

Odin/SMR limb observations of stratospheric trace gases: Validation of N₂O

J. Urban,^{1,2} N. Lautié,³ E. Le Flochmoën,^{1,4} C. Jiménez,^{3,5} P. Eriksson,³ J. de La Noë,¹ E. Dupuy,¹ L. El Amraoui,¹ U. Frisk,⁶ F. Jégou,¹ D. Murtagh,³ M. Olberg,⁷ P. Ricaud,^{1,4} C. Camy-Peyret,⁸ G. Dufour,⁸ S. Payan,⁸ N. Huret,⁹ M. Pirre,⁹ A. D. Robinson,¹⁰ N. R. P. Harris,¹¹ H. Bremer,¹² A. Kleinböhl,¹² K. Küllmann,¹² K. Kunzi,¹² J. Kuttippurath,¹² M. K. Ejiri,¹³ H. Nakajima,¹³ Y. Sasano,¹³ T. Sugita,¹³ T. Yokota,¹³ C. Piccolo,¹⁴ P. Raspollini,¹⁵ and M. Ridolfi¹⁶

Received 29 August 2004; revised 10 December 2004; accepted 19 January 2005; published 3 May 2005.

[1] The Sub-Millimetre Radiometer (Odin/SMR) on board the Odin satellite, launched on 20 February 2001, performs regular measurements of the global distribution of stratospheric nitrous oxide (N₂O) using spectral observations of the $J = 20 \rightarrow 19$ rotational transition centered at 502.296 GHz. We present a quality assessment for the retrieved N₂O profiles (level 2 product) by comparison with independent balloonborne and aircraftborne validation measurements as well as by cross-comparing with preliminary results from other satellite instruments. An agreement with the airborne validation experiments within 28 ppbv in terms of the root mean square (RMS) deviation is found for all SMR data versions (v222, v223, and v1.2) under investigation. More precisely, the agreement is within 19 ppbv for N₂O volume mixing ratios (VMR) lower than 200 ppbv and within 10% for mixing ratios larger than 150 ppbv. Given the uncertainties due to atmospheric variability inherent to such comparisons, these values should be interpreted as upper limits for the systematic error of the Odin/SMR N₂O measurements. Odin/SMR N₂O mixing ratios are systematically slightly higher than nonvalidated data obtained from the Improved Limb Atmospheric Spectrometer-II (ILAS-II) on board the Advanced Earth Observing Satellite-II (ADEOS-II). Root mean square deviations are generally within 23 ppbv (or 20% for VMR-N₂O > 100 ppbv) for versions 222 and 223. The comparison with data obtained from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on the Envisat satellite yields a good agreement within 9–17 ppbv (or 10% for VMR-N₂O > 100 ppbv) for the same data versions. Odin/SMR version 1.2 data show somewhat larger RMS deviations and a higher positive bias.

Citation: Urban, J., et al. (2005), Odin/SMR limb observations of stratospheric trace gases: Validation of N₂O, *J. Geophys. Res.*, 110, D09301, doi:10.1029/2004JD005394.

¹Observatoire Aquitain des Sciences de l'Univers, L3AB, Floirac, France.

²Now at Department of Radio and Space Science, Chalmers University of Technology, Göteborg, Sweden.

³Department of Radio and Space Science, Chalmers University of Technology, Göteborg, Sweden.

⁴Now at Laboratoire d'Aérodynamique, Observatoire de Midi-Pyrénées, Toulouse, France.

⁵Now at Institute of Atmospheric and Environmental Science, School of Geosciences, University of Edinburgh, Edinburgh, UK.

⁶Swedish Space Corporation, Solna, Sweden.

⁷Onsala Space Observatory, Chalmers University of Technology, Onsala, Sweden.

⁸Laboratoire de Physique Moléculaire et Applications, Université Pierre et Marie Curie, Paris 6, Paris, France.

⁹Laboratoire de Physique et Chimie de l'Environnement, Université d'Orléans, Orléans, France.

¹⁰Centre for Atmospheric Science, Department of Chemistry, University of Cambridge, Cambridge, UK.

¹¹European Ozone Research Coordinating Unit, Centre for Atmospheric Science, Department of Chemistry, University of Cambridge, Cambridge, UK.

¹²Institute of Environmental Physics, University of Bremen, Bremen, Germany.

¹³National Institute for Environmental Studies, Tsukuba, Ibaraki, Japan.

¹⁴Atmospheric, Oceanic and Planetary Physics, Department of Physics, University of Oxford, Oxford, UK.

¹⁵Istituto di Fisica Applicata "Nello Carrara" del CNR, Firenze, Italy.

¹⁶Dipartimento di Chimica Fisica ed Inorganica, Università di Bologna, Bologna, Italy.

1. Introduction

[2] Nitrous oxide in the atmosphere is produced by biological and industrial processes at the ground and is well mixed in the troposphere. From preindustrial values of ~ 270 ppbv, its abundance has been increasing steadily at the nearly linear growth rate of ~ 0.75 ppbv yr^{-1} during the past decades, reaching volume mixing ratios (VMR) of 315–317 ppbv in 2001 [World Meteorological Organization (WMO), 2003]. Entering the stratosphere mainly through the tropical tropopause, it is destroyed at higher altitudes by photodissociation ($\lambda \approx 185\text{--}210$ nm, $\sim 90\%$) and reaction with electronically excited oxygen atoms ($\text{O}(^1\text{D})$, $\sim 10\%$) [e.g., Minschwaner et al., 1993; Yung and Miller, 1997], making it the main source of ozone-depleting nitrogen oxides (NO_x) in the stratosphere. Nitrous oxide also acts as an efficient greenhouse gas [Intergovernmental Panel on Climate Change (IPCC), 2001]. Its photochemical life time quickly changes with altitude and is in the order of ~ 100 years in the troposphere, ~ 1 year at ~ 33 km and ~ 1 month at 45 km [Brasseur and Solomon, 1986]. On shorter timescales its global distribution is therefore mainly determined by the Brewer-Dobson circulation, making it a useful tracer for transport processes throughout the lower and middle stratosphere, e.g., with respect to global transport studies [e.g., Randel et al., 1993, 1994] or polar vortex dynamics [e.g., Proffit et al., 1990, 1992; Plumb and Ko, 1992; Bremer et al., 2002; Urban et al., 2004b].

[3] Global observations of stratospheric nitrous oxide from space have first been conducted between 1979 and 1983 by the Stratospheric and Mesospheric Sounder (SAMS) on the Nimbus 7 satellite [e.g., Jones and Pyle, 1984], an infrared pressure-modulator radiometer employing gas correlation spectroscopy for the analysis of thermal limb emissions of N_2O around $7.8\text{ }\mu\text{m}$, and then in the early 1990s by two instruments on board the Upper Atmosphere Research Satellite (UARS). The Improved Stratospheric and Mesospheric Sounder (ISAMS), working in the 4.6 to 16.3 micron range, provided data from October 1991 through July 1992 [Taylor et al., 1993; Remedios et al., 1996]. The Cryogenic Limb Array Etalon Spectrometer (CLAES), a cryogenically cooled infrared spectrometer, measured thermal emission from the Earth's limb between 3.5 and 13 microns and performed observations from October 1991 to May 1993 [Roche et al., 1993, 1996]. Additionally, solar occultation measurements of N_2O at infrared wavelengths have been performed by the Atmospheric Trace Molecule Spectroscopy Experiment (ATMOS), flown during several short missions in 1985, 1992, 1993, and 1994 on board the Space Shuttle [Gunson et al., 1996; Irion et al., 2002], as well as by the Improved Limb Atmospheric Spectrometer (ILAS) on board the Advanced Earth Observing Satellite (ADEOS) in 1996–1997 [Kanzawa et al., 2003]. These last two instruments also provided profile measurements but with limited geographical coverage resulting from the solar occultation technique and the platform orbits.

[4] The Sub-Millimetre Radiometer (SMR) on board the Odin satellite, launched in February 2001, is the first spaceborne sensor using passive submillimeter-wave heterodyne spectroscopy for observations of the global distribution of stratospheric N_2O and regular measurements

started in November 2001, on the basis of about 10 observation days per month.

[5] This work focuses on the quality assessment of the Odin/SMR level 2 product for N_2O by comparing our data with independent correlative measurements. The Odin/SMR measurements are first described in section 2. In section 3.1 we evaluate Odin/SMR N_2O measurements against results obtained from well-established and validated in situ and remote sensors operated on stratospheric balloons and research aircraft, i.e., providing relatively accurate measurements but with rather limited coverage in space and time. In section 3.2 we focus on cross comparisons with preliminary (so far nonvalidated) results obtained from recent satellite observations, namely from the Improved Limb Atmospheric Spectrometer-II (ILAS-II), operated on board the Advanced Earth Observing Satellite-II (ADEOS-II) in 2003 [Nakajima et al., 2003], as well as from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on the Envisat satellite, launched in February 2002 [European Space Agency (ESA), 2000; Carli et al., 2004]. In section 4 we discuss the broad morphological features of the Odin/SMR N_2O data set. A comprehensive summary of the validation results is given in section 5 and conclusions are drawn in section 6.

2. Odin/SMR N₂O Measurements

[6] The Sub-Millimetre Radiometer (SMR) on board the Odin satellite, a Swedish-led project involving contributions from France, Canada and Finland, passively observes thermal emissions originating from the Earth's limb using a 1.1 m telescope. The instrument employs 4 tunable single-sideband Schottky diode heterodyne receivers operating within the 486–581 GHz spectral range as well as two high resolution autocorrelator spectrometers [Frisk et al., 2003; Olberg et al., 2003].

[7] Aeronomy mode measurements are dedicated to the investigation of stratospheric and mesospheric chemistry and dynamics [Murtagh et al., 2002]. The main target species are O_3 , ClO , N_2O , HNO_3 , H_2O , CO , and isotopes of H_2O and O_3 . Observations of the $J = 20 \rightarrow 19$ rotational transition of N_2O centered at 502.296 GHz are simultaneously performed with observations of rotational transitions of ClO (501.3 GHz), O_3 (501.5 GHz, 544.5 GHz, 544.9 GHz), and HNO_3 (around 544.4 GHz). These so-called stratospheric mode measurements are typically scheduled on one day out of three, time-shared with other aeronomy measurement modes as well as with astronomical observations. A typical stratospheric mode scan covers the altitude range from 7 to 70 km (in about 90 s) and the spectrometer readout interval corresponds to ~ 1.5 km in terms of tangent altitudes below roughly 50 km and to ~ 5.5 km above. Measurements usually cover the latitude range 82.5°S – 82.5°N , determined by Odin's Sun-synchronous near polar orbit and the nominal pointing characteristics.

[8] Profile information (level 2) is retrieved from the calibrated spectral measurements of a limb scan (level 1b) by inverting the radiative transfer equation for a nonscattering atmosphere. Optimized retrieval schemes based on the Optimal Estimation Method (OEM) [Rodgers, 1976] have been developed and implemented for level 2 processing

within the Swedish and French parts of the Odin/SMR ground segment [Baron, 1999; Baron *et al.*, 2001, 2002; Merino *et al.*, 2001, 2002; Lautié *et al.*, 2001; Lautié, 2003; Eriksson *et al.*, 2002, 2005; Urban *et al.*, 2002, 2004a, 2005].

3. Quality Assessment for N₂O

[9] We assess the quality of the Odin/SMR N₂O level 2 product for the three most recent data versions. Version 1.2 (or “Chalmers-v1.2”) is the official product of the Chalmers University of Technology, Göteborg (Sweden), based on a processing scheme which focuses on fast operational analysis of the Odin/SMR measurements. Versions 222 and 223 of the so-called Chaîne de Traitement Scientifique Odin (CTSO) are reference products developed at the Observatoire Aquitain des Sciences de l’Univers, Floirac (France), serving to evaluate the quality of the operational product and to assure consistency of the SMR data analysis (also “CTSO-v222,” “CTSO-v223”). Moreover, without obligation to systematically process the entire set of measurements, the CTSO has in the past been used within the Odin/SMR ground segment as a flexible tool for optimizing and adapting the retrieval methodology to the achieved quality and characteristics of the Odin/SMR limb measurements. This allowed in particular new improved versions of the CTSO processing chain to be implemented relatively quickly.

[10] Versions 222 and 1.2 have been used for first scientific [e.g., Urban *et al.*, 2004b; Ricaud *et al.*, 2005] and data assimilation studies [e.g., El Amraoui *et al.*, 2004], while the reference version 223 is the most recent and advanced version. The different retrieval schemes and the characteristics, errors and limitations of the resulting level 2 data products are described and illustrated in detail by Urban *et al.* [2005]. The main differences are related to the altitude resolution and altitude range of the measurements. For N₂O, version 223 provides the best altitude resolution of ~1.5 km in the lower stratosphere, mainly determined by the integration time for a spectral measurement during a limb scan, i.e., the tangent altitude step between consecutive spectrometer readouts. The corresponding single-scan precision due to measurement noise is in the order of 10–20% (15–45 ppbv). Profile information is obtained throughout the stratosphere down to roughly 14 km for limb scans at high latitudes. This lower limit is typically 2–4 km higher at middle and low latitudes due to increased water vapor absorption. Version 222 profiles are retrieved on a fixed 2 km grid in the stratosphere and are therefore somewhat less noisy than version 223 data which are calculated on the higher resolution altitude grid given by the tangent points of the limb views. The version 223 retrieval scheme is more robust since a number of additional instrumental features influencing the spectral data quality is taken into account in this version. Finally, the version 1.2 retrieval algorithm puts more weight on the a priori information, used by the OEM method for regularization of the inversion problem, leading to a slightly reduced altitude range. Also, smoothing in altitude by assuming correlations between adjacent levels results in a deterioration of the altitude resolution, while the noise is reduced accordingly. The estimated total systematic error of

the Odin/SMR N₂O measurement, derived for a midlatitude scenario, increases from values smaller than 3 ppbv above 30 km with decreasing altitude to values of 12 ppbv at 20 km and 32 ppbv at 15 km. In terms of relative units, the systematic error is lower than 5% between 20 and 40 km and of the order of 5–15% below 20 km (see Urban *et al.* [2005] for details).

[11] Correlative measurements of N₂O are available from balloonborne, aircraftborne, and spaceborne sensors. In order to assess systematic effects and biases smaller than the SMR measurement precision, the somewhat noisy Odin/SMR profiles, retrieved from individual scans, will in the following be averaged within appropriate intervals determined by the times and positions of the airborne validation measurements. Typically, averaging will be done in intervals of ± 24 hours, $\pm 7.5^\circ$ in latitude and $\pm 15^\circ$ in longitude for situations of low atmospheric variability. This corresponds to about 3 to 10 averaged profiles, depending mainly on the latitude range of the comparison. In the case of the intercomparison with satellite observations, differences between zonally averaged data are evaluated. In general, only SMR level 2 data with negligible a priori contribution, or in other words with measurement response close to unity, are used for the comparisons.

3.1. Airborne Sensors

3.1.1. Balloon Measurements

[12] Correlative measurements of balloonborne sensors are available from the Fourier-transform infrared spectrometer of the Laboratoire de Physique Moléculaire et Applications (LPMA), Paris (France), from the SPIRALE infrared tunable diode laser instrument of the Laboratoire de Physique et Chimie de l’Environnement (LPCE), Université d’Orléans (France), and from the DIRAC gas chromatograph of the University of Cambridge (UK).

[13] A balloon carrying the LPMA Fourier-transform infrared spectrometer [Camy-Peyret *et al.*, 1995] was launched on 21 August 2001 from Esrange in northern Sweden (67.5°N/21.1°E). Solar occultation measurements were performed during ascent, sunset, and sunrise. Owing to the limb observation geometry, the LPMA instrument averages over a similar horizontal absorption path as Odin/SMR. The vertical resolution is about 2 km and the overall accuracy of the N₂O measurement is of the order of about 10% [Payan *et al.*, 1999]. On this particular flight, profile information was obtained between 11 and 38 km. Results are shown in Figure 1 and are compared to correspondingly averaged Odin/SMR measurements. Individual balloon and SMR measurements indicate stable atmospheric high-latitude summer conditions, even though some variability was observed by the balloon below 20 km. Differences between the averaged SMR profile and the balloon sunset observation are lower than 25 ppbv over the whole measurement range of the LPMA observation, largest values are found at 16 km and above 35 km. Note that the differences shown in Figure 1 and in all following figures are always calculated as “validation experiment” minus “SMR” data.

[14] A midlatitude flight of the balloonborne SPIRALE instrument (Spectroscopie Infrarouge par Absorption de Lasers Embarqués) was performed on 2 October 2002 starting in Aire-sur-l’Adour, France (~43°N/0°E). SPIRALE

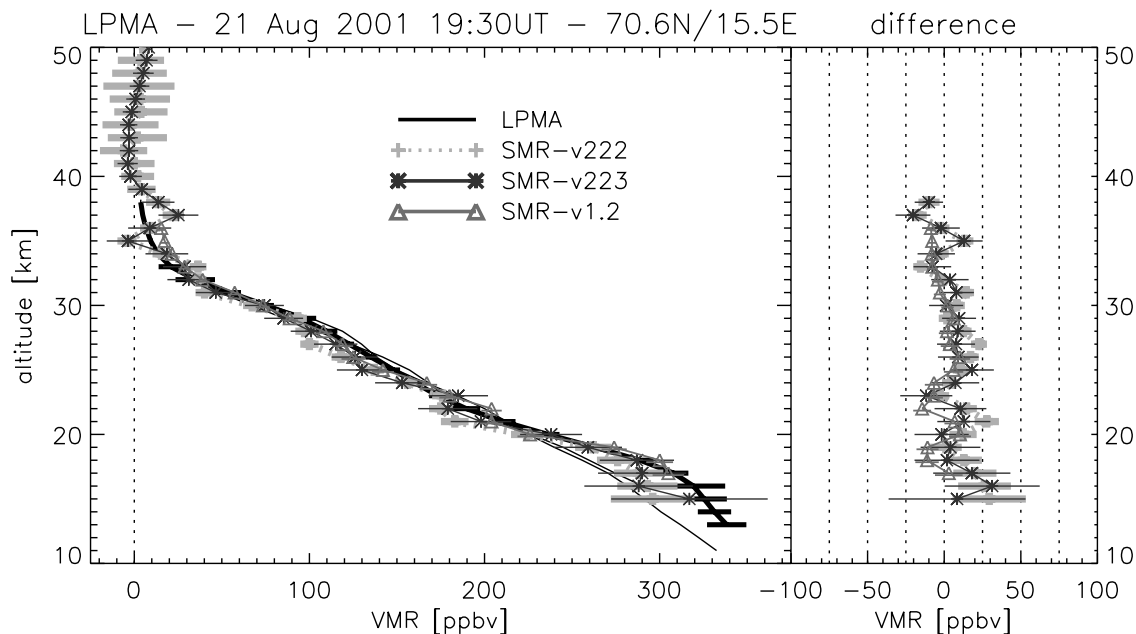


Figure 1. Comparison of N₂O from Odin/SMR with measurements of the balloon-borne LPMA Fourier-transform infrared spectrometer from 21 August 2001 over northern Sweden. Individual profile measurements of SMR for three different level 2 data versions were averaged within ± 1 day, $\pm 7.5^\circ$ in latitude and $\pm 15^\circ$ in longitude with respect to the average time and position of the validation experiment. The plot shows the comparison of the mean SMR profiles of the different versions, based on four limb scans, with the validation experiment. The thick black line with error bars is the LPMA profile measured during sunset on 21 August 2001 at 1930 UT, while the thin black lines indicate measurements during ascent of the balloon (21 August 2001, 1710 UT) and during sunrise (22 August 2001, 0143 UT). See legend for the line styles of the different SMR data versions shown (v222, v223, and v1.2). Differences of the SMR data with respect to the sunset profile are plotted on the righthand side, the error bars in the difference plot are the SMR errors (measurement precision).

measures N₂O using an in situ tunable diode laser technique which permits to obtain a precision of 3% and an altitude resolution of 5 m. Information on this instrument can be obtained from Moreau [1997, 2001]; Moreau *et al.* [2003]. The data used here are averaged to 250 m thick altitude bins and cover the altitude range ~ 12 –33 km. The measurements are shown in Figure 2 along with N₂O data from Odin/SMR. As indicated by the CH₄–N₂O correlation measured by SPIRALE (not shown), air masses measured at low altitudes had a different origin than the air measured at high altitudes. Accordingly, we find a reasonable agreement with Odin/SMR below ~ 23 km when averaging measurements at higher latitudes (northeastward of the average position of the validation measurement), and above ~ 29 km when averaging measurements taken at lower latitudes (toward the southwest). Agreement is within 25 ppbv from 17 to 23 km and from 29 to 33 km. Version 1.2 shows at the lower altitudes (17–20 km) a slightly larger positive bias compared to the reference versions 222 and 223.

[15] Figure 3 shows the comparison of Odin/SMR measurements of N₂O with a measurement taken by SPIRALE on 21 January 2003 at the edge of the Arctic polar vortex. According to Pirre *et al.* [2004], measurements of SPIRALE above 21 km (40 hPa) were taken inside the Arctic polar vortex, while measurements below sampled extravortex air. This is consistent with the Odin/SMR observations of the N₂O field in the Northern Hemisphere,

indicating a horizontally inhomogeneous, relatively small vortex below 20 km which extended further to the South at higher altitudes, covering the latitude range of the balloon trajectory over Northern Scandinavia at 67 – 68° N above roughly 21 km. The comparison of N₂O from SPIRALE with Odin/SMR measurements inside the vortex, averaged applying the usual selection criteria, yields agreement within ± 25 ppbv above 24 km. Below, deviations up to 50 ppbv are found for versions 222 and 223 in this inhomogeneous atmospheric situation close to the vortex edge and version 1.2 tends to even slightly higher positive deviations.

[16] Odin/SMR measurements of N₂O are compared to results of the balloonborne DIRAC (Determination In-situ by Rapid Chromatography) sensor from a midlatitude flight launched on 4 October 2002 from Aire-sur-l'Adour, France (Figure 4). DIRAC is a gas chromatograph providing an overall measurement uncertainty of $\sim 4\%$ for samples with atmospheric pressures greater than 50 hPa and of $\sim 15\%$ for samples with pressures lower than 50 hPa. Required sampling times lead to an altitude resolution of typically a few hundred meters, depending on ascent and descent velocities of the balloon. Data are here averaged to about 1–2 km thick layers. For a description of the instrument the reader is referred to Robinson *et al.* [2000]. As for the SPIRALE flight launched two days earlier from the same site, the atmospheric situation is rather inhomogeneous. An analysis including back trajectory and potential vorticity data

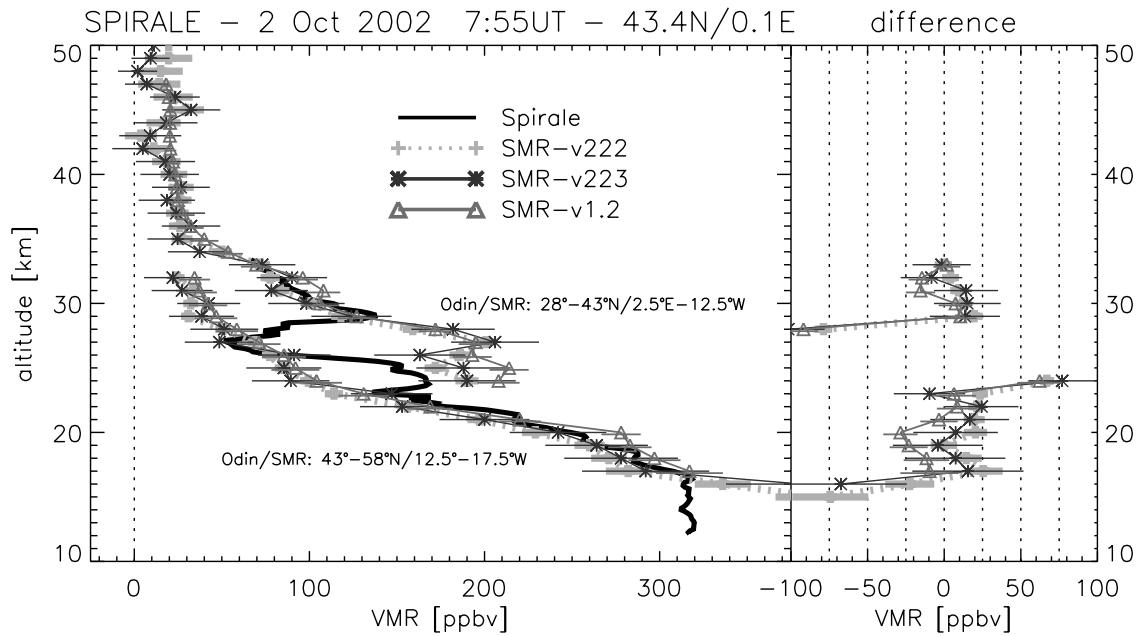


Figure 2. Comparison of Odin/SMR N₂O with measurements at northern midlatitudes of the balloonborne SPIRALE instrument, launched from Aire-sur-l'Adour (France) on 2 October 2002. The balloonborne instrument apparently observed air masses of different origin (above ~ 29 km and below ~ 23 km) during its flight, which can partly be reproduced by SMR measurements when averaged over different geographical regions, as indicated in the plot. The differences shown on the right-hand side are calculated accordingly from the corresponding mean profiles. The latter were calculated from three individual profiles in both cases.

revealed that two distinct air masses were encountered by the balloonborne sensor over the 23–29 km region. Air masses measured at lower altitudes had a high latitude origin (high PV, low N₂O), while air masses at higher

altitudes originated from low latitudes (low PV, high N₂O). Nevertheless, below 25 km an agreement within 25 ppbv is found with correspondingly averaged Odin/SMR measurements of version 222. The differences with versions

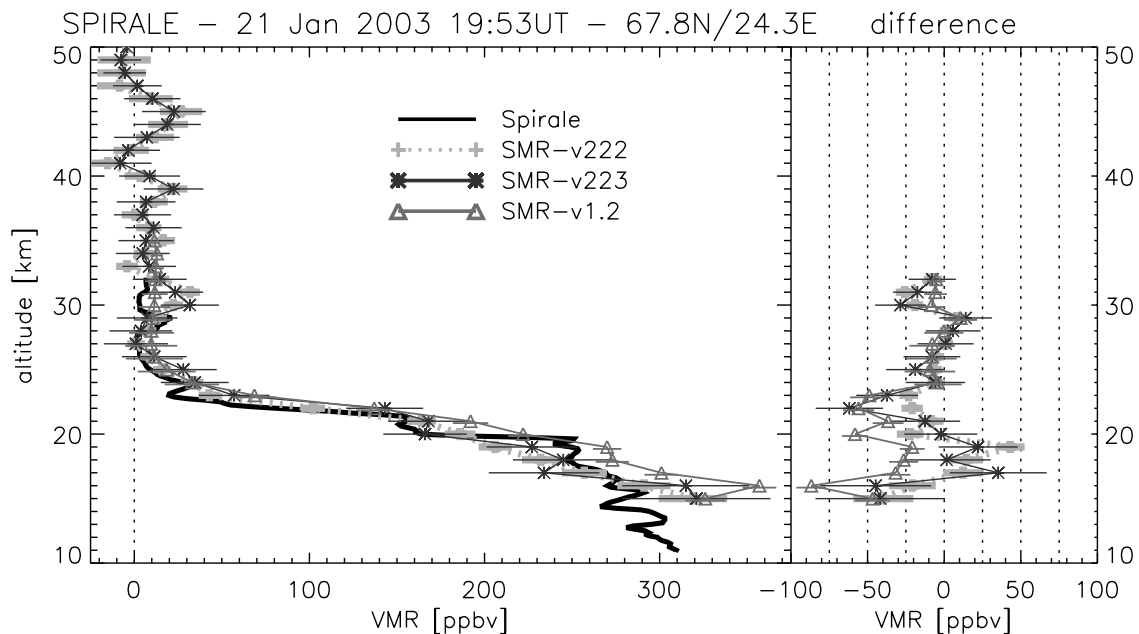


Figure 3. Comparison of Odin/SMR N₂O with measurements of the balloonborne SPIRALE instrument taken at the edge of the Arctic polar vortex on 21 January 2003 over northern Scandinavia. The average SMR profiles used for the comparison with SPIRALE (averaged within ± 1 day, $\pm 7.5^\circ$ in latitude, $\pm 15^\circ$ in longitude), are based on three limb scans sounding air masses inside the polar vortex.

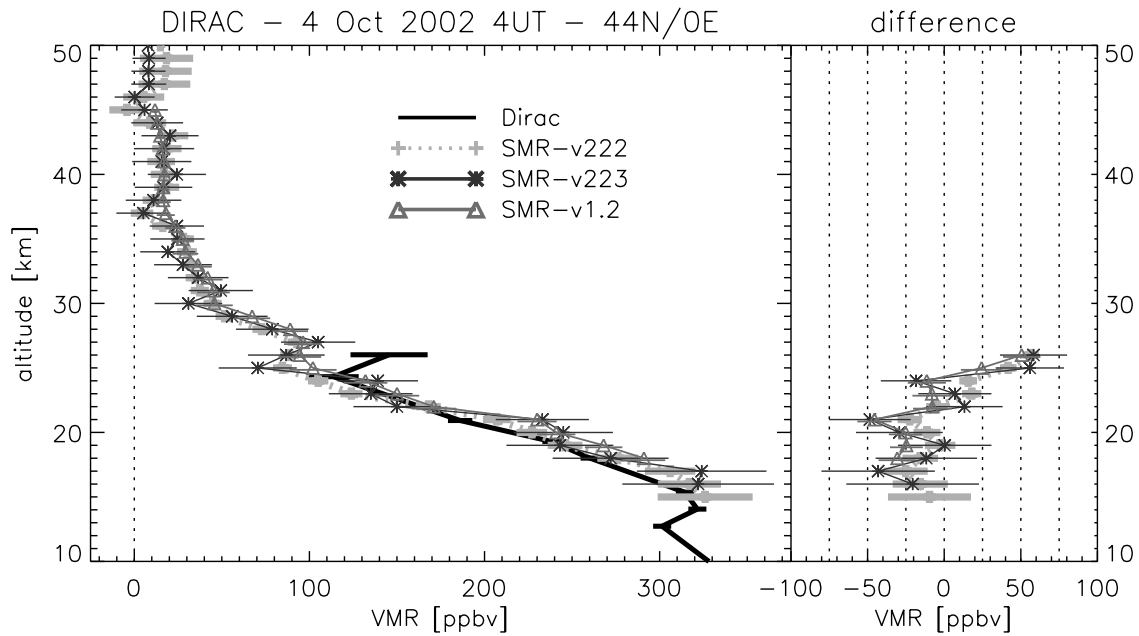


Figure 4. Comparison of Odin/SMR N₂O with measurements of the balloonborne DIRAC instrument, launched from Aire-sur-l'Adour (France) on 4 October 2002. SMR profiles, measured within ± 1 day, $\pm 7.5^\circ$ in latitude and $\pm 15^\circ$ in longitude of the DIRAC measurement time and position, were averaged (three profiles). The balloonborne sensor measures apparently air masses of different origin below and above 25 km, similar to the measurements performed by SPIRALE two days before (see Figure 2). Odin/SMR measurements match the balloonborne results below 25 km.

223 and 1.2 are slightly larger, but the agreement is still within 50 ppbv.

[17] Finally, Figure 5 compares Odin/SMR measurements taken at low latitudes (15° – 30° S) with N₂O measurements of DIRAC from a flight launched on 18 February 2003 in Bauru, Brasil (22.4° S/ 49.0° E). Despite of the fact that the

atmospheric variability seems to be fairly low according to the individual SMR measurements (not shown), the comparison with DIRAC yields large differences up to 75 ppbv at 24 km. Between 17 and 22 km both data sets are within 50 ppbv, but above the DIRAC measurement indicates a strong atmospheric layering precluding the use of these data

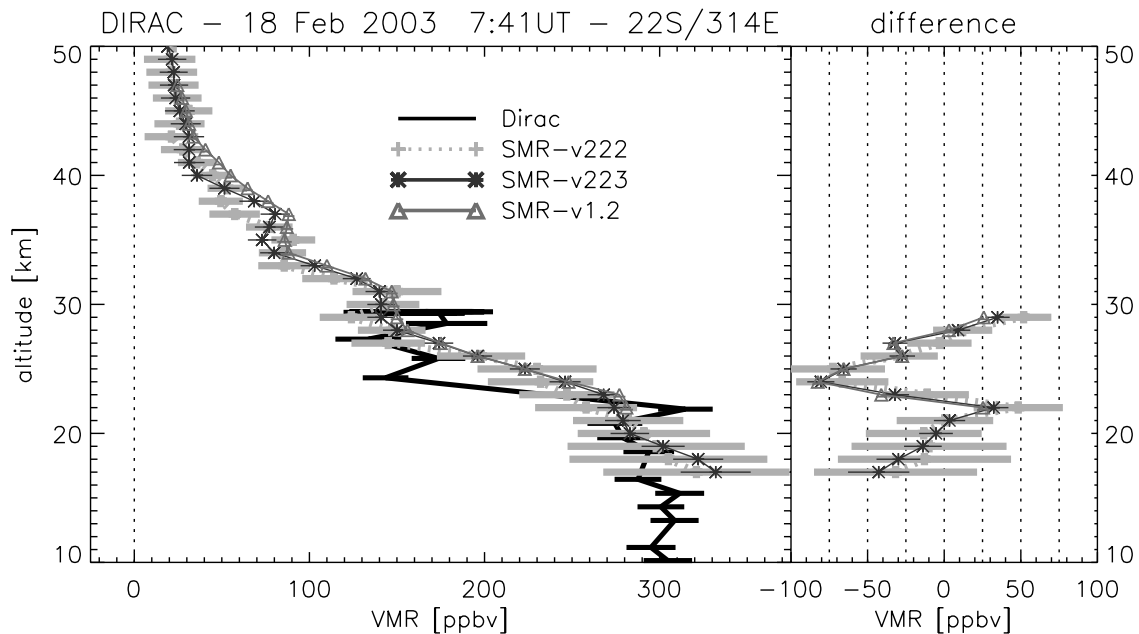


Figure 5. Comparison of Odin/SMR N₂O with DIRAC measurements at low latitudes. The balloon was launched in Bauru (Brasil) on 18 February 2003. SMR data are averaged within ± 1 day, $\pm 7.5^\circ$ in latitude and $\pm 15^\circ$ in longitude with respect to the validation measurement (four profiles).

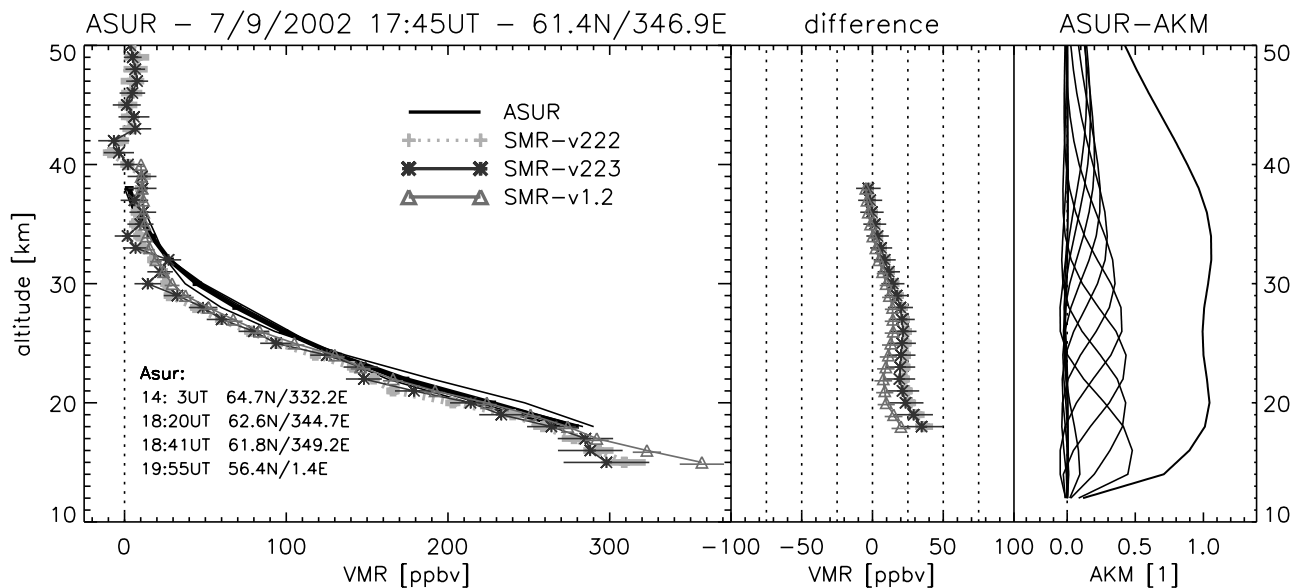


Figure 6. Comparison of Odin/SMR N₂O with measurements of the airborne ASUR radiometer taken during a flight at northern high latitudes on 7 September 2002. The thick black line is the average ASUR profile, thin black lines are the individual ASUR retrievals indicating the low variability of the ASUR measurements, i.e., clearly showing here the stability of the atmosphere. SMR profiles within ± 1 day, $\pm 7.5^\circ$ in latitude and $\pm 15^\circ$ in longitude are averaged for this comparison (10 profiles). To calculate the difference (middle), the Odin/SMR profiles have been smoothed using the averaging kernel functions of the ASUR measurements (right).

for validation purposes. A considerable variability in potential vorticity was found in the Odin/SMR sampling region around Bauru, which could explain why much of the structure locally observed by the in situ sensor is not captured by the spatially averaged remote measurement.

3.1.2. Research Aircraft

[18] We also compare Odin/SMR retrievals with N₂O measurements of the Airborne Submillimetre SIS Radiometer (ASUR), an aircraftborne single-sideband heterodyne receiver developed by the University of Bremen (Germany) and the Space Research Organization of the Netherlands (SRON). ASUR passively observes a thermal emission line of N₂O at 652.8 GHz using the up-looking observation geometry (elevation angle $\sim 12^\circ$). The typical integration time for an individual N₂O measurement is of the order of 150 s, leading to a horizontal resolution of about 30 km along the flight path of the research aircraft. The corresponding precision of a measurement is about 10 ppbv, while the accuracy (also including systematic uncertainties) is estimated to be 30 ppbv or 15%, whichever is higher. Information on N₂O can be retrieved between 15 and 45 km and the altitude resolution degrades with height from 8 km in the lower stratosphere up to 16 km in the upper stratosphere. The horizontal resolution in the direction of the line-of-sight, i.e., perpendicular to the flight direction, depends geometrically on the altitude resolution and varies from 40 to 80 km. Detailed information about the ASUR instrument can be obtained from Mees *et al.* [1995]; de Valk *et al.* [1997]; Urban [1998]; Urban *et al.* [1999]. The measurements and data analysis of N₂O are for example described by Bremer *et al.* [2002].

[19] A first comparison was conducted for a measurement flight on 7 September 2002 at northern high latitudes.

Results are shown in Figure 6. For a stable late summer situation, an agreement within 25 ppbv is found for all SMR level 2 data versions, with smallest deviations found for version 1.2. For calculating the differences, the average SMR profile was first convolved with the averaging kernel functions of the ASUR retrieval in order to account for the limited altitude resolution of the up-looking sensor. Odin/SMR mixing ratios are systematically slightly lower than ASUR data up to about 35 km.

[20] On 25 September 2002, an ASUR measurement flight parallel to the equator was performed starting from Nairobi, Kenya. Results are shown in Figure 7. Here we find considerable disagreement with respect to the profile shape of the Odin/SMR and ASUR measurements. The shape of the tropical ASUR profile must not necessarily be very realistic, given the limited altitude resolution of ASUR compared to Odin/SMR. However, when the convolved Odin/SMR profile is evaluated, the differences are still slightly larger than 25 ppbv.

[21] Neither the fairly stable atmospheric situations nor the limited altitude resolution of the up-looking ASUR sensor can be made responsible for the systematic disagreement which was found in both cases. This points toward systematic errors (calibration, spectroscopy) in at least one of the compared data sets.

3.1.3. Assessment

[22] In order to evaluate the SMR data quality against the airborne validation experiments, we combine all data in the form of a scatterplot. Results are shown in Figure 8. For a more quantitative estimation of the systematic effects in the data, the root mean square (RMS) deviation is calculated for each SMR level 2 data version and 3 ranges of the validation experiments N₂O mixing ratio: 0–75 ppbv,

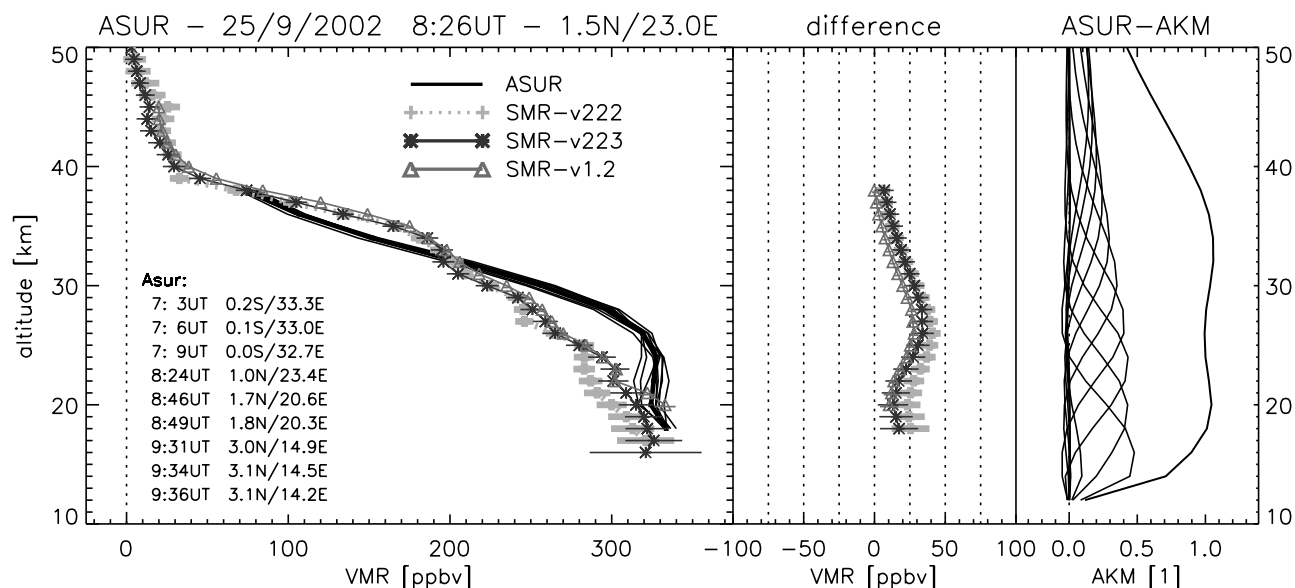


Figure 7. Same as Figure 6, but for measurements of the air-borne ASUR radiometer from a flight close to the equator performed on 25 September 2002. Ten individual SMR profiles within the usual coincidence limits were averaged for this comparison. To calculate the difference (middle), the unsmoothed Odin/SMR profiles (left) have been convoluted with the averaging kernel functions of the ASUR measurements (right).

75–225 ppbv, and 225–325 ppbv. For this estimation we exclude those data from SPIRALE and DIRAC where large differences were identified as arising from inhomogeneous atmospheric situations. In order to account for the limited altitude resolution of the ASUR measurements, only the average RMS differences with Odin/SMR within two distinct layers (18–25 km and 25–35 km) were considered. For low mixing ratios of N₂O (range 0–75 ppbv), i.e., at high altitudes, we find a RMS deviation smaller than 11, 12, and 8 ppbv for versions 222, 223, and 1.2, respectively. SMR shows here larger mixing ratios than the validation measurements. In the intermediate range (75–225 ppbv), SMR measurements result in slightly lower mixing ratios than the validation experiments and the RMS deviation is of the order of 13–19 ppbv. In the highest N₂O mixing ratio range (225–325 ppbv), i.e., at lowest altitudes, SMR still exhibits slightly lower values and we find a RMS deviation of 26–28 ppbv. The results are similar for all SMR data versions. Smallest RMS deviations are found for version 1.2, which is partly due to the lower noise level of version 1.2 data. Moreover, version 1.2 shows a slightly smaller bias compared to the reference versions 222 and 223. In summary, SMR data agree with the validation experiments roughly within 28 ppbv (RMS) in the whole exploitable altitude range.

3.2. Satellite Sensors

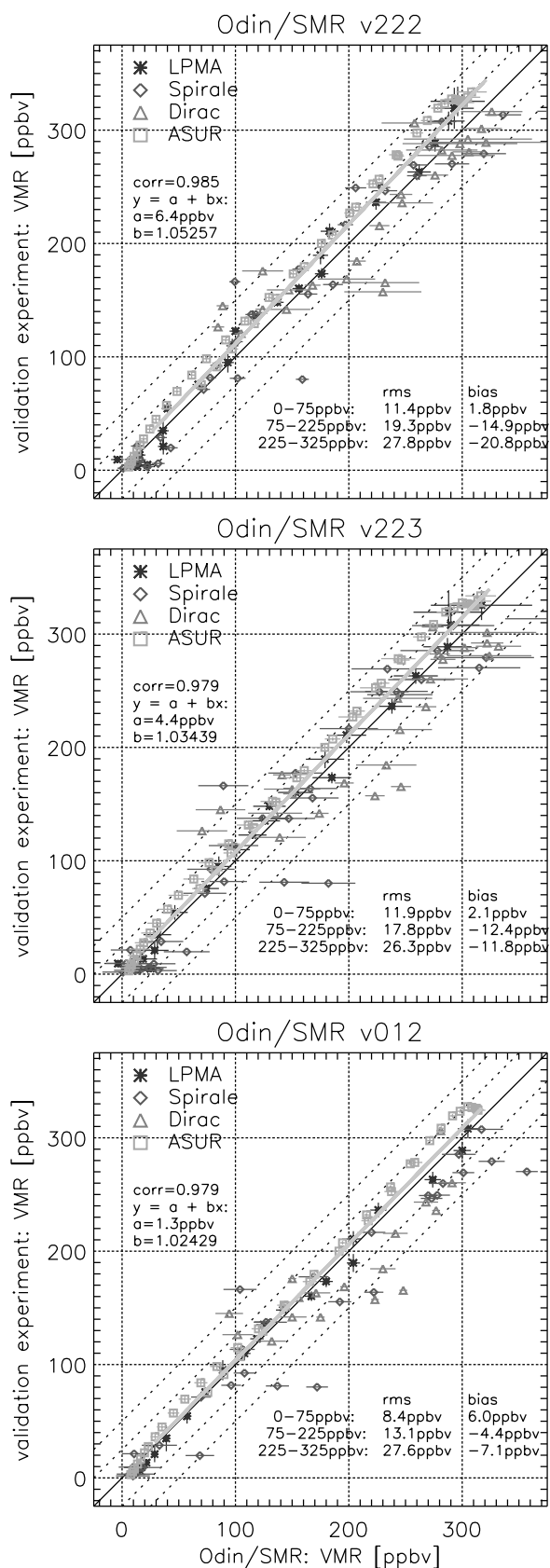
3.2.1. Comparison With ILAS-II

[23] The ILAS-II instrument on board the ADEOS-II satellite performed measurements from January to October 2003 [Nakajima *et al.*, 2003]. The instrument consists of 4 grating spectrometers with array detectors. ILAS-II measures the N₂O infrared absorption around 7.8 microns by means of the solar occultation technique. Owing to the Sun-synchronous polar orbit with an equator crossing time

at 1030 UT for the descending node, sunset and sunrise occur at middle and high latitudes in both hemispheres in very narrow latitude bands. There are about 14 observation points per day and hemisphere and the latitude of observation gradually shifts with the seasons within the ranges 54°–71°N and 64°–88°S. For the cross comparison with Odin/SMR, we exclusively use the most recent version 1.4 profiles. The ILAS-II retrieval method is similar to that of ILAS as described by Yokota *et al.* [2002], i.e., it is based on a spectral fit using a nonlinear least squares method in combination with an onion peeling approach for vertical profile reconstitution. Profiles are determined between ~10–50 km with a vertical resolution deteriorating with altitude: 1.3 km at 15 km, 1.6 km at 20 km, 1.9 km at 25 km, 2.2 km at 30 km, 2.7 km at 40 km, and 2.9 km at 50 km. A detailed error analysis and a validation paper for N₂O are currently under preparation (M. Ejiri *et al.*, manuscript in preparation, 2004).

[24] For the comparison with Odin/SMR, we choose 3 stratospheric mode observation days during the ILAS-II measurement period: 20–21 March, 18–19 June, and 7–8 September 2003 (1200–1200 UT). Zonal averages are calculated from the Odin/SMR measurements taken in a thin latitude band of $\pm 2.5^\circ$ around the ILAS-II observation latitude. Since Odin/SMR and ILAS-II measure at different times and geographical positions, we chose to compare zonal averages based on about 15 orbits per day rather than averages of the few coinciding individual profile measurements in order to improve the statistics of the comparison.

[25] Figure 9 shows the results for the Northern Hemisphere measurements on 18–19 June 2003. Individual measurements of both instruments indicate relatively stable atmospheric conditions at 54.4°N. All Odin/SMR data versions result in systematically higher N₂O mixing ratios below ~43 km with maximum deviations of up to 25 ppbv



for version 222 and 223 at 15–20 km. An even higher positive bias of up to ~50 ppbv is found for version 1.2 in this altitude range.

[26] The corresponding comparison for the Southern Hemisphere is shown in Figure 10. ILAS-II observations are conducted at 65.1°S. Owing to the presence of the Antarctic polar vortex, the atmospheric situation is rather inhomogeneous and measurements taken inside and outside the vortex have to be distinguished. We define the vortex edge using the observations of nitrous oxide: limb scans resulting in retrieved N₂O volume mixing ratios smaller than 70 ppbv at the potential temperature surface of 550 K (and at higher levels) are considered to be vortex measurements, while mixing ratios larger than 125 ppbv at 550 K (and at lower levels) indicate measurements outside the vortex [e.g., Urban *et al.*, 2004b]. Inside the vortex we find the Odin/SMR versions 222 and 223 to be systematically higher by up to ~25 ppbv between roughly 15–28 km. Version 1.2 shows a slightly larger positive bias of up to 50 ppbv below 20 km. Above 28 km all data sets agree within 5 ppbv up to the stratopause. Outside the vortex, we find similar results: a 25–30 ppbv offset of the SMR data between roughly 18 and 28 km. Unreasonably low values are found in version 1.2 retrievals below 20 km for this particular case.

[27] The same kind of picture is found in the Northern Hemisphere on 20–21 March, where the distinction was made with respect to the Arctic polar vortex (not shown). ILAS-II measurements are performed at 66.1°N. SMR retrievals of versions 222 and 223 are systematically higher by ~25 ppbv, with maximum deviations of up to 75 ppbv below 20 km. Version 1.2 retrievals tend even to slightly higher differences. At southern high latitudes (85.2°S), the atmospheric variability is low and SMR yields larger N₂O mixing ratios by 25–50 ppbv. Note that the highest latitude reached by the SMR measurements is 83°S and the SMR zonal mean profile corresponds in this particular case to the latitude range 80°–83°S, what might partly explain the systematic offset.

[28] Finally, the comparison for 7–8 September 2003 (not shown) confirms that SMR retrievals yield systematically higher mixing ratios than ILAS-II and that SMR version 1.2 has a trend toward unrealistically high values below 20 km. The atmospheric variability is relatively low for both hemispheres. All Southern Hemisphere measurements of ILAS-II on this day are taken inside the Antarctic vortex at

Figure 8. Scatterplot of Odin/SMR N₂O versus balloon-borne and aircraftborne validation experiments. (top) Odin/SMR v222 data. (middle) Odin/SMR v223 data. (bottom) Odin/SMR v1.2 data. Root mean square (RMS) deviations as well as biases are indicated for different ranges of the N₂O mixing ratio. Data with identified large discrepancies due to the atmospheric inhomogeneousness, such as from SPIRALE on 2 October 2002, and DIRAC on 4 October 2002 and 18 February 2003 are excluded from the RMS and bias calculation. The dotted lines delimit deviations of ±25 and ±50 ppbv for clarity. The solid gray lines indicate linear fits to the data in the interval 25 to 325 ppbv.

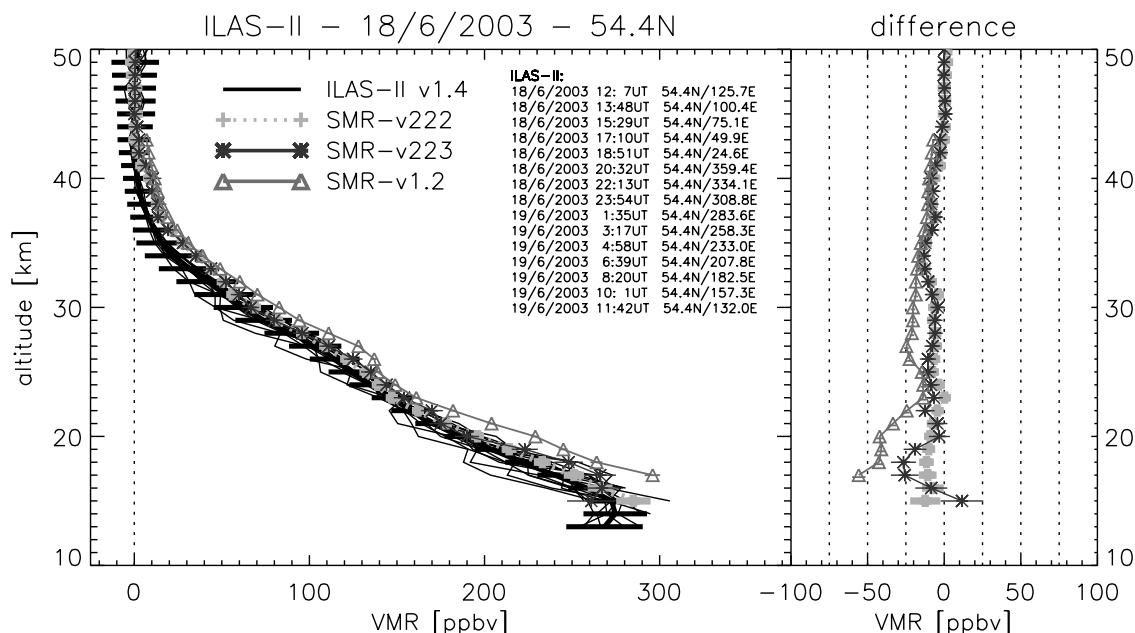


Figure 9. Comparison of Odin/SMR N₂O with spaceborne ILAS-II infrared solar occultation measurements taken at northern middle latitudes on 18–19 June 2003 (1200–1200 UT). Odin/SMR measurements were averaged within a latitude band of $54.4 \pm 2.5^\circ\text{N}$. The plot shows the comparison of the zonal mean SMR profiles with the zonal mean ILAS-II profile (thick black line with 1σ error bars). The thin black lines are the individual ILAS-II measurements.

a latitude of 84.1°S , while the Northern Hemisphere late summer measurements are taken at 66.3°N .

[29] We combine the results obtained from the comparisons with ILAS-II data in Figure 11. As pointed out before, SMR measurements result in higher mixing ratios compared to ILAS-II retrievals. Root mean square deviations are again calculated for the three ranges of the N₂O mixing ratio. At low N₂O mixing ratios (range 0–75 ppbv), we find differences of 7–8 ppbv for SMR versions 222 and 223, while version 1.2 shows a considerably larger RMS deviation of 14 ppbv. For the intermediate range (75–225 ppbv N₂O), RMS differences of 20, 23, and 47 ppbv are found for versions 222, 223, and 1.2, respectively. At high N₂O mixing ratios (225–325 ppbv), the corresponding differences are 19, 23, and 66 ppbv and version 1.2 data show a somewhat larger variability compared to the reference data versions. In summary, the comparison with ILAS-II yields globally an agreement within 23 ppbv for versions 222 and 223, while version 1.2 is characterized by a systematically larger RMS deviation of roughly a factor of two. The $\ln(\text{VMR})$ retrieval scheme of version 1.2, constraining profiles to positive mixing ratios, might possibly be at the origin of the larger positive bias of the version 1.2 zonal averages compared to the reference versions 222 and 223. However, one should also note that the comparison with airborne validation experiments (Figure 8) revealed only a relatively small systematic difference between the SMR data versions, amongst a set of correlative data characterized by a somewhat larger variability which might hide part of the effect. Different effects such as uncertainties of spectroscopic or instrumental parameters used in the forward models as well as systematic errors of calibration and altitude registration might contribute to the systematic

deviations of the N₂O mixing ratios measured by Odin/SMR and ILAS-II. These effects are under investigation and shall be discussed elsewhere (e.g., Urban *et al.* [2005] for Odin/SMR), while in this work we restrict ourselves to the quantitative evaluation of the differences between the N₂O level 2 products.

3.2.2. Comparison With Envisat/MIPAS

[30] The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard the Envisat satellite measures high-resolution emission spectra at the Earth's limb in the near to midinfrared wavelengths range, allowing continuous global observations of nitrous oxide during day and night [ESA, 2000]. Profiles are retrieved from the spectral measurements of a limb scan using an algorithm based on the global fit technique [Ridolfi *et al.*, 2000; Raspollini *et al.*, 2003]. We compare Odin/SMR measurements with level 2 (off-line) data of the European Space Agency (ESA). The ESA off-line processor is an optimized version of the operational near-real time processor described by Carli *et al.* [2004]. Compared to the near-real time processor, it uses consolidated level 1b data based on more accurate orbital state parameters and addresses a number of additional retrieval issues, for example with respect to the vertical range of the profile retrieval (down to 6 km instead of 12 km) and the iteration and convergence scheme. It also includes a systematic treatment of clouds in the line-of-sight. For N₂O, the altitude resolution of the retrieved profile is of the order of 3 km and the total retrieval error is estimated to be in the range 10–20% in the stratosphere below 40 km.

[31] The thermal emission measurements of Envisat/MIPAS and Odin/SMR have a similar global coverage allowing the global distribution of nitrous oxide to be

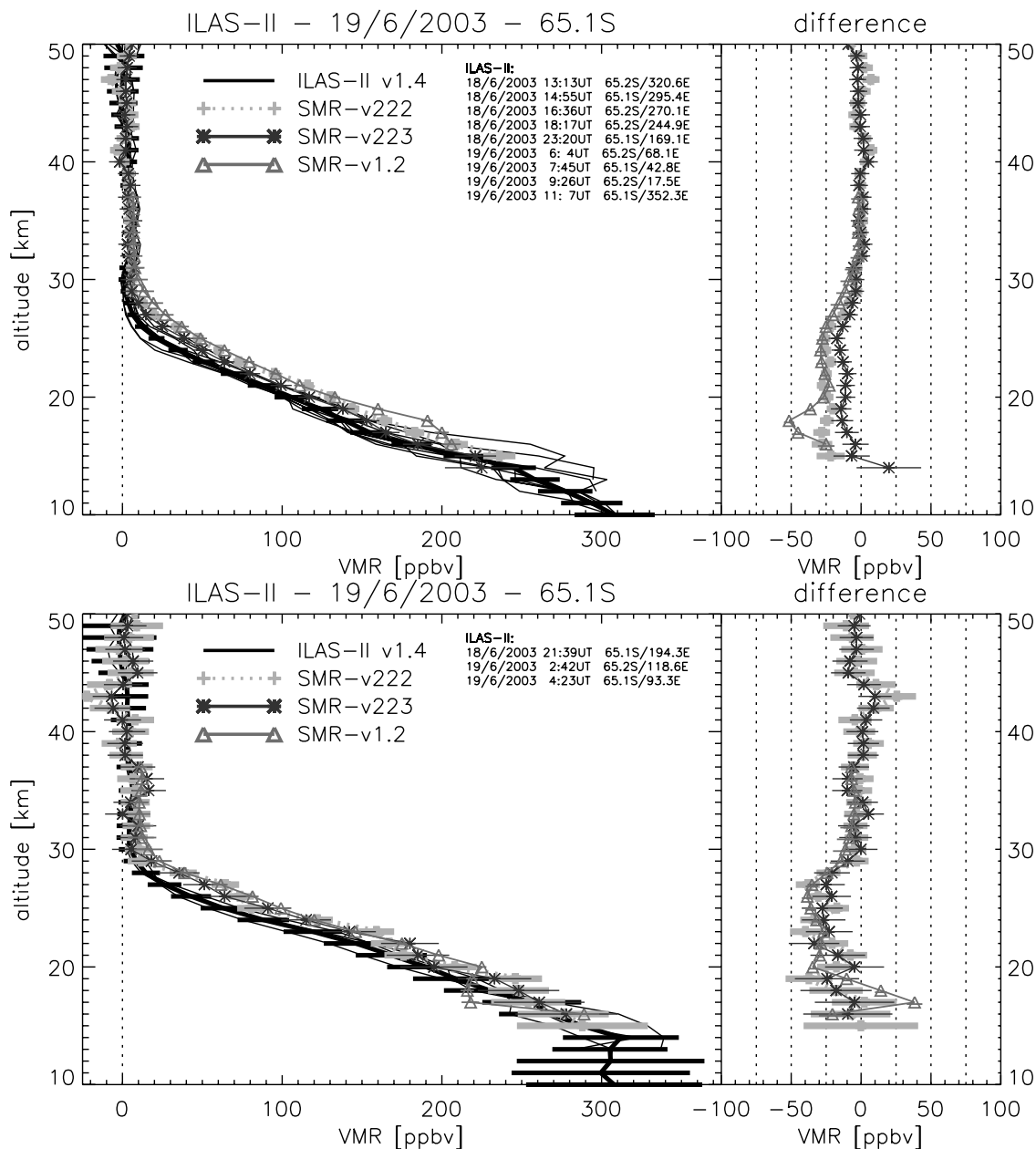


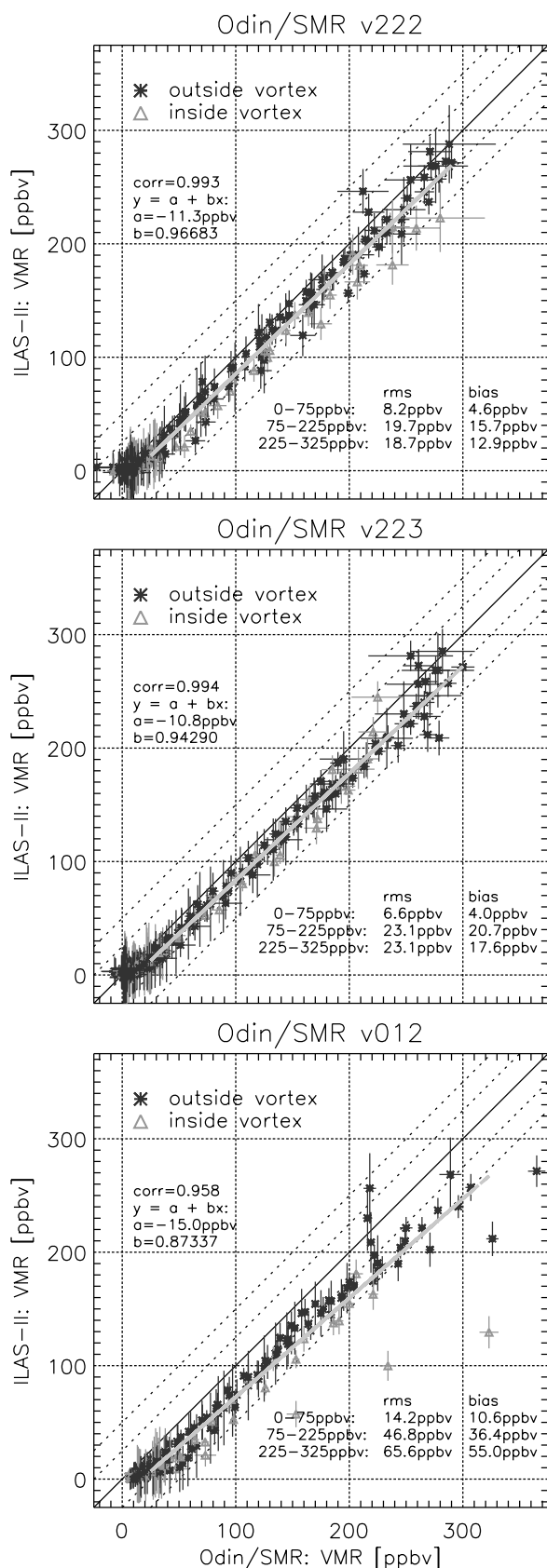
Figure 10. Same as Figure 9, but for Odin/SMR and ILAS-II N₂O observations from 18–19 June 2003 (1200–1200 UT) at southern high latitudes in a latitude band around $65.1 \pm 2.5^\circ\text{S}$. (top) Comparison of measurements taken inside the Antarctic polar vortex. (bottom) Comparison of outside vortex averages.

compared. Figure 12 shows global N₂O fields interpolated on the 50 hPa pressure level as observed by Odin/SMR and Envisat/MIPAS on 25–26 September 2002 and 20–21 March 2003 (from 1200 to 1200 UT). Again, 3 versions of SMR data are shown along with the off-line processor data from MIPAS.

[32] On 25–26 September 2002, just after a major stratospheric warming occurred in the Southern Hemisphere winter, the Antarctic polar vortex had a very elongated shape at this level which is well captured in the observations of both instruments. Small differences may be attributed to the different times and positions of the individual measurements as well as to the smaller number of profiles available for MIPAS on this particular day. Globally, SMR versions 222

and 223 are in reasonable qualitative agreement with MIPAS. However, differences are found for Odin/SMR version 1.2 in the tropics, where unrealistically high N₂O values are retrieved from a number of limb scans. We find the same qualitative agreement for 20–21 March 2003. The Northern Hemisphere polar vortex is well captured by all data versions. In the tropics, we again find very high mixing ratios in some Odin/SMR version 1.2 profiles.

[33] For a more quantitative comparison, we calculate zonal averages for the latitude bands $80^\circ\text{--}60^\circ\text{S}$, $10^\circ\text{S--}10^\circ\text{N}$, $60^\circ\text{--}80^\circ\text{N}$ as well as for 10° wide bands at middle latitudes. In the high latitude winter cases, we only use measurements taken inside the polar vortex. For the midlatitude bands, only measurements taken outside the vortex are



considered. The results are shown for selected bands in Figure 13. If one excludes the cases where the atmospheric variability due to the presence of the elongated Antarctic vortex is large (at high and middle southern latitudes on 25–26 September 2002), agreement is roughly within ± 25 ppbv down to about 100 hPa for SMR data versions 222 and 223. Version 1.2 shows a tendency toward slightly higher positive biases than version 222 and 223. At higher pressures, systematic differences become larger which might be caused by the sensitivity of the MIPAS measurements to clouds and aerosols but also by other systematic uncertainties of the MIPAS and SMR measurements. Note that the preliminary MIPAS data used for this comparison are not accompanied by a quality flag, i.e., bad and good data cannot be properly distinguished. Consequently, the averages shown here might be biased by a few incorrect profiles.

[34] Finally, the differences between SMR and MIPAS are estimated quantitatively. Figure 14 combines the results obtained from the zonal mean comparisons for 25–26 September 2002 and 20–21 March 2003. Obviously bad data points are shown in the figure, but are excluded from the RMS calculation when the reason for the disagreement was identified, e.g., if arising from a highly variable atmospheric situation. For low mixing ratios of N₂O (range 0–75 ppbv), we find small RMS differences within 6 ppbv for SMR versions 222 and 223, respectively, with SMR measurements on the high side. Version 1.2 shows a considerably larger RMS deviation of 15 ppbv. In the intermediate range (75–225 ppbv), the RMS deviations are ~ 10 ppbv for versions 222 and 223 and 23 ppbv for version 1.2. Largest RMS differences are found for measurements of high N₂O mixing ratios (range 225–325 ppbv): 17, 13, and 17 ppbv for versions 222, 223, and 1.2, respectively. Versions 222 and 223 mixing ratios are slightly smaller than MIPAS data and version 1.2 data are slightly larger. To conclude, Odin/SMR retrievals are in relatively good agreement with N₂O mixing ratios obtained from the MIPAS level 2 off-line processor. Version 1.2 shows a small positive bias, but root mean square deviations are roughly within the estimated systematic errors of the MIPAS and SMR observations. Please also note that the comparison with MIPAS near-real time data resulted in general in considerably larger RMS deviations.

4. Morphology of Global N₂O Data Set

[35] The global Odin/SMR N₂O data set shows the same broad morphological features previously observed by the SAMS instrument on Nimbus-7 [e.g., Jones and Pyle, 1984] and by CLAES and ISAMS on UARS [e.g., Roche *et al.*, 1996; Remedios *et al.*, 1996], but shows considerable improvements with respect to global and temporal coverage.

Figure 11. Scatterplot of zonal mean N₂O from Odin/SMR versus zonal mean N₂O from ILAS-II. (top) Odin/SMR v222 data. (middle) Odin/SMR v223 data. (bottom) Odin/SMR v1.2 data. Data taken inside or outside the polar vortices are shown with different symbols. Root mean square (RMS) deviations as well as biases are indicated for three different ranges of the N₂O mixing ratio. The dotted lines represent deviations of ± 25 and ± 50 ppbv, and the solid gray lines are linear fits within the interval 25 to 325 ppbv.

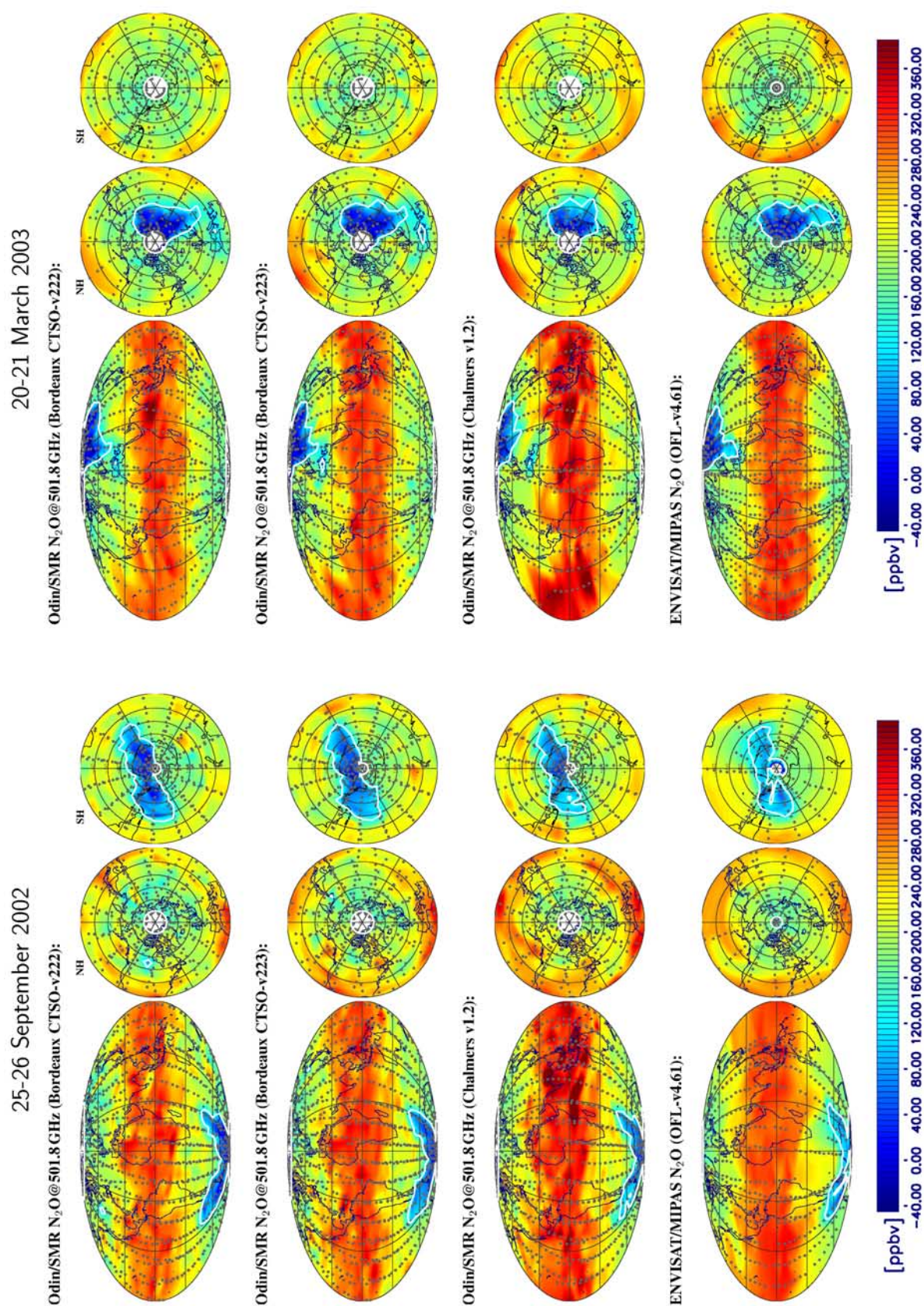


Figure 12

25–26 September 2002

20–21 March 2003

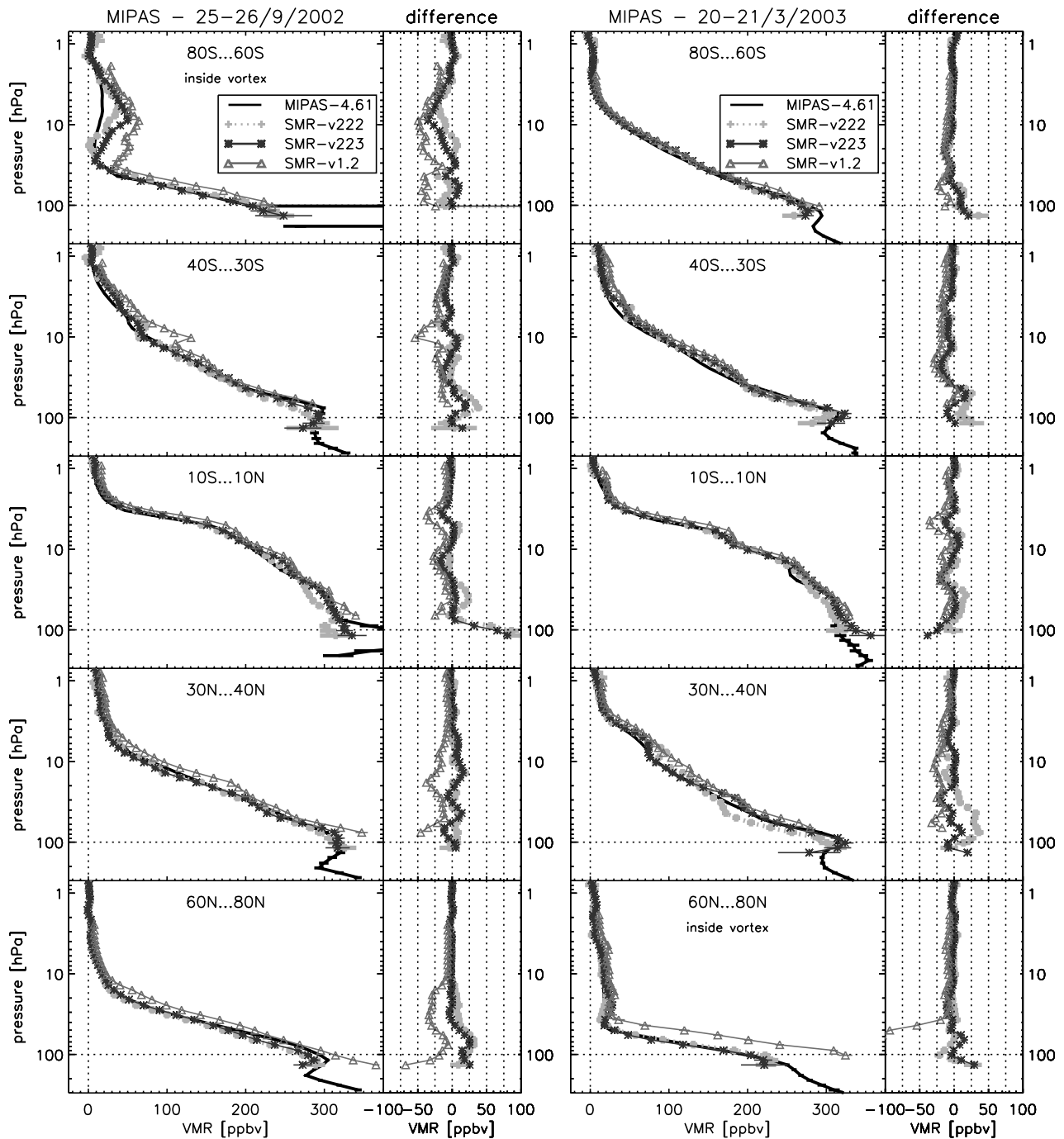
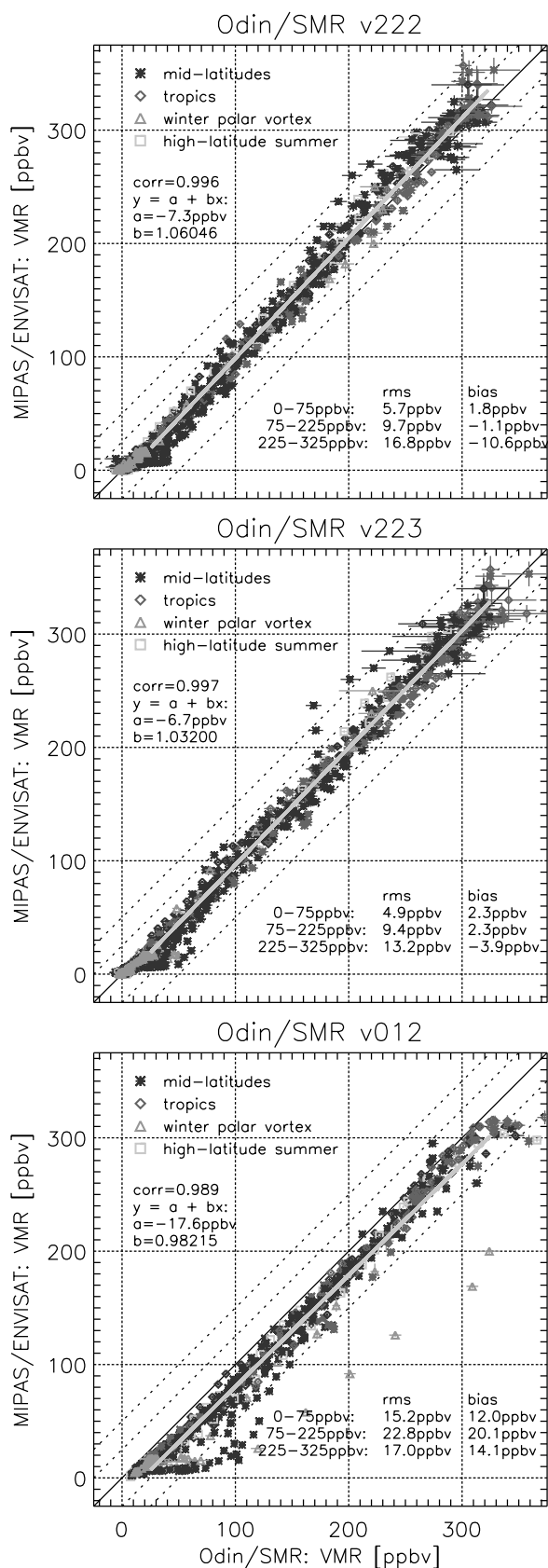


Figure 13. (left) Comparison of zonal mean N₂O from Odin/SMR for 25–26 September 2002 (1200–1200 UT) for selected latitude bands with zonal means derived from measurements of the MIPAS instrument on board the Envisat satellite for the same period (v4.61 off-line data). (right) Same but for 20–21 March 2003 (1200–1200 UT).

Figure 12. (left) Global maps of N₂O according to Odin/SMR level 2 products and Envisat/MIPAS version 4.61 (off-line) data for 25–26 September 2002 (1200–1200 UT) at the 50 hPa pressure level. Positions of the individual limb-scans are marked (crosses). The white line is the 125 ppbv contour of N₂O, chosen to indicate the edge of the polar vortex at 50 hPa. Lowest latitudes shown in the polar projections are 30°N (NH) or 30°S (SH). (right) Same but for 20–21 March 2003 (1200–1200 UT).



The Odin data set starts in November 2001 and N₂O measurements are performed in average on one day out of three and at least once per week. Due to Odin's Sun-synchronous near polar orbit, measurements cover the latitude range 82.5°S–82.5°N, except for a few periods when the satellite was pointing slightly out off the orbit direction. About 65 limb scans are performed per orbit and the orbit period of 97 min leads to about 15 orbits per day. N₂O measurements are meaningful roughly within the pressure range ~100 hPa to ~1 hPa (~15–50 km).

[36] As an example, zonal mean fields derived from Odin/SMR version 222 data are shown in Figure 15 for selected days in the 15 month period from October 2002 up to December 2003. Basic features of the zonal mean fields are the maximum in the tropical lower stratosphere, and decreasing mixing ratios with altitude primarily due to photodissociation by shortwave radiation. The global distribution of N₂O as a long-lived trace gas is determined by transport out of the tropics toward the winter hemispheres governed by the mean meridional circulation and its seasonal variation. Particular features of the zonal mean fields are the occurrence of steep gradients of the N₂O isopleths at high latitudes in the winter hemisphere (see, e.g., October 2002, December 2002 to March 2003, June to October 2003), indicating strong subsidence of air within the polar vortices [e.g., Urban *et al.*, 2004b], as well as relative flat isopleths in the well mixed “surf zone” at middle latitudes equatorward from the vortex, caused by quasi-horizontal mixing by planetary wave breaking [e.g., Randel *et al.*, 1993]. The previously reported “double peak” structure in the tropics at pressures below 5–10 hPa occurring during Southern Hemisphere fall is also present in the Odin/SMR measurements in April–June 2003. The peak in the Southern Hemisphere winter moves gradually poleward and downward during the Southern Hemisphere fall to spring seasons and disappears with the vortex breakup in November 2003. In contrast to SAMS and CLAES observations, we also see an equivalent “double-peak” feature during Northern Hemisphere winter in November 2002. However, the effect is much less pronounced in the November data of 2001 and 2003, which are more typical Southern Hemisphere winters with the vortex breakdown occurring later in November than during 2002. To summarize, the global Odin/SMR N₂O data set shows all major morphological features reported from previous measurements. This consistency and the overall data quality makes it certainly very

Figure 14. Scatterplot of zonal mean N₂O from Odin/SMR for the observation days 25–26 September 2002 and 20–21 March 2003 versus zonal mean N₂O derived from Envisat/MIPAS version 4.61 (off-line) data for the same dates. (top) Odin/SMR v222 data. (middle) Odin/SMR v223 data. (bottom) Odin/SMR v1.2 data. Root mean square (RMS) deviations and biases are again indicated for the three ranges of N₂O. Data with obvious large discrepancies arising from situations of high atmospheric variability, such as from high and middle southern latitudes on 25–26 September 2002, are excluded from the RMS and bias calculation. The dotted lines indicate deviations of ±25 and ±50 ppbv and the solid gray lines are the results of a linear fit.

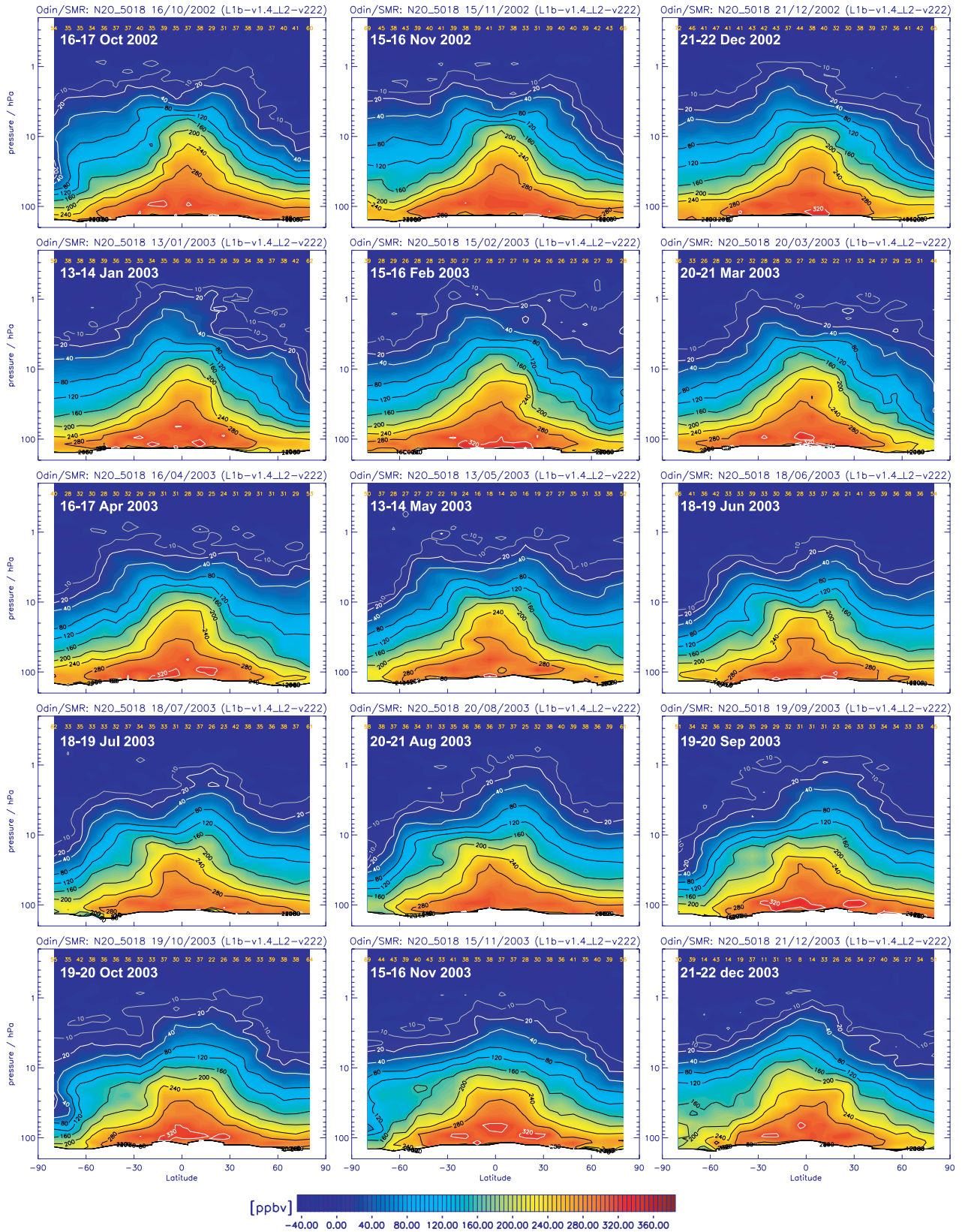
Odin/SMR N₂O@501.8GHz (v222) – 2003

Figure 15. Global zonal mean N₂O distribution derived from Odin/SMR measurements for selected observation days during a 15 month period from October 2002 up to December 2003 (version 222 data). Data are binned to 7.5° wide latitude intervals.

Table 1. Root Mean Square Deviations of Odin/SMR N₂O (Level 2) Data From Airborne Validation Experiments and Spaceborne Sensors ILAS-II (Level 2 Version 1.4) and MIPAS (ESA Off-Line Processor, Version 4.61)^a

N ₂ O Range, ppbv	SMR Version		
	v222, ppbv	v223, ppbv	v1.2, ppbv
<i>Balloon and Aircraft</i>			
0–75	11 (+2)	12 (+2)	8 (+6)
75–225	19 (–15)	18 (–12)	13 (–4)
225–325	28 (–21)	26 (–12)	28 (–7)
<i>ILAS-II</i>			
0–75	8 (+5)	7 (+4)	14 (+11)
75–225	20 (+16)	23 (+21)	47 (+36)
225–325	19 (+13)	23 (+18)	66 (+55)
<i>MIPAS</i>			
0–75	6 (+2)	5 (+2)	15 (+12)
75–225	10 (–1)	9 (+2)	23 (+20)
225–325	17 (–11)	13 (–4)	17 (+14)
<i>Odin/SMR Single-Scan Precision^b</i>			
0–75	10–20	15–25	5–10
75–225	10–15	25–30	10–15
225–325	15–45	30–45	15–25
<i>Odin/SMR Total Systematic Error^c</i>			
0–75	≤3		
75–150	3–12 (4–8%)		
>150	12–35 (8–15%)		

^aThe bias (or average difference) of Odin/SMR data with respect to the correlative measurements is given in parentheses. Also indicated are typical values for the single-scan precision and an estimation for the total systematic error of the Odin/SMR N₂O measurement.

^bCorresponding altitude resolution: v222 ~ 2 km, v223 ~ 1.5 km, v1.2 ≤ 4 km.

^cAdopted from Urban *et al.*, [2005].

useful for more detailed scientific studies with respect to stratospheric dynamics. Note also that Odin/SMR measurements of CO, performed on a regular basis of ~2 observation days per month since October 2003, will in the future provide another useful tool for transport studies, extending the vertical range of Odin tracer measurements up to the mesosphere/lower thermosphere region [Dupuy *et al.*, 2004].

5. Summary

[37] The Sub-Millimetre Radiometer (SMR) on board the Odin satellite, launched in February 2001, provides a quasi-continuous global data set of stratospheric nitrous oxide starting in November 2001. We presented an assessment of the quality of the Odin/SMR N₂O profile measurements by comparison of the latest level 2 data versions with correlative measurements from balloonborne, aircraftborne, and spaceborne sensors. The results of this assessment are summarized in Table 1 and Figure 16.

[38] An agreement with airborne validation experiments within a RMS deviation of 28 ppbv is found for all data versions under investigation. For N₂O mixing ratios lower than 200 ppbv, the agreement is within 19 ppbv. The results are based on validation measurements at middle and high northern latitudes and in the tropics. In terms of relative units, we find an agreement within 10% for

mixing ratios larger than about 150 ppbv. Results are roughly consistent with estimations of the systematic instrumental and spectroscopic error of 12–32 ppbv (8–14%) for the same range. For lower N₂O mixing ratios or higher altitudes, the validation analysis indicates slightly larger deviations than expected from the formal error analysis. Given the uncertainties due to (1) the atmospheric variability, and (2) the remaining noise in the averaged SMR profiles, the resulting values of the here presented validation analysis should therefore be interpreted as upper limits for the systematic errors of the SMR measurements.

[39] The cross comparison with the infrared solar occultation measurements of the ILAS-II instrument on ADEOS-II at middle and high southern and northern latitudes shows a positive bias of Odin/SMR N₂O measurements compared to nonvalidated ILAS-II v1.4 data. Root mean square (RMS) deviations are generally within 23 ppbv for Odin/SMR retrievals of version 222 and 223. The agreement is within 10 ppbv for VMR-N₂O < 50 ppbv, or, in terms of relative units, within 20% for VMR-N₂O > 100 ppbv. Version 1.2 retrievals show roughly a factor of 2 larger deviations. The systematic bias between the SMR data versions is thus larger than for the comparison with the airborne validation experiments. Also note in this context that for mixing ratios larger than 75 ppbv the ILAS-II v1.4 data were found to be systematically smaller by up to 30% compared to correlative balloonborne measurements, according to preliminary results of the validation analysis for this instrument (M. Ejiri *et al.*, manuscript in preparation, 2004).

[40] Odin/SMR retrievals are in relatively good agreement with (nonvalidated) N₂O mixing ratios obtained from the ESA Envisat/MIPAS level 2 off-line processor. For versions 222 and 223, RMS differences are within ~10–15 ppbv and the agreement is better than 10% for N₂O mixing ratios larger than ~100 ppbv. Version 1.2 retrievals show a small positive bias and give slightly larger RMS differences up to 25 ppbv, but still within the estimated systematic errors of the MIPAS and SMR observations. Note that MIPAS off-line processing yields sometimes unrealistical results for N₂O, without that those profiles were flagged as bad quality profiles. Although the most obvious cases were not used for the estimation of the systematic effects, some of the incorrect data might still influence the quantitative result of this comparison. Odin/SMR and MIPAS measurements both capture the global distribution of N₂O in reasonable qualitative agreement, in particular with respect to the N₂O gradients at the edge of the polar vortices. Good agreement is also found for the vertical distribution of N₂O at low latitudes. Note that MIPAS measurements are expected to be very sensitive to the presence of aerosols and clouds at low altitudes.

6. Conclusions

[41] To conclude, we described the status of the Odin/SMR N₂O level 2 data product by evaluating the quality of the 3 presently available data versions: Chalmers-v1.2, CTSO-v222, and CTSO-v223. A users choice of a version would very much depend on the application, even though

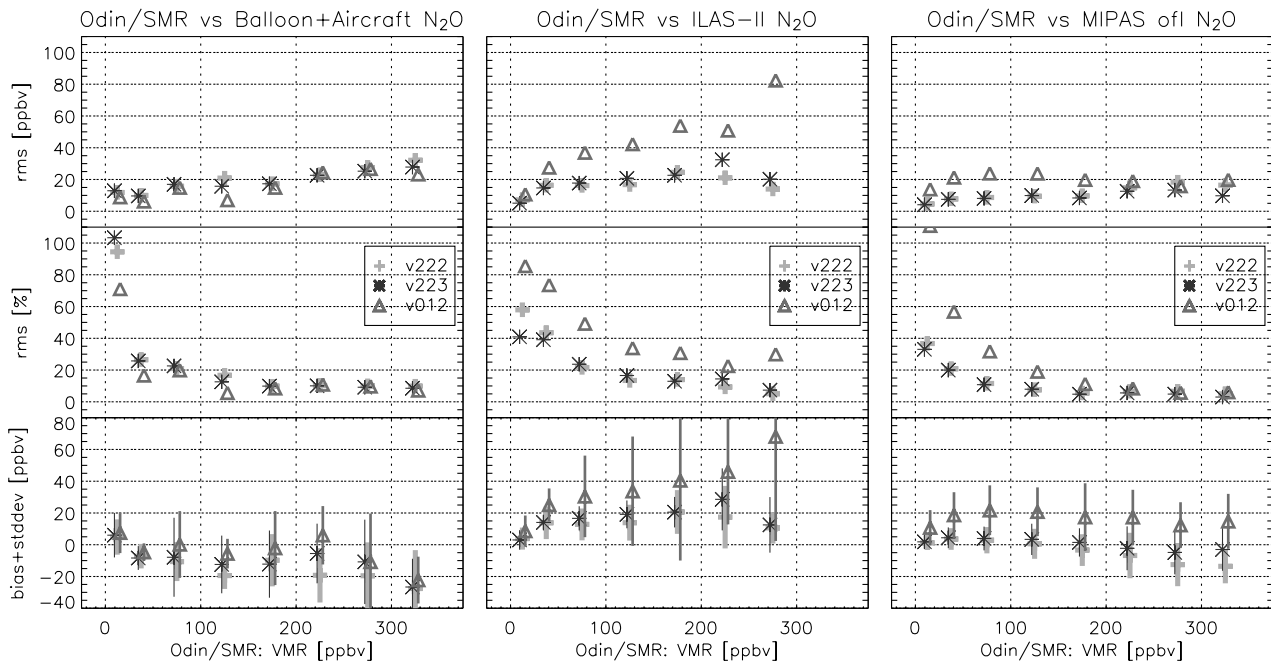


Figure 16. Differences between Odin/SMR N₂O data (versions 222, 223, and 1.2) and correlative measurements. The top and middle panels of each part show absolute and relative root mean square deviations. The bottom panels show bias and corresponding standard deviation (bars). Data are binned to 50 ppbv wide intervals. (left) Airborne validation experiments. (middle) ILAS-II onboard ADEOS-II (level 2 version 1.4). (right) Envisat/MIPAS (off-line processor, version 4.61).

version 223, the most recent and advanced version, appears to be slightly better according to this validation study. Chalmers-v1.2 data, the operational level 2 product, has the advantage to be systematically processed and covers therefore the whole Odin measurement period. The versions of the reference processor (CTSO-v222, CTSO-v223) serve in the first place to assure internal consistency and are therefore not produced systematically. However, version 222 data (based on a relatively slow processor) are already available for various periods of particular scientific interest (Arctic winter 2002–2003, Antarctic vortex split 2002), and version 223 data could be produced on request for selected observation days, if the best possible altitude resolution is required to answer a particular scientific question.

[42] In general, only good quality Odin/SMR profiles (assigned flag QUALITY = 0) shall be used for scientific studies and the measurement response associated to each retrieved mixing ratio shall be larger than ~ 0.9 , a measure to assure that the information comes entirely from the measurement and the contribution of the climatological a priori profile used by the OEM retrieval is negligible. Both values, quality flag and measurement response, are provided in the Odin/SMR level 2 HDF data files. See Urban *et al.* [2005] for a more detailed discussion. Known caveats of the Odin/SMR N₂O data are a systematic positive bias of the version 1.2 data with respect to the reference versions 222 and 223. In particular, care should be taken for the lowest retrieval altitudes where unrealistically high N₂O mixing ratios are sometimes found in version 1.2 data.

[43] Work on an improved “unified” Odin/SMR level 2 data product is underway and future releases of the operational product will address most of the issues raised

in this paper by further optimization of the retrieval methodology.

[44] **Acknowledgments.** Odin is a Swedish-led satellite project funded jointly by the Swedish National Space Board (SNSB), the Canadian Space Agency (CSA), the National Technology Agency of Finland (Tekes) and the Centre National d’Etudes Spatiales (CNES) in France. The Swedish Space Corporation (SSC) has been the industrial prime contractor. This research was supported in France by contracts from CNES and from the Programme National de Chimie Atmosphérique (PNCA) and in Sweden by SNSB. The authors wish to thank P. Baron and F. Merino for their early work on the implemented retrieval algorithms, C. Boone and F. Girod of the French database “Ether” for their contribution to level 2 data processing as well as Y. Kasai and J. P. Pommereau for their assistance to this validation effort. We are most grateful to the work of the Swedish team assuring the successful operation of the satellite. We also like to acknowledge that the ILAS-II instrument was funded and developed by the Ministry of the Environment of Japan (MOE) and was launched on board the ADEOS-II satellite by the National Space Development Agency of Japan (NASDA). ILAS-II data used in this paper were processed at the ILAS-II Data Handling Facility at the National Institute for Environmental Studies (NIES), Tsukuba, Japan. Envisat/MIPAS data were processed by the European Space Agency (ESA) and the development of the MIPAS retrieval system was supported by ESA contract 11717/95/NL/CN.

References

- Baron, P. (1999), Développement et validation du code MOLIERE: Chaîne de traitement des mesures micro-ondes du satellite Odin, Ph.D. thesis, Univ. of Bordeaux 1, Bordeaux, France.
- Baron, P., F. Merino, and D. Murtagh (2001), Simultaneous retrievals of temperature and volume mixing ratio constituents from non-oxygen Odin submillimeter bands, *Appl. Opt.*, 40(33), 6102–6110.
- Baron, P., P. Ricaud, J. de La Noë, J. E. P. Eriksson, F. Merino, M. Ridal, and D. P. Murtagh (2002), Studies for the Odin sub-millimetre radiometer: II. Retrieval methodology, *Can. J. Phys.*, 80(4), 341–356.
- Brasseur, G., and S. Solomon (1986), *Aeronomy of the Middle Atmosphere*, 2nd ed., Springer, New York.
- Bremer, H., M. von König, A. Kleinböhl, H. Küllmann, K. Künzi, K. Bramstedt, J. P. Burrows, K.-U. Eichmann, M. Weber, and A. P. H. Goede (2002), Ozone depletion observed by the Airborne Sub-

- millimeter Radiometer (ASUR) during the Arctic winter 1999/2000, *J. Geophys. Res.*, **107**(D20), 8277, doi:10.1029/2001JD000546.
- Camy-Peyret, C., P. Jeseke, T. Hawat, G. Durry, S. Payan, G. Berubé, L. Rochette, and D. Huegeni (1995), The LPMA balloon-borne FTIR spectrometer for remote sensing of atmospheric constituents, in *Proceedings of the 12th ESA Symposium on European Rocket and Balloon Programmes and Related Research*, Eur. Space Agency Spec. Publications, vol. SP-370, edited by B. Kaldeich-Schuermann, pp. 323–328, Eur. Space Ag., Paris.
- Carli, B., et al. (2004), First results of MIPAS/ENVISAT with operational Level 2 code, *Adv. Space Res.*, **33**, 1012–1019.
- de Valk, P., et al. (1997), Airborne heterodyne measurements of stratospheric ClO, HCl, O₃ and N₂O during SESAME-I over northern Europe, *J. Geophys. Res.*, **102**(D1), 1391–1398.
- Dupuy, E., et al. (2004), Strato-mesospheric measurements of carbon monoxide with the Odin Sub-Millimetre Radiometer: Retrieval and first results, *Geophys. Res. Lett.*, **31**, L20101, doi:10.1029/2004GL020558.
- El Amraoui, L., et al. (2004), Assimilation of Odin/SMR O₃ and N₂O measurements in a three-dimensional chemistry transport model, *J. Geophys. Res.*, **109**, D22304, doi:10.1029/2004JD004796.
- Eriksson, P., F. Merino, D. Murtagh, P. Baron, P. Ricaud, and J. de La Noë (2002), Studies for the Odin sub-millimetre radiometer: I. Radiative transfer and instrument simulation, *Can. J. Phys.*, **80**(4), 321–340.
- Eriksson, P., C. Jiménez, and S. Buehler (2005), Qpack, a tool for instrument simulation and retrieval work, *J. Quant. Spectrosc. Radiat. Transfer*, **91**, 47–61, doi:10.1016/j.jqsrt.2004.05.050.
- European Space Agency (ESA) (2000), *Envisat-MIPAS: The Michelson Interferometer for Passive Atmospheric Sounding, An Instrument for Atmospheric Chemistry and Climate Research*, vol. SP-1229, Paris.
- Frisk, U., et al. (2003), The Odin satellite: I. Radiometer design and test, *Astron. Astrophys.*, **402**(3), 27–34.
- Gunson, M., et al. (1996), The Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment: Deployment on the ATLAS Space Shuttle missions, *Geophys. Res. Lett.*, **23**, 2333–2336.
- Intergovernmental Panel on Climate Change (IPCC) (2001), *Climate Change 2001: The Scientific Basis, Contribution of Working Group I to the Third Assessment Report of the IPCC*, edited by J. T. Houghton et al., Cambridge Univ. Press, New York.
- Irion, F., et al. (2002), Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment version 3 data retrievals, *Appl. Opt.*, **41**(33), 6968–6979.
- Jones, R., and J. Pyle (1984), Observations of CH₄ and N₂O by the Nimbus 7 SAMS—A comparison with in situ data and two-dimensional numerical model calculations, *J. Geophys. Res.*, **89**(18), 5263–5279.
- Kanzawa, H., et al. (2003), Validation and data characteristics of nitrous oxide and methane profiles observed by the Improved Limb Atmospheric Spectrometer (ILAS) and processed with the Version 5.20 algorithm, *J. Geophys. Res.*, **108**(D16), 8003, doi:10.1029/2002JD002458.
- Lautié, N. (2003), Traitement des mesures satellitaires sub-millimétriques effectuées par Odin/SMR: Étude non-linéaire de la vapeur d'eau, Étude stratosphérique de HCN au moyen de mesures micro-ondes, Ph.D. thesis, Univ. of Paris VI, Paris.
- Lautié, N., et al. (2001), Retrieval of trace gas profiles from Odin/SMR limb measurements: Non-linear retrieval scheme for H₂O at 556.9 GHz, in *International Symposium on Submillimeter Wave Earth Observation From Space—III*, 8–9 Oct. 2001, Delmenhorst, Germany, edited by S. Bühler, pp. 93–105, Springer, New York.
- Mees, J., S. Crewell, H. Nett, G. de Lange, H. van de Stadt, J. Kuipers, and R. Panhuyzen (1995), ASUR—An airborne SIS receiver for atmospheric measurements of trace gases at 625 to 760 GHz, *IEEE Trans. Microwave Theory Tech.*, **43**(11), 2543–2548.
- Merino, F., et al. (2001), The Odin operational code (an optimized forward and retrieval code), *Rep. AP-39*, Arrhenius Lab., Meteorol. Inst. of Stockholm Univ., Stockholm.
- Merino, F., D. P. Murtagh, M. Ridal, J. E. P. Eriksson, P. Baron, P. Ricaud, and J. de La Noë (2002), Studies for the Odin sub-millimetre radiometer: III. Performance simulations, *Can. J. Phys.*, **80**(4), 357–373.
- Minschwaner, K., R. J. Salawitch, and M. B. McElroy (1993), Absorption of solar radiation by O₂: Implications for O₃ and lifetimes of N₂O, CFC₁₃, and CF₂Cl₂, *J. Geophys. Res.*, **98**(17), 10,543–10,561.
- Moreau, G. (1997), A new balloon-borne instrument for in situ measurements of stratospheric trace species using infrared laser diodes, in *Proceedings of the 13th ESA Symposium on European Rocket and Balloon Programmes and Related Research*, 26–29 May 1997, Öland, Sweden, Eur. Space Agency Special Publ., vol. SP-397, pp. 421–426, Paris.
- Moreau, G. (2001), Results and goals of SPIRALE after the flight of Gap in June 1999, in *Proceedings of the 15th ESA Symposium on European Rocket and Balloon Programmes and Related Research*, 28–31 May 2001, Biarritz, France, Eur. Space Agency Special Publ., vol. SP-471, edited by B. Warmbein, pp. 309–314, Paris.
- Moreau, G., M. Pirre, F. Taupin, C. Robert, and C. Camy-Peyret (2003), ENVISAT validation with SPIRALE from 2002 autumn mid-latitude and 2003 winter Arctic flights, in *Proceedings of the 16th ESA Symposium on European Rocket and Balloon Programmes and Related Research*, 2–5 June 2003, St. Gallen, Switzerland, Eur. Space Agency Special Publ., vol. SP-530, edited by B. Warmbein, pp. 481–486, Paris.
- Murtagh, D., et al. (2002), An overview of the Odin atmospheric mission, *Can. J. Phys.*, **80**(4), 309–319.
- Nakajima, H., T. Sugita, T. Yokota, and Y. Sasano (2003), Current status and early result of the ILAS-II onboard the ADEOS-II satellite, in *Proceedings of The International Society for Optical Engineering (SPIE): Sensors, Systems, and Next Generation Satellites IX*, 8–12 September 2003, Barcelona, Spain, vol. 5234, edited by R. Meynart et al., pp. 36–45, Int. Soc. Opt. Eng., Bellingham, Wash.
- Olberg, M., et al. (2003), The Odin satellite: II. Radiometer data processing and calibration, *Astron. Astrophys.*, **402**(3), 35–38.
- Payan, S., C. Camy-Peyret, P. Jeseke, T. Hawat, M. Pirre, J. Renard, C. Renard, F. Lefèvre, H. Kanzawa, and Y. Sasano (1999), Diurnal and nocturnal distribution of stratospheric NO₂ from solar and stellar occultation measurements in the Arctic vortex: Comparison with models and ILAS satellite measurements, *J. Geophys. Res.*, **104**, 21,585–21,593.
- Pirre, M., N. Huret, G. Moreau, C. Robert, V. Catoire, F. Lefèvre, G. Berthet, and J. Urban (2004), Subsidence and chlorine activation at the edge of the Arctic polar vortex observed by SPIRALE on January 21, 2003, in *Proceedings of XX Quadrennial Ozone Symposium Kos, Greece*, edited by C. Zerefos, pp. 1021–1022, Int. Ozone Comm., Kos, Greece.
- Plumb, A., and M. Ko (1992), Interrelationships between mixing ratios of long-lived stratospheric constituents, *J. Geophys. Res.*, **97**(D9), 10,145–10,156.
- Proffit, M., J. Margitan, K. Kelly, M. Loewenstein, J. Podolske, and K. Chan (1990), Ozone loss in the Arctic polar vortex inferred from high-altitude aircraft measurements, *Nature*, **347**, 31–36.
- Proffit, M., M. Loewenstein, and S. Solomon (1992), Comparison of 2-D model simulations of ozone and nitrous oxide at high latitudes with stratospheric measurements, *J. Geophys. Res.*, **97**, 939–944.
- Randel, W., J. Gille, A. Roche, J. Kumer, J. Mergenthaler, J. Waters, E. Fishbein, and W. Lahoz (1993), Stratospheric transport from the tropics to middle latitudes by planetary-wave mixing, *Nature*, **365**, 533–535.
- Randel, W., B. Boville, J. Gille, P. Bailey, S. Massie, J. Kumer, J. Mergenthaler, and A. Roche (1994), Simulation of stratospheric N₂O in the NCAR CCM2: Comparison with CLAES data and global budget analyses, *J. Atmos. Sci.*, **51**, 2834–2845.
- Raspollini, P., et al. (2003), Level 2 near-real-time analysis of MIPAS measurements on ENVISAT, in *Remote Sensing of Clouds and the Atmosphere VII*, vol. 4882, edited by K. Schäfer et al., pp. 324–334, Int. Soc. Opt. Eng., Bellingham, Wash.
- Remedios, J., et al. (1996), Measurements of CH₄ and N₂O distributions by the ISAMS: Retrieval and validation, *J. Geophys. Res.*, **101**, 9843–9871.
- Ricaud, P., et al. (2005), Polar vortex evolution during the 2002 Antarctic major warming as observed by the Odin satellite, *J. Geophys. Res.*, **110**, D05302, doi:10.1029/2004JD005018.
- Ridolfi, M., et al. (2000), Optimized forward model and retrieval scheme for MIPAS near-real-time data processing, *Appl. Opt.*, **39**(8), 1323–1340.
- Robinson, A., J. McIntyre, N. Harris, J. Pyle, P. Simmonds, and F. Danis (2000), A lightweight balloon-borne gas chromatograph for in situ measurements of atmospheric halocarbons, *Rev. Sci. Instrum.*, **71**(12), 4553–4560.
- Roche, A., J. Kumer, J. Mergenthaler, G. Ely, W. Uplinger, J. Potter, T. James, and L. Sterritt (1993), The Cryogenic Limb Array Etalon Spectrometer (CLAES) on UARS: Experiment description and performance, *J. Geophys. Res.*, **98**, 10,763–10,775.
- Roche, A., et al. (1996), Validation of CH₄ and N₂O measurements by the CLAES instrument on the Upper Atmosphere Research Satellite, *J. Geophys. Res.*, **101**, 9679–9710.
- Rodgers, C. D. (1976), Retrieval of atmospheric temperature and composition from remote measurements of thermal radiation, *Rev. Geophys. Space Phys.*, **14**(4), 609–624.
- Taylor, F., et al. (1993), Remote sensing of atmospheric structure and composition by pressure modulator radiometry from space: The ISAMS experiment on UARS, *J. Geophys. Res.*, **98**, 10,799–10,814.
- Urban, J. (1998), Measurements of the stratospheric trace gases ClO, HCl, O₃, N₂O, H₂O, and OH using air-borne submm-wave radiometry at 650 and 2500 GHz, Ph.D. thesis, *Rep. Pol. Res. 264*, Univ. of Bremen, Bremen.
- Urban, J., et al. (1999), Recent airborne heterodyne receivers for the sub-millimeter-wave range, in *Proceedings of the International Workshop on Submm-Wave Observation of Earth's Atmosphere From Space*, edited by

- H. Masuko, M. Shiotani, and K. Shibasaki, pp. 151–163, Earth Observ. Res. Cent., Natl. Space Devel. Agency of Jpn., Tokyo.
- Urban, J., et al. (2002), Trace gas retrieval from Odin/SMR limb observations: Stratospheric mode, in *Proceedings of the 6th European Symposium on Stratospheric Ozone*, 2–6 Sept. 2002, Göteborg, Sweden, edited by N. Harris, G. Amanatidis, and J. Levine, pp. 474–477, Eur. Union, Brussels.
- Urban, J., P. Baron, N. Lautié, K. Dassas, N. Schneider, P. Ricaud, and J. de La Noë (2004a), MOLIERE (v5): A versatile forward and inversion model for the millimeter and sub-millimeter wavelength range, *J. Quant. Spectrosc. Radiat. Transfer*, 83(3–4), 529–554.
- Urban, J., et al. (2004b), The Northern Hemisphere stratospheric vortex during the 2002–03 winter: Subsidence, chlorine activation and ozone loss observed by the Odin Sub-Millimetre Radiometer, *Geophys. Res. Lett.*, 31, L07103, doi:10.1029/2003GL019089.
- Urban, J., et al. (2005), Odin/SMR limb observations of stratospheric trace gases: Level 2 processing of ClO, N₂O, O₃, and HNO₃, *J. Geophys. Res.*, doi:10.1029/2004JD005741, in press.
- World Meteorological Organization (WMO) (2003), Scientific assessment of ozone depletion: 2002, *Global Ozone Res. and Monit. Proj. Rep.* 47, Geneva.
- Yokota, T., H. Nakajima, T. Sugita, H. Tsubaki, Y. Itou, M. Kaji, M. Suzuki, H. Kanzawa, J. H. Park, and Y. Sasano (2002), Improved Limb Atmospheric Spectrometer (ILAS) data retrieval algorithm for Version 5.20 gas profile products, *J. Geophys. Res.*, 107(D24), 8216, doi:10.1029/2001JD000628.
- Yung, Y., and C. Miller (1997), Isotopic fractionation of stratospheric nitrous oxide, *Science*, 278, 1778–1780.
- H. Bremer, A. Kleinböhl, K. Küllmann, K. Künzi, and J. Kuttippurath, Institute of Environmental Physics, University of Bremen, Otto-Hahn-Allee 1, D-28359 Bremen, Germany.
- C. Camy-Peyret, G. Dufour, and S. Payan, Laboratoire de Physique Moléculaire et Applications/CNRS, Université Pierre et Marie Curie (Paris 6), Case 76, 4, place Jussieu, F-75252 Paris Cedex 05, France.
- M. K. Ejiri, H. Nakajima, Y. Sasano, T. Sugita, and T. Yokota, National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba-Shi, Ibaraki, 305-8506, Japan.
- J. de La Noë, E. Dupuy, L. El Amraoui, and F. Jégou, Observatoire Aquitain des Sciences de l'Univers, CNRS, L3AB, Université Bordeaux 1, F-33270 Floirac, France.
- P. Eriksson, N. Lautié, D. Murtagh, and J. Urban, Department of Radio and Space Science, Chalmers University of Technology, Hörsalsvägen 11, CE-412 96, Göteborg, Sweden. (urban@obs.u-bordeaux1.fr; jo.urban@rss.chalmers.se)
- U. Frisk, Swedish Space Corporation, Box 4207, 171 04 Solna, Sweden.
- N. R. P. Harris, European Ozone Research Coordinating Unit, Centre for Atmospheric Science, University of Cambridge, Department of Chemistry, Lensfield Road, Cambridge CB2 1EW, UK.
- N. Huret and M. Pirre, Laboratoire de Physique et Chimie de l'Environnement, CNRS, Université d'Orléans, 3A, Avenue de la Recherche Scientifique, F-45071 Orléans Cedex 2, France.
- C. Jiménez, Institute of Atmospheric and Environmental Science, School of Geosciences, University of Edinburgh JCMB, Mayfield Road, Edinburgh EH9 3JZ, UK.
- E. Le Flochmoën and P. Ricaud, Laboratoire d'Aérodynamique, Observatoire de Midi-Pyrénées, F-31400 Toulouse, France. (lefe@aero.obs-mip.fr; philippe.ricaud@aero.obs-mip.fr)
- M. Olberg, Onsala Space Observatory, Chalmers University of Technology, SE-439 92 Onsala, Sweden.
- A. D. Robinson, Centre for Atmospheric Science, University of Cambridge, Department of Chemistry, Lensfield Road, Cambridge CB2 1EW, UK.
- C. Piccolo, Atmospheric, Oceanic and Planetary Physics, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, UK.
- P. Raspollini, Istituto di Fisica Applicata “Nello Carrara” del CNR, Via Panciatichi, 64, I-50127 Firenze, Italy.
- M. Ridolfi, Dipartimento di Chimica Fisica ed Inorganica, Università di Bologna, Viale Risorgimento, 4, I-40136 Bologna, Italy.