio (ppmv) and x the N₂O mixing ratio (ppbv). This correlation curve needs to be validated using other measurements. Data of the last SPIRALE flight in the equatorial region in June 2005 will be useful to this end.

[38] High values of CH₄ and N₂O mixing ratios in the low levels have been previously observed by other balloon-borne measurements [*Engel et al.*, 1996; *Herman et al.*, 1998; *Kanzawa et al.*, 2003; *Michelsen et al.*, 1999b]. *Engel et al.* [1996] for example have deduced from their in situ measurements a correlation line slightly above the M1998a midlatitude and vortex curves. Their set of data is based on all their mid and high-latitude measurements from 1988 to 1992. Nevertheless no detailed investigations on the air mass origin and classification associated have been made while M1998a have excluded from their study the mixed profiles corresponding to semi tropical, subtropical or vortex edge air masses.

[39] From our study we can conclude that the M1998a correlation line above 293 ppbv, is a baseline for midlati-

tude data when mixing processes through the barriers have not occurred but that such mixing could be very usual in the “tropically controlled transition region” pointed out by *Rosenlof et al.* [1997].

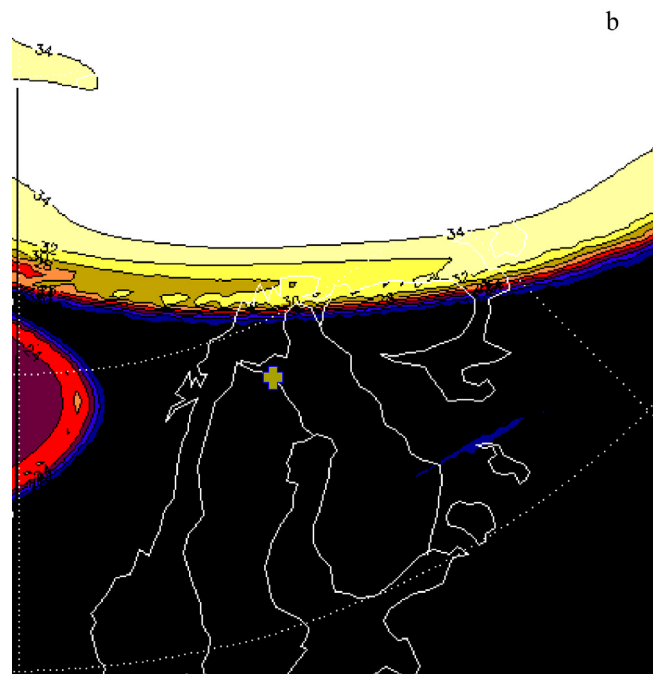


Figure 4. (continued)

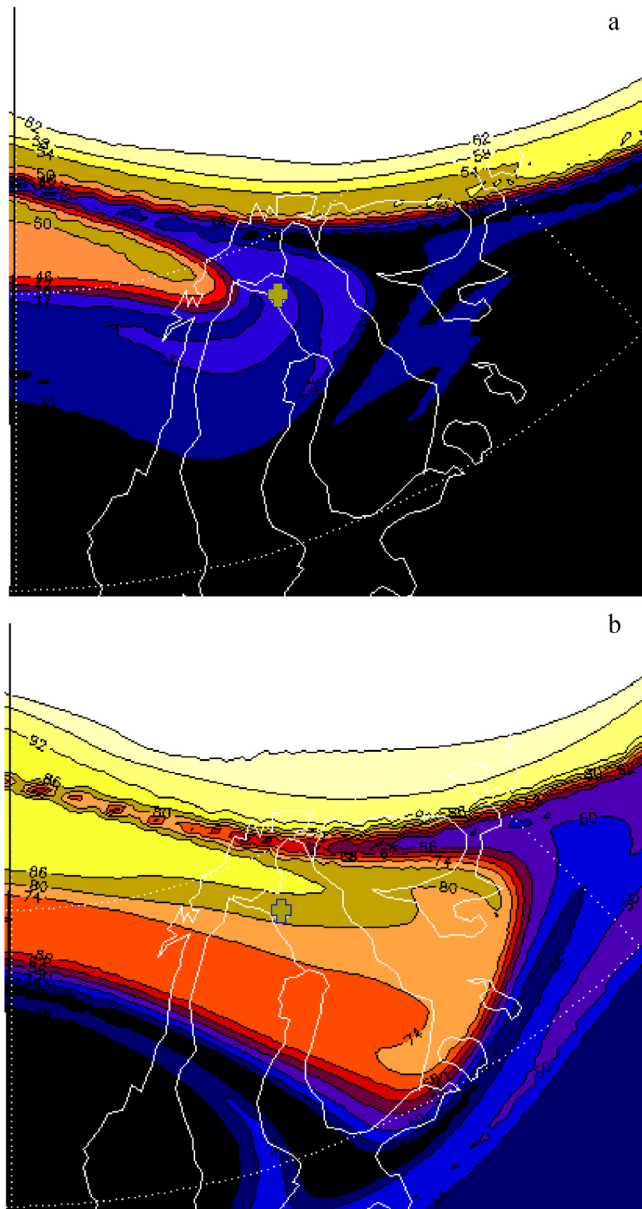


Figure 8. Same as Figure 4 but on 21 January 2003 at high latitudes at (a) 500 K (21 km) and (b) 547 K (23 km). Black regions denote air mass outside the vortex.

SMR N_2O averaged over the geographical region 43° – $58^\circ N/12.5^\circ$ – $17.5^\circ W$, and above 29 km by SMR N_2O in the region 28° – $43^\circ N/2.5^\circ E$ – $12.5^\circ W$. This is in agreement with our $N_2O:CH_4$ interpretation and with the PV map, that above 29 km the air mass is more tropical. This region appears to be affected by very intense and large exchanges between the tropics and the polar region occurring during the first stage of the polar vortex formation [Manney *et al.*, 1996] and followed by mixing processes. Vertical wind shear is probably responsible for non monotony of both profiles. Air masses from different regions have been transported to midlatitudes. In the layers where the mixing ratios increase the mixing processes is occurring, but vertical homogenization is not achieved.

[42] The dynamical vertical structure of the midlatitude stratosphere on 2 October 2002 revealed by the SPIRALE measurements is therefore very disturbed. SPIRALE has sampled air mass coming from the tropical troposphere in the lowermost stratosphere below 16 km. between 16 and 18 km air mass sampled is probably typical of the tropically controlled transition region, with a correlation line above the tropics and midlatitude curve recommended by M1998a. Above 18 km and below 23 km the layer composition appears also to be influenced by tropical stratospheric air although these layers are above the tropically controlled transition region highlighted by Rosenlof *et al.* [1997]. Above 23 km the situation is different because of the unusual and nonmonotonic N_2O and CH_4 mixing ratio profiles. Correlation curves are well ordered in each isolated layer observed in the profiles but significantly different from each other. This is probably due to the phenomena which prevail at the beginning of the vortex formation [Michelsen *et al.*, 1998b] when large horizontal transport between the tropics and high latitudes occurs followed by mixing processes.

4.2. High-Latitude Flight

4.2.1. $CH_4:N_2O$ Below 23 km

[43] As shown before (section 4.1.2), comparing $CH_4:N_2O$ correlations with M1998a midlatitudes/protovortex curves (Figure 3) and with potential vorticity calculations from MIMOSA it appears that SPIRALE measurements have been made outside the vortex below 18 km (Figure 4b). At 21 km (500 K) the measurements have been made in the vortex edge region where the PV contour gradient is maximum (Figure 8a). Above 22.5 km the air mass sampled is inside the vortex (Figure 8b, 547 K). This is consistent with the very large decrease of the N_2O mixing ratio between 19.5 km and 22.5 km observed on Figure 1b. In the range 200–30 ppbv of N_2O value, correlation points measured by SPIRALE (Figure 2) are slightly above well-formed vortex curve from M1998a. However, the major part of the measurements has been made very close to the vortex edge; outside the vortex Figure 8a or inside the vortex Figure 8b. In addition M1998a correlation curve refers to a vortex well formed later in the season.

4.2.2. $CH_4:N_2O$ Above 23 km

[44] In Figure 9, $CH_4:N_2O$ correlations above 23 km are compared with those derived from ATMOS at midlatitudes and in the well-formed vortex between 23 and 31 km. Correlation points derived from SPIRALE can be sorted into three sets as a function of altitude: set 1 from 23 to 27.8 km, set 2 from 27.8 to 29.5 km and set 3 above 29.5 km. Referring to Figure 1a, in set 1, the N_2O mixing ratio is decreasing down to very low values (1.5 ppbv). Set 2 corresponds to a layer with higher N_2O mixing ratio (up to 22 ppbv) corresponding to the nonmonotonic part of the profiles and, in set 3, low N_2O mixing ratios are again observed. Set 1 is in agreement with the vortex curve derived from ATMOS measurements, at least in the N_2O mixing ratio range measured by ATMOS (N_2O larger than 10 ppbv). We can notice that the maximum altitude of this set (27.8 km) is lower than the maximum altitude of the ATMOS measurements used to establish the curve in the well-formed vortex (31 km). SPIRALE measures lower N_2O and CH_4 mixing ratios than

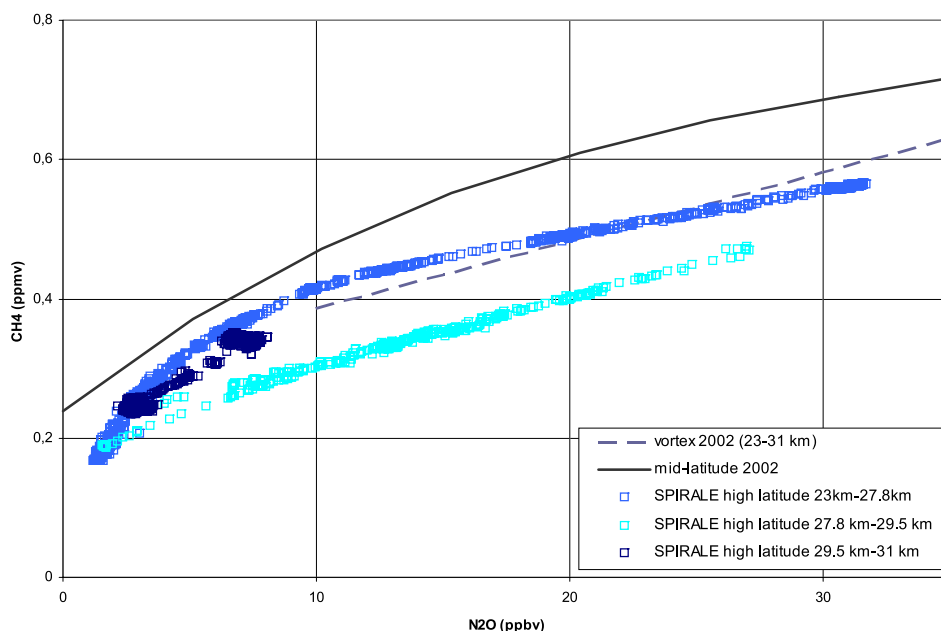


Figure 9. Same as Figure 3 in the range 0–35 ppbv for N_2O , for the high-latitude flight in the range 23–27.8 km layer (set 1), 27.8–29.5 km (set 2) layer (clear blue squares), and 29.5–31 km (set 3) layer (dark and very dark blue squares). M1998a correlation curves for vortex conditions between 23 km and 31 km (dashed gray line) and midlatitude conditions (black solid line) are shown.

ATMOS. Because of CO high value measured it appears that the diabatic descent is strong in January 2003. As mentioned by M1998a, $CH_4:N_2O$ correlation in polar region is variable year to year depending strongly on the intensity of the mesospheric descent. It also means that the correlation curves derived from ATMOS do not cover all possible situations inside the vortex. SPIRALE measurements therefore extend the correlation values to lower N_2O values thanks to the large mesospheric descent observed by the SPIRALE measurements. This mesospheric descent has also been observed on March 2003 by other instruments [Engel *et al.*, 2005].

[45] The correlation points derived from SPIRALE measurements above 29.5 km (set 3) are close to set 1. Correlations obtained (set 2) for the region between those two layers are significantly different at the level 5% with respect to set 1 and set 3 and the M1998a vortex curve. This means that the history of the air mass sampled in this layer is very different to those sampled above and below. This layer corresponds to the layer previously identified in Figure 1b with an increase of both N_2O and CH_4 .

[46] This layer seems to be made of mixed midlatitude stratospheric and mesospheric air masses. PV maps calculated every 25 K show that SPIRALE crosses a midlatitude PV filament between 670 and 750 K (Figure 10). Such a filament indicates that the polar vortex barrier is permeable [Nakamura, 1996], which could explain that this layer extends inside the polar vortex with respect to PV map. In addition, during the same period N_2O measurements from ODIN/SMR averaged inside the polar vortex show a similar layer between 29 km and 32 km [Urban *et al.*, 2005].

[47] We have deduced from our measurements two correlation curves. The first one uses values from set 1 and set 3. This correlation curve represents low amount of N_2O and

CH_4 that can be encountered inside the vortex due to a mesospheric descent.

[48] For N_2O lower than 35 ppbv in the altitude range 23–31 km:

$$y = -1.10 \cdot 10^{-6} x^4 + 9.10 \cdot 10^{-5} x^3 - 3.10 \cdot 10^{-3} x^2 + 5.02 \cdot 10^{-2} x + 1.2 \cdot 10^{-1}$$

where y represents the CH_4 mixing ratio (ppmv) and x the N_2O mixing ratio (ppbv).

[49] The second correlations curve deduced represents the mixed layer observed (set 2) when the instrument crosses the midlatitude PV filament.

[50] For N_2O lower than 28 ppbv:

$$y = 1.04 \cdot 10^{-2} x + 1.9172 \cdot 10^{-1}$$

where y represents the CH_4 mixing ratio (ppmv) and x the N_2O mixing ratio (ppbv).

[51] These curves are presented on Figure 11 as well as midlatitude and vortex curves from M1998a. The data from set 2 fall on a line in between the SPIRALE vortex curve deduced and the midlatitude curve from M1998a, this reveals that the strong subsidence in the vortex occurs together with quasi horizontal mixing with air masses from outside the vortex [Ray *et al.*, 2002; Waugh *et al.*, 1997]. An extrapolation of SPIRALE “mixed” correlation line has been made to the midlatitude correlation curve (point C) given by M1998a. We assume that point C corresponds to the air mass composition outside vortex (64.5 ppbv for N_2O and 0.86 ppmv for CH_4) and that point A corresponds to the air mass composition in the mesospheric descent (2 ppbv for N_2O and 0.22 ppbv for CH_4). The SPIRALE measurement point B is the point where the maximum amount of midlatitude air is mixed with mesospheric air. Calculating

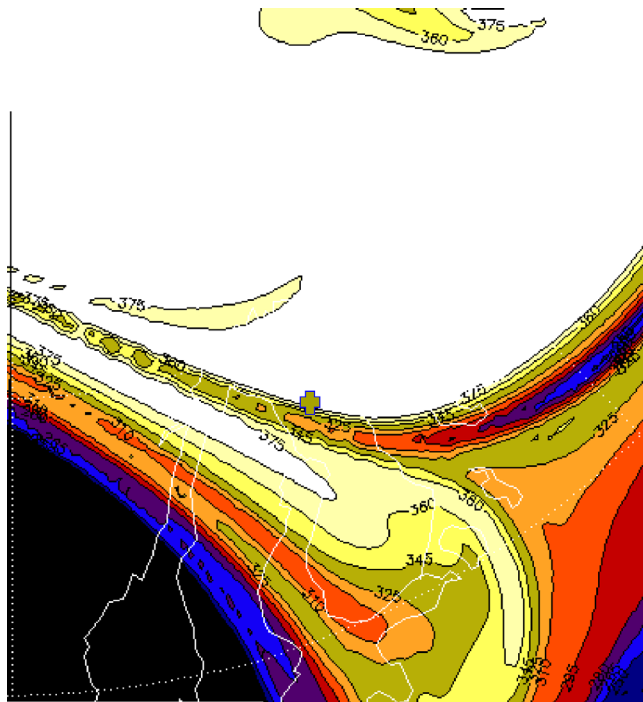


Figure 10. Same as Figure 4 on 21 January 2003 at high latitudes at 700 K; black region denotes air mass outside the vortex.

the ratio between $(CH_4(B) - CH_4(A)) / (CH_4(C) - CH_4(A))$ we found roughly 40% of stratospheric midlatitude air mixed with 60% of mesospheric air at point B. Comparing the CH_4 mixing ratio obtained with the January

HALOE climatology and the 0.86 ppmv CH_4 mixing ratio deduced from the midlatitude correlation curve (point C), it appears that the filament comes from a midlatitude layer located between 28 and 29 km. This has to be explored with the help of modeling study.

[52] For the high-latitude flight, SPIRALE has detected a large mesospheric descent with very low mixing ratio of N_2O and CH_4 not reported by M1998a. A well-identified layer between 27.8 and 29.5 km with high N_2O and CH_4 mixing ratio and a straight correlation curve has been observed. This layer is due to mixing between midlatitude air and mesospheric air inside the vortex. Note that this mixing process is present inside the vortex showing that the edge of the vortex is permeable as the PV filament suggests also.

5. Conclusion

[53] CH_4 and N_2O measured by the SPIRALE in situ TDLAS balloonborne instrument has been used to study the dynamical structure of the stratosphere at midlatitudes and in the polar region. The measurements have been compared with $CH_4:N_2O$ correlation curves derived from ATMOS measurements by Michelsen *et al.* [1998a].

[54] The high vertical resolution of the instrument allows us to highlight specific layers with thickness less than 1 km in the profiles. Tracer-tracer plots of the two species as well as potential vorticity maps from the MIMOSA model have been used to interpret in detail the measurements in terms of air mass origin. In both flights the instrument has sampled specific layers of different origin and affected by mixing processes.

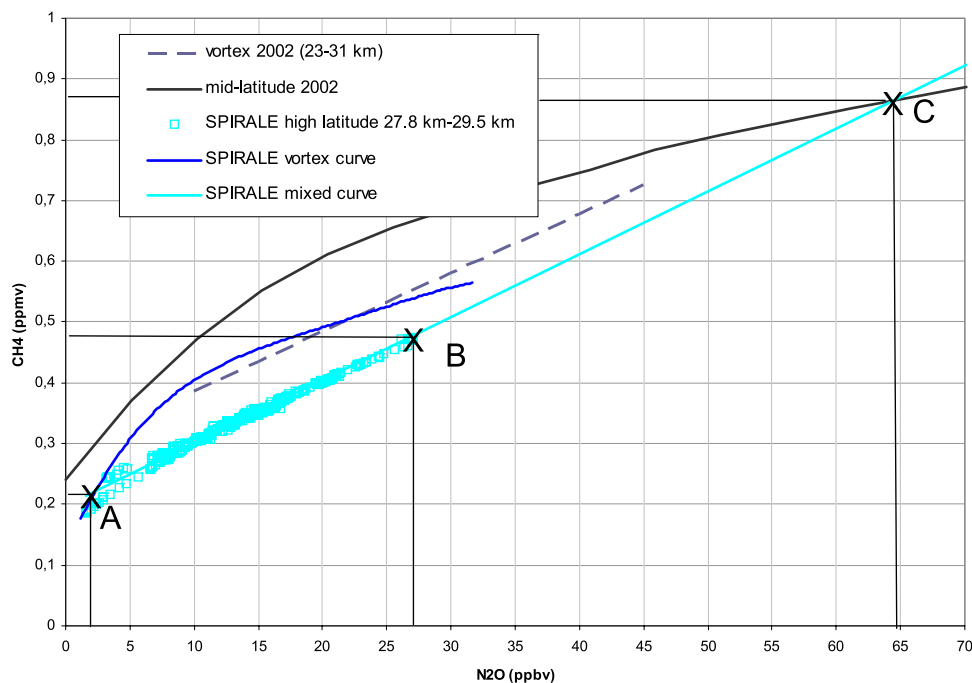


Figure 11. Same as Figure 3 in the range 0–70 ppbv for N_2O , but for the high-latitude flight in the range 27.8–29.5 km layer (clear blue squares). Correlation curves deduced from SPIRALE measurements for vortex in the ranges 23–27.8 km and 29.5–31 km layer (dark blue curve), and mixed curve in the range 27.8–29.5 km layer (clear blue curve). M1998a correlation curves for vortex conditions between 23 and 31 km (dashed gray line) and midlatitude conditions (black solid line) are shown.

[55] At midlatitudes below 16 km SPIRALE has sampled a tropospheric air mass coming from the tropics with a composition close to the Earth surface value. From 16 km to 23 km air masses with composition more and more typical of midlatitudes/protovortex composition have been observed, but with a layer coming from the stratospheric tropical region appearing between 16 km and 18 km. This layer probably corresponds to the “tropically controlled transition region” highlighted by *Rosenlof et al.* [1997]. This tropical stratospheric layer differs significantly from correlation curves recommended by *Michelsen et al.* [1998a]. A new specific linear correlation curve has been deduced from the measurements. This curve needs to be compared with tropical measurements in future studies. Above 18 km the air masses are composed of mixed tropical and midlatitude stratospheric air and mixing appears irreversible. Above 23 km, the nonmonotonic nature of the vertical profiles observed in association with significantly different $\text{CH}_4:\text{N}_2\text{O}$ correlations points indicate air masses of different origin. Above 29 km the air mass is more tropical than below it. Because the profiles are clearly nonmonotonic, mixing processes are probably not complete and could be reversible. Wind shear due to the first stage of the polar vortex explains the nonmonotonic nature of the profiles and why exchanges between polar region and tropical region have occurred. This needs to be verified using trajectory investigations.

[56] At high latitudes the SPIRALE instrument flew outside the polar vortex at the lowest heights and air masses typical of the midlatitude region composition were observed. Above 23 km, the measurements show a very intense mesospheric descent compared to those referred to by *Plumb et al.* [2002] because the mixing ratio of CO observed is as large as 600 ppbv. This large mesospheric descent has also been observed later, in March 2003 [*Engel et al.*, 2005]. Measurements allow us to determine new $\text{CH}_4:\text{N}_2\text{O}$ correlation values. Between altitudes of 27.8 km and 29.5 km, a layer with an increase in both N_2O and CH_4 is observed. These high mixing ratios for both species are due to mixing between midlatitude air and mesospheric air. Using tracer-tracer plots we deduce that this layer is composed of a maximum of 40% of midlatitude air and a minimum of 60% of mesospheric air present in the vortex. The dynamical situation revealed by PV maps shows that SPIRALE has crossed a midlatitude filament in this range of altitude. This midlatitude filament inside the vortex shows that the edge of the vortex is permeable and chemical composition is also modified inside the vortex. Measurements from the SMR instrument on the Odin satellite [*Urban et al.*, 2005] confirm this feature.

[57] For each flight the instrument has observed different layers in which mixing processes have occurred. These mixing processes appear to take place in a different manner; they may be complete or incomplete, reversible or irreversible. Knowledge of the different mixing processes is a key for the understanding of the chemical composition of the stratosphere. The SPIRALE measurements could help test and improve the efficiency of 3-D CTMs model in representing such mixing processes. In the future, we plan to use NO_2 , HNO_3 , HCl and HOCl mixing ratios also measured during these two flights to investigate the impact of such

mixing processes on the chemical partitioning of species in the NO_y and Cl_y families.

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