Mixing Processes during the Antarctic Vortex Split in September–October 2002 as Inferred from Source Gas and Ozone Distributions from ENVISAT–MIPAS

N. Glatthor, T. von Clarmann, and H. Fischer
IMK, Forschungszentrum Karlsruhe, and Universität Karlsruhe, Karlsruhe, Germany

B. Funke
Instituto de Astrofisica de Andalucia (CSIC), Granada, Spain

IMK, Forschungszentrum Karlsruhe, and Universität Karlsruhe, Karlsruhe, Germany

(Manuscript received 26 May 2003, in final form 5 January 2004)

ABSTRACT

In late September 2002, an Antarctic major stratospheric warming occurred, which led to a strong distortion of the southern polar vortex and to a split of its mid- and upper-stratospheric parts. Such an event had never before been observed since the beginning of regular Antarctic stratospheric temperature observations in the 1950s. The split is studied by means of nonoperational level-2 CH4, N2O, CFC-11, and O3 data, retrieved at the Institute for Meteorology and Climate Research Karlsruhe (IMK) from high-resolution atmospheric limb emission spectra from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on board the European research satellite, Environmental Satellite (ENVISAT). Retrieved horizontal and vertical distributions of CH4 and N2O show good consistency with potential vorticity fields of the European Centre for Medium-Range Weather Forecasts (ECMWF) analysis for the entire period under investigation, even for fine structures such as vortex filaments. Tracer correlation analysis suggests that mixing into the vortex had already occurred before the major warming and that vortex fragments were transported into the surrounding air masses on potential temperature levels above 400 K during the split. Correlation analysis of ozone with the source gases indicates slight ongoing ozone destruction in the lower-stratospheric vortex (below ~500 K) after the beginning of the warming event.

1. Introduction

In comparison to normal Antarctic winters, the winter of 2002 was very unusual, with three minor warmings during August and early September and a major warming at the end of September, which caused an increasing distortion of the vortex after 20 September and a split of its midstratospheric part (~ above the 650-K level of potential temperature, θ) around 25 September (Varotsos 2002). On the 10-hPa level, the major warming was characterized by a reversal of the zonal-mean wind into easterlies and by an increase of the zonal-mean temperature poleward of 60°S. Such an event had never been observed during Antarctic temperature observations since the 1950s (Roscoe et al. 2005; Simmons et al. 2005). After 25 September, one of the midstratospheric vortex fragments dissolved into the midlatitudes, whereas the lower-stratospheric part of the vortex and the remaining fragment at the beginning of October returned to the South Pole and persisted until November. Detailed descriptions of the vortex split are given in previous studies (e.g., Allen et al. 2003; Krüger et al. 2005 and references therein).

Tracer–tracer correlations have been investigated by various authors (e.g., Kondo et al. 1999; Rex et al. 1999; Plumb et al. 2000; Wetzel et al. 2002; Ray et al. 2002). Michelsen et al. (1998a,b) performed a comprehensive study using Atmospheric Trace Molecule Spectroscopy Instrument (ATMOS) data. For particular latitudinal regions, they found compact correlations for CH4 and N2O, only varying little between seasons and years. However, they observed significant differences between correlations representing the Tropics, midlatitudes, and polar regions. For the Tropics and for the midlatitudes, they measured a curved relationship; for
the springtime Arctic vortex, they measured a linear relationship. They explained the latter as mixing of the subsided vortex air with intruded midlatitude air masses.

Quasi-horizontal mixing, that is, mixing along isentropes, of air masses containing chemically inert trace gases with different volume mixing ratios (VMRs) can be observed if the undisturbed long-lived tracer–tracer correlation is compact and nonlinear. Incomplete mixing along an isentrope is manifested by a so-called linear “mixing line,” which connects different sections on the reference correlation (Waugh et al. 1997). In case of mixing of polar and midlatitude air masses the lower-VMR end of the line is called “inner-vortex end member” and the other end is called “out-of-vortex end member.” Determination of the height of the inner-vortex end member on a midlatitude VMR profile gives a rough estimate of subsidence inside the vortex. When the air masses are finally completely mixed, the mixing line contracts to a mixing point or cluster, the distance of which to the end members on the reference curve reflects the relative fractions of the original air masses. This picture has been refined by Plumb et al. (2000), who show that mixing lines can also be nonlinear, and by Ray et al. (2002), who propose differential subsidence at various locations inside the vortex prior to mixing to explain their measurements.

In this paper, we present an analysis of mixing processes in the Antarctic stratosphere in September and October 2002 from data measured with the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS).

2. The Michelson Interferometer for Passive Atmospheric Sounding

On 1 March 2002, MIPAS was brought into space on board the Environmental Satellite (ENVISAT), which is a sun-synchronous polar-orbiting satellite at about 800-km altitude with 98.55° inclination, performing 14.4 orbits per day. MIPAS is a limb-viewing Fourier transform infrared (FTIR) emission spectrometer with 0.035 cm⁻¹ spectral resolution (unapodized), covering the midinfrared region in five spectral bands, namely from 685 to 970 cm⁻¹ (band A), from 1020 to 1170 cm⁻¹ (band AB), from 1215 to 1500 cm⁻¹ (band B), from 1570 to 1750 cm⁻¹ (band C), and from 1820 to 2410 cm⁻¹ (band D) (Fischer and Oelhaf 1996; Harris 2000). The field of view (FOV) of MIPAS is 30 km in the horizontal across-orbit direction and approximately 3 km in the vertical direction. The data used in this investigation were taken in the standard observation mode, which consists in rearward limb scans covering the altitude range from 6 to 68 km within 17 steps. The step width is 3 km for the 13 lowermost tangent altitudes and increases to 8 km at the upper end of the limb scan.

Operational geophysical data products of MIPAS are provided under European Space Agency (ESA) responsibility, but only for the main targets: temperature, H₂O, CH₄, N₂O, O₃, HNO₃, and NO₂. Since we want to investigate additional target species during the vortex split, and in order to have full diagnostic metadata available, we created an independent and self-consistent dataset at the Institute for Meteorology and Climate Research Karlsruhe (IMK), which is used in this paper.

Unfortunately, MIPAS was switched off during the vortex split because of a failure of the Stirling cooler. Thus, radiance spectra were only available for 20–27 September, which covers the initial split on 25 September (cf. Allen et al. 2003), and for 11–13 October. Moreover, for these days, only subsets of the maximum number of ~14 orbits per day were available, amounting to 59 orbits in total. For this study, only the measurements south of ~30° geographic latitude were analyzed at IMK, which accounted for more than 1100 limb scans.

3. Data processing

MIPAS limb radiance spectra are inverted to vertical profiles of atmospheric state parameters by constrained nonlinear least squares fitting of modeled to measured spectra. A detailed description of the overall approach is given in von Clarmann et al. (2003a). Prior to retrieval of trace gases, temperature and tangent altitudes are retrieved (von Clarmann et al. 2003b). As customary in infrared spectroscopy, retrievals were not performed using the whole spectral range but only in small microwindows, that is, spectral regions with strong signatures of the target species and as-small-as-possible contributions of interfering species. These microwindows were not defined empirically, but via a quantitative mathematical method by minimizing the estimated retrieval error, and were collected in databases (von Clarmann and Echle 1998). Then, for each target species, height-dependent combinations of microwindows with a trade-off between computation time and retrieval error were calculated using an optimization scheme proposed by Echle et al. (2000). The microwindows selected for joint retrieval of CH₄ and N₂O VMRs lie in the spectral region 1230–1305 cm⁻¹ (Table 1), and those used for ozone retrieval are situated in the spectral regions 741–799 and 1062–1108 cm⁻¹. For CFC-11 evaluation, one large analysis window ranging from 838 to 853 cm⁻¹ was taken. Retrieval parameters were the respective target, for example, the N₂O and CH₄ VMRs, a microwindow-dependent empirically fitted background radiation continuum term, and a microwindow-dependent but height-independent radiometric zero-level calibration correction. The continuum was forced to zero above 32-km altitude, where its contribution becomes negligible. The retrieval grid used for the targets had a 1-km altitude spacing up to
44-km altitude and a 2–10-km spacing up to 120 km with up to 60 levels, which is considerably finer than the tangent height spacing. Thus, to avoid instabilities, a smoothing constraint was applied for the retrieval of profiles, using Tikhonov’s first-derivative operator as implemented by Steck and von Clarmann (2001). For CH\(_4\) and N\(_2\)O, the resulting vertical resolution, that is, the half-width of the response of the retrieval to a delta perturbation of the profile, was typically between 4 and 5 km in the altitude range 20–50 km and decreased to more than 10 km below and above this region. The degradation of the altitude resolution compared to the measurement step size is caused by the smoothing constraint selected, which is considered as an optimal trade-off between vertical resolution and precision of the data.

Because of the opacity of limb paths through the lower atmosphere, which amplifies most kinds of retrieval errors, the two lowermost tangent altitudes of MIPAS were excluded from analysis in case of CH\(_4\) and N\(_2\)O retrievals, and the retrieval grid started from 10 km. Because of its rapid decrease in the stratosphere, CFC-11 was only retrieved from the nine lowermost tangent altitudes (6–30 km).

### 4. Error estimation

For all profiles presented here, the following diagnostic parameters were calculated: retrieval error caused by measurement noise, vertical height resolution, and number of degrees of freedom, that is, number of independent pieces of information in the retrieved profile. Moreover, for representative scans, the total retrieval error was determined, which contains, besides measurement noise, retrieval errors related to uncertainties in the temperature profile, the instrument line-of-sight pointing, the VMR profile of each interfering gas, spectroscopic data, spectral shift, gain calibration, instrumental line shape (ILS), and errors induced by radiative transfer calculations without considering non-local thermodynamic equilibrium (NONLTE). All diagnostics are based on the actual retrieved temperatures, tangent heights, source gas mixing ratios, and simulated spectra and Jacobians from the final iteration. For species that were not jointly retrieved, the climatological vertical profiles and variabilities were used. The error analysis follows the methodology proposed by Rodgers (1990). In Tables 2 and 3, we show total estimated retrieval errors of CH\(_4\) and N\(_2\)O as well as noise-induced random errors and errors induced by uncertain parameters for a scan of 26 September from southern midlatitudes. For CH\(_4\), the total error ranges

### Table 1. Microwindows used for joint retrieval of CH\(_4\) and N\(_2\)O (right) Logical matrix shows at which heights between 5 and 68 km the respective microwindow is used (T) and not used (=).

<table>
<thead>
<tr>
<th>Microwindow</th>
<th>Altitude coverage</th>
</tr>
</thead>
<tbody>
<tr>
<td>cm(^{-1})</td>
<td>5–68 km, 3 km steps</td>
</tr>
</tbody>
</table>
| 1231.6750–1231.9250 | TTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTTT...
from 0.07 to 0.28 ppmv, and for N$_2$O, it ranges from 1 to 43 ppbv. The noise errors are considerably lower and lie between 0.02 and 0.14 ppmv for CH$_4$ and between less than 1 and 12 ppbv for N$_2$O, respectively. For both gases, the dominant errors are uncertainties in temperature, spectroscopic data, and the ILS. In section 7, these retrieval errors are compared with the scatter of the CH$_4$-N$_2$O data to check the compactness of the correlation.

Total CH$_4$ errors for scans inside the vortex fall from 0.3 ppmv at 15 km to 0.03–0.05 ppmv in the mid-and upper stratosphere, whereas the noise errors fall from 0.1 to 0.02 ppmv. For N$_2$O, the total error inside the vortex decreases from 50 ppbv to less than 1 ppbv, and the noise error decreases from 10 to less than 1 ppbv for the same altitude range. The total O$_3$ error in the region of the ozone maximum is 1.5 and 1 ppmv for midlatitude and inner-vortex profiles, respectively, and decreases considerably toward lower and higher altitudes. The random error is much lower for the midlatitudes as well as for the polar region, namely between 0.1 and 0.2 ppmv over the entire altitude range. The total CFC-11 error falls from about 0.02 to 0.005 ppbv from the lower to the midstratosphere, respectively, for both midlatitude and polar conditions, and the noise error is about 0.004 ppbv. The retrieval errors listed in this chapter can be used to judge the scatter of all other correlations shown in section 7.

Beyond the errors assessed above, retrieved mixing ratios of N$_2$O and CH$_4$ are biased high with respect to the climatological tropospheric values of about 320 ppbv and 1.8 ppmv, presumably caused by a gain calibration problem that for some reason affects particularly the MIPAS band B. This high bias also appears in the operational ESA data product. Since there seems to be an additional gain calibration drift, relevant to the time period under investigation in this paper, one has to be careful in drawing conclusions from the temporal development of N$_2$O or CH$_4$ values alone, while, as will be discussed below, the correlations of these species prove to be reasonably robust against this problem.

### 5. Horizontal shape of the vortex on the 550-K level

In this section, we compare our retrieved tracer fields with potential vorticity (PV) of the European Centre for Medium-Range Weather Forecasts (ECMWF) to gain insight into the period of the vortex split and into possible mixing of air masses from inside and outside of the vortex. For this reason, we defined the vortex boundary region to extend from −12 to −20 PV units (PVU; 1 PVU = 1.0 × 10$^{-6}$ m$^2$ s$^{-1}$ K kg$^{-1}$) on the 400-K level of potential temperature, from −30 to −45 PVU on the 475-K level, from −55 to −90 PVU on the 550-K level, from −90 to −155 PVU on the 625-K level, and from −150 to −270 PVU on the 700-K level (cf. Fig. 3). We did this by inspection of the PV gradients in combination with the following analytical scaling factor (Lait 1994):

$$\text{PV}(\Theta) = \text{PV}_0 \times \left(\Theta/\Theta_0\right)^{4.5},$$

where PV$_0$ is the value for the inner or outer edge of the vortex boundary on $\Theta_0 = 475$ K, that is, −45 and −30 PV units, respectively. In our definition, the boundary region between $\Theta = 400$ K and $\Theta = 700$ K is rather wide to classify measurements safely as outside or inside the vortex. For levels above this range of potential temperatures, we scaled the edge values from $\Theta = 475$ K...
strictly according to Eq. (1) to define the boundary region. For the lowermost altitudes, we used the vortex boundary definition from the 400-K level.

In Fig. 1, the temporal and spatial development of the Antarctic vortex according to the N$_2$O distribution and to potential vorticity is illustrated for the periods 20–27 September and 11–13 October on the 550-K level of potential temperature, which corresponds to heights between 18.5 and 23 km. As mentioned above, radiance spectra were not available for the time in between. The red quasi-oval bands indicate the vortex boundary, derived from potential vorticity fields of the ECMWF for 1200 UTC. The color-coded symbols represent the retrieved N$_2$O VMRs in ppmv, whereby symbols enclosed by boxes mark the beginning of the partial orbits on the dayside. Orbits depicted by triangles are also shown as vertical cross sections in Fig. 2. Data gaps are geolocations, where the retrieval did not converge. According to the shape of its boundary, the vortex is still relatively undisturbed on 20 and 21 September, but between 22 and 27 September, it is displaced toward the southern Atlantic and becomes oblong and bipolar. The strongest constriction is visible around 25 September; however, according to PV, the vortex did not completely split at this altitude. As shown by Allen et al. (2003) and Hoppel et al. (2003) as well as by Konopka et al. (2005), Krüger et al. (2005), and Manney et al. (2005), a complete split occurred for the mid- and upperstratospheric part of the vortex (above ~650 K), and one of these fragments dissolved into midlatitude air masses. On 26 and 27 September, the vortex started to recover on the 550-K level, and on 11–13 October, it was centered above the South Pole again, however, considerably smaller than before the split.

During the whole period, the N$_2$O VMRs are low inside (≈100 ppbv) and significantly higher (~200 ppbv) outside the vortex. Thus, the horizontal shape of the vortex agrees well in both PV and MIPAS data. Moreover, there is even agreement in some finer structures; in the region of the strips of high PV, visible on 11 and 13 October above the southern Indian Ocean, South America, and the southern Pacific, lower N$_2$O-values were also measured. Thus, here both datasets correspondingly indicate vortex filaments.

6. Vertical sections through the vortex along representative orbits

In Fig. 2, retrieved N$_2$O, CH$_4$, and O$_3$ VMRs as well as potential vorticity from the ECMWF are plotted versus relative latitude and altitude along representative partial orbits, whereby relative latitude is the coordinate along ENVISAT’s orbit. Geographical latitude corresponds to relative latitude between −30° and −90° on the satellite’s daytime downward leg and to (−180°-relative latitude) on the nighttime upward leg. For clearer illustration, PV from all Θ levels was scaled to the 475-K level according to Eq. (1). The black lines denote the 400-, 550-, 700-, 850-, 1000-, 1250-, and 1500-K levels of potential temperature, derived from preceding pressure and temperature retrievals. The orbits chosen are orbit 2915 (20 September), orbit 2972 (24 September), and orbit 3246 (13 October), that is, from the beginning, during, and after the vortex split. As already mentioned in section 4, the N$_2$O and CH$_4$ VMRs of the lower altitudes (~10–20 km) are too high, especially for the September data.

On orbit 2915, both N$_2$O and CH$_4$ VMRs (left column) are considerably lower between −60° and −110° relative latitude in the height region above 15 km as compared to the surrounding lower latitudes. In good agreement with potential vorticity, this shows the location of the vortex, where high-altitude air masses containing low CH$_4$ and N$_2$O have subsided. The horizontal gradients are rather steep, especially between 20 and 40 km, corresponding to a well-defined vortex boundary. According to both source gases as well as to PV, the vortex boundary is tilted to the left, which shows the beginning deformation of the vortex. The O$_3$ VMRs are considerably lower inside the vortex than the surrounding midlatitude values throughout the stratosphere. Below about 20–25 km (600–650 K), this is caused by chemical ozone destruction by activated chlorine, whereas in the midstratosphere the main reason for lower ozone is the dynamical isolation of the vortex. This is discussed in more detail in sections 7c and 7d.

Because of the displacement and deformation of the vortex, on orbit 2972 (middle column) vortex air masses were observed at much lower latitudes between −35° and −65°. From −65° to −85°, midlatitude air masses were encountered at all altitudes. However, both CH$_4$ and N$_2$O VMRs indicate a second encounter of polar air masses from −85° to −125° between the 475 and 700-K isentropes (altitude region 15 to 25 km) and midlatitude air masses above. Even these complex N$_2$O and CH$_4$ cross sections are in good agreement with PV data (third row, middle column). Accordingly, the respective plot in Fig. 1 (orbit of 24 September marked by triangles) shows that these measurements were well inside the vortex on the 550 K isentrope. However, according to PV, on 700 K, extravortex air was observed. Thus, the “blob-like” feature is caused by the tilted vortex edge. The vertical tilt of the vortex was also shown by Allen et al. (2003), for the following day (25 September), when the situation was still rather similar. Manney et al. (2005) and Randall et al. (2005) also discuss in detail the vertical structure and evolution of the vortex, as seen in modeled CH$_4$ and N$_2$O (Manney et al. 2005) and in modeled and measured ozone (Randall et al. 2005). In agreement with the source gases, MIPAS observed low O$_3$ VMRs between −35° and −65° and between −90° and −125° around 20 km.

On orbit 3246 (right column), low N$_2$O and CH$_4$ VMRs are centered around the South Pole again, which shows the recovery of the vortex (cf. Fig. 1). Again,
FIG. 1. Southern hemispheric N2O distribution (filled squares and triangles) at the 550-K level of potential temperature, measured by MIPAS–ENVISAT on 20–27 Sep and 11–13 Oct 2002. Symbols enclosed by open squares mark the beginning of each partial orbit on the dayside. The red ovals indicate the vortex boundary, derived from PV fields of the ECMWF. Partial orbits marked by triangles are shown as vertical cross sections in Fig. 2.
there is very good agreement with location and extension of the vortex as seen in potential vorticity. Furthermore, even fine structures in PV like the minima at 35-km altitude and −40° and −120° relative latitude, indicating filamentation, correspond to low source gas VMRs. In comparison to orbit 2915, the area of low CH₄ and N₂O VMRs and of low PV values is smaller (cf. Fig. 1) and decreases with altitude, stretching from −65° to −110° at the 475-K level of potential temperature, but only from −80° to −100° at the 850-K level.
Above this altitude, the vortex is less pronounced, and at the 1500-K isentropic level it disappears, which is especially evident in the PV plot. The ozone distribution is also symmetrical around the South Pole again, but inside the vortex, the O$_3$ VMRs are somewhat lower between 25- and 40-km altitude than on orbit 2915.

In summary, there is good agreement between vertical cross sections of N$_2$O, CH$_4$, and potential vorticity for general structures like the vortex boundary, but also for more complex features like the blob observed on orbit 2972 and for the upper-stratospheric fine structure from orbit 3246.

7. Correlations

To check data consistency and to gain more information on horizontal mixing of midlatitude and polar air masses caused by the vortex split, we correlated the retrieved trace gases with potential vorticity and with each other. We used the midlatitude trace gas correlations of 20 and 21 September as reference, that is, from the beginning of the vortex split, and compared them with midlatitude correlations of 22–27 September and 11–13 October, and with polar correlations from 20–21 September and 11–13 October.

a. N$_2$O versus potential vorticity

In Fig. 3, the retrieved N$_2$O VMRs are plotted versus potential vorticity for the 400-, 475-, 550-, 625-, and 700-K levels of potential temperature. The blue, green, and red data points are from 20–21 September, 22–27 September, and 11–13 October, respectively. On each level of potential temperature, the dense clusters of
values with high VMRs at low absolute PV values are extravortex measurements, whereas the point clouds with low VMRs at high absolute PV values are from inside the vortex. The more sparsely covered gradient region in between is the vortex boundary region, whereby the vertical lines mark its inner and outer edge according to our definition (cf. section 5). Because of the weakened vortex, the inner-vortex PV values of October are much lower on the 550-, 625-, and 700-K levels than before the split. The systematic decrease in N2O from September to October, mainly visible on the 400- to 550-K levels, is caused by an artifact discussed more in detail in section 7b(2).

b. CH4 versus N2O

1) DISCUSSION OF 20–21 SEPTEMBER CORRELATIONS

The left diagram in Fig. 4 shows the reference CH4–N2O correlation of 20–21 September for all extravortex air masses south of −30°, whereby the points are color coded with respect to altitude. This correlation is very compact. Chi-square analysis associated with the polynomial fit (black line) resulted in an χ²-value above 1 (χ² = 2.08), when the scatter of the correlation was related to the measurement noise of N2O and CH4 (cf. section 4). Comparison of the scatter of the data points with the total retrieval errors of N2O and CH4 resulted in χ² below 1 (χ² = 0.15). Thus, the scatter of the correlation lies in between the estimated noise and total errors but closer to the former. This means that the spread of the dataset is mainly governed by measurement noise and not strongly affected by other uncertain parameters, which is consistent with the analytical error analysis in section 4, because some, but not all, of the errors beyond measurement noise also have random variability. Furthermore, a sensitivity study based on perturbed temperature profiles showed that the resulting changes of the N2O and CH4 VMRs were correlated with a slope of −6, while the CH4–N2O correlation itself has a slope of −3.6, for N2O values larger than 50 ppbv, respectively. Thus, such error components would move a data point with a rather small component perpendicular to the correlation line. This may explain why the correlation is somewhat less compact than expected from measurement noise alone. Evidence for systematic drifts of retrieved temperatures has not been found (D.-Y. Wang 2003, unpublished manuscript).

The correlation is nonlinear between zero and 250 ppbv N2O and nearly linear above. A similar feature

Fig. 3. N2O VMRs from MIPAS–ENVISAT vs PV of the ECMWF for 20–21 Sep (blue points), 22–27 Sep (green points), and 11–13 Oct (red points). Top to bottom: 400-, 475-, 550-, 625-, and 700-K levels of potential temperature. The vertical lines mark the inner and outer edge of the vortex boundary region.
has been observed in the midlatitudes by Michelsen et al. (1998a,b) and is plotted as a blue curve. The solid green curve is a linear regression fit to in situ measurements between 44° and 68° N from Engel et al. (1996).

In comparison to these two datasets, the MIPAS–ENVISAT correlation is slightly shifted toward higher CH$_4$. Furthermore, as discussed in section 4, the N$_2$O and CH$_4$ VMRs are biased high: they extend up to 400 ppbv and 2.5 ppmv, whereas the climatological tropospheric values, which are the maximum values expected, are about 320 ppbv and 1.8 ppmv, respectively.

The correlation of 20–21 September for vortex air masses (right diagram of Fig. 4) has a larger scatter. Furthermore, it is more linear for N$_2$O values between 50 and 300 ppbv and thus is hardly suited as a reference for the detection of additional mixing into the vortex during the September–October period. The stronger linearity and larger scatter is an indication for mixing of midlatitude air into the vortex at any time during the winter prior to the investigated period. The blue and green curves are fits to vortex ATMOS data (Michelsen et al. 1998a,b) and to Arctic MIPAS balloon FTIR measurements (Wetzel et al. 2002). The offset between the MIPAS–ENVISAT data and these measurements is smaller than in the midlatitudes.

2) Comparison of midlatitude correlations

In Fig. 5 (top left), all extravortex CH$_4$ measurements south of −30° of 20–21 September (red points) and 22–27 September (blue points) are correlated with the respective N$_2$O data. The majority of the 22–27 September data lie atop the 20–21 September correlation, but a certain amount covers the region below, especially for N$_2$O VMRs between 0 and 250 ppmv (above ~450 K). This more linear portion indicates vortex air masses drawn out into the midlatitudes, probably not yet homogeneously mixed but as filaments. According to Konopka et al. (2005), the time constants for homogenous mixing were of the order of 3 and 6 weeks at 900 and 700 K, respectively. Filamentation has also been shown for this period by Allen et al. (2003) on the 460- and 850-K levels, by Randall et al. (2005) for 600 K and 850 K, and by Konopka et al. (2005) at 500 and 800 K.

In the top right diagram of Fig. 5, we show all extravortex CH$_4$ and N$_2$O measurements of 20–21 September (red points) and 11–13 October (blue points). Here, even the majority of points form different curves between N$_2$O values of 50 and 230 ppbv; the September correlation exhibits a kind of bulge, whereas the October correlation is more linear in this region. This is an indication for more completed mixing of vortex filaments, which had been stripped off the vortex during the split, into the midlatitudes. As mentioned above, this is supported by the dissolution of the second vortex fragment into the midlatitudes. However, the October data at lower altitudes are, especially for N$_2$O, systematically lower than the September data and no longer significantly exceed the climatological tropospheric values. It turned out that this systematic deviation is proportional to VMR (cf. Fig. 3) and extends up into the stratosphere, which means that the October N$_2$O and CH$_4$ axes are contracted in comparison to the September axes. This makes quantitative conclusions about mixing between 20–21 September and 11–13 October difficult. Nevertheless, scaling of the N$_2$O and CH$_4$ data of September to the October axes would not remove the differences between 50 and 230 ppbv.
In the bottom diagram of Fig. 5, the midlatitude correlation of October is separately displayed for several theta intervals between 320 and 820 K along with the reference correlation of 20–21 September. The data between 320 and 340 K are centered on the reference but shifted downward along the reference. Since this theta range covers altitudes where no pronounced vortex is identifiable, the shift is caused by the bias of the September data discussed earlier. At 400–420 K, most October data also cover the reference curve, with a slight indication of a mixing line below the reference. October data between 460 and 480, 540 and 560, and 640 and 660 K form clusters well outside the reference correlation, with an indication of some remains of mixing lines. The data of 800–820 K form a cluster at the lower edge of the reference correlation. The shifts of the clusters between 460–480 and 800–820 K have a larger negative component along the CH4 axis than the shift at 320–340 K. Therefore, we conclude that, despite the systematic bias, there remains indication for intrusion of vortex air above 400 K into the midlatitudes between 20–21 September and 11–13 October.

3) COMPARISON OF MIDLATITUDE AND POLAR CORRELATIONS

Figure 6 (left) contains the midlatitude reference correlation of 20–21 September and vortex measurements of the same time period for the same theta ranges as in the last section. Since these datasets are from the same days, they are free of the systematic bias between September and October. The vortex data between 320 and 340 K are centered on the reference correlation for the same reason as explained above, whereas on the higher theta levels the vortex data extend into the area below the reference curve. This result indicates that midlatitude air had already been mixed into the vortex at any time before the split on theta levels above 400 K. Comparison of the midlatitude reference with vortex measurements of 11–13 October (Fig. 6, right) shows a rather similar picture. Thus, additional mixing of midlatitude air into the vortex during the vortex split cannot be proven from our data. However, evidence of this process is given by Manney et al. (2005) and Randall et al. (2005).

c. O3 versus N2O

Figure 7 shows the vortex O3 measurements of 20–21 September (red points) and 11–13 October (blue points) versus N2O. Because of the shapes of these distributions (mainly vertical for N2O < 30 ppbv and nearly horizontal for N2O > 50 ppbv), quantitative conclusions can be drawn from this diagram, because the shift of a data point along the N2O axis caused by the bias drift would not significantly affect observed differ-
ences in ozone VMRs. The midstratospheric maximum O3 VMRs (around 20 ppbv N2O) of September range up to 6.5 ppmv, whereas in October they do not exceed 5 ppmv. This is true even if the inner-vortex edge is relaxed to −1000 PV units on the 1000-K level of potential temperature (corresponds to −35 scaled PV units; cf. Fig. 2) to account for the weakened vortex in October. The difference between O3 VMRs of September and October is confirmed by Karlsruhe SImulation of the Middle Atmosphere (KASIMA) model calculations (R. Ruhnke 2004, personal communication). The reason for this is probably ozone reduction by enhanced Antarctic NOx in October (B. Funke 2003, unpublished manuscript), which will be further investigated.

Because of ozone depletion by activated chlorine during Antarctic spring, the lower-stratospheric ozone nearly forms uniform distributions of very low ozone for N2O VMRs larger than 50 ppbv. This feature is in good agreement with ATMOS correlation analysis (Michelsen et al. 1998a,b). Furthermore, ozone VMRs in the lower stratosphere in October exhibit even less ozone (∼0.5 ppmv) than those of 20–21 September (∼0.8 ppmv). This observation is confirmed by Polar Ozone and Aerosol Measurement III (POAM III) measurements, which show a persisting ozone loss of 25%–50% from 17 September to 7 October (albeit less than in previous Antarctic winters) between about 350 and 570 K (Hoppel et al. 2003). This decrease indicates a certain amount of ongoing chemical ozone depletion after 21 September. This is in agreement with model calculations of Grooß et al. (2005), which resulted in ozone depletion rates persisting, even though rapidly decreasing, until about 26 September for theta levels between 425 and 550 K.

d. O3 versus CFC-11

To gain unbiased information on ozone changes between 20–21 September and 11–13 October, we also correlated ozone with CFC-11 (Fig. 8). For the midlatitudes (left), both datasets nearly overlap, and ozone values reach maximum values of 9 ppmv. However, some October data points are found below the September correlation and form an L-shaped distribution similar to the vortex correlation (right). This, again, hints at intrusion of vortex air into the surrounding air masses. The right diagram of Fig. 8 shows vortex ozone for 20–21 September and 11–13 October. In good agreement with the respective O3–N2O correlation, the midstratospheric vortex O3 data of October are restricted to lower VMRs than the September data. Furthermore, the lower-stratospheric O3 data are also slightly lower in October than in September, indicating some ongoing chemical ozone destruction. Lower maximum ozone VMRs as compared to the extravortex measurements are primarily caused by the dynamical isolation of the vortex.

FIG. 6. (left) All extravortex CH4 and N2O data of 20–21 Sep (black crosses) and CH4 and N2O data of the same time period from inside the vortex on different levels of potential temperature. Red circles: 320–340 K; blue circles: 400–420 K; green circles: 460–480 K; yellow circles: 540–560 K; red squares: 640–660 K; and blue triangles: 800–820 K. (right) All extravortex CH4 and N2O data of 20–21 Sep (red points) and all vortex data of 11–13 Oct (blue points).

FIG. 7. All vortex O3 and N2O data of 20–21 Sep (red points) and 11–13 Oct (blue points).
8. Summary and conclusions

In September–October 2002, a split of the Antarctic polar vortex occurred, an event never observed until that point. To study the vortex split, at IMK, a self-consistent dataset of various atmospheric trace gases was inferred from MIPAS radiance spectra for the periods 20–27 September and 11–13 October. In this paper, we analyzed the retrieved N$_2$O, CH$_4$, CFC-11, and O$_3$ distributions by comparing them with each other and with potential vorticity of the ECMWF.

During the whole analyzed period, there was good agreement between the horizontal distribution of N$_2$O VMRs and the shape of the vortex, derived from potential vorticity. Comparison of vertical cross sections through the vortex showed good agreement between N$_2$O, CH$_4$, and potential vorticity, even for smaller and complex features. Thus, in both horizontal and vertical cross sections, the intermittent distortion and skewness of the vortex was consistently visible.

Correlation of N$_2$O VMRs with potential vorticity provided a check of the definition of the vortex boundary region used. For correlations between trace gases, the extravortex data of 20–21 September were taken as a relatively undisturbed reference. The reference CH$_4$–N$_2$O correlation turned out to be very compact and had a similar nonlinear shape between 0 and 250 ppbv N$_2$O as seen in ATMOS data (Michelsen et al. 1998a,b). The 20–21 September correlation from inside the vortex showed a larger scatter and was nearly linear between 50 and 250 ppbv N$_2$O. This is an indication that mixing into the vortex had already happened during the winter prior to the investigated period. Closer comparison with the extravortex reference suggested mixing mainly in the altitude range above 400 K. Comparison of the midlatitude 20–21 September reference with vortex data from 11–13 October practically showed the same state, that is, mixing into the vortex above 400 K, which means that no further significant mixing into the vortex can be inferred from these data for the investigated period.

The extravortex CH$_4$–N$_2$O correlation of 22–27 September showed a linear portion of data points below the reference curve, and the extravortex correlation of 11–13 October showed a generally more linear course between 50 and 250 ppbv N$_2$O. Both observations indicate mixing of vortex air into the midlatitudes above 400 K. Correlations of vortex O$_3$ with N$_2$O and CFC-11 for September and October data showed a reduction by 0.3–0.5 ppmv in the lower stratosphere caused by ongoing chemical ozone destruction.

Acknowledgments. Part of this work has been funded under EC Contract EVG1–CT–1999–00015 (AMIL2DA), ESA Contract 15530/01/NL/SF (INFLIC), and BMBF Contracts 07 ATF 43/44 (KODYACS), 07 ATF 53 (SACADA), and 01 SF 9953 (HGF Vernetzungsfonds). B. Funke has been supported through a European Community Marie Curie Fellowship. The authors would like to thank ESA for giving access to MIPAS level-1 data. Meteorological analysis data have been provided by ECMWF. Furthermore, the authors acknowledge the thorough and helpful comments of a reviewer.

REFERENCES


Fischer, H., and H. Oelhaf, 1996: Remote sensing of vertical pro-

Fig. 8. (left) All extravortex O$_3$ and CFC-11 data of 20–21 Sep (red points) and 11–13 Oct (blue points). (right) Same as left, but for vortex data.
Kondo, Y., and Coauthors, 1999: NOy \textendash} N$_2$O correlation observed inside the Arctic vortex in February 1997. Dynamical and chemical effects. J. Geophys. Res., 104, 8215–8224.