Atmos. Chem. Phys. Discuss., 12, 22629–22685, 2012
www.atmos-chem-phys-discuss.net/12/22629/2012/
doi:10.5194/acpd-12-22629-2012

Atmospheric Chemistry and Physics Discussions

Assessment of the interannual variability and impact of the QBO and upwelling on tracer-tracer distributions of N_2O and O_3 in the tropical lower stratosphere

F. Khosrawi[1], R. Müller[2], J. Urban[3], M. H. Proffitt[4], G. Stiller[5], M. Kiefer[5], S. Lossow[5], D. Kinnison[6], F. Olschewski[7], M. Riese[2], and D. Murtagh[3]

[1]Department of Meteorology, Stockholm University, Stockholm, Sweden
[2]IEK-7: Stratosphere, Forschungszentrum Jülich, Jülich, Germany
[3]Department of Radio and Space Science, Chalmers University of Technology, Göteborg, Sweden
[4]Proffitt Instruments, Austin, Texas, USA
[5]Karlsruhe Institute of Technology, Karlsruhe, Germany
[6]National Center for Atmospheric Research, Boulder, Colorado, USA
[7]Physics Department, Universität Wuppertal, Wuppertal, Germany

Received: 2 August 2012 – Accepted: 14 August 2012 – Published: 3 September 2012

Correspondence to: F. Khosrawi (farah@misu.su.se)

Published by Copernicus Publications on behalf of the European Geosciences Union.

ACPD

12, 22629–22685, 2012

Abstract

A modified form of tracer-tracer correlations of N_2O and O_3 has been used as a tool for the evaluation of atmospheric photochemical models. Applying this method monthly averages of N_2O and O_3 are derived for both hemispheres by partitioning the data into altitude (or potential temperature) bins and then averaging over a fixed interval of N_2O. In a previous study, the method has been successfully applied to the validation of two Chemical Transport Models (CTMs) and one Chemistry-Climate Model (CCM) using 1-year climatology derived from the Odin Sub Millimetre Radiometer (Odin/SMR). However, the applicability of a 1-year climatology of monthly averages of N_2O and O_3 has been questioned due to the inability of some CCMs to simulate a specific year for the evaluation of CCMs. In this study, satellite measurements from Odin/SMR, the Aura Microwave Limb Sounder (Aura/MLS), the Michelson Interferometer for Passive Atmospheric Sounding on ENVISAT (ENVISAT/MIPAS), and the Cryogenic Infrared Spectrometers and Telescopes for the Atmosphere (CRISTA-1 and CRISTA-2) as well as model simulations from the Whole Atmosphere Community Climate Model (WACCM) are considered. By using seven to eight years of satellite measurements derived between 2003 and 2010 from Odin/SMR, Aura/MLS, ENVISAT/MIPAS and six years of model simulations from WACCM the interannual variability of lower stratospheric monthly averages of N_2O and O_3 is assessed. It is shown that the interannual variability of the monthly averages of N_2O and O_3 is low and thus can be easily distinguished from model deficiencies. Further, it is investigated why large differences between Odin/SMR observations and model simulations from the Karlsruhe Simulation Model of the Middle Atmosphere (KASIMA) and the atmospheric general circulation model ECHAM5/Messy1 are found for the Northern and Southern Hemisphere tropics (0° to 30° N and 0° to -30° S, respectively). The differences between model simulations and observations are most likely caused by an underestimation of the quasi-biennial oscillation and tropical upwelling by the models as well as due to biases and/or instrument noise from the satellite instruments. Finally, an inter-comparison between

ACPD
12, 22629–22685, 2012

Odin/SMR, Aura/MLS, ENVISAT/MIPAS and WACCM was performed. The comparison shows that these data sets are generally in good agreement but that also some known biases of the data sets are clearly visible in the monthly averages, thus showing that this method is not only a valuable tool for model evaluation but also for satellite inter-comparisons.

1 Introduction

N_2O is a long-lived species in the troposphere and lower stratosphere and thus can be used as a tracer for atmospheric transport. N_2O is produced in the troposphere by natural (soils, wetlands) as well as by anthropogenic sources (industrial emissions, biomass burning) and is destroyed photochemically in the stratosphere. N_2O abundances in the troposphere have rapidly increased due to anthropogenic emissions during the last centuries. Pre-industrial values were around 270 ± 7 ppbv and have risen to 319 ± 0.12 ppbv in 2005 (Forster et al., 2007). The lifetime of N_2O is 100 years in the troposphere and decreases with altitude from several years in the lower stratosphere to ~8 months in the middle stratosphere (Stanford and Ziemke, 1991). Throughout the lower stratosphere, the mean N_2O mixing ratio decreases with increasing altitude. N_2O is transported via the Brewer-Dobson circulation to the polar regions after entering the lower stratosphere at the tropical tropopause (Brewer, 1949; Dobson, 1956). The temporal and spatial distribution of N_2O can thus be used as diagnostic of global-scale transport processes at different timescales, from seasons to decades (e.g. Ricaud et al., 2009). Measurements of N_2O show that the zonally averaged lower stratospheric N_2O mixing ratios vary systematically with season, latitude and altitude (Strahan et al., 1999). Strahan et al. (1999) also demonstrated that local changes from the seasonal zonal mean profiles can be interpreted as the result of recent stratospheric transport.

Ozone (O_3) is rather short lived in the troposphere (days to weeks). Although O_3 has a longer lifetime in the lower stratosphere ozone is not photochemically conserved and thus has a limited application as tracer of transport. O_3 mixing ratios in the lower

ACPD
12, 22629–22685, 2012

stratosphere are highly variable. This is due to both the strong variability of O_3 photochemistry with latitude, altitude and season as well as the seasonality of the atmospheric transport vertically and horizontally (e.g. Dobson et al., 1973; Proffitt et al., 2003). O_3 is throughout the year produced photochemically primarily in the tropical stratosphere. The peak production rate of O_3 occurs at ~30 km (Perliski et al., 1989). The photochemical destruction of ozone is particularly pronounced at polar latitudes during late winter/early spring and summer. Ozone changes due to dynamical processes are caused by ascent of stratospheric air due to the Brewer Dobson circulation in the tropical stratosphere together with descent over the Arctic and Antarctic. This results in a seasonal transport of stratospheric air from the tropics to the polar regions where the strength of the ozone transport is dependent on season (Brewer, 1949; Dobson, 1956).

The inter-annual variability of the equatorial stratosphere (~17–50 km) is dominated by the quasi-biennial oscillation (QBO); downward propagating easterly and westerly wind regimes that occur with an average period of 28 months (Baldwin et al., 2001) while the semi-annual oscillation (SAO) is the strongest mode of annual variability above 35 km altitude (e.g. Randel et al., 1998; Lossow et al., 2008). Satellite measurements (e.g. Randel et al., 1994; Ricaud et al., 2009; Jin et al., 2009) and model simulations (e.g. Sassi et al., 1993; Jin et al., 2009) have shown that the equatorial N_2O fields exhibit a SAO in the mid and upper stratosphere. In the lower stratosphere several studies have shown that the signature of the annual oscillation (AO) is found in the trace gas distributions (e.g. Randel et al., 1998; Schoeberl et al., 2008; Ricaud et al., 2009). Further, based on satellite measurements, the influence of the QBO on long-lived species in the tropical regions has been shown (e.g. O'Sullivan and Dunkerton, 1997).

In the recent decade a lot of effort has been put into the evaluation and improvement of Chemistry-Climate Models (CCMs) to provide reliable predictions of future changes in climate (WMO, 2010). Evaluation of CCMs with measurements, using a variety of diagnostics and ensuring that appropriate diagnostics are used, is thus essential

ACPD
12, 22629–22685, 2012

(SPARC CCMVal, 2010). The simulation of the QBO in CCMs is among other processes still a challenge (e.g. Giorgetta et al., 2006). The difficulties in simulating the QBO have various reasons which all result in serious biases in the representation of the wave-mean flow interaction which is the key process in the QBO forcing (Holton and Lindzen, 1972). While some models get the oscillations with realistic time scales and thus resemble the observed QBO, other models get periods deviating from the observed ones and are typically faster (Giorgetta et al. (2006) and references therein). Giorgetta et al. (2006) showed that for producing a realistic QBO in CCM simulations a very high vertical resolution is required (∼1 km) so that the vertical structures of the waves are resolved sufficiently well. Further, a realistic simulation of the QBO improves the simulation of tropical upwelling and the atmospheric tape recorder (Baldwin et al., 2001; Giorgetta et al., 2006). The atmospheric tape recorder is caused by the imprint of the tropopause temperature on trace gases as e.g. H_2O, CO and HCN and their transport into the stratosphere with the upwelling branch of the Brewer-Dobson circulation. The tape recorder signature was first discussed based on satellite-borne water vapour measurements by Mote et al. (1996).

In a recent model evaluation study (Khosrawi et al., 2009) a method based on ozone (O_3) and nitrous oxide (N_2O) measurements was used to evaluate two CTMs and one CCM in the lower stratosphere. In this method, monthly averages of O_3 and N_2O binned by potential temperature are calculated in the N_2O/O_3 tracer space. The method applied in Khosrawi et al. (2009) was introduced by Proffitt et al. (2003) for the northern hemisphere lower stratosphere based on aircraft and balloon-borne measurements and helps to separate O_3 variability due to latitudinal transport from photochemical changes. In our follow-up studies this method had been extended to greater altitudes and to the southern hemisphere (Khosrawi et al., 2004, 2006; Khosrawi et al., 2008) using satellite data from the Improved Limb Atmospheric Spectrometers (ILAS and ILAS-II) and the Odin Sub-Millimetre Radiometer (Odin/SMR), respectively.

The model evaluation (Khosrawi et al., 2009) was performed for the potential temperature levels of 500 ± 25 K and 650 ± 25 K. In the northern hemisphere tropics (0°

ACPD
12, 22629–22685, 2012

to 30° N) differences between models and satellite observations were found at both levels. At 500 ± 25 K, in both models a steeper negative correlation (decreasing O_3 with increasing N_2O) was found. In Khosrawi et al. (2009) these differences were explained to be probably caused partly by the large vertical O_3 gradients occurring in the tropics which cannot entirely be resolved by the rather coarse vertical resolution of Odin/SMR of 3 km and partly by inaccuracies in the model simulations of transport in the tropical stratosphere. In the frame of this study it will be shown that the latter reason is applicable and that the former has no direct influence on the results. Though the method suggested by Proffitt et al. (2003) focuses on O_3, a discrepancy in N_2O between models and satellite observations had been found as well. In the northern hemisphere tropics, monthly averages of N_2O values at 650 ± 25 K (~25 km) were much higher from Odin/SMR observations (reaching up to 330 ppbv) than simulated by the models. Though not discussed in detail by Khosrawi et al. (2009), the same discrepancies were found in the southern hemisphere tropics (0° to −30° S). These N_2O values are ~10 ppbv higher than the highly accurate ground-based observations of N_2O (319 ppbv in 2005) derived in the troposphere which is the only source region of this trace gas. Satellite data sets tend in general to have somewhat too high N_2O mixing ratios exceeding >320 ppbv as can be seen in the tracer-tracer correlations and probability density functions (PDFs) derived from ACE-FTS (Hegglin and Shepherd, 2007) and vertical profiles from e.g. Aura-MLS and ENVISAT/MIPAS (Barrett et al., 2006; Lambert et al., 2007; Griesfeller et al., 2008; Payan et al., 2009).

Here, monthly averages of N_2O and O_3 from different satellite data sets are derived and compared to the Odin/SMR monthly averages. The satellite data sets that have been chosen are the ones which have a high temporal and spatial coverage in the tropics as the Aura Microwave Limb Sounder (MLS), the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on ENVISAT and the Cryogenic Infrared Spectrometers and Telescopes for the Atmosphere (CRISTA). Further, monthly averages of N_2O and O_3 derived from the satellite data sets are compared to the monthly averages derived from model simulations from the Whole Atmosphere Community Climate

ACPD
12, 22629–22685, 2012

Model (WACCM). By applying eight years of Odin/SMR (2003–2010), seven years of Aura/MLS (2004–2010), seven years of ENVISAT/MIPAS (2003-2009) measurements and six years of WACCM simulations (2005–2010) the inter-annual variability of monthly averages of N_2O and O_3 can be assessed. This assessment allows the criticism on e.g. Khosrawi et al. (2009) to be addressed that the application of solely one year of monthly averages of N_2O and O_3 for the evaluation of CCMs is not sufficient. By applying additionally CRISTA-1 and CRISTA-2 data the cause of the exceptionally high N_2O bins derived from Odin/SMR at 650 ± 25 K that were found in our recent model evaluation study (Khosrawi et al., 2009) are investigated. Finally, a comparison between Odin/SMR, Aura/MLS, ENVISAT/MIPAS and WACCM is performed for the years 2003, 2006 and 2009 to test the applicability of the monthly averages of N_2O and O_3 also for satellite-satellite inter-comparisons.

2 Satellite Data

In the following the satellite data sets used in this study will be briefly described. A summary of the used satellite data sets, their vertical resolution, their precision as well as the number of profiles they measured per day is given in Table 1.

2.1 Odin/SMR

Odin was launched on 20 February 2001 and carries two instruments, the Optical Spectrograph and Infrared Imaging System (OSIRIS) (Llewellyn et al., 2004) and the Sub-Millimetre Radiometer (SMR) (Frisk et al., 2003). Observations of the thermal emission of trace gases originating from the Earth's limb were performed by Odin/SMR in a time-sharing mode with astronomical observations until 2007 and solely in aeronomy mode thereafter. In aeronomy mode, various target bands are dedicated to profile measurements of trace constituents relevant to stratospheric and mesospheric chemistry and dynamics such as O_3, ClO, N_2O, HNO_3, H_2O, CO, HO_2 and NO, as well as

ACPD
12, 22629–22685, 2012

minor isotopologues of H_2O and O_3 (e.g., Murtagh et al., 2002). Stratospheric mode measurements were performed every third day until April 2007 and every other day thereafter. A typical stratospheric mode scan covers the altitude range from 7 to 70 km with a resolution of ~1.5 km in terms of tangent altitude below 50 km and of ~5.5 km above. Usually, the latitude range between 82.5° S and 82.5° N is observed by the measurements (Urban et al., 2005a,b). Here, we use Chalmers Odin/SMR version 2.1 data of N_2O and O_3 from the 501.8 GHz band. N_2O and O_3 profiles are retrieved from ~12 to 60 km and ~13 to 65 km, respectively, with an altitude resolution of 1.5 km and 3 km, respectively. The systematic error of the N_2O measurements is estimated to be ≤12 ppbv above 20 km and in the range of 12-35 ppbv (up to 10-15 %) below (Urban et al., 2005a). The single profile precision has been estimated to be 10-30 ppbv. Extensive validation of Odin/SMR has been conducted, especially with the space-borne sensors ENVISAT/MIPAS, Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS) and Aura/MLS. The Odin/SMR N_2O data have been validated in the altitude range of ~15-50 km. The comparison of Odin/SMR N_2O with ENVISAT/MIPAS Oxford processor showed a good overall agreement within 4–7 ppbv (Urban et al., 2005b, 2006). The systematic error of Odin/SMR O_3 measurements is estimated to be lower than 0.6 ppmv. Odin/SMR measurements of O_3 derived with the Chalmers Version 2.1 retrieval algorithm were validated by Jégou et al. (2008) and showed a good agreement of Odin/SMR Version 2.1 data with ground-based (-0.15 ± 0.3 ppmv), balloon-borne (-0.7 ± 1 ppmv) and space-borne sensors (-0.3 ± 0.2 ppmv). The inter-comparison of Odin/SMR ozone measurements with ENVISAT/MIPAS and balloon sonde data by Jones et al. (2007) showed an agreement within 10 % between 17 and 55 km (a maximum deviation of 0.42 ppmv) and 5–10 % between 25 and 35 km (less than 0.5 ppmv below 33 km).

2.2 Aura/MLS

The Microwave Limb Sounder (MLS) on the Earth Observing System Aura satellite was launched in July 2004. The Aura/MLS instrument is an advanced successor to

ACPD
12, 22629–22685, 2012

the MLS instrument on the Upper Atmosphere Research satellite (UARS) that was launched in 1991 and provided measurements until 1999. MLS is a limb sounding instrument that measures the thermal emission at millimeter and submillimetre wavelengths using seven radiometers to cover five broad spectral regions (Waters et al., 2006). Measurements are performed from the surface to 90 km with a global latitude coverage from 82° S and 82° N. Here, we use MLS v2.2 data. The estimated single profile precision of a retrieved profile of N_2O is ∼13–25 ppbv (7–38 %) and the estimated accuracy 3–70 ppbv (9–25 %) for the pressure range 100-4.6 hPa. The scientifically useful range of the N_2O data is from 100 to 1 hPa. A detailed validation of the MLS N_2O data can be found in Lambert et al. (2007). A good agreement with correlative data was found. The agreement with ACE-FTS was within ±5 % for pressures 100–1 hPa, with the ENVISAT/MIPAS within ±5 % for pressures 32–1 hPa and for Odin/SMR between 0 % and −5 % for pressures 68-4.6 hPa. Validation studies of MLS O_3 were performed by Jiang et al. (2007); Livesey et al. (2008) and Froidevaux et al. (2008). The precision of O_3 has been estimated to be ±40 ppbv in the upper troposphere and lower stratosphere (215–100 hPa). The accuracy has been estimated to be ±20 ppbv in the UT/LS between 215 and 147 hPa. A comparison of MLS O_3 with SAGE (Froidevaux et al., 2008) and radiosondes (Jiang et al., 2007) showed a ∼20 % bias in MLS O_3 at 215 hPa at mid and high latitudes. However, this bias was not found in the comparison of MLS O_3 with ground-based lidar measurements (Jiang et al., 2007).

2.3 ENVISAT/MIPAS

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) was launched in March 2002 onboard the ESA's ENVIronmental SATellite (ENVISAT) research satellite. MIPAS is a middle infrared Fourier Transform spectrometer measuring the atmospheric emission spectrum in the limb sounding geometry. MIPAS operated in its nominal observation mode from June 2002 to March 2004, thus, approximately two years. Measurements during this time period were performed in its full spectral resolution measurement mode with a designated spectral resolution of 0.035 cm^{-1}.

ACPD
12, 22629–22685, 2012

Measurements were performed covering the altitude range from the mesosphere to the troposphere with a high vertical resolution (about 3 km in the stratosphere). After a failure of the interferometer slide in the end of March 2004, MIPAS resumed measurements in January 2005 with a reduced spectral resolution of 0.0625 cm^{-1} but with improved spatial resolution. Target products of MIPAS are the trace gases as e.g. H_2O, O_3, HNO_3, CH_4, N_2O and NO_2 as well as temperature (Fischer and Oelhaf, 1996). Here, we use the N_2O and O_3 data retrieved with the IMK/IAA (Institut für Klimatologie und Klimaforschung in Karlsruhe/Instituto de Astrofisica de Andalucia) processor, namely version V3O_N2O_11 and V3O_O3_9, respectively, for the years 2003 and 2004 (full resolution measurements) as well as V4O_N2O_202 and V4O_O3_201, respectively for 2005 onwards measurements (reduced resolution measurements). A detailed description of the MIPAS retrieval and error analysis for observations with the reduced spectral resolution can be found in von Clarmann et al. (2009). For N_2O the total retrieval error varies between 8 and 16 % in major parts of the stratosphere and reaches 30.5 % at the stratopause. The precision has been estimated to be 8.8 ppbv (4.5 %) at 20 km, 13.3 ppbv (8.3 %) at 25 km and 3.9 ppbv (6.7 %) at 30 km. The total error of O_3 is 12–13 % and precision for O_3 varies between 50 and 275 ppbv (140 ppbv or 3.8 % at 20 km, 275.9 ppbv or 4.4 % at 25 km and 256.7 ppbv or 4.3 % at 30 km). The IMK/IAA full spectral resolution retrievals of N_2O and O_3 are described in Glatthor et al. (2005) and Glatthor et al. (2006). In both versions a positive bias is found in the N_2O data in the lowermost stratosphere. The total error for the full spectral resolution retrieval lies between 18 to 25 ppbv (10–12 %) for N_2O at altitudes between 20 and 30 km. The MIPAS N_2O data were validated by Payan et al. (2009) and the O_3 data by Steck et al. (2007). A very good agreement with correlative data has been found for O_3 with differences within ±10 %. The MIPAS N_2O measurements are in good agreement with correlative data. No bias was found in the middle stratosphere while a small positive bias of 4 % was found in the lower stratosphere (Payan et al., 2009).

ACPD
12, 22629–22685, 2012

2.4 CRISTA

The CRISTA (CRyogenic Infrared Spectrometers and Telescopes for the Atmosphere) instrument was launched aboard the NASA space shuttles "Atlantis" and "Discovery", respectively, into an orbit of 300 km altitude and 57° inclination. The CRISTA-1 mission was conducted from 4–12 November 1994 (Offermann et al., 1999) and the CRISTA-2 mission from 8–16 August 1997 (Grossmann et al., 2002). The CRISTA instrument was mounted on the CRISTA Shuttle Pallet Satellite (SPAS) platform which operates at a distance of 20–100 km behind the shuttle. CRISTA was a limb scanning instrument which measured the thermal emission (4–71 µm) of 15 trace gases as well as of aerosols and clouds. CRISTA has a high spatial resolution in all three dimensions, with a resolution of typically 6° in longitude, 3° in latitude and 2 km vertical. The horizontal distance of two adjacent measurement points is about 650 km across the flight track and 200 to 400 km along the flight track. The latitudinal coverage of the observations was from 57° S to 67° N for the CRISTA-1 mission. CRISTA-2 measurements were performed with an increased latitudinal coverage ranging from 74° S to 74° N. Measurements were made for photochemically active gases as e.g. O_3, $ClONO_2$, HNO_3, NO_2, N_2O_5 as well as of long-lived trace gases as CFC-11, N_2O and CH_4 (Riese et al., 1997, 1999; Offermann et al., 1999). Here, we use CRISTA-1 version 3 and CRISTA-2 version 1 measurements of N_2O and O_3. The CRISTA-1 systematic and statistical errors for N_2O are 26 % and 3 %, respectively, and for O_3 10 % and 2 %, respectively, at 25 km. The CRISTA-2 s ystematic and statistical error for N_2O are 12 % and 5.3 %, respectively, and for O_3 10 % and 2.1 %, respectively, at 25 km. A description of the CRISTA error analyses can be found in Riese et al. (1999).

ACPD
12, 22629–22685, 2012

3 Model data

3.1 WACCM

The Whole Atmosphere Community Climate Model, Version 4 (WACCM 4) is a fully interactive chemistry climate model, where the radiatively active gases affect heating and cooling rates and therefore dynamics (Garcia et al., 2007). Recently, a new version of the WACCM 4 model has been developed that allows the model to be run with "external" specified dynamical (SD) fields (Lamarque et al., 2012). These meteorological fields come from the NASA Global Modeling and Assimilation Office (GMAO) Goddard Earth Observing System Model, Version 5 (GEOS–5). Here, temperature, zonal and meridional winds, and surface pressure are used to drive the physical parameterization that control boundary layer exchanges, advective and convective transport, and the hydrological cycle. In this study, the WACCM 4 meteorological fields are 'nudged' with the SD meteorological fields using the approach described in Kunz et al. (2011). The chemical module of WACCM 4 is based upon the 3-D chemical transport Model of OZone and Related Tracers, Version 3 (MOZART-3) (Kinnison et al., 2007). WACCM 4 includes a detailed representation of the chemical and physical processes in the troposphere through the lower thermosphere. The species included within this mechanism are contained within the O_x, NO_x, HO_x, ClO_x, and BrO_x chemical families, along with CH_4 and its degradation products. In addition, fourteen primary non–methane hydrocarbons and related oxygenated organic compounds are included (Emmons et al., 2010). This mechanism contains 122 species, more than 220 gas-phase reactions, 71 photolytic processes, and 18 heterogeneous reactions on multiple aerosol types. The SD-WACCM simulation employed here corresponds to the time period from 1 January 2005 through the end of 2010. For this simulation the model had a spin-up time from 1980 through 2003 using the fully interactive mode, i.e., WACCM without SD. On 1 January 2004 the model was switched to SD with the nudging approach as described in Kunz et al. (2011). The horizontal resolution is $1.9^\circ \times 2.5^\circ$ (210 × 270 km), with a vertical

ACPD
12, 22629–22685, 2012

resolution of <1 km in the troposphere, 1 km in the lower stratosphere, and ~2 km in the upper stratosphere.

4 Method

The method applied here has been put forward by Proffitt et al. (2003) based on balloon and aircraft data for the northern hemisphere and has been applied to satellite data and extended to southern hemisphere data by Khosrawi et al. (2004). The first application of the method to satellite data was done for the polar regions using ILAS and ILAS-II data (Khosrawi et al., 2004, 2006) and then extended to other latitude regions using Odin/SMR data (Khosrawi et al., 2008). In our latest study the monthly averages of N_2O and O_3 were applied for the evaluation of atmospheric chemical models in the tropics, midlatitudes and polar regions (Khosrawi et al., 2009). An assessment of the usage of different data sets as well as an assessment of different vertical resolutions used in the model simulations was given. Detailed descriptions of the method including discussions on the influence of limited sampling, the influence of diabatic descent on the N_2O/O_3 distribution can be found in the above references. Here, the method will be only briefly described.

4.1 General characteristics

The application of a modified form of tracer-tracer correlations of N_2O and O_3, namely by using the method as suggested by Proffitt et al. (2003) has the advantage that it helps separating O_3 variability due to latitudinal transport from photochemical changes. Monthly averages of N_2O and O_3 are calculated by binning the data by altitude or potential temperature and then averaging over a fixed interval of N_2O (20 ppbv). This is exemplary shown in Fig. 1 for the month August and November 2005 applying the method on measurements derived from the Atmospheric Chemistry Experiment Fourier Transform

ACPD
12, 22629–22685, 2012

Spectrometer (ACE-FTS) in the northern hemisphere.[1] First, the N_2O and O_3 is data is separated into potential temperature bins (here from 400 ± 25 K to 650 ± 25 K as given by the color coding in Fig. 1). The separation into potential temperature (or altitude) bins makes a typical feature of the N_2O/O_3 relationship visible, namely the decrease of N_2O and increase of O_3 with increasing potential temperature (or altitude). Second, N_2O and O_3 are averaged over 20 ppbv N_2O resulting in a set of curves that lie within these potential temperature bins.

Due to the limited spatial sampling of ACE-FTS caused by the applied measurement technique and chosen satellite orbit, a separation of the monthly averages of N_2O and O_3 derived from ACE-FTS into latitude regimes is not possible. Thus, measurements for the entire hemisphere are considered for describing our method. However, applying the May midlatitude (ATMOS Shuttle 1985), April high-latitude (ATMOS Shuttle 1993) as well as the November tropics (ATMOS Shuttle 1994) reference curves (Proffitt et al., 1990; Michelsen et al., 1998b), we can differentiate between air of tropical, midlatitude and polar character (Proffitt et al., 2003; Khosrawi et al., 2008). These reference curves were derived from the high resolution spectrometer ATMOS flown on space shuttle mission with durations of approximately two weeks each (Michelsen et al., 1998a). The ACE-FTS measurements in August 2005 are centered around the tropical reference curve indicating that the measurements were primarily performed in the tropics while in November the observations are centered around the midlatitude and polar reference curve indicating air of midlatitudinal and polar character as can be expected from the ACE-FTS coverage during these months.

The general characteristic of the families of curves derived from monthly averages of N_2O and O_3 binned by altitude or potential temperature in the polar regions is a positive

[1] ACE-FTS is a solar occultation instrument (Bernath et al., 2005; Boone et al., 2005). A seasonally varying coverage of the globe is provided, with an emphasis on midlatitudes and the polar regions. Here, we use ACE-FTS version 2.2. ACE-FTS measurements are used here since the method applied in this study can be much easier be visualized using data from a solar occultation instrument.

ACPD
12, 22629–22685, 2012

correlation (increasing N_2O with increasing O_3) at potential temperature levels above 500 ± 25 K and a negative correlation (decreasing N_2O with increasing O_3) at potential temperature levels below 500 ± 25 K (Fig. 1, right panel). The positive correlation at the levels above 500 ± 25 K is caused by diabatic descent of air from above the O_3 maximum (Proffitt et al., 2003; Khosrawi et al., 2009). At and below 500 ± 25 K the curves are influenced by a combination of diabatic descent and polar winter ozone loss. Descent at potential temperature levels $\leq 500 \pm 25$ K is visible in these curves as an extension of the curves to N_2O mixing ratios <50 ppbv and ozone loss by a change of slope (Fig. 1). However, as discussed by Khosrawi et al. (2008) diabatic descent is in the monthly averages to a certain extent masked by tropical ozone production when the entire hemisphere is considered. Thus, the separation in photochemical and dynamical processes can be most easily performed when only the polar regions are considered. As in Khosrawi et al. (2008) and Khosrawi et al. (2009) the monthly averages are separated into latitude regimes (tropics, midlatitudes and polar regions).

4.2 N_2O/O_3 distributions in the tropics

In this study, the method is applied to measurements and model simulations derived in the northern hemisphere tropics (0°–30° N) as previously done in Khosrawi et al. (2008, 2009). In the tropics, a flat to positive correlation is found at potential temperature levels above 700 ± 25 K (Khosrawi et al., 2008). The positive correlation is caused by the photochemical production of ozone in the tropics. The flat correlation below 700 ± 25 K is caused by the general distribution of N_2O and O_3. Both species have their maxima in the lower stratosphere and thus exhibit no latitudinal gradients in the tropics. At levels below 550 ± 25 K the tropical air is influenced by midlatitude air (Khosrawi et al., 2008). In fact, the tropics are not truly isolated from the midlatitudes and these are not truly isolated from the high latitudes as has been shown in previous studies (e.g., Randel et al., 1993; Proffitt et al., 2003; Konopka et al., 2009). The O_3/N_2O distribution in the tropics is similar in both hemispheres. Further, seasonal changes are small and equal in both hemispheres (Khosrawi et al., 2008).

ACPD
12, 22629–22685, 2012

In our recent model comparison (Khosrawi et al., 2009) we evaluated the Karlsruhe Simulation Model of the Middle Atmosphere (KASIMA, Ruhnke et al., 1999; Reddmann et al., 2001) as well as the atmospheric general circulation model ECHAM5/Messy1 (E5M1, now better known as EMAC (Jöckel et al., 2006)). Large differences between model simulations and observations were found in the tropics. The model evaluation was performed at 500 ± 25 K and 650 ± 25 K using Odin/SMR observations as reference. Figure 2 shows the comparison of the monthly averages of N_2O and O_3 derived from model simulations by KASIMA (blue) and E5M1 with T40L90MA resolution (green) with the averages derived from Odin/SMR (grey) observations for two potential temperature levels (500 ± 25 K and 650 ± 25 K) for January 2003 (NH, tropics).

A reasonable agreement was found at 500 ± 25 K for both models. At 500 ± 25 K in both models a steeper negative correlation as for the Odin/SMR observations was found which leads to higher O_3 mixing ratios at N_2O mixing ratios smaller than 250 ppbv and lower O_3 mixing ratios at N_2O mixing ratios larger than 250 ppbv. Therefore, differences between models and observations in averaged O_3 mixing ratios were changing from +40 % to −40 % (not shown). Indeed, it could be that the strong vertical ozone gradients in the tropical lower stratosphere are difficult to resolve with the relatively coarse altitude resolution of Odin/SMR O_3 observations (3 km). However, to assess the effect of this aspect we degraded as an exemplary test highly resolved SD-WACCM data onto the altitude resolution of the MIPAS O_3 observations (which is essentially the same as for Odin/SMR) using a larger set (more than 2000) of averaging kernel and a priori information from the MIPAS O_3 retrieval (see e.g. Connor et al. (1994); equation 4). This test revealed only a small difference between the original and degraded SD-WACCM O_3 data, i.e. 0.05 ppmv at 650 K and virtually zero at 500 K. Hence the altitude resolution of the satellite data is not the cause of the discrepancies between the model and satellite data. The differences could rather be due to the fact that N_2O and O_3 were retrieved with different altitude resolutions and that we use the relationship between these two species. On the other hand, transport processes in the tropical lower stratosphere are difficult to represent in models (e.g. Hegglin and Shepherd, 2007)

ACPD
12, 22629–22685, 2012

and vertical velocities are overestimated by models in the lower tropical stratosphere (Ploeger et al., 2010), so that the model vs Odin/SMR differences could also be partly due to model deficiencies (Khosrawi et al., 2009).

Though a satisfactory agreement between models and observations was found at 650 ± 25 K (differences generally within ±20 %) unusually (unrealistically) high N_2O mixing ratios ($N_2O > 320$ ppbv) were found in the Odin/SMR data that were not found in the model simulations. The reported statistical uncertainty of a single Odin/SMR data point on 650 ± 25 K level is of the order of the chosen N_2O bin size (1σ ~25 ppbv). This may lead to an artificial extension of the correlation curves at their ends since certain N_2O/O_3 pairs will then be sorted into the 320 to 340 ppbv bin. Further, the number of data points present in the bins for $N_2O > 320$ ppbv is rather small, i.e. typically only ~10 % of the data points that are found in other bins (Khosrawi et al., 2009). In this study we investigate if these high N_2O mixing ratios ($N_2O > 320$ ppbv) are caused by the influence of the statistical uncertainty on the N_2O/O_3 averages or by measurement uncertainties or if these values are caused by a physical process which is not well represented or difficult to simulate in the models as e.g. the QBO. We also investigated the cause of differences between models and measurements at 500 ± 25 K and assess the inter-annual variability of monthly averages of N_2O and O_3 as well as test their applicability for satellite-satellite inter-comparisons. For this purpose, we apply our method to measurements in the tropics derived from several satellite data sets as Odin/SMR, Aura/MLS and ENVISAT/MIPAS, CRISTA-1, CRISTA-2, as well as to model simulations from a further CCM, the SD-WACCM model.

5 Results

5.1 Assessment of the inter-annual variability

To assess the inter-annual variability of monthly averages of N_2O and O_3 we consider eight years of Odin/SMR (2003-2010) measurements, seven years of ENVISAT/MIPAS

ACPD
12, 22629–22685, 2012

measurements (2003-2009), seven years of Aura/MLS measurements (2004–2010) as well as 6 years of model simulations from SD-WACCM (2005–2010). Figure 3 shows the monthly averages of N_2O and O_3 derived from Odin/SMR for the northern hemisphere tropics (0-30° N) at 500 ± 25 K and 650 ± 25 K for the years 2003 to 2010. The figure shows that the inter-annual variability is low and can easily be distinguished from model deficiencies (see e.g. differences between model simulations and Odin/SMR observations in Fig. 2).

Small inter-annual variations in the monthly averages of N_2O and O_3 are seen as an extension of the curves in the N_2O space and slightly varying O_3 mixing ratios (max 1 ppmv) at both ends of the curves. The slight variations in O_3 at 650 ± 25 K for N_2O ≥ 300 ppbv are most likely caused by the quasi-biennial oscillation (QBO). In fact, the QBO dominates the variability of the equatorial stratosphere with a peak amplitude at around 25 km (Baldwin et al., 2001). The slight variations in O_3 at 500 ± 25 K for N_2O ≥ 300 ppbv are most likely caused by the inter-annual variability of tropical upwelling in connection with the prevailing QBO phase. There is a clear modulation of tropical ascent associated with the QBO (Punge et al., 2009). Upwelling is enhanced when the vertical wind shear caused by the QBO winds at a selected level is easterly but reduced or even turned to subsidence in westerly shear conditions at the equator (Plumb and Bell, 1982; Punge et al., 2009). Thus, higher N_2O values at e.g. 650 K can be expected during the QBO east phase. During the time period considered in this study the QBO was in its easterly phase in 2003, 2005, 2007 and 2009. The extension to $N_2O \leq$ 220 ppbv and $N_2O \leq$ 150 ppbv, respectively, and the variations in O_3 at these N_2O values can be attributed to air of mid-latitude origin (see reference curves in Fig. 1).

The monthly averages of N_2O and O_3 derived from Aura/MLS observations for the northern hemisphere midlatitude tropics are shown in Fig. 4. Note that 2005 is used here as reference since measurements of MLS started at the end of 2004. Aura/MLS is a microwave instrument like Odin/SMR and similar trace gas distributions are derived from both instruments (Barrett et al., 2006; Lambert et al., 2007). However, there are differences between Aura/MLS and Odin/SMR in the geographical and temporal

ACPD
12, 22629–22685, 2012

sampling as well as in the vertical resolution of the measurements. N_2O is measured by Odin/SMR with a vertical resolution of 1.5 km and O_3 with a vertical resolution of 2.5-3.5 km while N_2O is measured by Aura/MLS with a vertical resolution of 4–6 km and O_3 with a vertical resolution of 3 km (Table 1). The much lower standard deviations of monthly averages from Aura/MLS compared to the monthly averages derived from Odin/SMR are caused by five times higher number of observations and the coarser vertical resolution (Table 1) of the Aura/MLS measurements which results in a lower variability of the measured values.

Further, due to the coarser spatial resolution of Aura/MLS compared to Odin/SMR the inter-annual variability in the monthly averages of N_2O and O_3 is even lower. As for Odin/SMR the slight variations in O_3 at 650 ± 25 K at $N_2O \geq 300$ ppbv are most likely caused by the inter-annual variations in trace gas concentrations caused by the AO and QBO and the differences at 500 ± 25 K are most likely caused by different strengths in tropical upwelling from year to year (which however is also steered by the QBO). The QBO and tropical upwelling are connected (Punge et al., 2009) but the effect of the QBO is stronger at 650 K since this is the altitude where the QBO amplitude peaks and 500 K is a altitude where upwelling is most pronounced. Since these processes are coupled differences in upwelling may also cause differences in the QBO and vice versa (e.g. Punge et al., 2009).

The ENVISAT/MIPAS N_2O measurements have a better vertical resolution than the Aura/MLS observations, but not as good as the Odin/SMR observations of (Table 1). Further, the temporal resolution of the measurements from ENVISAT/MIPAS is not as high as from Aura/MLS and not as low as from Odin/SMR. Thus, the standard deviations of the monthly averages of N_2O and O_3 derived from ENVISAT/MIPAS are not as low as the ones derived from Aura/MLS and not as high as the ones derived from Odin/SMR (Note: these are not the sole reasons, instruments precision plays also a role). The monthly averages of N_2O and O_3 derived from ENVISAT/MIPAS for the years 2003 to 2010 are shown in Fig 5. As for Odin/SMR and Aura/MLS the inter-annual variability of monthly averages of N_2O and O_3 is also low for ENVISAT/MIPAS. A bias with

ACPD
12, 22629–22685, 2012

respect to the observed N_2O mixing ratios between the high spectral resolution (2003-2004) and the low spectral resolution observations (2005 onwards) at 650 ± 25 K is in some months clearly visible. The bias between these two data versions can be mainly attributed to the differences in the retrieval set-up. Slight variations in the monthly averages are caused as for Odin/SMR and Aura/MLS by air of midlatitude origin, the inter-annual variability in N_2O and O_3 most likely caused by the QBO and different strengths of tropical upwelling from year to year due to the prevailing QBO phase.

In addition to the consideration of seven to eight years of monthly averages of N_2O and O_3 derived from three different satellite data sets the inter-annual variability of these averages as derived from a Chemistry-Climate model is investigated. For the purpose of a model evaluation study it is important that not only the inter-annual variability in the satellite data sets is low but also that the inter-annual variability in the model simulations is low. The monthly averages of N_2O and O_3 derived from SD-WACCM for the years 2005 to 2010 are shown in Fig 6. The inter-annual variability in the SD-WACCM monthly averages of N_2O and O_3 is as low as for the monthly averages derived from the satellite data sets.

In the SD-WACCM simulation the QBO is realistically represented and arises solely from the nudging of the WACCM dynamics with GEOS5 meteorological fields. As in our recent model evaluation study the curves of monthly averages of N_2O and O_3 derived from SD-WACCM are at 500 ± 25 K steeper (but not as steep as E5M1 and KASIMA) than the ones derived from the satellite data which can most likely be attributed to a stronger tropical upwelling in the model simulation than observed. At 650 ± 25 K the slight variations form year to year in the monthly averages are caused most likely by the QBO. In the SD-WACCM data in April and extension of the curves to much lower N_2O values than in the monthly averages derived from the satellite observations are found which can be attributed to a stronger in-mixing of midlatitude air into the tropics by the model.

ACPD
12, 22629–22685, 2012

5.2 Observation of exceptionally high N_2O mixing ratios

5.2.1 Analysis of Odin/SMR observations

In the tropics, monthly averages of N_2O values at 650 ± 25 K were much higher derived from Odin/SMR observations (reaching up to 330 ppbv) than simulated by KASIMA and E5M1. These values are even higher than the highly accurate ground-based observations of N_2O (319 ppbv in 2005) derived in the troposphere (Forster et al., 2007). In the Odin/SMR data these high N_2O mixing ratios occur solely in the tropics and with a seasonal cycle. In the northern hemisphere a maximum is found during the winter months and a minimum during summer months (e.g. January and July 2003, respectively, see Supplement). This summer/winter variation is consistent with tropical upwelling which is stronger during northern hemisphere winter (e.g. Randel et al., 2007; Ploeger et al., 2010). Further, besides a seasonal variability in the observation of these high values we also found an inter-annual variability. N_2O mixing ratios exceeding 320 ppbv are found in January 2003, 2004, 2006 and 2010 but not in January 2005, 2007 and 2009, as well as in October 2003, 2006 and 2007 but not in October 2004, 2005 and 2008-2010 (Fig. 3 and 7).

The Odin/SMR N_2O anomaly fields (daily zonal-mean minus a multi-year zonal mean) for the tropics at latitudes between 10° N and 10° S are shown in Fig. 8 top and second panel. The N_2O anomaly fields derived from Odin/SMR show a clear signature of the quasi-biennial oscillation in the stratosphere. The positive anomalies propagate downward in the upper stratosphere with an interval of approximately two years. The QBO was in its westerly phase during 2002, 2004, 2006, 2008 and 2010 while the QBO was in its easterly phase during 2003, 2005, 2007 and 2009 (FU Berlin database http://www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo/ and Jin et al. (2009)).

The occurrence of the maxima and minima in the maximum N_2O mixing ratios of the averaged bins 650 ± 25 K (see Supplement) agree quite well with the positive and negative anomalies found at around 650 K in the N_2O anomaly field (Fig. 8 second panel). For example, distinct negative anomalies are found during summer 2008 and

ACPD
12, 22629–22685, 2012

2009 where we also find distinct minima in the maximum N_2O mixing ratios of the averaged bins. Further, the N_2O fields for the stratosphere (Fig. 8 third panel) show that due to a stronger upwelling N_2O was transported higher up in 2002, 2004, 2006, 2008 and 2010 which is in agreement with the maxima of the maximum N_2O mixing ratios of the averaged bins we found (Fig. 7 as well as Supplement).

5.2.2 Analysis of Aura/MLS, CRISTA and ENVISAT/MIPAS observations

The same data sets as in Sect. 5.1 are applied here plus additionally the CRISTA-1 and CRISTA-2 measurements. The CRISTA data sets have been included in this study though they only provide measurements for two weeks in November 1994 and August 1997, respectively. However, CRISTA-1 and CRISTA-2 is one of the few data sets having a very high temporal and spatial resolution in the tropics. Comparing the results from Odin/SMR with the results derived from Aura/MLS generally lower maximum N_2O mixing ratios are found in Aura/MLS than in Odin/SMR (Fig. 4 and 7 as well as tables in Supplement). A low bias of Aura/MLS N_2O relative to Odin/SMR N_2O has already been shown in recent validation studies (Barrett et al., 2006; Lambert et al., 2007). Barrett et al. (2006) found that maximum absolute differences between Aura/MLS and Odin/SMR N_2O can reach up to 20–30 ppbv.

Though the highest N_2O averages from Aura/MLS are ~20 ppbv lower than the monthly averages from Odin/SMR, a similar structure of higher N_2O values during winter months is found (see Fig. 7 and tables in Supplement). The summer minima, however, are not as strongly pronounced in the Aura/MLS data as in Odin/SMR data. The fact that lower N_2O averages than Odin/SMR (330 ppbv) are found in the Aura/MLS observation is likely caused due to the coarser vertical resolution of Aura/MLS as can be seen from Fig. 9. Figure 9 shows the Probability Density Functions (PDFs) calculated from Odin/SMR with the original Odin/SMR resolution of N_2O measurements (1.5 km) and for the Odin/SMR N_2O measurements smoothed to the Aura/MLS vertical resolution (4 km). The application of PDFs of long-lived tracers has been introduced by Sparling (2000) to quantify mixing regions and mixing barriers. The winter hemisphere PDF

ACPD
12, 22629–22685, 2012

has three modes while the summer hemisphere PDF has two modes. The peaks in the winter hemispheric PDF correspond to the polar vortex, mid-latitude surf-zone and tropics and the minima define the vortex edge and the subtropical boundary. The summer hemisphere PDF peaks correspond to the tropics and summer extra-tropics with a broader minimum between them corresponding to the subtropical barrier (Palazzi et al., 2011). As can be seen from the PDFs shown in Fig. 9, the high N_2O mixing ratios ($N_2O > 300$ ppbv) are smoothed out due to the coarser resolution of Aura/MLS. Further, small temporal and spatial differences in the structure of the SAO and QBO as observed by Odin/SMR and Aura/MLS were found by Jin et al. (2009).

For considering a data set which has a similar vertical resolution as Odin/SMR (1.5 km) we apply our method to the CRISTA-1 and CRISTA-2 observations (2 km) for November 1994 and August 1997, respectively. Though CRISTA-1 and CRISTA-2 provide only data for two week periods it is to our knowledge the only satellite instrument with such a high vertical and horizontal resolution in the tropics. Figure 10 shows the monthly averages of N_2O and O_3 derived from all CRISTA-1 measurements performed from 4 to 12 November 1994 at altitude levels between 25 and 45 km and derived from CRISTA-2 measurements from 8 to 16 August 1997 at altitudes between 20 and 50 km (NH, tropics). Although these measurements were performed several years earlier than the Odin/SMR measurements, also here N_2O mixing ratios up to 330 ppbv at 25 km (~650 K) are found that are in good agreement with the Odin/SMR monthly averages (CRISTA-1, November 1994), despite of the large systematic errors of CRISTA-1 (see Table 1). The mean tropospheric N_2O mixing ratio has increased during this time period from 314 ppmv in 1998 to 319 ppbv in 2005 (IPCC, 2007; Forster et al., 2007).

The ENVISAT/MIPAS N_2O measurements have a better vertical resolution (3–4 km) than the Aura/MLS observations (4–6 km), but not as good as the Odin/SMR observations (1.5–3 km). In 2003 and 2004, before the intermission in the ENVISAT/MIPAS operation, maximum N_2O mixing ratios of 330 ppbv are found between February and April. From 2005 onwards when ENVISAT/MIPAS continued its operation with a lower spectral resolution much lower N_2O mixing ratios (Fig 5 and 7) are found than in 2003

ACPD
12, 22629–22685, 2012

and 2004 (e.g. Palazzi et al., 2011). These low values are a result of the efforts to reduce the well-known bias in MIPAS N_2O observations of the period before. However, these values are also lower than the ones measured by Aura/MLS and Odin/SMR. As in the Odin/SMR monthly averages higher N_2O values are found in the ENVISAT/MIPAS monthly averages during the winter months. Especially, besides 2003 and 2004 between 2008 and 2010 higher N_2O values are found, thus a similar structure as in the Aura/MLS and SD-WACCM monthly averages (Sect. 5.2.3). Thus, from this satellite data comparison applying monthly averages of N_2O and O_3 a seasonal and inter-annual cycle in the occurrence of enhanced N_2O mixing ratios is consistently found in all data sets, however, more or less pronounced dependent on which data set is considered. We found that these values occur preferentially in the satellite data sets with high vertical resolution. However, the degradation of the Odin/SMR data onto the lower vertical resolution of the ENVISAT/MIPAS data set, using their averaging kernel and a priori information (Connor et al., 1994), resulted in only small differences in the order of a few ppbv between the highly resolved and degraded Odin/SMR data. Thus, the preferential occurrence of these high N_2O values in the data set with high vertical resolution must be a coincidence. Concerning the reliability of these values it is very likely that the absolute N_2O values exceeding 320 ppbv are caused due to some bias problems or instrument noise. To entirely understand the reason for occurrence of these high N_2O values in satellite measurements further studies are necessary which are beyond the scope of this study.

5.2.3 Analysis of SD-WACCM model simulations

Since these high N_2O mixing ratios in the tropical lower stratosphere were found while performing a model evaluation (Khosrawi et al., 2009) we also consider model simulations in this study. Due to the fact that these high values occur with a seasonal and inter-annual variability it was presumed that the QBO that is usually not well represented in many models (e.g. Giorgetta et al., 2006; SPARC CCMVal, 2010) could be

ACPD
12, 22629–22685, 2012

the cause for the occurrence of the high values. [1] In fact, in the simulation used for our recent model evaluation study (Khosrawi et al., 2009) the meteorological fields in the KASIMA simulation where nudged toward operational ECMWF analyses between 7 and 48 km. In the E5M1 low resolution simulation the QBO was nudged while in the high resolution E5M1 simulation the QBO is generated internally. We do not intend to perform an additional model evaluation study, thus, only simulations from one model are considered.

SD-WACCM simulations for the years 2005 to 2010 including a realistic representation of the QBO are taken into account (Fig 6). The simulation of a realistic QBO improves also the simulation of the tropical upwelling and the atmospheric tape recorder compared to a model without a QBO. Further, the SAO is only simulated realistically if the QBO is represented (Giorgetta et al., 2006). SD-WACCM N_2O mixing ratios are of a comparable value as the ENVISAT/MIPAS mixing ratios derived from the low spectral resolution observations (from 2005 onwards). Thus, SD-WACCM maximum N_2O mixing ratios are ~20 ppbv lower than Aura/MLS and Odin/SMR maximum N_2O mixing ratios.

Although the QBO has been fully considered in SD-WACCM and the model simulations were performed with a high vertical resolution in the lower stratosphere (1 km), no structure as pronounced as in the satellite data sets showing higher maximum N_2O mixing ratios of the averaged bins during winter months is found. However, higher values are found during Mai 2007, February 2009 and from January to June 2010. The QBO in SD-WACCM is realistically simulated and in good agreement with the QBO derived from Odin/SMR (not shown). However, the curves from SD-WACCM at 550 ± 25 K are not as steep as the ones derived from KASIMA and E5M1 and thus agree much better with the ones derived from the satellite data as will be discussed in the next section. Further, in SD-WACCM the QBO signal does not propagate as far down as in

[1] Note: As discussed in the previous section the absolute values are most likely caused by satellite biases and instrument noises, but the winter/summer variation on the occurrence of higher/lower N_2O values we could attribute to the QBO

ACPD
12, 22629–22685, 2012

in Odin/SMR. This may explain why we do not see the structure of higher N_2O values in winter than in summer as pronounced in SD-WACCM as in the satellite data.

5.3 Inter-comparison

For investigating if the monthly averages of N_2O and O_3 can not only be applied for model evaluations but also for satellite inter-comparison a comparison of Odin/SMR, Aura/MLS, ENVISAT/MIPAS and SD-WACCM averages derived for the tropics is performed. The comparison is performed for all years but is shown here for the years 2003 and 2009 (years with higher N_2O) and 2006 (years with lower N_2O). The QBO in 2003 and 2009 was in its easterly phase, while it was in its westerly phase in 2006.

In these data comparisons the Odin/SMR data is taken as the reference. In Fig. 11 the monthly averages derived from Odin/SMR and ENVISAT/MIPAS for the year 2003 are shown (the other data sets were not available for that year). ENVISAT/MIPAS O_3 mixing ratios are somewhat higher than Odin/SMR at 650 ± 25 K and somewhat lower than Odin/SMR at 500 ± 25 K. The O_3 differences between these two instruments are within ± 20 % (not shown). The differences between MIPAS and Odin/SMR O_3 are similar to the ones found in the validation study by Jones et al. (2007).

The curves of both data sets are very similar concerning their N_2O mixing ratios. However, Odin/SMR extends generally to a somewhat larger N_2O range at both potential temperature levels. High N_2O mixing ratios (330 ppbv) are found in the Odin/SMR monthly averages between January to March and October to December. In the ENVISAT/MIPAS monthly averaged values are as high as the ones from Odin/SMR are found between February and April (Fig. 11 as well as Fig. 7).

The comparison of Odin/SMR with ENVISAT/MIPAS, Aura/MLS and SD-WACCM for the year 2006 (Fig. 12) shows a good overall agreement at 500 ± 25 K. At 650 ± 25 K Odin/SMR O_3 mixing ratios seem to be somewhat lower (0.5-1-1 pmv), indicating a negative bias of Odin/SMR O_3 measurements compared to ENVISAT/MIPAS and Aura/MLS as described above and as reported in the validation studies by Barrett et al. (2006). N_2O mixing ratios as high as 330 ppbv are found in January and October in

ACPD
12, 22629–22685, 2012

the Odin/SMR data, but not in the other three data sets where maximum values range between 270 and 290 ppbv. Although the other data sets do not reach maximum N_2O mixing ratios as high as Odin/SMR still higher maximum N_2O mixing ratios than in the other months are found in January (except for SD-WACCM). The fact, that Aura/MLS N_2O is biased low by 20–30 ppbv between 68.1 and 10 hPa compared to Odin/SMR has already been shown by Barrett et al. (2006). However, the Odin/SMR N_2O single profile precision was estimated to be 10–30 ppbv and the estimated systematic error 12–25 ppbv (Urban et al., 2006; Lambert et al., 2007). Thus, the differences in the absolute values of the maximum mixing ratios may be caused by the instrument biases.

Another interesting feature in this comparison is that the curves at 500 ± 25 K from ENVISAT/MIPAS, Aura/MLS and SD-WACCM are not as flat as the ones derived from Odin/SMR. The flat N_2O/O_3 curve from Odin/SMR was, besides the overestimation of tropical upwelling by the models, the reason that large differences between models and observations were found at 500 ± 25 K in our recent model comparison (Khosrawi et al., 2009). Thus, if we would repeat our model comparison with ENVISAT/MIPAS or Aura/MLS and SD-WACCM we would not find these large discrepancies between model and measurements. The flat relationship in the Odin/SMR at 500 ± 25 K is probably caused by an underestimation of O_3 in the tropics and subtropics. Jones et al. (2007) found the largest systematic differences with Odin/SMR O_3 being lower than ozone sonde measurements at tropical latitudes below 35 km. An underestimation of Odin/SMR O_3 compared to the MIPAS ESA version 4.61 was found between 19 and 25 km. The underestimation of Odin/SMR O_3 compared to ozone sondes was found to be more than 1 ppmv between 20–25 km in the tropics (Jones et al., 2007).

The comparison of Odin/SMR with ENVISAT/MIPAS, Aura/MLS and SD-WACCM for the year 2009 (Fig. 13) shows a better agreement of the satellite data sets at 650 ± 25 K. However, somewhat larger differences than for 2009 are found for 2003 and 2006 are found at at 500 ± 25 K. Especially, the differences increase with increasing N_2O. Palazzi et al. (2011) derived PDFs from Odin/SMR, Aura/MLS and ENVISAT/MIPAS and found that the PDFs derived from these three instruments have

ACPD
12, 22629–22685, 2012

the same structure which can be expected for instruments which have a comparable coverage. The PDFs show a variation from year to year which is a function of the QBO phase (e.g Randel et al., 1998; Palazzi et al., 2011). The wintertime subtropical barrier is shifted toward the summer hemisphere when the QBO is in its westerly phase. In 2003 and 2009 the QBO was in its easterly phase and in its westerly phase in 2006. Figure 14 shows the PDFs derived from Odin/SMR at 650 K for the southern and northern hemisphere for 2003, 2006 and 2009. In 2006 when the QBO was in its westerly phase a smaller subtropical edge (minima in the PDF at ~200 ppbv) and broader surf-zone (peak in the PDF around 100-200 ppbv) accompanied with somewhat larger surf-zone and vortex peak values are found during northern hemisphere winter compared to 2003 and 2009 when the QBO was in its easterly phase. This results in N_2O/O_3 curves which span over a shorter N_2O range in 2006 when the QBO was in its westerly phase.

6 Conclusions

We applied a modified form of tracer-tracer correlations of N_2O and O_3 where the data is organized monthly for both hemispheres by partitioning the data into altitude or potential temperature bins and then averaging over a fixed interval of N_2O. By applying this method to satellite data it becomes a quite valuable tool for the evaluation of atmospheric chemical models as well as for satellite inter-comparisons. By applying eight years of Odin/SMR measurements (2003–2010), seven years of Aura/MLS measurements (2004–2010) and seven years of ENVISAT/MIPAS measurements (2003–2009) as well as six years of model simulations from SD-WACCM the inter-annual variability of monthly averages of N_2O and O_3 has been assessed. We found that the inter-annual variability is low and can easily be distinguished from model deficiencies.

In a recent model evaluation study where two CTMs and one CCM were evaluated (Khosrawi et al., 2009) large differences between model simulations and Odin/SMR observations in the northern and southern hemisphere tropics (0° to 30° N and 0° to

ACPD
12, 22629–22685, 2012

−30° S, respectively) were found. The model evaluation was performed for the potential temperature levels of 500 ± 25 K and 650 ± 25 K. At 500 ± 25 K in both models a steeper negative correlation (decreasing O_3 with increasing N_2O) was found than in the observations. In Khosrawi et al. (2009) these differences were explained to be probably partly caused by the large vertical O_3 gradients occurring in the tropics which cannot entirely be resolved by the rather coarse vertical resolution of Odin/SMR of 3 km and partly by inaccuracies in the model simulations of transport in the tropical stratosphere. While it could be shown here that the latter reason is applicable, we found that the vertical resolution itself is not a reason for the differences between model and observations. We attribute the steeper correlation in the model simulations to a incorrect simulation of tropical upwelling which is due to a missing or incorrect simulation of the QBO. In fact, the meteorological fields in the KASIMA simulation were nudged toward operational ECMWF analyses for generating the QBO in the simulation. In the E5M1 low resolution simulation the QBO was nudged. Though in the E5M1 high resolution simulation the QBO was generated internally and a realistic QBO was simulated the modelled QBO did not always represent the observed QBO phase (Jöckel et al., 2006). As we have shown in this study applying model simulations of SD-WACCM with a high vertical resolution and thus a realistic representation of the QBO results in smaller differences between model and measurements. However, an underestimation of O_3 from Odin/SMR found by Jones et al. (2007) could be the reason for the rather flat correlation of the monthly N_2O/O_3 averages that contributed to the large differences found in Khosrawi et al. (2009) between models and satellite data.

The N_2O monthly averages derived from Odin/SMR observations at the potential temperature level of 650 ± 25 K were much higher (20 to 40 ppbv) than the N_2O values that were derived from the model simulations. Further, these values were also higher than the highly accurate ground-based measurements of N_2O derived in the troposphere which is the only source for this trace gas (330 ppbv compared to the tropospheric average of 319 ppbv ± 0.12 ppbv in 2005 Forster et al., 2007). However, the bias we found in our monthly averages is much higher than the differences to the

ACPD
12, 22629–22685, 2012

measured ground-based N_2O mixing ratios (20-40 ppbv between model and satellite measurements compared to 10 ppbv between satellite and ground-based measurements). Such a high positive bias was not found in validation studies performed applying Odin/SMR N_2O observations and thus this difference is probably caused by the combination of too low N_2O values derived from the model simulation as well as somewhat too high N_2O mixing ratios measured by Odin/SMR.

Since in our method the data is averaged over bins of fixed N_2O we found that the maximum bins with $N_2O \geq 330$ ppbv contain a relatively low number of data points compared to other bins. Further, these values occur with a seasonal dependence showing a maximum in winter and a minimum in summer. Furthermore, by considering eight years (2003–2010) of data it was found that these high values occur also with an inter-annual variability. We found that the exceptionally high values of N_2O averages found in Odin/SMR are also found in the monthly averages derived from CRISTA and ENVISAT/MIPAS which like Odin/SMR provide N_2O measurements with a high vertical resolution.

Thus, the exceptionally high absolute values of N_2O found in the data of instruments with high vertical resolution might be related to some (partly well-known) bias problems or instrument noise. However, the degradation of the Odin/SMR data onto the lower vertical resolution of the ENVISAT/MIPAS data set, using their averaging kernel and a priori information, resulted in only small differences in the order of a few ppbv between the highly resolved and degraded Odin/SMR data. Thus, the preferential occurrence of these high N_2O values in the data set with high vertical resolution must be a coincidence. However, the seasonal and inter-annual variability of these values is most probably caused by local dynamical processes in the tropics as in particular the seasonal cycle of tropical upwelling and the quasi-biennial oscillation (QBO), where the latter usually is not well represented in model simulations. The monthly averages of N_2O and O_3 derived from Odin/SMR, Aura/MLS, ENVISAT/MIPAS and SD-WACCM were compared with each other for the years 2003, 2006 and 2009. The comparison showed that these data sets are generally in good agreement but that also known

ACPD
12, 22629–22685, 2012

biases of the satellite data sets are clearly visible in the monthly averages, thus showing that this method is not only a valuable tool for model evaluation but also for satellite inter-comparisons.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/12/22629/2012/acpd-12-22629-2012-supplement.pdf.

Acknowledgements. We are grateful to the European Space Agency (ESA) for providing the Odin/SMR data. Odin is a Swedish led project funded jointly by Sweden (SNSB), Canada (CSA), Finland (TEKES), and France (CNES) and the European Space Agency (ESA). We would like to thank the ACE-FTS team for providing their data. Funding for the ACE mission is provided by the Canadian Space Agency (CSA) and the Natural Sciences and Engineering Research Council of Canada (NSERC). We also would like to thank the MLS team for providing their data. MLS data were obtained from the NASA Goddard Earth Sciences Data and Information Center. The WACCM modeling work was conducted at the National Center for Atmospheric Research (NCAR) and sponsored by the U.S. National Science foundation. We would like to thank the UK Met Office (UKMO) and European Centre for Medium-Range Weather Forecasts (ECMWF) for providing their meteorological analyses and the National Computer Center (NSC) Linköping, Sweden for providing computer resources. We also would like to acknowledge P. Braesicke and R. Hommel for helpful discussions, J.-U. Grooß for helpful discussions and help with the processing of the Odin/SMR and Aura/MLS data as well as N. Thomas for her help with processing the UKMO data. We are also grateful to the Swedish Research Council and the Swedish Space Board for funding F. Khosrawi.

ACPD
12, 22629–22685, 2012

References

Baldwin, M. P., Gray, L. J., Dunkerton, T. J., Hamilton, K., Haynes, P. H., Randel, W. J., Holton, J. R., Alexander, M. J., Hirota, I., Horinouchi, T., Jones, D. B. A., Kinnesley, J. S., Marquardt, C., Sato, K., and Takahashi, M.: The quasi-biennal oscillation, Rev. Geophys., 39, 179–229, 2001. 22633, 22634, 22647

Barrett, B., Ricaud, P., Santee, M. L., Attié, J.-L., Urban, J., Le Flochmoën, E., Berthet, G., Murtagh, D. P., Eriksson, P., Jones, A., de La Noë, J., Dupuy, E., Froidevaux, L., Livsey, N. J., Waters, J. W., and Filipiak, M. J., Intercomparison of trace gas profiles from the Odin/SMR and AURA/MLS limb sounders, J. Geophys. Res., 111, doi:10.1029/2006JD007305, 2006. 22635, 22647, 22651, 22655, 22656

Bernath, P. F., McElroy, C. T., Abrams, M. C., Boone, C. D., Butler, M., Camy-Peyret, C., Carleer, M., Clerbaux, C., Coheur, P.-F., Colin, R., DeCola, P., DeMaziére, M., Drummond, J. R., Dufour, D., Evans, W. F. J., Fast, H., Fussen, D., Gilbert, K., Jennings, D. E., Llewellyn, E. J., Lowe, R. P., Mahieu, E., McConnell, J. C., McHugh, M., McLeod, S. D., Michaud, R., Midwinter, C., Nassar, R., Nichitiu, F., Nowlan, C., Rinsland, C. P., Rochon, Y. J., Rowlands, N., Semeniuk, K., Simon, P., Skelton, R., Sloan, J. J., Soucy, M.-A., Strong, K., Tremblay, P., Turnbull, D., Walker, K. A., Walkty, I., Wardle, D. A., Wehrle, V., Zander, R., and Zou, J.: Atmospheric Chemistry Experiment (ACE): Mission overview, Geophys. Res. Lett., 32, L15S01, doi:10.1029/2005GL022368, 2005. 22643

Boone, C. D., Nassar, R., Walker, K. A., Rochon, Y., McLeod, S. D., Rinsland, C. P., and Bernath, P. F.: Retrievals for the Atmospheric Chemistry Experiment Fourier Transform Spectrometer, Appl. Optics, 44, 7218–7231, 2005. 22643

Brewer, A. W.: Evidence for a world circulation provided by the measurements of helium and water vapor distribution in the stratosphere, Quart. J. R. Meteorol. Soc., 75, 351–363, 1949. 22632, 22633

Connor, B. J., Siskind, D. E., Tsou, J. J., Parrish, A., and Remsberg, E. E.: Ground-based microwave observations of ozone in the upper stratosphere and mesosphere, J. Geophys. Res., 99, 16757–16770, 1994. 22645, 22653

Dobson, G. M. B.: Origin and distribution of polyatomic molecules in the atmosphere, Proc. R. Soc. London, Ser. A, 236, 187–193, 1956. 22632, 22633

Dobson, G. M. B., Harrison, D. N., and Lawrence, J.: The laminated structure of ozone in the atmosphere, Q. J. R. Meteorol. Soc., 99, 599–607, 1973. 22633

ACPD
12, 22629–22685, 2012

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010. 22641

Fischer, H. and Oelhaf, H.: Remote sensing of vertical profiles of atmospheric trace constituents with MIPAS limb-emission spectrometers, Appl. Optics, 35, 2787–2796, 1996. 22639

Forster, P. M., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and Van Dorland, R., Changes in Atmospheric Constituents and in Radiative Forcing, in Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by S. Solomon, D. Quin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor, and H. L. Miller, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2007. 22632, 22650, 22652, 22658

Frisk, U., Gastrom, M., Ala-Laurinaho, J., Andersson, S., Berges, J. C., Chabaud, J. P., Dahlgren, M., Emrich, A., Florin, G., Fredrixon, M., Gaier, T., Haas, R., Hjalmarsson, T. H. A., Jakobsson, B., Jukkala, P., Kildal, P. S., Kollberg, E., Lecacheux, J. L. A., Lehikoinen, P., Lehto, A., Mallat, J., Marty, C., Michet, D., Narbonne, J., Nexon, M., Olberg, M., Olofsson, A. O. H., Olofsson, G., Origne, A., Petersson, M., Piirone, P., Pouliquen, D., Ristorcelli, I., Rosolen, C., Rouaix, G., Raisanen, A. V., Serra, G., Sjoberg, F., Stenmark, L., Torchinsky, S., Tuovinen, J., Ullberg, C., Vinterhav, E., Wadefalk, N., Zirath, H., Zimmermann, P., and Zimmermann, R.: The Odin satellite: I. Radiometer design and test, Astron. Astrophys., 403, 27–34, 2003. 22636

Froidevaux, L., Jiang, Y. B., Lambert, A., Livesey, N. J., Read, W. G., Waters, J. W., Browell, E. V., Hair, J. W., Avery, M. A., McGee, T. J., Twigg, L. W., Sumnicht, G. K., Jucks, K. W., Margitan, J. J., Sen, B., Stachnik, R. A., Toon, G. C., Bernath, P. F., Boone, C. D., Walker, K. A., Filipiak, M. J., Harwood, R. S., Fuller, R. A., Manney, G. L., Schwartz, M. J., Daffer, W. H., Drouin, B. J., Cofield, R. E., Cuddy, D. T., Jarnot, R. F., Knosp, B. W., Perun, V. S., Snyder, W. V., Stek, P. C., Thurstans, R. P., and Wagner, P. A., Validation of Aura Microwave Limb Sounder stratospheric ozone measurements, J. Geophys. Res., 113, D15S20, doi:10.1029/2007JD008771, 2008. 22638

ACPD
12, 22629–22685, 2012

Garcia, R. R., Marsh, D., Kinnison, D. E., Boville, B., and Sassi, F.: Simulations of secular trends in the middle atmosphere, 1950–2003, J. Geophys. Res., 112, D09301, doi:10.1029/2006JD007485, 2007. 22641

Giorgetta, M. A., Manzini, E., Roeckner, E., Esch, M., and Bengtsson, L., Climatology and forcing of the quasi-biennial oscillation in the MAECHAM5 model, J. Clim., 19, 3882–3901, 2006. 22634, 22653, 22654

Glatthor, N., von Clarmann, T., Fischer, H., Funke, B., Grabowski, U., Höpfner, M., Kellmann, S., Kiefer, M., Linden, A., Milz, M., Steck, T., Stiller, G. P.: Mengistu Tsidu, G., and Wang, D.-Y., Mixing processes during the Antarctic vortex split in September-October 2002 as inferred from source gas and ozone distributions from ENVISAT-MIPAS, J. Atmos. Sci, 62, 787–800, 2005. 22639

Glatthor, N., von Clarmann, T., Fischer, H., Funke, B., Gil-López, S., Grabowski, U., Höpfner, M., Kellmann, S., Linden, A., López-Puertas, M., Mengistu Tsidu, G., Milz, M., Steck, T., Stiller, G. P., and Wang, D.-Y.: Retrieval of stratospheric ozone profiles from MIPAS/ENVISAT limb emission spectra: a sensitivity study, Atmos. Chem. Phys., 6, 2767–2781, doi:10.5194/acp-6-2767-2006, 2006. 22639

Griesfeller, A., von Clarmann, T., Griesfeller, J., Höpfner, M., Milz, M., Nakajima, H., Steck, T., Sugita, T., Tanaka, T., and Yokota, T.: Intercomparison of ILAS-II version 1.4 and version 2 target parameters with MIPAS-Envisat measurements, Atmos. Chem. Phys., 8, 825–843, doi:10.5194/acp-8-825-2008, 2008. 22635

Grossmann, K. U., Offermann, D., Gusev, O., Oberheide, J., Riese, M., and Spang, R.: The CRISTA-2 Mission, J. Geophys. Res., 107, 8173, doi:10.1029/2001JD000667, 2002. 22640

Hegglin, M. I. and Shepherd, T. G., O_3-N_2O correlations from Atmospheric Chemistry Experiment: Revisiting a diagnostic of transport and chemistry in the stratosphere, J. Geophys. Res., 112, D19301, doi:10.1029/2006JD008281, 2007. 22635, 22645

Holton, J. R. and Lindzen, R. S.: An updated theory for the quasi-biennal cycle of the tropical stratosphere, J. Atmos. Sci., 29, 1076–1080, 1972. 22634

IPCC, Intergovernmental Panel on Climate Change: Climate Change 2007, Cambridge University Press, Cambridge, UK, 2007. 22652

Jégou, F., Urban, J., de La Noë, J., Ricaud, P., Le Flochmoën, E., Murtagh, D. P., Eriksson, P., Jones, A., Petelina, S., Llewellyn, E. J., Lloyd, N. D., Haley, C., Lumpe, J., Randall, C., Bevilacqua, R. M., Catoire, V., Huret, N., Berthet, G., Renard, J. B., Strong, K., Davies, J., Mc Elroy, C. T., Goutail, F., and Pommereau, J. P.: Technical Note: Validation

ACPD
12, 22629–22685, 2012

of Odin/SMR limb observations of ozone, comparisons with OSIRIS, POAM III, ground-based and balloon-borne instruments, Atmos. Chem. Phys., 8, 3385–3409, doi:10.5194/acp-8-3385-2008, 2008. 22637

Jiang, Y. B., Froidevaux, L., Lambert, A., Livesey, N. J., Read, W. G., Waters, J. W., Bojkov, B., Leblanc, T., McDermid, I. S., Godin-Beekmann, S., Filipiak, M. J., Harwood, R. S., Fuller, R. A., Daffer, W. H., Drouin, B. J., Cofield, R. E., Cuddy, D. T., Jarnot, R. F., Knosp, B. W., Perun, V. S., Schwartz, M. J., Snyder, W. V., Stek, P. C., Thurstans, R. P., Wagner, P. A., Allaart, M., Andersen, S. B., Bodeker, G., Calpini, B., Claude, H., Coetzee, G., Davies, J., De Backer, H., Dier, H., Fujiwara, M., Johnson, B., Kelder, H., Leme, N. P., Konig-Langlo, G., Kyro, E., Laneve, G., Fook, L. S., Merrill, J., Morris, G., Newchurch, M., Oltmans, S., Parrondos, M. C., Posny, F., Schmidlin, F., Skrivankova, P., Stubi, R., Thompson, D. T. A., Thouret, V., Viatte, P., Vömel, H., von Der Gathen, P., Yela, M., and Zablocki, G., Validation of Aura Microwave Limb Sounder Ozone by ozonesonde and lidar measurements, J. Geophys. Res., 112, D24S34, doi:10.1029/2007JD008776, 2007. 22638

Jin, J. J., Semeniuk, K., Beagley, S. R., Fomichev, V. I., Jonsson, A. I., McConnell, J. C., Urban, J., Murtagh, D., Manney, G. L., Boone, C. D., Bernath, P. F., Walker, K. A., Barret, B., Ricaud, P., and Dupuy, E.: Comparison of CMAM simulations of carbon monoxide (CO), nitrous oxide (N_2O), and methane (CH4) with observations from Odin/SMR, ACE-FTS, and Aura/MLS, Atmos. Chem. Phys., 9, 3233–3252, doi:10.5194/acp-9-3233-2009, 2009. 22633, 22650, 22652

Jöckel, P., Tost, H., Pozzer, A., Brühl, C., Buchholz, J., Ganzeveld, L., Hoor, P., Kerkweg, A., Lawrence, M. G., Sander, R., Steil, B., Stiller, G., Tanarhte, M., Taraborrelli, D., van Aardenne, J., and Lelieveld, J.: The atmospheric chemistry general circulation model ECHAM5/MESSy1: consistent simulation of ozone from the surface to the mesosphere, Atmos. Chem. Phys., 6, 5067–5104, doi:10.5194/acp-6-5067-2006, 2006. 22645, 22658

Jones, A., Murtagh, D., Urban, J., Eriksson, P., and Rösevall, J.: Intercomparison of Odin/SMR ozone measurements with MIPAS and ballon sonde data, Can. J. Phys., 85, 1111–11 123, 2007. 22637, 22655, 22656, 22658

Khosrawi, F., Müller, R., Proffitt, M. H., and Nakajima, H., Monthly averaged ozone and nitrous oxide from the Improved Limb Atmospheric Spectrometer (ILAS) in the Northern and Southern Hemisphere polar regions, J. Geophys. Res., 109, D10301, doi:10.1029/2003JD004,365, 2004. 22634, 22642

ACPD
12, 22629–22685, 2012

Khosrawi, F., Müller, R., Proffitt, M. H., and Nakajima, H.: Monthly averages of nitrous oxide and ozone for the northern and southern hemisphere high latitudes: A "1-year climatology" derived from ILAS/ILAS-II observations, J. Geophys. Res., 111, D11S11, doi:10.1029/2005JD006,384, 2006. 22634, 22642

Khosrawi, F., Müller, R., Proffitt, M. H., Urban, J., Murtagh, D. P., Ruhnke, R., Grooß, J.-U., and Nakajima, H., Seasonal cycle of averages of nitrous oxide and ozone in the Northern and Southern Hemisphere polar, midlatitude and tropical regions derived from ILAS/ILAS-II and Odin/SMR observations, J. Geophys. Res., 113, D18305, doi:10.1029/2007JD009,556., 2008. 22634, 22642, 22643, 22644

Khosrawi, F., Müller, R., Proffitt, M. H., Ruhnke, R., Kirner, O., Jöckel, P., Grooß, J.-U., Urban, J., Murtagh, D., and Nakajima, H.: Evaluation of CLaMS, KASIMA and ECHAM5/MESSy1 simulations in the lower stratosphere using observations of Odin/SMR and ILAS/ILAS-II, Atmos. Chem. Phys., 9, 5759–5783, doi:10.5194/acp-9-5759-2009, 2009. 22634, 22635, 22636, 22642, 22644, 22645, 22646, 22653, 22654, 22656, 22657, 22658

Kinnison, D. E., Brasseur, G. P., Walters, S., Garcia, R. R., Sassi, F., Boville, B. A., Marsh, D., Harvey, L., Randall, C., Randel, W., Lamarque, J. F., Emmons, L. K., Hess, P., Orlando, J., Tyndall, J., and Pan, L.: Sensitivity of chemical tracers to meteorological parameters in the MOZART-3 chemical transport model, J. Geophys. Res., 112, D20302, doi:10.1029/2006JD007879, 2007. 22641

Konopka, P., Grooß, J.-U., Plöger, F., and Müller, R.: Annual cycle of horizontal in-mixing into the lower tropical stratosphere, J. Geophys. Res., 114, D19111, doi:10.1029/2009JD011955, 2009. 22644

Kunz, A., Pan, L., Konopka, P., Kinnison, D., and Tilmes, S., Chemical and dynamical discontinuity at the extratropoical tropopause based on START08 and WACCM analysis, J. Geophys. Res., 116, D24 302, doi:10.1029/2011JD016,686, 2011. 22641

Lamarque, J.-F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E. A., Lauritzen, P. H., Neu, J., Orlando, J. J., Rasch, P., and Tyndall, G.: CAM-chem: description and evaluation of interactive atmospheric chemistry in CESM, Geosci. Model Dev., 5, 369–411, doi:10.5194/gmd–5–369–2012, 2012. 22641

Lambert, A., Read, W. G., Livesey, N. J., Santee, M. L., Manney, G. L., Froidevaux, L., Wu, D. L., Schwartz, M. J., Pumphrey, H. C., Jimenez, C., Nedoluha, G. E., Cofield, R. E., Cuddy, D. T., Daffer, W. H., Drouin, B. J., Fuller, R. A., Jarnot, R. F., Knosp, B. W., Pickett, H. M., Perun, V. S., Snyder, W. V., Stek, P. C., Thurstans, R. P., Wagner, P. A., Waters, J. W., Jucks,

ACPD
12, 22629–22685, 2012

K. W., Toon, G. C., Stachnik, R. A., Bernath, P. F., Boone, C. D., Walker, K. A., Urban, J., Murtagh, D., Elkins, J. W., and Atlas, E., Validation of the Aura Microwave Limb Sounder middle atmosphere water vapor and nitrous oxide measurements, J. Geophys. Res., 112, D24S36, doi:10.1029/2007JD008,724, 2007. 22635, 22638, 22647, 22651, 22656

Livesey, N. J., Filipiak, M. J., Froidevaux, L., Read, W. G., Lambert, A., Santee, M. L., Jiang, J. H., Pumphrey, H. C., Waters, J. W., Cofield, R. E., Cuddy, D. T., Daffer, W. H., Drouin, B. J., Fuller, R. A., Jarnot, R. F., Jiang, Y. B., Knosp, B. W., Li, Q. B., Perun, V. S., Schwartz, M. J., Snyder, W. V., Stek, P. C., Thurstans, R. P., Wagner, P. A., Avery, M., Browell, E. V., Cammas, J.-P., Christensen, L. E., Diskin, G. S., Gao, R.-S., Jost, H.-J., Loewenstein, M., Lopez, J. D., Nedelec, P., Osterman, G. B., Sachse, G. W., and Webster, C. R., Validation of Aura microwave limb sounder O_3 and CO observations in the upper troposphere and lower stratosphere, J. Geophys. Res., 113, D15S02, doi:10.1029/2007JD008,805, 2008. 22638

Llewellyn, E. J., Lloyd, N. D., Degenstein, D. A., Gattinger, R. L., Petelina, S. V., Bourassa, A. E., Wiensz, J. T., Ivanov, E. V., Dade, I. C. M., Solhem, B. H., Connell, J. C. M., Haley, J. C., von Savigny, C., and et al., The OSIRIS instrument on the Odin spacecraft, Can. J. Phys., 82, 411–422, 2004. 22636

Lossow, S., Urban, J., Gumbel, J., Eriksson, P., and Murtagh, D.: Observations of the mesospheric semi-annual oscillation (MSAO) in water vapour by Odin/SMR, Atmos. Chem. Phys., 8, 6527–6540, doi:10.5194/acp-8-6527-2008, 2008. 22633

Michelsen, H. A., Manney, G. L., Gunson, M. R., Rinsland, C. P., and Zander, R.: Correlations of stratospheric abundances of CH_4 and N_2O derived from ATMOS measurements, Geophys. Res. Lett., 25, 2777–2780, 1998a. 22643

Michelsen, H. A., Manney, G. L., Gunson, M. R., and Zander, R.: Correlations of stratospheric abundances of NO_y, O_3, N_2O, and CH_4 derived from ATMOS measurements, J. Geophys. Res., 103, 28347–28359, 1998b. 22643, 22672

Mote, P. W., Rosenlof, K. H., McIntyre, M. E., Carr, E. S., Gille, J. G., Holton, J. R., Kinnersley, J. S., Pumphrey, H. C., Russell III, J. M., and Waters, J. W.: An atmospheric tape recorder: The imprint of tropical tropopause temperatures on stratospheric water vapor, J. Geophys. Res., 101, 3989 – 4006, 1996. 22634

Murtagh, D., Frisk, U., Merino, F., Ridal, M., Jonsson, A., Stegman, J., Witt, G., Eriksson, P., Jimenez, C., Megie, G., de la Noe, J., Ricaud, P., Baron, P., Pardo, J. R., Hauchcorne, A., Llewellyn, E. J., Degenstein, D. A., Gattinger, R. L., Lloyd, N. D., Evans, W. F. J., McDade, I. C., Haley, C. S., Sioris, C., von Savigny, C., Solheim, B. H., McConnell, J. C., Strong,

ACPD
12, 22629–22685, 2012

K., Richardson, E. H., Leppelmeier, G. W., Kyrola, E., Auvinen, H., and Oikarinen, L.: An overview of the Odin atmospheric mission, Can. J. Phys., 80, 309–319, 2002. 22637
Offermann, D., Grossmann, K.-U., Barthol, P., Knieling, P., Riese, M., and Trant, R., Cryogenic Infrared Spectrometers and Telescopes for the Atmosphere (CRISTA) experiment and middle atmosphere variability, J. Geophys. Res., 104, 16,311–16,325, 1999. 22640
O'Sullivan, D. and Dunkerton, T. J.: The influence of the quasi-biennal oscillation on global constituent distributions, J. Geophys. Res., 102, 21,731–21,743, 1997. 22633
Palazzi, E., Fierli, F., Stiller, G. P., and Urban, J.: Probability density functions of long-lived tracer observations from satellite in the subtropical barrier region: data intercomparison, Atmos. Chem. Phys. Discuss., 11, 18385–18432, doi:10.5194/acpd-11-18385-2011, 2011. 22652, 22653, 22656, 22657
Payan, S., Camy-Peyret, C., Oelhaf, H., Wetzel, G., Maucher, G., Keim, C., Pirre, M., Huret, N., Engel, A., Volk, M. C., Kuellmann, H., Kuttippurath, J., Cortesi, U., Bianchini, G., Mencaraglia, F., Raspollini, P., Redaelli, G., Vigouroux, C., De Mazière, M., Mikuteit, S., Blumenstock, T., Velazco, V., Notholt, J., Mahieu, E., Duchatelet, P., Smale, D., Wood, S., Jones, N., Piccolo, C., Payne, V., Bracher, A., Glatthor, N., Stiller, G., Grunow, K., Jeseck, P., Te, Y., and Butz, A.: Validation of version-4.61 methane and nitrous oxide observed by MIPAS, Atmos. Chem. Phys., 9, 413–442, doi:10.5194/acp-9-413-2009, 2009. 22635, 22639
Perliski, L. S., Solomon, S., and London, J., On the interpretation of seasonal variations of stratospheric ozone, Planet. Space Sci., 37, 1527–1538, 1989. 22633
Ploeger, F., Konopka, P., Günther, G., Grooß, J.-U., and Müller, R., Impact of the vertical velocity scheme on modelling transport in the tropical tropopause layer, J. Geophys. Res., 115, D03301, doi:10.1029/2009JD012,023, 2010. 22646, 22650
Plumb, R. A. and Bell, R. C.: A model of the quasi-biennal oscillation on an equatorial beta-plane, Q. J. Meteorol. Soc., 108, 335–352, 1982. 22647
Proffitt, M. H., Margitan, J. J., Kelly, K. K., Loewenstein, M., Podolske, J. R., and Chan, K. R.: Ozone loss in the Arctic polar vortex inferred from high altitude aircraft measurements, Nature, 347, 31–36, 1990. 22643, 22672
Proffitt, M. H., Aikin, K., Tuck, A. F., Margitan, J. J., Webster, C. R., Toon, G. C., and Elkins, J. W.: Seasonally averaged ozone and nitrous oxide in the Northern Hemisphere lower stratosphere, J. Geophys. Res., 108, 4110, doi:10.1029/2002JD002657, 2003. 22633, 22634, 22635, 22642, 22643, 22644

ACPD
12, 22629–22685, 2012

Punge, H. J., Konopka, P., Giorgetta, M. A., and Müller, R.: Effects of the quasi-biennal oscillation on low-latitude transport in the stratosphere derived from trajectory calculations, J. Geophys. Res., 114, D03102, doi:10.1029/2008JD010518, 2009. 22647, 22648

Randel, W. J., Gille, J. C., Roche, A. E., Kumer, J. B., Mergenthaler, J. L., Waters, J. W., Fishbein, E. F., and Lahoz, W. A.: Stratospheric transport from the tropics to middle latitudes by planetary-wave mixing, Nature, 365, 533–535, 1993. 22644

Randel, W. J., Boville, B. A., and Gille, J. C.: Simulation of stratospheric N_2O in the NCAR CCM2: comparison with CLAES data and global budget analyses, J. Atmos. Sci, 51, 2834–2845, 1994. 22633

Randel, W. J., Wu, F., Russell III, J. R., Roche, A., and Waters, J. W.: Seasonal cycles and QBO variations in stratospheric CH_4 and H_2O observed in UARS HALOE data, J. Atmos. Sci., 55, 163–185, 1998. 22633, 22657

Randel, W. J., Park, M., Wu, F., and Livesey, N.: A large annual cycle in ozone above the tropical tropopause linked to the Brewer-Dobson circulation, J. Atmos. Sci., 64, 4479–4488, 2007. 22650

Reddmann, T., Ruhnke, R., and Kouker, W.: Three-dimensional model simulations of SF_6 with mesospheric chemistry, J. Geophys. Res., 106, 14525–14537, 2001. 22645

Ricaud, P., Pommereau, J.-P., Attié, J.-L., Le Flochmoen, E., El Amraoui, Ricaud, P., Pommereau, J.-P., Attié, J.-L., Le Flochmoën, E., El Amraoui, L., Teyssèdre, H., Peuch, V.-H., Feng, W., and Chipperfield, M. P.: Equatorial transport as diagnosed from nitrous oxide variability, Atmos. Chem. Phys., 9, 8173–8188, doi:10.5194/acp-9-8173-2009, 2009. 22632, 22633

Riese, M., Preusse, P., Spang, R., Ern, M., Jarisch, M., Grossmann, K., and Offermann, D.: Measurements of trace gases by the cryogenic infrared spectrometers and telescopes for the atmosphere (CRISTA) experiment, Adv. Space Res., 19, 563–566, 1997. 22640

Riese, M., Spang, R., Preusse, P., Ern, M., Jarisch, M., Offermann, D., and Grossmann, K.-U.: Cryogenic infrared spectrometers and telescopes for the atmosphere (CRISTA) data processing and atmospheric temperature and trace gas retrieval, J. Geophys. Res., 104, 16349–16347, 1999. 22640

Ruhnke, R., Kouker, W., Reddmann, T., Berg, H., Hochschild, G., Kopp, G., Krupa, R., and Kuntz, M.: The vertical distribution of ClO at Ny-Ålesund during March 1997, Geophys. Res. Lett., 26, 839–842, 1999. 22645

ACPD
12, 22629–22685, 2012

Sassi, F., Garcia, R. R., and Boville, B. A.: The stratopause semiannual oscillation in the NCAR Community Climate Model, J. Atmos. Sci., 50, 3608–3624, 1993. 22633

Schoeberl, M. R., Douglass, A. R., Newman, P. A., Lait, L. R., Lary, D., Waters, J., Livesey, N., Froidevaux, L., Lambert, A., Read, W., Filipiak, M. J., and Pumphrey, H. C., QBO and annual cycle variations in tropical lower stratosphere trace gases from HALOE and Aura MLS observations, J. Geophys. Res., 113, D05301, doi:10.1029/2007JD008678, 2008. 22633

SPARC CCMVal: SPARC report on the evaluation of chemistry-climate models, 2010. 22634, 22653

Sparling, L. C.: Statistical perspectives on stratospheric transport, Rev. Geophys., 38, 417–436, 2000. 22651

Stanford, J. L. and Ziemke, J. R.: CH_4 and N_2O photochemical lifetimes in the upper stratosphere: In situ estimates using SAMS data, Geophys. Res. Lett., 18, 677–680, 1991. 22632

Steck, T., von Clarmann, T., Fischer, H., Funke, B., Glatthor, N., Grabowski, U., Höpfner, M., Kellmann, S., Kiefer, M., Linden, A., Milz, M., Stiller, G. P., Wang, D. Y., Allaart, M., Blumenstock, Th., von der Gathen, P., Hansen, G., Hase, F., Hochschild, G., Kopp, G., Kyrö, E., Oelhaf, H., Raffalski, U., Redondas Marrero, A., Remsberg, E., Russell III, J., Stebel, K., Steinbrecht, W., Wetzel, G., Yela, M., and Zhang, G.: Bias determination and precision validation of ozone profiles from MIPAS-Envisat retrieved with the IMK-IAA processor, Atmos. Chem. Phys., 7, 3639–3662, doi:10.5194/acp-7-3639-2007, 2007. 22639

Strahan, S., Loewenstein, M., and Podolske, J.: Climatology and small-scale structure of lower stratospheric N_2O based on in-situ observations, J. Geophys. Res., 104, 2195–2208, 1999. 22632

Urban, J., Lautié, N., Le Flochmoën, E., Jiménez, C., Eriksson, P., De La Noë, J., Dupuy, E., Ekström, M., El Amraoui, L., Frisk, U., Murtagh, D., Olberg, M., and Ricaud, P., Odin/SMR limb observations of stratospheric trace gases: Level 2 processing of ClO, N_2O, HNO_3, and O_3, J. Geophys. Res., 110, D14 307, doi:10.1029/2005JD005741, 2005a. 22637

Urban, J., Lautié, N., Le Flochmoën, E., Jiménez, C., Eriksson, P., De La Noë, J., Dupuy, E., El Amraoui, L., Frisk, U., Jégou, F., Murtagh, D., Olberg, M., Ricaud, P., Camy-Peyret, C., Dufour, G., Payan, S., Huret, N., Pirre, M., Robinson, A. D., Harris, N. R. P., Bremer, H., Kleinböhl, A., Küllmann, K., Künzi, K., Kuttipurath, J., Ejiri, M. K., Nakajima, H., Sasano, Y., Sugita, T., Yokota, T., Piccolo, C., Raspollini, P., and Ridolfi, M.: Odin/SMR limb obser-

ACPD
12, 22629–22685, 2012

vations of stratospheric trace gases: Validation of N_2O, J. Geophys. Res., 110, D09301, doi:10.1029/2005JD005394, 2005b. 22637

Urban, J., Murtagh, D., Lautié, N., Barret, B., Dupuy, E., De La Noë, J., Eriksson, P., Frisk, U., Jones, A., Le Flochmoën, E., Olberg, M., Piccolo, C., Ricaud, P., and Rösevall, J.: Odin/SMR limb observations of trace gases in the polar lower stratosphere during 2004–2005, in: Proceedings of the ESA First Atmospheric Science Conference, 8–12 May 2006, Frascati, Italy, edited by: Lacoste, H., Eur. Space Agency Spec. Publ., 2006. 22637, 22656

von Clarmann, T., Höpfner, M., Kellmann, S., Linden, A., Chauhan, S., Funke, B., Grabowski, U., Glatthor, N., Kiefer, M., Schieferdecker, T., Stiller, G. P., and Versick, S.: Retrieval of temperature, H_2O, O_3, HNO_3, CH_4, N_2O, $ClONO_2$ and ClO from MIPAS reduced resolution nominal mode limb emission measurements, Atmos. Meas. Tech., 2, 159–175, doi:10.5194/amt-2-159-2009, 2009. 22639

Waters, J., Froidevaux, L., Harwood, R. S., Jarnot, R. F., Pickett, H. M., Read, W. G., Siegel, P. H., Cofield, R. E., Filipiak, M. J., Flower, D. A., Holden, J. R., Lau, G. K., Livesey, N. J., Manney, G. L., Pumphrey, H. C., Santee, M. L., Wu, D. L., Cuddy, D. T., Lay, R. R., Loo, M. S., Perun, V. S., Schwartz, M. J., Stek, P. C., Thurstans, R. P., Boyles, M. A., Chandra, K. M., Chavez, M. C., Chen, G. S., Chudasama, B. V., Dodge, R., Fuller, R. A., Girard, M. A., Jiang, J. H., Jiang, Y. B., Knosp, B. W., LaBelle, R. C., Lam, J. C., Lee, K. A., Miller, D., Oswald, J. E., Patel, N. C., Pukala, D. M., Quintero, O., Scaff, D. M., Van Snyder, W., Tope, M. C., Wagner, P. A., and Walch, M. J., The Earth Observing System Microwave Limb Sounder (EOS MLS) on the aura satellite, IEEE Trans. Geosci. Remote Sens., 44, 1075–1092, 2006. 22638

WMO: Scientific assessment of ozone depletion: 2010, Report No. 52, Geneva, 2010. 22633

ACPD
12, 22629–22685, 2012

Table 1. Satellite measurements used in this study.

Instrument	Version	Measurements since	N_2O vertical resolution	N_2O precision	O_3 vertical resolution	O_3 precision	profiles per day
Odin/SMR	V2.1	2001	1.5 km	10–20 %	2.5-3.5 km	20 %	~650
CRISTA-1	V3	Nov 1994[1]	2 km	26 %(3 %)[3]	2 km	12 %(2 %)[3]	~4300
CRISTA-2	V1	Aug 1997[1]	2 km	12 %(5.3 %)[3]	2 km	10 %(2.1 %)[3]	~4300
MLS	V2.2	2004	4–6 km	9–25 %	3 km	3–10 %	~3300
MIPAS	V3/V4[2]	2002	3–4 km	5–10 %	2.5–4.5 km	3–8 %	~1200

[1] Measurements only performed during this month.
[2] V3O_N2O_11 and V3O_O3_9, V4O_N2O_202 and V4O_O3_201
[3] Errors are given for the tropics only.
[3] The Precision error analyses was separated into systematic (include uncertainties of spectroscopy as well) and random errors.

ACPD
12, 22629–22685, 2012

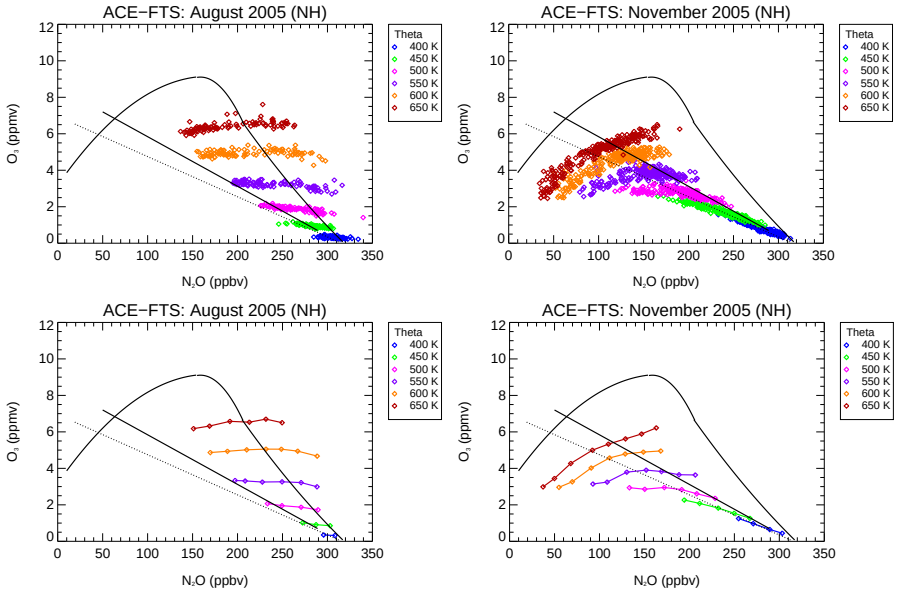

Fig. 1. N_2O versus O_3 for August and November 2005 derived from ACE-FTS observations (NH). Top: N_2O/O_3 correlation separated into potential temperature bins from 400 ± 25 K to 650 ± 25 K given by the colour coding. Bottom: O_3/N_2O correlation after the averaging over 20 ppbv N_2O has been applied within the potential temperature bins. Additionally, the May mid-latitude (ATMOS Shuttle 1985, solid line), April high-latitude (ATMOS Shuttle 1993, dashed line), and November tropics (ATMOS Shuttle 1994, solid curve) reference curves are shown (Proffitt et al., 1990; Michelsen et al., 1998b).

ACPD
12, 22629–22685, 2012

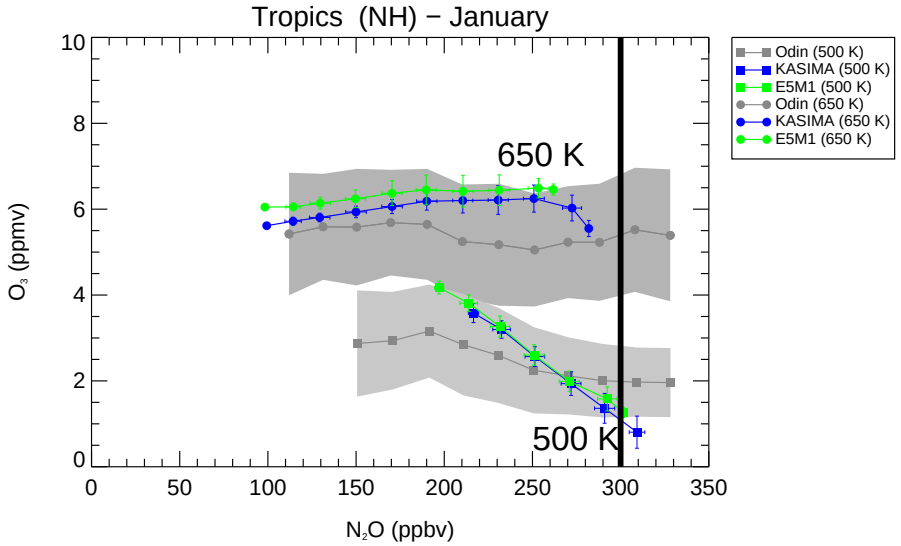

Fig. 2. Comparison of monthly averages of N_2O and O_3 derived from KASIMA (blue) and E5M1 T40L90MA (green) with Odin/SMR (grey) at 500 ± 25 K and 650 ± 25 K in the northern hemisphere tropics (0°-30° N, January 2003). The grey shaded area marks the range of the standard deviations of the monthly averages derived from Odin/SMR. The black solid line marks where N_2O mixing ratios exceed 300 ppbv.

ACPD
12, 22629–22685, 2012

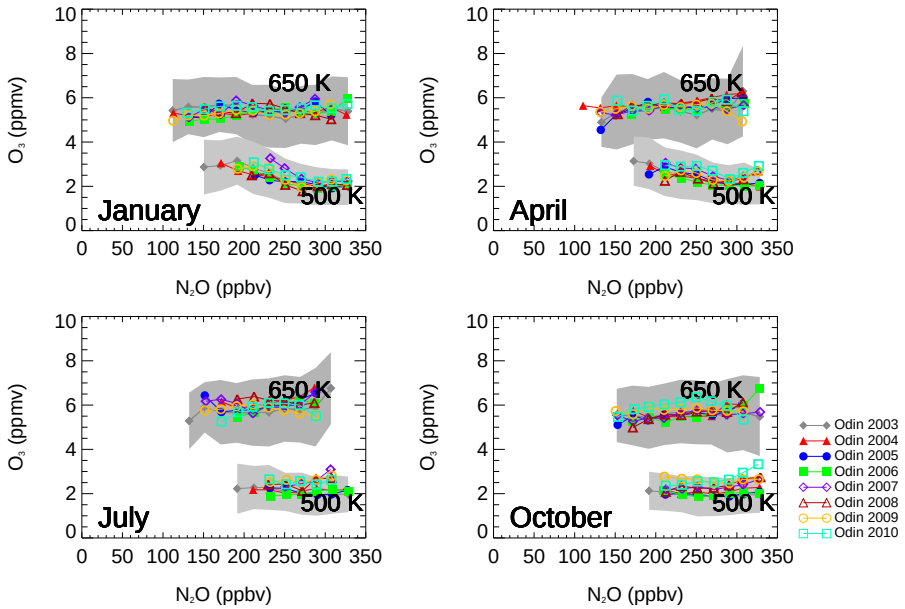

Fig. 3. Comparison of monthly averages of O_3 and N_2O derived from Odin/SMR for the years 2004 (red triangles), 2005 (blue circles), 2006 (green squares), 2007 (purple diamonds), 2008 (dark red triangles), 2009 (gold circles) and 2010 (green squares) with monthly averages derived for the year 2003 (grey diamonds) at 500 ± 25 K and 650 ± 25 K. Shown are the monthly averages for the northern hemisphere tropics (0°-30° N) for the months January, April, July and October. The grey shaded area marks the range of standard deviations of the monthly averages of O_3 derived from Odin/SMR data for the year 2003.

ACPD
12, 22629–22685, 2012

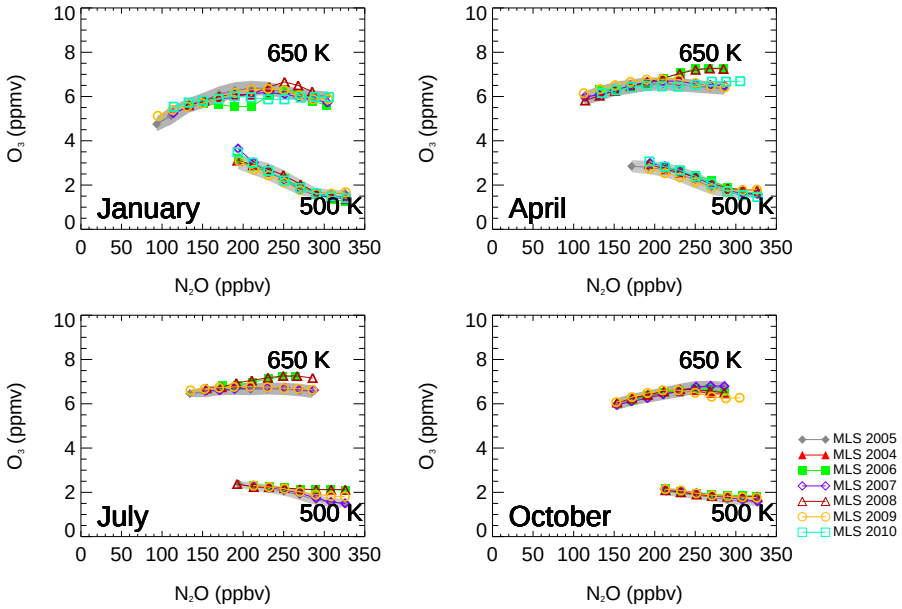

Fig. 4. Same as Fig 3 but for MLS observations for the years 2004–2010: 2004 (red triangles), 2005 (grey diamonds) and 2006 (blue circles), 2007 (green squares), 2008 (violet diamonds), 2009 (dark red triangles), 2010 (gold circles). Note: 2005 is used here as reference since measurements of MLS started in mid-2004.

ACPD
12, 22629–22685, 2012

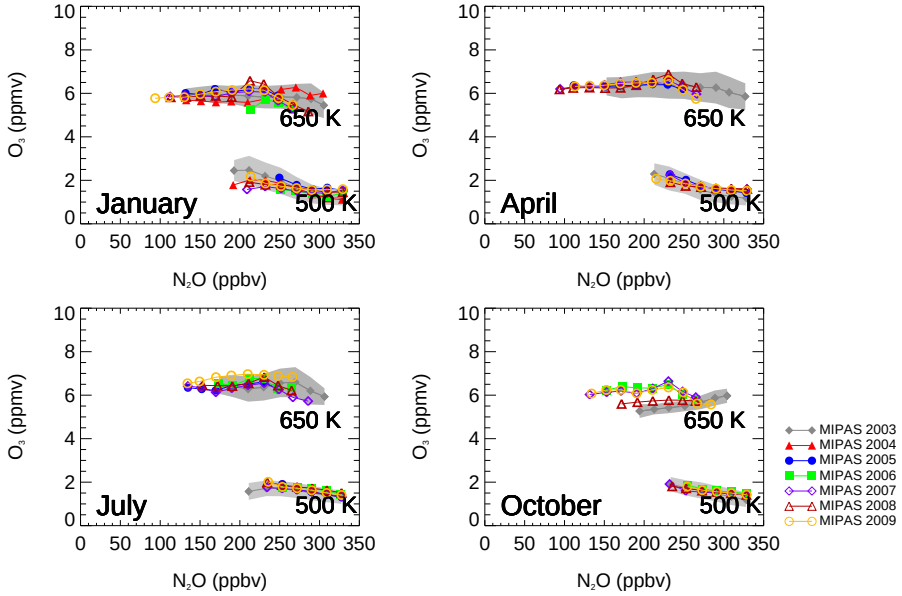

Fig. 5. Same as Fig 3 but for MIPAS observations for the years 2003–2009: 2003 (grey diamonds), 2004 (red triangles), 2005 (blue circles) and 2006 (green squares), 2007 (violet diamonds), 2008 (dark red triangles), 2009 (gold circles).

ACPD
12, 22629–22685, 2012

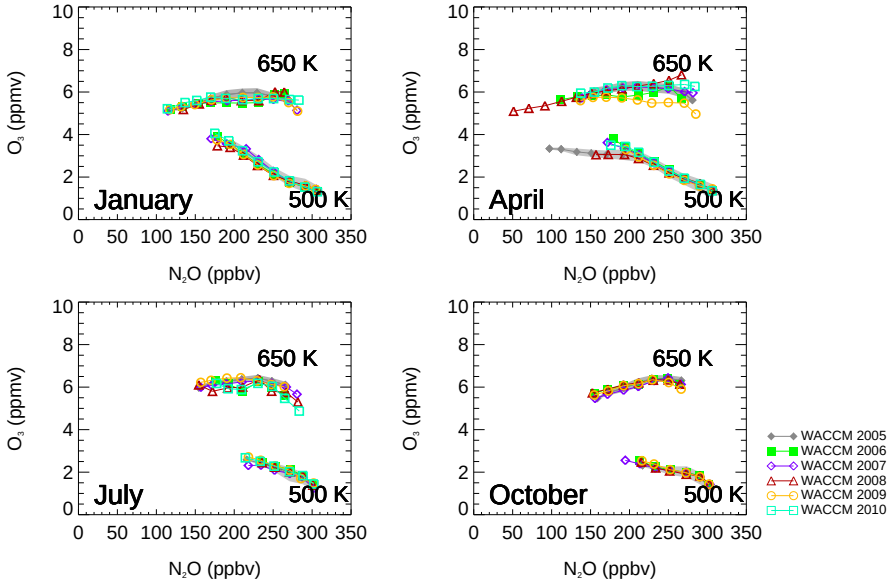

Fig. 6. Same as figure 3 but for SD-WACCM simulations for the years 2005–2010: 2005 (grey diamonds) and 2006 (blue circles), 2007 (green squares), 2008 (violet diamonds), 2009 (dark red triangles), 2010 (gold circles). Note: 2005 is used here as reference.

ACPD
12, 22629–22685, 2012

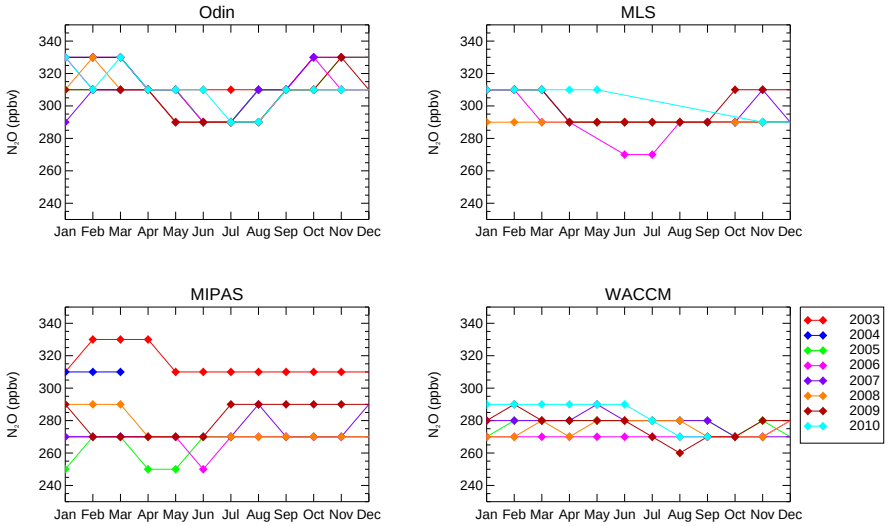

Fig. 7. Maximum monthly averaged N_2O mixing ratios (averaged mixing ratio of the last N_2O bin) at 650 ± 25 K are shown for Odin/SMR, Aura/MLS, ENVISAT/MIPAS and SD-WACCM (Visualization of the data presented in Tables 1 to 4 of the Supplement).

ACPD
12, 22629–22685, 2012

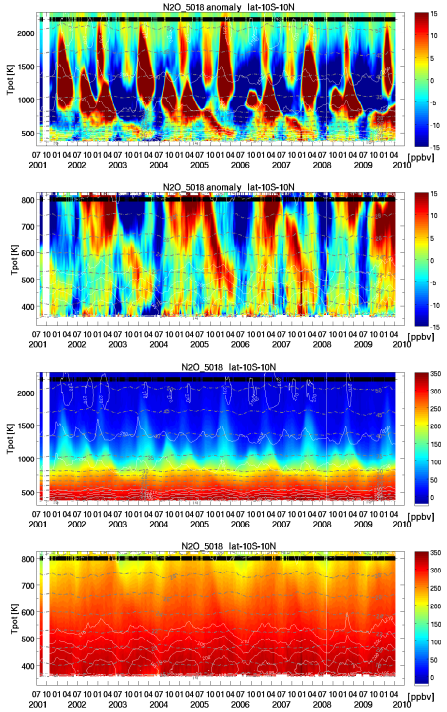

Fig. 8. Top and second row: Odin/SMR N_2O anomaly (daily zonal-mean minus multi-year zonal mean) for the years 2001-2010 for the entire stratosphere and lower stratosphere for the equivalent latitude range 10° N-10° S, respectively. Third row and bottom: N_2O mean for the years 2001–2010 for the entire stratosphere and lower stratosphere for the equivalent latitude range 10° N–10° S. Altitude and temperature (from ECMWF) are given as grey and white lines, respectively.

ACPD
12, 22629–22685, 2012

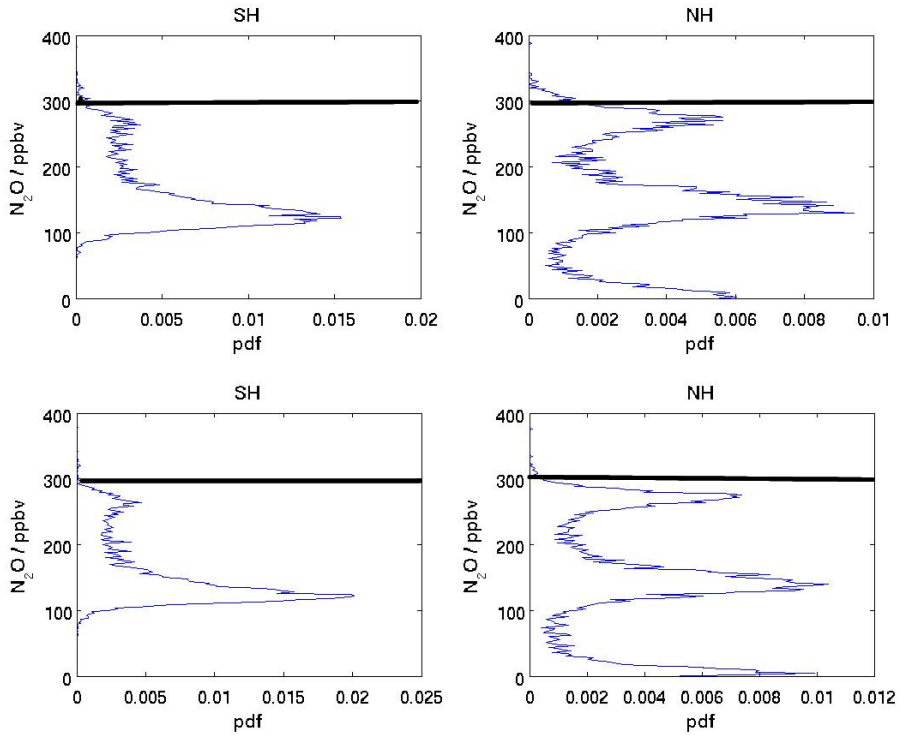

Fig. 9. Probability Density functions (PDFs) derived for Odin/SMR measurements (NH and SH) with Odin/SMR vertical resolution (1.5 km, top) and reduced to MLS vertical resolution (4 km, bottom) for January 2003 at 650 K. The black solid lines mark where N_2O exceeds 300 ppbv.

ACPD
12, 22629–22685, 2012

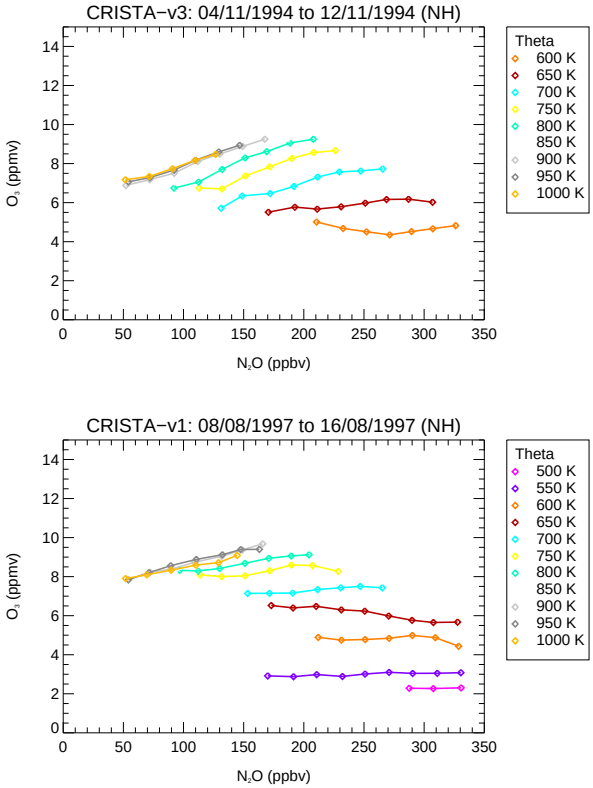

Fig. 10. Averages of N_2O and O_3 derived from CRISTA-1 measurements for November 1994 (4–12 November 1994) for potential temperatures levels between 600 ± 25 and 1000 ± 25 K (top) and CRISTA-2 measurements for August 1997 (8–16 August 1997) for potential temperatures levels between 500 ± 25 and 1000 ± 25 K (bottom) for the northern hemisphere tropics (0°–30°).

ACPD
12, 22629–22685, 2012

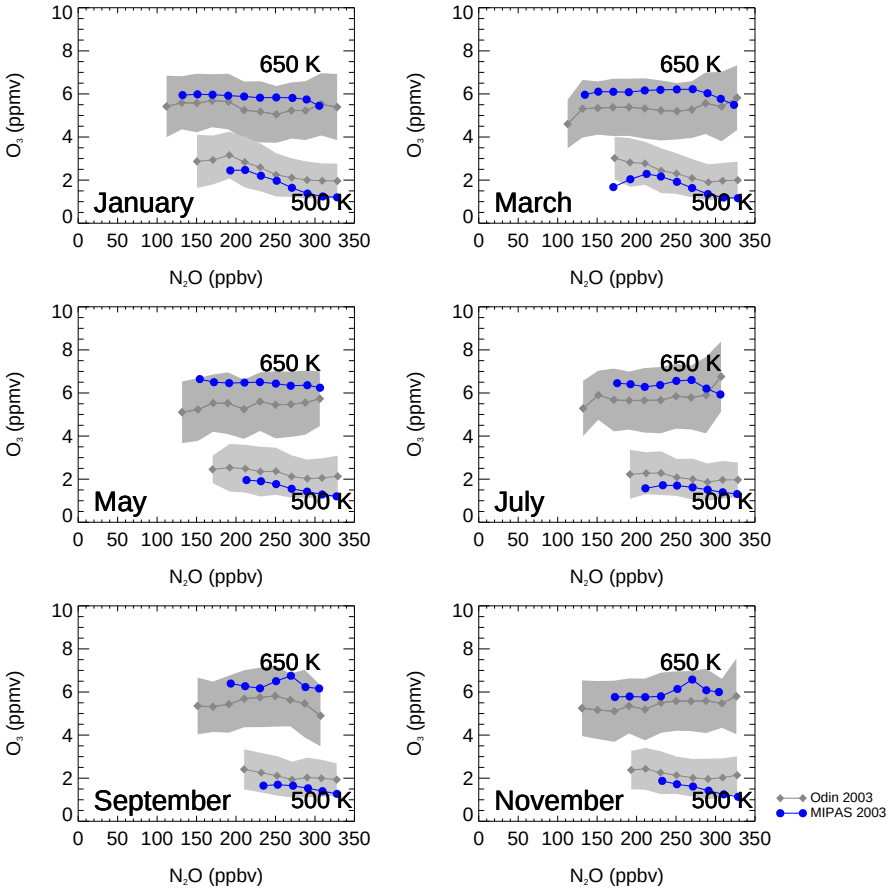

Fig. 11. Comparison of Odin/SMR and ENVISAT/MIPAS monthly averages of N_2O and O_3 for January, March, May, July, September and November 2003: Odin (grey diamonds) and MIPAS (blue circles).

ACPD
12, 22629–22685, 2012

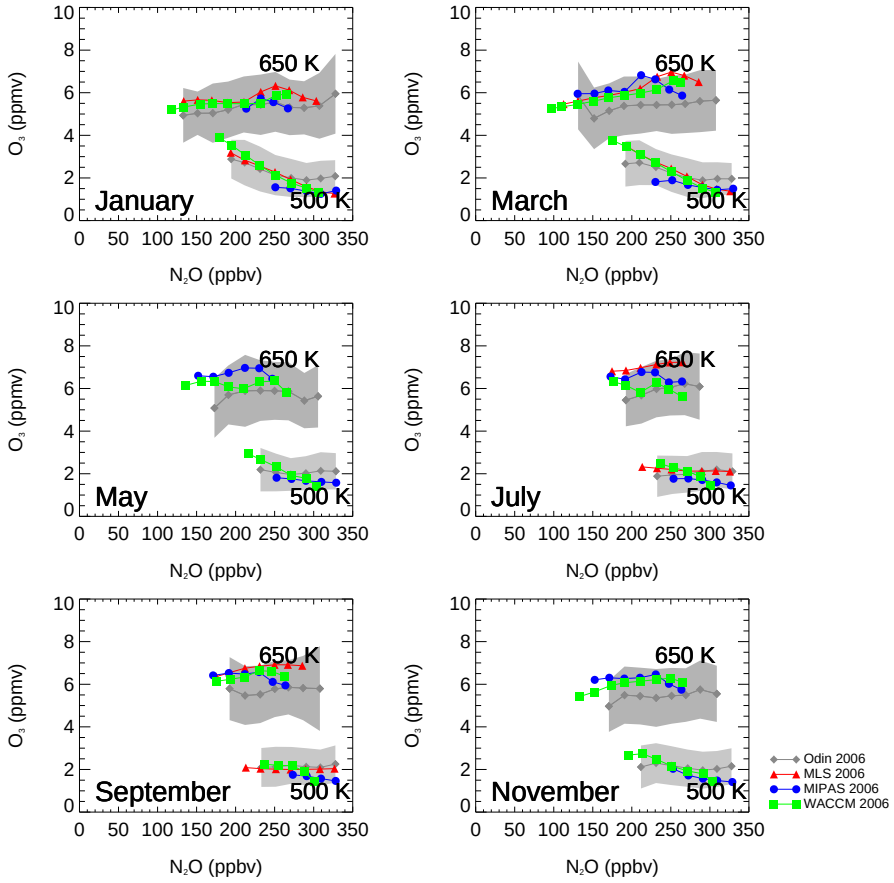

Fig. 12. Same as figure 11, but for 2006 and including Aura/MLS data and SD-WACCM data. Odin (grey diamonds), MLS (red triangles), MIPAS (blue circles) and SD-WACCM (green squares). Note: no MLS data available for May and November 2006.

ACPD
12, 22629–22685, 2012

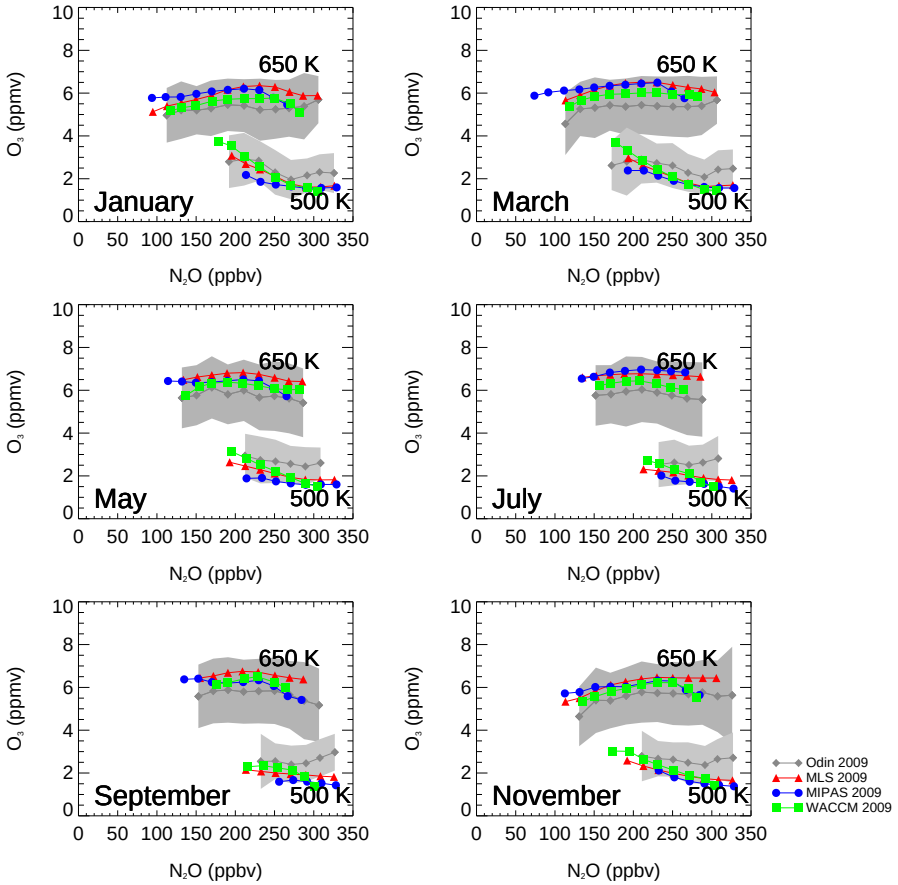

Fig. 13. Same as Fig. 11, but for the year 2009 and including Aura/MLS and SD-WACCM data. Odin/SMR (grey diamonds), Aura/MLS (red triangles) and SD-WACCM (green squares).

ACPD
12, 22629–22685, 2012

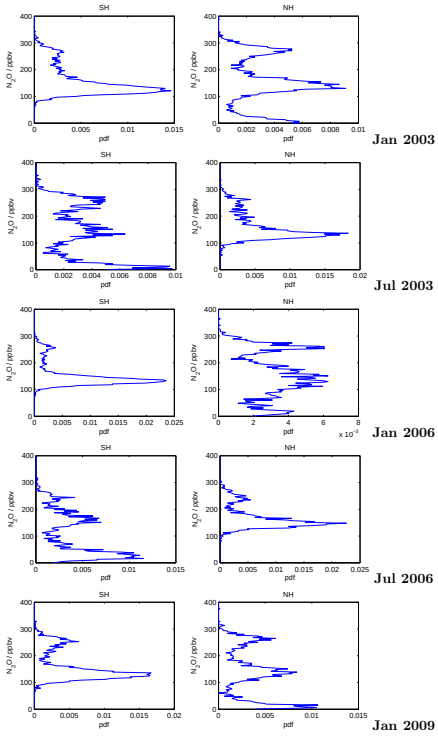

Fig. 14. Probability Density functions (PDFs) derived for Odin/SMR measurements (NH and SH) with Odin/SMR at 650 K for January and July 2003, January and July 2006 and January 2009. Note: the x-scale is not the same for all figures.

ACPD
12, 22629–22685, 2012