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Assessment of the interannual variability and influence 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 Earth and Space Science, Chalmers University of Technology, Gothenburg, 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

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

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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 evaluation of two chemical transport models (CTMs) and one chemistry–climate model (CCM) using a 1 yr climatology derived from the Odin Sub-Millimetre Radiometer (Odin/SMR). However, the applicability of a 1 yr 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. Furthermore, it is investigated why large differences are found 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 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. A realistic consideration of the QBO in the model reduces the differences between model simulation and observations significantly. Finally, an intercomparison between Odin/SMR, Aura/MLS, ENVISAT/MIPAS and WACCM was performed. The comparison shows that these data sets are generally in good agreement, although some known biases of the data sets are clearly visible in the monthly averages. Nevertheless, the differences caused by the uncertainties of the satellite data sets are sufficiently small and can be clearly distinguished

from model deficiencies. Thus, the method applied in this study is not only a valuable tool for model evaluation, but also for satellite data intercomparisons.

1 Introduction

Nitrous oxide (N_2O) is a long-lived species in the troposphere and lower stratosphere and can therefore 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. The N_2O abundance in the troposphere has increased rapidly due to anthropogenic emissions during the last centuries. Preindustrial values were around 270 ± 7 ppbv and had risen substantially to 319 ± 0.12 ppbv by 2005 (Forster et al., 2007). The global mean lifetime of N_2O is 122 ± 24 yr in the troposphere (Volk et al., 1997) 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). Thus, the temporal and spatial distribution of stratospheric N_2O can be used as a diagnostic tool of global-scale transport processes at different timescales, from seasonal to decadal (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 it has a longer lifetime in the lower stratosphere. The lifetime of O_3 is about 1 month in the lowermost stratosphere and in the winter hemisphere high latitudes, but decreases strongly with altitude in the stratosphere, particularly during summer months (Garcia and Solomon, 1985). Accordingly, O_3 mixing ratios in the lower stratosphere are highly variable, due to both the strong variability of O_3 photochemistry with latitude, altitude, and season as well as the seasonality of atmospheric transport vertically and horizontally (e.g. Dobson et al., 1973). Since O_3 is not photochemically conserved it has limited application as a tracer of transport (Proffitt et al., 2003). Throughout the year, O_3 is produced photochemically primarily in the tropical stratosphere with the peak production rate of O_3 occurring at ~ 30 km (Perliski et al., 1989). The photochemical destruction of O_3 is particularly pronounced at polar latitudes during late winter/early spring and summer. O_3 changes due to dynamical processes are characterized by the Brewer–Dobson circulation, consisting of ascent of tropospheric air into the stratosphere in the tropics, followed by quasi-horizontal poleward transport in the stratosphere, then descent back into the troposphere at the poles with strength of the transport dependent on season (Brewer, 1949; Dobson, 1956).

The interannual variability of the equatorial stratosphere ($\sim$ 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). The strongest mode of annual variability above 35 km altitude is the semi-annual oscillation (SAO) (e.g. Randel et al., 1998; Lossow et al., 2008). Both 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 the signature of the annual oscillation (AO) is also evident in other trace gas distributions (e.g. Randel et al., 1998; Schoeberl et al., 2008; Ricaud et al., 2009). Furthermore, based upon satellite measurements, the influence of the QBO on long-lived species has been demonstrated within the tropical regions (e.g. O'Sullivan and Dunkerton, 1997).

In the recent decade major efforts have been made to evaluate and improve chemistry–climate models (CCMs) in order to provide reliable predictions of future climate changes (WMO, 2010). To evaluate CCMs with measurements careful use of appropriate diagnostics is essential (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 come from various sources which together result in major biases in the representation of the wave mean flow interaction – the key process in QBO forcing (Holton and Lindzen, 1972). While some models generate the oscillations with realistic time scales, and thus resemble the observed QBO, other models get periods deviating from those observed, and are typically faster (Giorgetta et al. (2006) and references therein). Giorgetta et al. (2006) showed that to produce a realistic QBO in CCM simulations, a very high vertical resolution is required (~ 1 km) in order to sufficiently resolve the vertical structures of the waves. 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 signature, as first discussed using satellite-borne H_2O measurements in Mote et al. (1996), is caused by the imprint of the tropopause temperature on H_2O and its transport into the stratosphere within the upwelling branch of the Brewer–Dobson circulation. A tape recorder signature has also been found in other trace gases, e.g. CO, CO_2 and HCN and is caused by the seasonal variability of tropospheric source gases (e.g. Schoeberl et al., 2006; Pommrich et al., 2010; Andrews et al., 1999).

In a recent model evaluation study (Khosrawi et al., 2009), a method was used based on O_3 and N_2O measurements 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 and based on aircraft and balloon-borne measurements. This type of analysis helps to separate O_3 variability due to latitudinal transport from photochemical changes. In our follow-up studies this method has 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 presented for the potential temperature levels of 500 ± 25 K and 650 ± 25 K. In the Northern Hemisphere tropics (0° to 30° N) differences between models and satellite observations were found at both levels. In both models a steeper negative correlation (decreasing O_3 with increasing N_2O) than observed was found at 500 ± 25 K. Khosrawi et al. (2009) explained these differences as due in part to the large vertical O_3 gradients occurring in the tropics which cannot be resolved by the rather coarse 3 km vertical resolution of Odin/SMR and partly by inaccuracies of transport in the tropical stratosphere in the model simulations. In the context of this study it will be shown that the latter reason is applicable and that the former has no direct influence on the results. Although 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 those 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 tropospheric observations of N_2O (319 ppbv in 2005), whereas the troposphere is the only source region of this trace gas. Satellite data sets in general tend to derive higher N_2O mixing ratios, exceeding > 320 ppbv compared to in situ measurements. These unrealistic high values in satellite N_2O measurements can also be seen in the tracer–tracer correlations and probability density functions (PDFs) derived from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) (Hegglin and Shepherd, 2007) and vertical profiles from the Aura Microwave Limb Sounder (Aura-MLS) and the Michelson Interferometer for Passive Atmospheric Sounding onboard the ESA's Environmental Satellite (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 using Odin/SMR as a reference. Odin/SMR has been used as reference to facilitate comparison with the results derived in Khosrawi et al. (2009). The satellite data sets considered here are the ones with a high temporal and spatial coverage in the tropics are 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 Model (WACCM). The main purposes of this study are the following: (1) to apply 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) so that the interannual 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. (2) To utilize additionally CRISTA-1 and CRISTA-2 data to examine the cause of the large differences in N_2O at 650 ± 25 K that were found between Odin/SMR and model simulations in our recent model evaluation study (Khosrawi et al., 2009). (3) Odin/SMR, Aura/MLS, ENVISAT/MIPAS and WACCM are compared for the years 2003, 2006 and 2009 in order to test the applicability of the monthly averages of N_2O and O_3 also for satellite data intercomparisons.

2 Satellite data

In the following the satellite data sets used in this study will be briefly described. A summary of these 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 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 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

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] Given errors are for the tropics only. Precision error analysis separated into systematic (include uncertainties of spectroscopy as well) and random errors.

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 (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, 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 the 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), which show 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 intercomparison of Odin/SMR O_3 measurements with ENVISAT/MIPAS and balloon sonde data by Jones et al. (2007) showed 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 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 millimetre 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), where 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 upper troposphere/lower stratosphere between 215 and 147 hPa. A comparison of MLS O_3 with the Stratospheric Aerosol and Gas Experiment (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

MIPAS was launched in March 2002 onboard the ESA's 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}. 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, 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 measurements 2005 onwards (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).

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, 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 respective CRISTA-1 systematic and statistical errors for N_2O and O_3 at 25 km are 26 % and 3 % for N_2O, and 10 % and 2 % for O_3. The respective CRISTA-2 systematical and statistical errors for N_2O and O_3 at 25 km are 12 % and 5.3 % for N_2O, and 10 % and 2.1 % for O_3. A description of the CRISTA error analyses can be found in Riese et al. (1999).

3 Model data

3.1 WACCM

The Whole Atmosphere Community Climate Model, Version 4 (WACCM 4) is a fully interactive CCM, 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 (GEOS5). Here, temperature, zonal and meridional winds, and surface pressure are used to drive the physical parameterization that controls 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 to the end of 2010. For this simulation the model had a spin-up time from 1980 through to 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 $1.9° \times 2.5°$ (210×270 km), with a vertical 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, mid-latitudes, 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, and of diabatic descent on the N_2O/O_3 distribution can be found in the above references. Here the method will be described only briefly.

4.1 General characteristics

A modified form of tracer–tracer correlations of N_2O and O_3 was first used by Proffitt et al. (2003) for estimating O_3 loss during the Arctic winter months. This method has the advantage of separating O_3 variability due to latitudinal transport from photochemical changes. We follow this method using monthly averages of N_2O and O_3 that are calculated by binning the data by altitude or potential temperature, then averaging over a fixed interval of N_2O (20 ppbv). This is shown in Fig. 1 for the months of August and November 2005 using measurements derived from ACE-FTS in the Northern Hemisphere[1]. First, the N_2O and O_3 data are 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, the N_2O and O_3 binned by potential temperature are averaged over 20 ppbv N_2O resulting in a set of curves. Therefore, each curve lies within its potential temperature bin (e.g. one curve at 400 ± 25 K, 450 ± 25 K, and so on).

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 used. However, using the May mid-latitude (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, mid-latitude 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 missions with durations of approximately two weeks each (Michelsen et al., 1998a). The ACE-FTS measurements in August 2005 are centred around the tropical reference curve indicating that the measurements were primarily performed in the tropics, while in November the observations are centred around the mid-latitude and polar reference curve indicating air of mid-latitudinal 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 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 O_3 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), in the monthly averages, diabatic descent is to a certain extent masked by tropical O_3 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, mid-latitudes 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

[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 mid-latitudes 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.

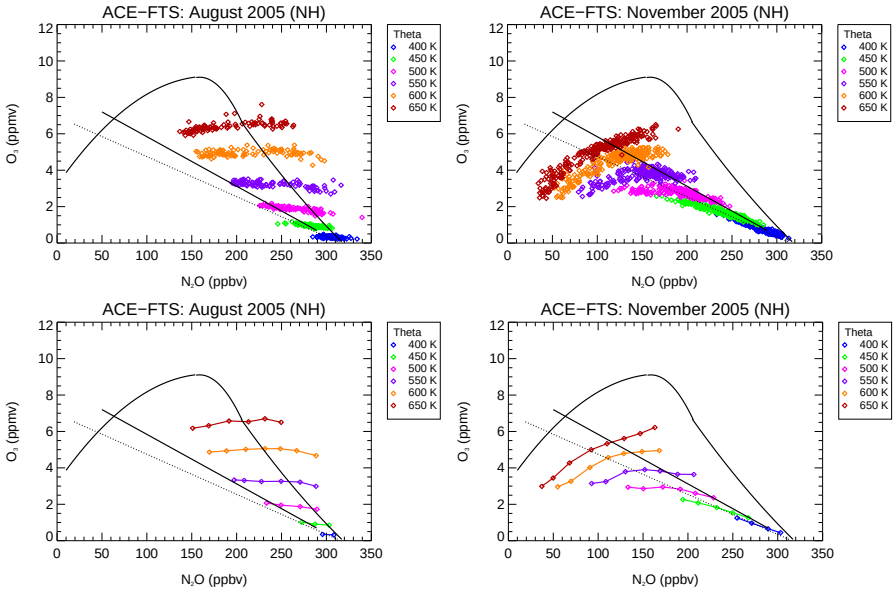

Fig. 1. N_2O versus O_3 for August and November 2005 derived from ACE-FTS observations (NH). Top: N_2O/O_3 correlation separated into potential temperature bins from 400 ± 25 K to 650 ± 25 K given by the colour coding. Bottom: O_3/N_2O correlation after the averaging over 20 ppbv N_2O has been applied 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).

(Khosrawi et al., 2008). The positive correlation is caused by the photochemical production of O_3 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 mid-latitude air (Khosrawi et al., 2008). In fact, the tropics are not truly isolated from the mid-latitudes 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 tropical O_3/N_2O distribution is similar in both hemispheres. Further, seasonal changes are small and equal in both hemispheres (Khosrawi et al., 2008).

In our recent study (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) during January 2003 (NH, tropics).

In both models a steeper negative correlation than for the Odin/SMR observations was found at 500 ± 25 K, which leads to higher O_3 mixing ratios at N_2O mixing ratios less than 250 ppbv and lower O_3 mixing ratios at N_2O mixing ratios greater 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 O_3 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, as an exemplary test to assess the effect of this aspect, we degraded the 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; Eq. 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 we conclude that 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

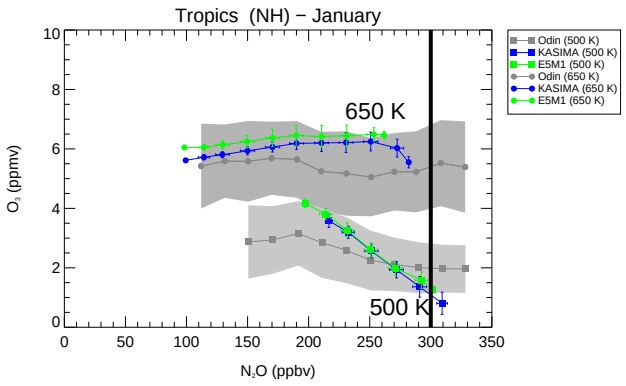

Fig. 2. Comparison of monthly averages of N_2O and O_3 derived from KASIMA (blue) and E5M1 T40L90MA (green) with Odin/SMR (grey) at 500 ± 25 K and 650 ± 25 K in the Northern Hemisphere tropics (0–30° N, January 2003). The grey shaded area marks the range of the standard deviations of the monthly averages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

models (e.g. Hegglin and Shepherd, 2007). This is due to the difficulties in deriving the vertical velocities in the tropical upper troposphere and lower stratosphere. Ploeger et al. (2010) and Schoeberl et al. (2012) have shown that vertical velocities in models can be both overestimated and underestimated, resulting thus in faster or slower tropical upwelling compared to measurements. The model vs Odin/SMR differences could thus 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 ($1\sigma \sim 25$ 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 perhaps difficult to simulate in the models, e.g. the QBO. We also investigated the cause of differences between models and measurements at 500 ± 25 K and assess the interannual variability of monthly averages of N_2O and O_3 as well as test their applicability for satellite–satellite intercomparisons. 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 interannual variability

To assess the interannual 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 measurements (2003–2009), seven years of Aura/MLS measurements (2004–2010) as well as 6 yr 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 interannual 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 interannual 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 $N_2O \geq 300$ 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 around 25 km (Baldwin et al., 2001). The slight variations in O_3 at 500 ± 25 K for $N_2O \geq 300$ ppbv are most likely caused by the interannual 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 mid-latitude 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 sampling as

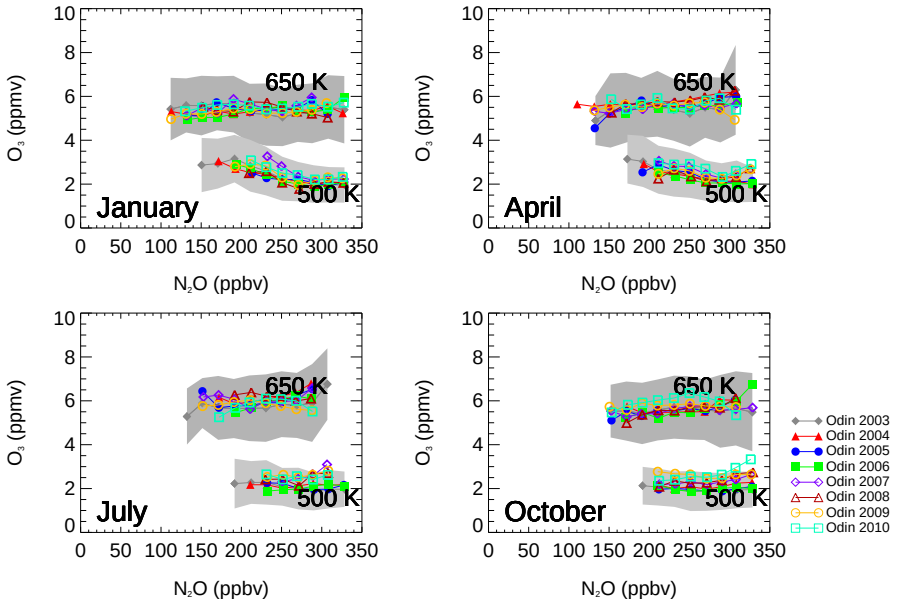

Fig. 3. Comparison of monthly averages of O_3 and N_2O derived from Odin/SMR for the years 2004 (red triangles), 2005 (blue circles), 2006 (green squares), 2007 (purple diamonds), 2008 (dark red triangles), 2009 (gold circles) and 2010 (green squares) with monthly averages derived for the year 2003 (grey 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.

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 a 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 interannual 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 where $N_2O \geq 300$ ppbv are most likely caused by the interannual variations in trace gas concentrations caused by the AO and QBO. 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 level where the QBO amplitude peaks, and 500 K is a level 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 are not as good as the Odin/SMR observations (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; instrumental precision also plays 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 interannual variability of monthly averages of N_2O and O_3 is also low for ENVISAT/MIPAS. A bias with 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 Odin/SMR and Aura/MLS averages are caused by air mid-latitude origin; the interannual variability in N_2O and O_3 is most likely due to 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 interannual variability of these averages as derived from a CCM is investigated. For the purpose of a model evaluation study, it is important that not only

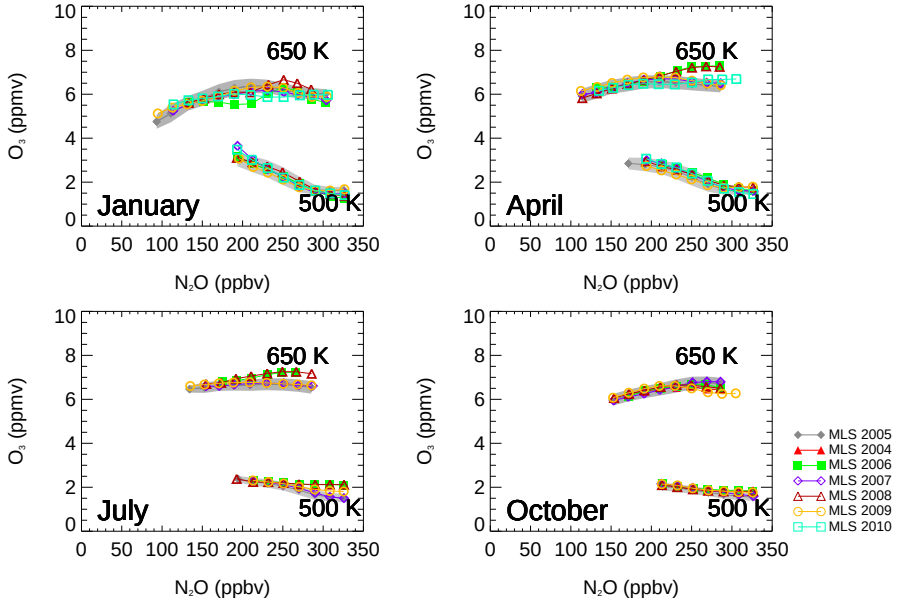

Fig. 4. Same as Fig. 3, but for MLS observations for the years 2004–2010: 2004 (red triangles), 2005 (grey diamonds) and 2006 (blue circles), 2007 (green squares), 2008 (violet diamonds), 2009 (dark red triangles), 2010 (gold circles). Note: 2005 is used here as reference since measurements of MLS started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

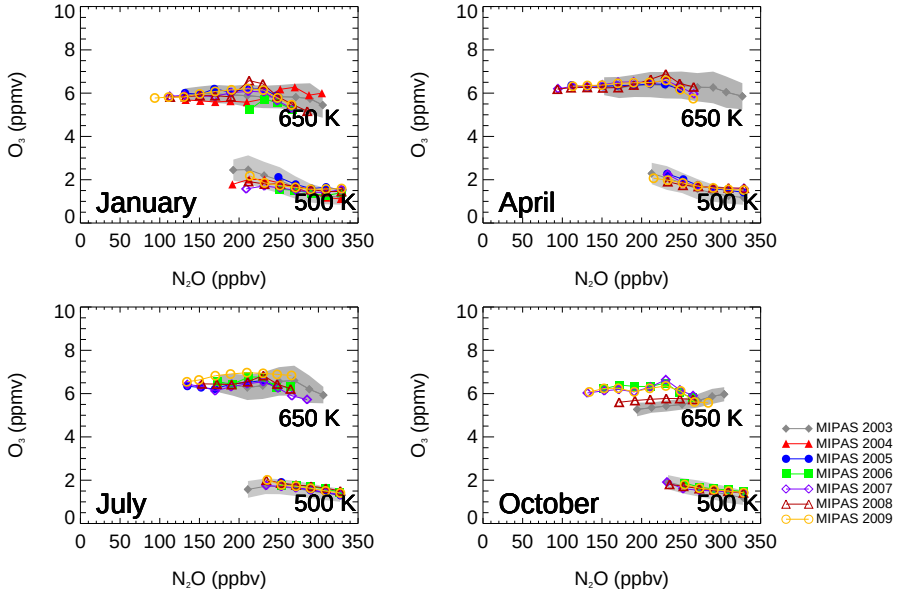

Fig. 5. Same as Fig. 3, but for MIPAS observations for the years 2003–2009: 2003 (grey diamonds), 2004 (red triangles), 2005 (blue circles) and 2006 (green squares), 2007 (violet diamonds), 2008 (dark red triangles), 2009 (gold circles). Note: MIPAS measurements in 2003–2004 were performed with the nominal spectral resolution and from 2005 onwards with reduced spectral resolution.

the interannual variability in the satellite data sets is low, but also that the interannual 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 interannual variability in the SD-WACCM monthly averages of N_2O and O_3 is as low as 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

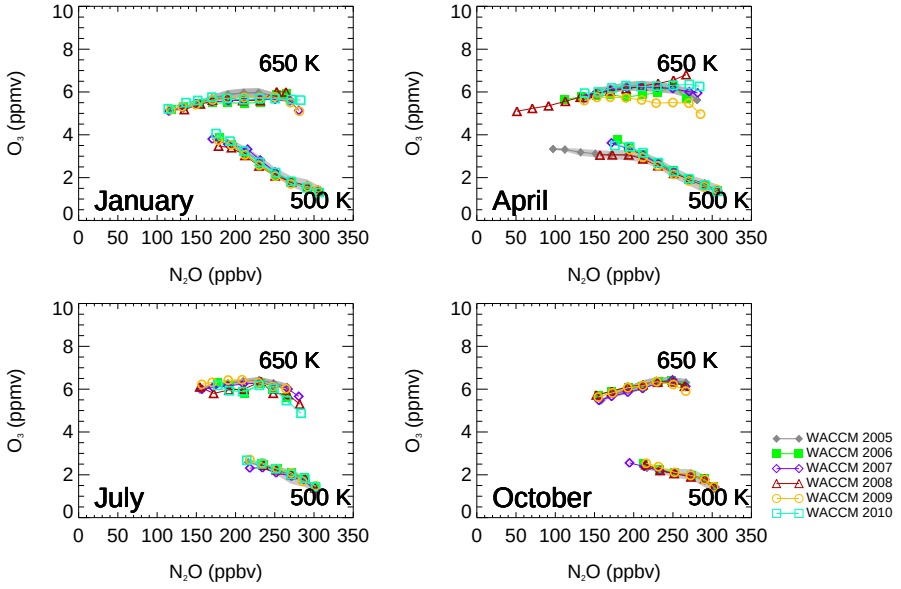

Fig. 6. Same as Fig. 3, but for SD-WACCM simulations for the years 2005–2010: 2005 (grey diamonds) and 2006 (blue circles), 2007 (green squares), 2008 (violet diamonds), 2009 (dark red triangles), 2010 (gold circles). Note: 2005 is used here as reference.

averages of N_2O and O_3 derived from SD-WACCM at 500 ± 25 K are steeper than (but not as steep as E5M1 and KASIMA) the ones derived from the satellite data, which can most likely be attributed to a stronger tropical upwelling in the model simulation than was actually observed (see Sect. 5.3). In the case of stronger upwelling, low O_3 and high N_2O will be brought up to higher altitudes than during weak upwelling and thus increase the steepness of the N_2O/O_3 curves. Weak upwelling will have the opposite effect and cause a flattening of the curves. At 650 ± 25 K the slight variations from year to year in the monthly averages are caused most likely by the QBO. In the SD-WACCM data in April an 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 mid-latitude air into the tropics by the model.

5.2 Large N_2O differences between model and measurements

5.2.1 Analysis of Odin/SMR observations

In the tropics, monthly averages of N_2O values derived from Odin/SMR observations at 650 ± 25 K were much higher (reaching up to 330 ppbv) than simulated by KASIMA and E5M1. These values are also much higher than the lower stratospheric measurements by in situ instruments on aircraft or balloons (e.g. Strahan et al., 1999; Herman et al., 1998; Proffitt et al., 2003; Moore et al., 2003). Further, these values are even higher than the highly accurate ground-based measurements of N_2O, of 319 ppbv in 2005 (Forster et al., 2007). This indicates that satellite data generally tend to derive too high N_2O mixing ratios due to instrument noise and/or biases. In the Odin/SMR data these high N_2O mixing ratios occur solely in the tropics and with a seasonal cycle which indicates that the occurrence of higher/lower values (ignoring the absolute values) may be caused by a physical process. 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, apart from a seasonal variability in the observation of these high values we also found an interannual 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 (Figs. 3 and 7 and Supplement).

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). Further, the QBO easterly

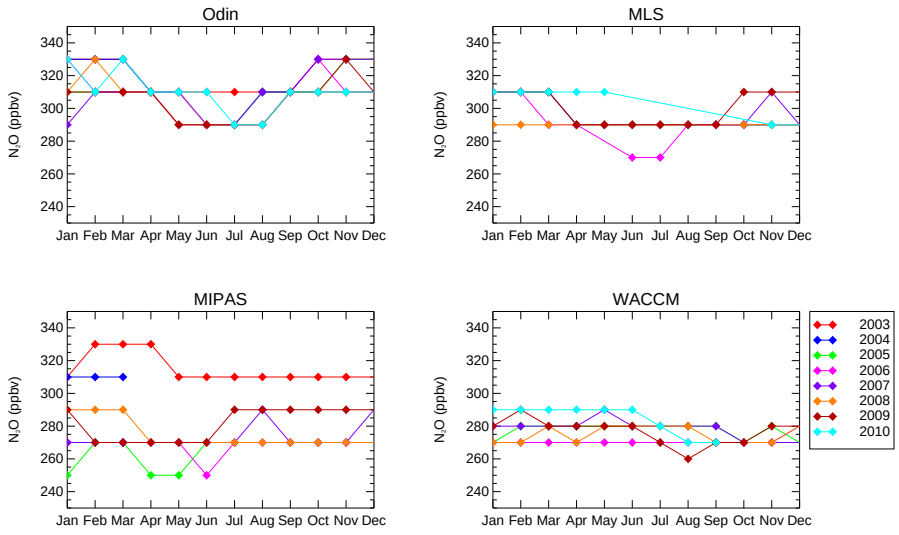

Fig. 7. Maximum monthly averaged N_2O mixing ratios (averaged mixing ratio of the last N_2O bin) at 650 ± 25 K are shown for Odin/SMR, Aura/MLS, ENVISAT/MIPAS and SD-WACCM (Visualization of the data presented in Tables 1 to 4 of the Supplement).

(westerly) shear phase coincides with enhanced (reduced) upwelling (Punge et al., 2009). The QBO N_2O and O_3 analysis with a linar regression model shows a variation of 10–15 ppbv in absolute values and an amplitude of ± 6 ppbv in N_2O and a variation of 1 ppmv in absolute values and an amplitude of 0.5 ppmv in O_3 for the Odin/SMR data. These variations are in agreement with the variations in N_2O and O_3 we see in the monthly averages derived from the Odin/SMR data (see Supplement).

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 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 to greater altitudes in 2002, 2004, 2006, 2008, and 2010. This 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). Thus, though the absolute values are caused by instrument noise and biases, the winter/summer variation of the occurrence of higher/lower N_2O values can be attributed to the QBO.

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, although 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 (Figs. 4 and 7 as well as tables in Supplement). This 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 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 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

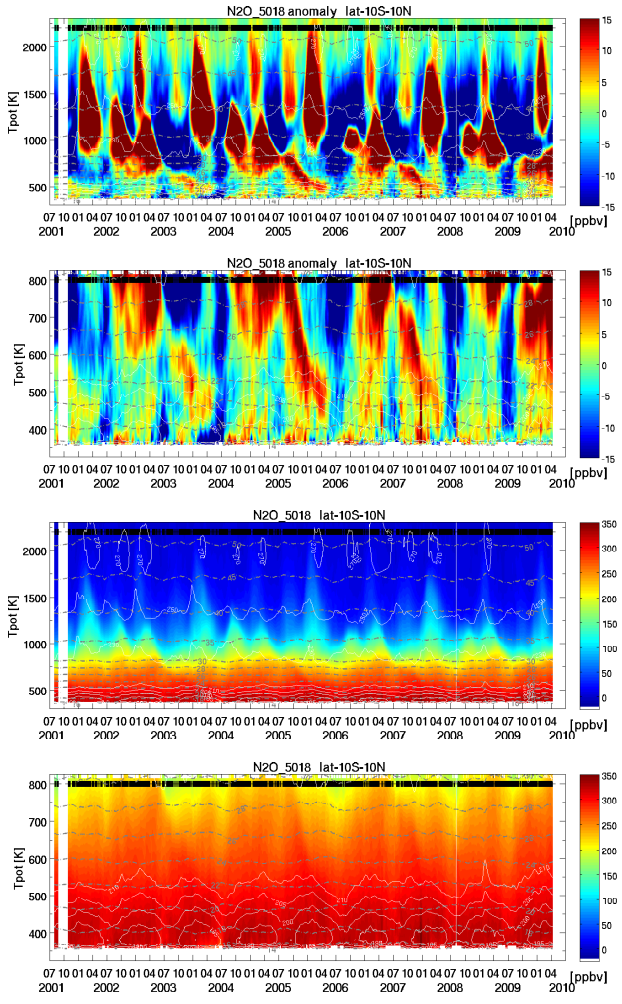

Fig. 8. Top and second row: Odin/SMR N_2O anomaly (daily zonal mean minus multi-year zonal mean) for the years 2001–2010 for the entire stratosphere and lower stratosphere for the latitude range 10° N–10° S, respectively. Third row and bottom: N_2O mean for the years 2001–2010 for the entire stratosphere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

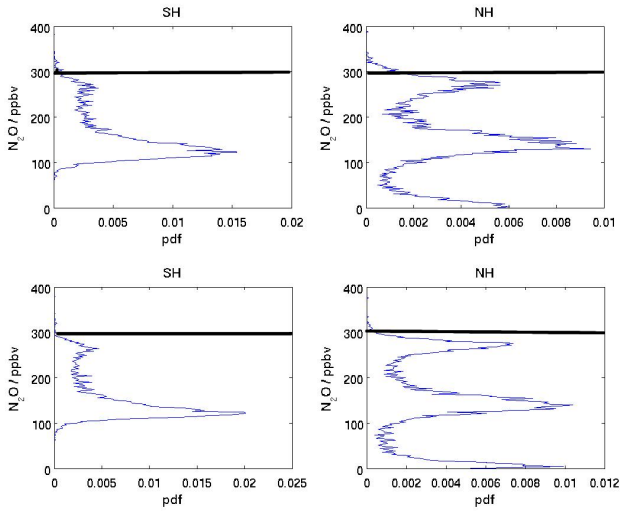

Fig. 9. Probability density functions (PDFs) derived for Odin/SMR measurements (NH and SH) with Odin/SMR vertical resolution (1.5 km, top) and reduced to MLS vertical resolution (4 km, bottom) for January 2003 at 650 K. The black solid lines mark where N_2O exceeds 300 ppbv.

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. Although 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, here we also find N_2O mixing ratios up to 330 ppbv at 25 km ($\sim$ 650 K). However, this does not mean that CRISTA-1 observations suggest N_2O values above the tropospheric background levels, since the systematic error of the CRISTA-1 observation is rather large in the tropical lower stratosphere (26 %, Table 1). In addition, it should be noted that 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). Nevertheless, this once again indicates that satellite data generally tend to derive too high N_2O mixing ratios due to instrument noises and/or biases.

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 (Figs. 5 and 7) are found than in 2003 and 2004 (e.g. Palazzi et al., 2011). These low values are a result of the efforts to reduce the well-known high bias in MIPAS N_2O observations during the earlier period. However, these values

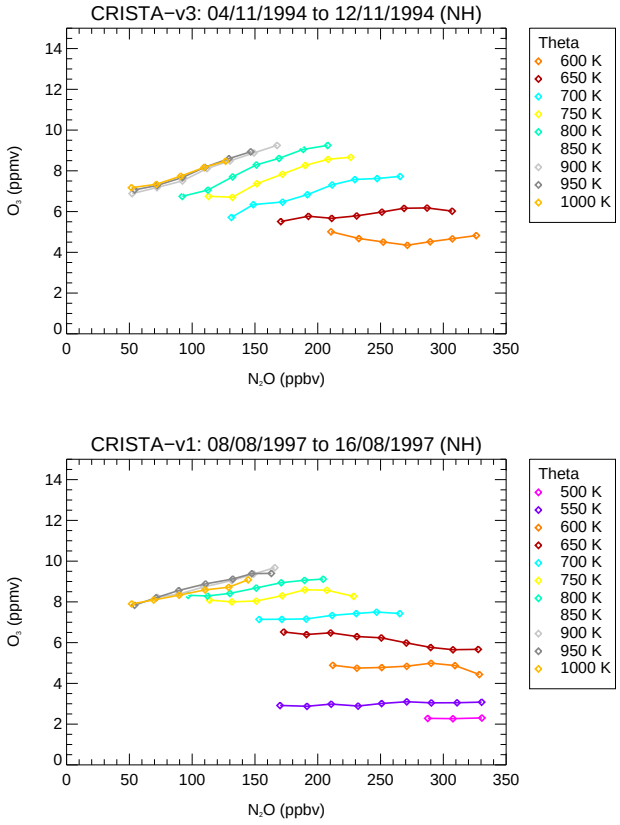

Fig. 10. Averages of N_2O and O_3 derived from CRISTA-1 measurements for November 1994 (4–12 November 1994) for potential temperatures levels between 600 ± 25 and 1000 ± 25 K (top) and CRISTA-2 measurements for August 1997 (8–16 August 1997) for potential temperatures levels between 500 ± 25 and 1000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

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, apart from 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 our satellite data comparison where we use monthly averages of N_2O and O_3, a seasonal and interannual cycle in the occurrence of enhanced N_2O mixing ratios is found in all data sets; however, it is more or less pronounced dependending on which data set is considered. We found that these values occur preferentially in the satellite data sets with high vertical resolution. Additionally, 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, we know 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. However, though the absolute values are too high, they do not conflict with the results derived in this study. Two sensitivity studies, one allowing averages only being calculated up to 320 ppbv and another one reducing the entire Odin/SMR data set by 20 ppbv (see Supplement), show that this bias/noise error does not affect the interannual variability. Thus, the monthly averages derived from the satellite data sets considered here are suitable for model evaluation and satellite intercomparisons.

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 interannual 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 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 were 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 also improves 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 to 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

[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

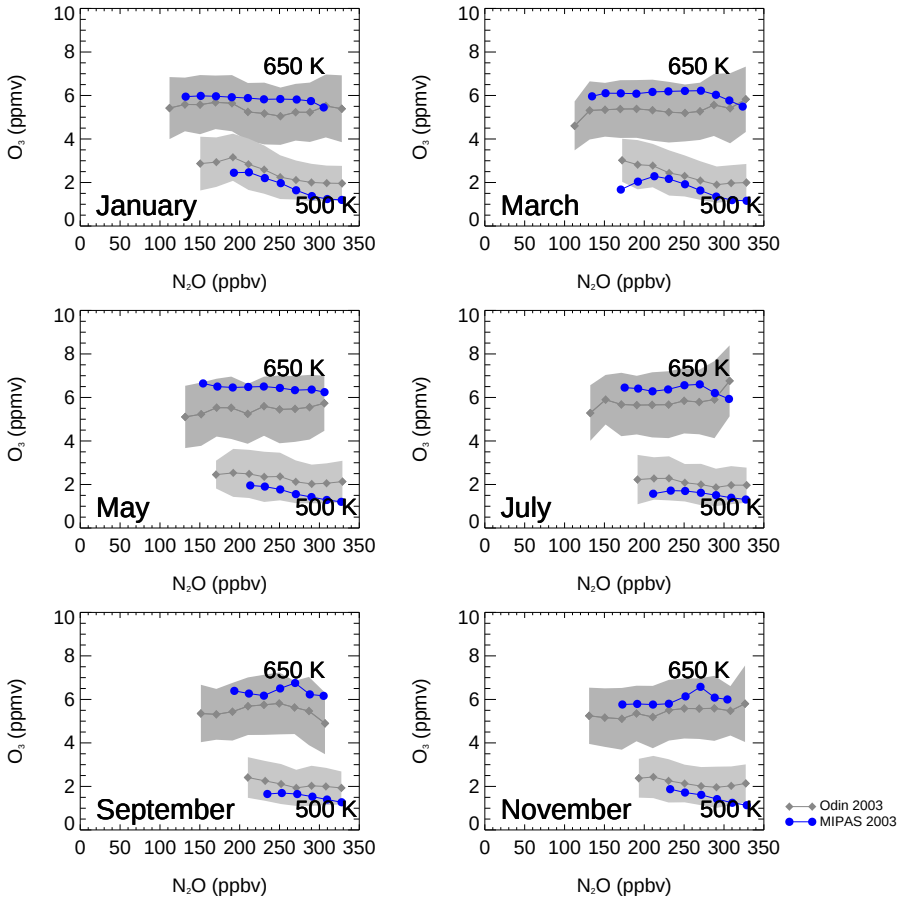

Fig. 11. Comparison of Odin/SMR and ENVISAT/MIPAS monthly averages of N_2O and O_3 for January, March, May, July, September and November 2003: Odin (grey diamonds) and MIPAS (blue circles).

bins during winter months is found. However, higher values are found during May 2007, February 2009, and from January to June 2010. The QBO in SD-WACCM is realistically simulated and the QBO signature found in the N_2O anomalies is in good agreement (not shown) with the ones derived from Odin/SMR (Fig. 8). 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 in Odin/SMR. This may explain why we do not see the feature of higher N_2O values in winter than in summer as pronounced in SD-WACCM as in the satellite data.

5.3 Intercomparison of model and satellite data

For investigating of whether the monthly averages of N_2O and O_3 can not only be applied for model evaluations, but also for satellite intercomparison, a comparison of Odin/SMR, Aura/MLS, ENVISAT/MIPAS and SD-WACCM averages derived for the tropics is performed. The comparison was made 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 are 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

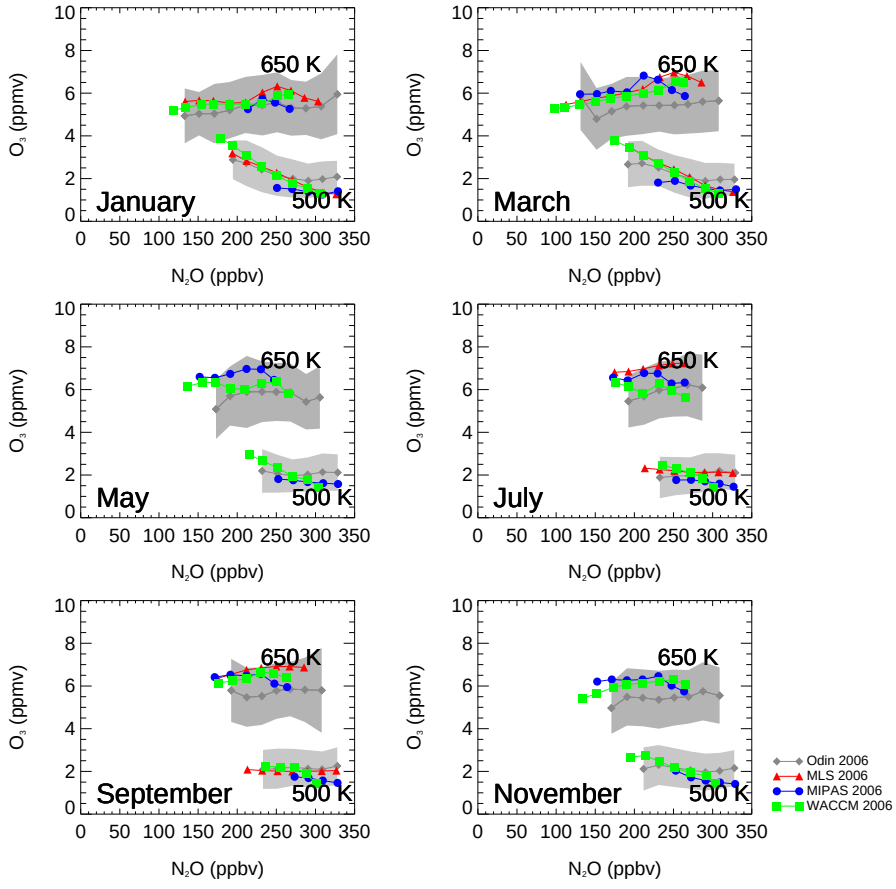

Fig. 12. Same as Fig. 11, but for 2006 and including Aura/MLS data and SD-WACCM data. Odin (grey diamonds), MLS (red triangles), MIPAS (blue circles) and SD-WACCM (green squares). Note: no MLS data available for May and November 2006.

averaged values 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 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 ppmv), 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 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 when 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, additionally to 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 evaluation (Khosrawi et al., 2009). Thus, if we would repeat our model evaluation 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 O_3 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 O_3 sondes was found to be

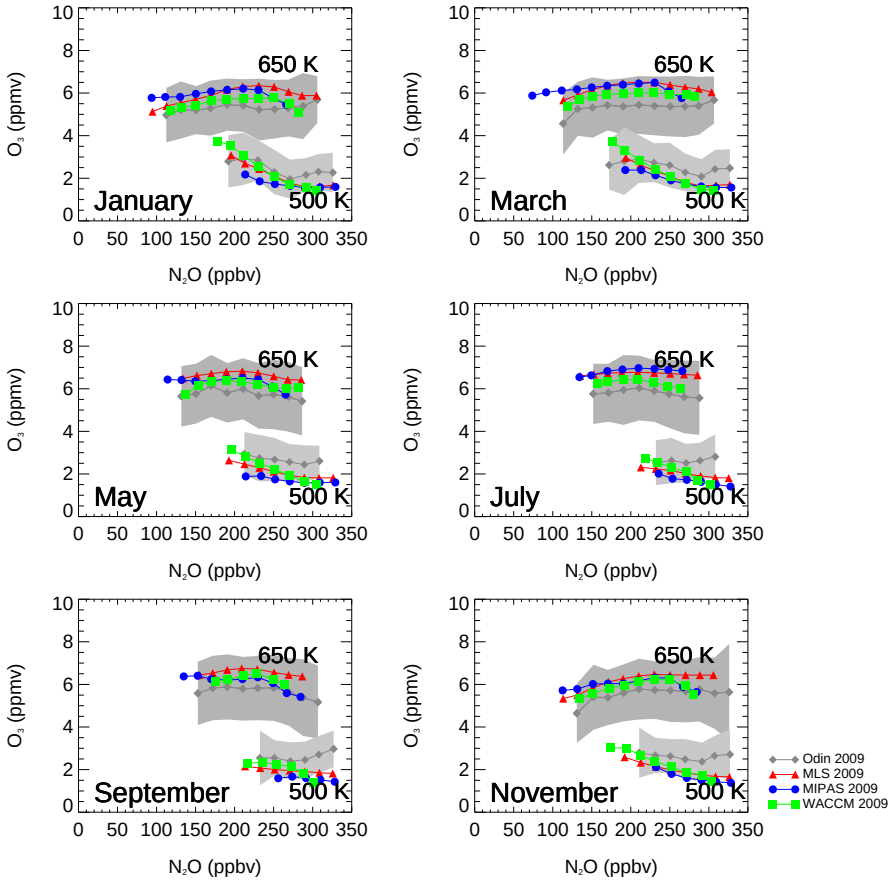

Fig. 13. Same as Fig. 11, but for the year 2009 and including Aura/MLS and SD-WACCM data. Odin/SMR (grey diamonds), Aura/MLS (red triangles) and SD-WACCM (green squares).

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 at 500 ± 25 K. In particular, 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 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 by a 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 intercomparisons. 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 interannual variability of monthly averages of

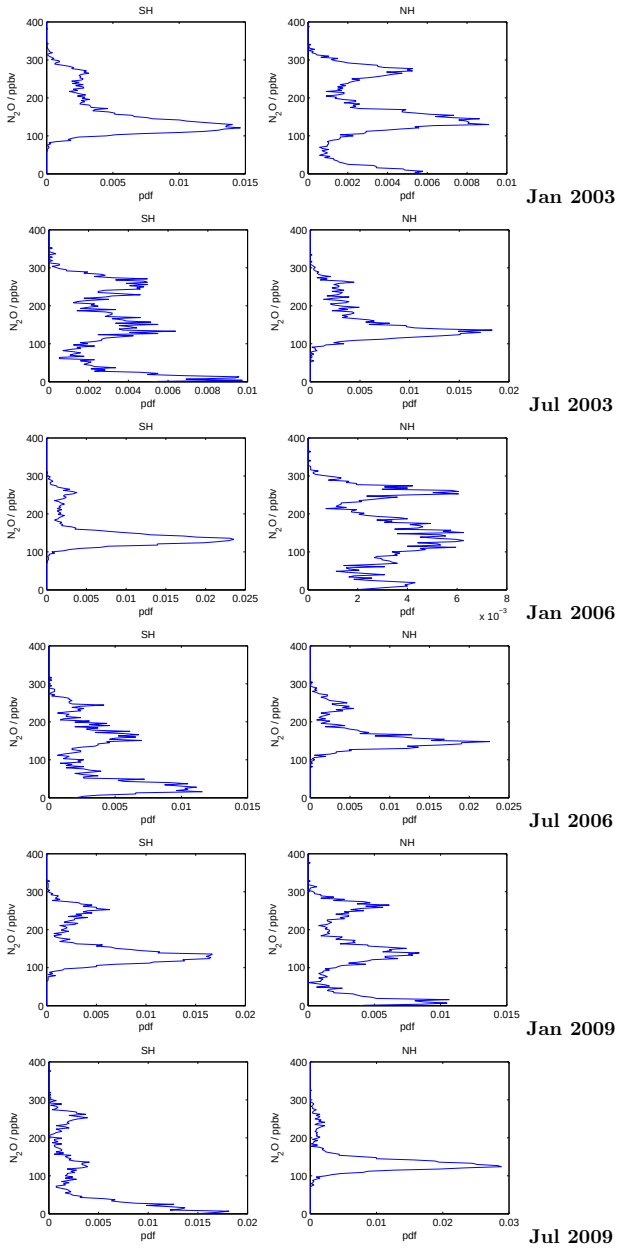

Fig. 14. Probability density functions (PDFs) derived from Odin/SMR measurements (NH and SH) at 650 K for January and July 2003, January and July 2006 and January and July 2009. The QBO was in its easterly phase in 2003 and 2009 and in its westerly phase in 2006. Note: the x-scale is not the same for all figures.

N_2O and O_3 has been assessed. We found that the interannual 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 −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 caused in part 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 an 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. Thus, the results of model evaluation or satellite data intercomparison can be to some part be influenced by uncertainties in the satellite data used as reference for such a study and must be taken into account.

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. Furthermore, 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 $\pm$ 0.12 ppbv in 2005 Forster et al., 2007). However, the difference we found in our monthly averages between models and observations is much higher than the difference to the ground-based N_2O measurements. That is, we found a difference of 20–40 ppbv between model and satellite measurements compared to 10–20 ppbv between satellite and ground-based measurements. N_2O is significantly destroyed in the tropical stratosphere at altitudes above 25 km; thus, below 25 km we can expect an N_2O mixing ratio that has decreased by 10-20 ppbv compared to the mixing ratio found in the troposphere (Warneck, 1988; Kuttipurath et al., 2010). Such a high positive bias between model and simulations (20–40 ppbv) was not found in validation studies applying Odin/SMR v2.1 N_2O

observations. We conclude that this difference is probably caused by the combination of N_2O values from model simulations being too low, and the mixing ratios measured by Odin/SMR being too high. In fact, the simulation of too low N_2O is a common feature of most models as it was discussed in e.g. Kuttipurath et al. (2010) and references therein. Note, though the satellites derive too high N_2O mixing ratios, this does not conflict with the results derived in this study. This bias/noise error in the satellite data sets does not affect the interannual variability. Further, the differences caused by the uncertainties in the satellite instruments can be clearly distinguished from model deficiencies. Thus, the monthly averages derived from the satellite data sets considered here are suitable for model evaluation and satellite intercomparisons.

Since in our method the data are 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 interannual 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 are 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 interannual variability of these values is most probably caused by local dynamical processes in the tropics such 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 not only that these data sets are generally in good agreement, but that also known 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 data intercomparisons.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/13/3619/2013/acp-13-3619-2013-supplement.pdf.

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