Atmos. Chem. Phys. Discuss., 10, 9153–9171, 2010
www.atmos-chem-phys-discuss.net/10/9153/2010/

Atmospheric Chemistry and Physics Discussions

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Response of tropical stratospheric O_3, NO_2 and NO_3 to the equatorial Quasi-Biennial Oscillation and to temperature as seen from GOMOS/ENVISAT

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Received: 29 January 2010 – Accepted: 28 March 2010 – Published: 9 April 2010

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

Abstract

The stellar occultation spectrometer GOMOS (Global Ozone Monitoring by Occultation of Stars) on ESA's Envisat satellite measures vertical profiles O_3, NO_2 and NO_3 with a high long-term stability due to the self-calibrating nature of the technique. More than 6 years of GOMOS data from August 2002 to end 2008 have been analysed to study the inter-annual variation of O_3, NO_2 and NO_3 in the tropics. It is shown that the QBO of the equatorial wind induces variations in the local concentration larger than 10% for O_3 and larger than 25% for NO_2.

Quasi-Biennial Oscillation signals can be found in the evolution of the three constituents up to at least 45 km. We found that NO_3 is positively correlated with temperature up to 40 km in the region where it is in chemical equilibrium with O_3. Above 40 km, NO_3 is no more in equilibrium during night and its concentration is correlated with both O_3 and NO_2. For O_3 and NO_2, our results confirm the existence of a transition from a dynamical control of O_3 below 28 km with O_3 correlated with NO_2 and temperature and a chemical/temperature control between 28 and 38 km with O_3 anti-correlated with NO_2 and temperature. Above 38 km and up to 50 km a regime never described before is found with both O_3 and NO_2 anti-correlated with temperature. For the NO_2/temperature anti-correlation, our proposed explanation is the modulation of the N_2O ascent in the upper stratosphere by the QBO and the modulation of the Brewer-Dobson circulation. The oxidation of N_2O is the main source of NO_y in this altitude region. An enhancement of the ascending motion will cool adiabatically the atmosphere and will increase the amount of N_2O concentration available for NO_y formation.

1 Introduction

The Quasi-Biennial Oscillation (QBO) of the equatorial wind is a main feature of the dynamics of the tropical stratosphere (see the review by Baldwin et al., 2001). Equatorial winds are observed to exhibit a downward propagating oscillation with a mean

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period of about 28 months. Lindzen and Holton (1968) showed in a 2-D model that the QBO could be driven by a spectrum of vertically propagating gravity waves. The QBO induces variations in the temperature field and in the vertical and meridional transport of constituents. The easterly QBO phase (winds blowing from east) in the lower stratosphere is associated with an upwelling at the equator inducing an adiabatic cooling (Reed et al., 1964) and a reduction in total column ozone. The opposite is found during the westerly QBO phase. A few studies were made to detect a QBO signal in observations of the concentration of ozone and other constituents in the tropical stratosphere. Logan et al. (2003) showed a QBO signature in tropical ozone sonde data. QBO signals in Stratospheric Aerosol and Gas Experiment (SAGE) O_3 and NO_2 data and their phase relationship were analysed by Zawodny and McCormick (1991), Hasebe (1994) and Chipperfield et al. (1994). Randel and Wu (1996) showed a ±4% modulation in the ozone equatorial column Dobson from Solar Backscatter Ultraviolet (SBUV) data. Schoeberl et al. (2008) looked at the QBO and annual cycle variations in trace gases from HALOE and Aura MLS observations. Fadnavis and Beig (2009) analysed temperature and ozone QBO using UARS MLS data. Gray and Pyle (1989) performed the first coupled 2-D QBO simulation of the O_3 QBO. Chipperfield et al. (1994) studied the link between QBO signals on O_3 and NO_2 using a 2-D model. Tian et al. (2006) used a fully coupled chemistry-climate model that generates its own QBO in wind and temperature to examine the QBO signals in stratospheric trace gases.

The goal of this study is to evaluate, using Global Ozone Monitoring by Occultation of Stars (GOMOS) data from August 2002 to December 2008, the QBO impact on O_3, NO_2 and NO_3 vertical profiles in the Tropics and their cross-correlation with temperature derived from European Centre for Medium-range Weather Forecasts (ECMWF) meteorological analyses. GOMOS instrument is described in Sect. 2. The data selection and processing strategy are presented in Sect. 3. The inter-annual variability of both O_3 and NO_2 is shown in Sect. 4 and their relationship with QBO, NO_3 and temperature in Sect. 5. The results are discussed in Sect. 6 and we conclude in Sect. 7.

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2 The GOMOS instrument

GOMOS, one of ten instruments on ESA's ENVISAT mission, is a stellar occultation spectrometer aimed to build a global climatology of ozone and related species in the middle atmosphere (15 to 100 km) with a very high accuracy using the technique of stellar occultation (Bertaux et al., 2004). Bertaux et al. (2010) give an overview of the scientific results. Each constituent can be identified by its absorption spectrum. The atmospheric transmission spectrum is equal to the ratio between the star spectrum absorbed and scattered by the atmosphere and the reference star spectrum measured outside the atmosphere. As the reference spectrum is measured at the beginning of each occultation, we can consider that GOMOS is a self-calibrated instrument, independent of any radiometric calibration. Furthermore the stellar occultation technique allows a perfect knowledge of the tangent altitude, depending only on the geometry of the light path between the star and the satellite. The 250–680 nm spectral domain is used for the determination of O_3, NO_2, NO_3 and aerosols from the upper troposphere to the mesosphere. In addition, two high spectral resolution channels centred at 760 and 940 nm allow measuring O_2 and H_2O and two fast photometers (1 kHz sampling rate) are used to correct star scintillation perturbations and to determine high vertical resolution temperature profiles. Global latitude coverage is obtained with up to 40 stellar occultations per orbit from South Pole to North Pole. The data quality depends on illumination conditions and on star brightness and temperature. Data acquired on dark limb (night-time) are of better quality than on bright limb (day-time) because of a smaller perturbation by background light. Hot stars have a strong UV emission and allow obtaining ozone profiles up to the lower thermosphere using UV absorption bands. Cold-star ozone profiles are limited to the stratosphere using visible Chappuis band. NO_2 profiles extend from 20 to 50 km in typical non-polar conditions and up to 70 km during polar winter when a strong NO_2 enhancement occurs. The typical accuracy is 5% for O_3 (Kyrölä et al., 2006) and 15–20% for NO_2 (Hauchecorne et al., 2005).

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3 Data selection and processing

One difficulty of the stellar occultation technique is the sensitivity of the data quality to star characteristics (brightness, temperature) and to viewing geometry (sun azimuth and elevation, occultation obliquity). Furthermore, for a given star, the latitude of the tangent point depends on the day of the year and the latitude coverage changes with time. In order to build 3-D NO_2 and NO_3 fields as homogeneous as possible, a 3-step selection and averaging procedure is applied:

(1) As explained in Hauchecorne et al. (2005) we use only the 70 brightest stars and we discard daylight observations (illumination flag "bright limb") to eliminate too noisy O_3, NO_2 and NO_3 profiles. Concentration profiles are converted in mixing ratio profiles using a combined atmospheric density profile from ECMWF analyses below 1 hPa (about 48 km) and from climatological MSIS-2000 model (Hedin, 1991) above 1 hPa.

(2) For a given star and during one day, the latitude of the tangent point is almost constant and the star is occulted 14 or 15 times (once per orbit) with a 25° westward longitude shift per orbit. A daily mean profile is computed as the weighted average of all available profiles amongst the 14–15 ones. Weights are inversely proportional to the squared error estimates. Each daily mean profile can be considered as a zonal average at the mean latitude of the tangent point.

(3) All daily mean profiles of ozone mixing ratio for one month and within the tropical region (15° S to 15° N latitude band) are averaged. The evolution of tropical ozone for the period August 2002 to December 2008 is thus obtained. A similar processing is applied to NO_2 and NO_3 data.

Finally, for each altitude a multi-parameter fit is performed to extract the semi-annual and annual component, the response to QBO wind at 30 hPa and 10 hPa and a linear trend. 30 hPa and 10 hPa levels are chosen because they are almost orthogonal and

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the wind evolution at 30 hPa follows those at 10 hPa with a delay of about a quarter of QBO period.

4 Interannual variability

For a given star, it is possible to obtain 1 profile per orbit with latitude of the tangent point varying slowly in time and the longitude shifted by 25° westward from one orbit to the following. It is then possible to obtain 14 to 15 profiles during one day at constant latitude around the globe and to derive a daily zonal mean vertical profile for each constituent. Furthermore, the evolution of the latitude of the tangent point is very reproducible from one year to another and it is possible to compare profiles obtained at the same date for consecutive years. It should be noted that, due to limitations in possible star pointing azimuth since 2005, we are not able to follow all stars between 2002 and 2008 in a periodic manner. The star S029 in GOMOS catalogue (Beta Carinae) has been chosen because it is always occulted in the tropical region. Figure 1 shows the evolution of NO_2 and O_3 at 30 km during 4 consecutive years. The day-to-day variation is quite low and probably within the instrumental noise but the year-to-year variation is quite large, up to 25% for NO_2 and up to 10% for O_3 with an apparent 2 years period. Furthermore, it is clear that the variations of O_3 and NO_2 are in opposite directions. This seems to indicate that the concentration of these 2 species are anti-correlated and linked with the QBO phase.

5 Relationship with QBO and temperature

Figure 2 presents the evolution of monthly averaged NO_2 and O_3 mixing ratios normalized by their mean seasonal value. The evolution of the 2 constituents is clearly correlated with the QBO signal in equatorial wind at 10 hPa (about 30 km, right scale in the figure). We consider the QBO wind at 10 hPa as a good proxy because this

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latitude corresponds to a peak of the ozone QBO (Fadnavis and Beig, 2009). The sign of the correlation with the 10 hPa wind is changing with altitude. For NO_2, there is a negative correlation between 22 and 27 km and a positive correlation above at least up to 40 km. For O_3 the correlation is positive between 22 and 28 km, negative between 28 and 36 km and positive again between 36 and 45 km. Below 22 km the signal is less clear, probably due to the increase of measurement uncertainties. Figure 2 shows clearly that the variation of O_3 and NO_2 are correlated or anti-correlated between them depending on the altitude. This O_3-NO_2 correlation (Fig. 3) is found to be positive between 20 and 24 km, not significant between 24 and 27 km, strongly negative between 27 and 38 km with a maximum of anti-correlation up to −0.85 at 30 km and positive again between 38 and 55 km. The relative variation of O_3 is between −40% to +40% of that of NO_2. These phase relationships will be discussed in Sect. 6.

The NO_3 concentration is known to be very sensitive to temperature (Marchand et al., 2007). It is then not surprising to observe that the evolution of monthly averaged NO_3 mixing ratios is well correlated with the evolution of ECMWF temperature interpolated at the location of observations (Fig. 4). The downward propagation of the QBO signal in both temperature and NO_3 can be followed from 45 km to 27 km.

In order identify all the phase relationships between variables, Fig. 5 presents the correlation coefficient of O_3, NO_2 and NO_3, with the other constituents, with temperature (T) and with the 2 QBO proxies, the equatorial wind at 10 hPa (U_{10}) and at 30 hPa (U_{30}). For O_3/NO_2 we found again the succession of positive and negative correlation altitude domains shown in Fig. 3. For O_3/NO_3 the correlation is found positive between 25 and 30 km and negative between 35 and 45 km and positive between 47 and 53 km. For NO_2/NO_3 the correlation is positive between 29 and 42 km and negative between 44 and 53 km. For O_3/T, the correlation is positive between 20 and 28 km and negative between 30 and 52 km in relation with the change between the ozone dynamically controlled region below 28–30 km and ozone chemically controlled region above 30 km (Chipperfield et al., 1994). O_3/U_{10} and O_3/U_{30} present a succession of positive and negative regions with a shift in altitude between the 2 corresponding to the altitude

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difference between the levels 10 hPa (~30 km) and 30 hPa (~24 km). NO_2 is positively correlated with U_{10} from 28 to 45 km, with U_{30} from 25 to 32 km, showing again the altitude shift between the two wind levels and with T from 32 to 38 km. NO_3 is positively correlated with T between 25 and 46 km. This is due the strongly positive temperature dependence of the chemical reaction forming NO_3.

$$NO_2 + O_3 + O \longrightarrow NO_3 + O_2 \quad (1)$$

Marchand et al. (2007) showed that the temperature can be derived from O_3 and NO_3 GOMOS measurements between 30 and 40 km where NO_3 is in chemical equilibrium during night. Above ~40 km, NO_3 is no more in equilibrium because the formation time is longer than the time interval between sunrise and GOMOS observations (~4 h) and is positively correlated with both NO_2 and O_3.

6 Discussion

There are only few studies concerning the relation between tropical NO_2, O_3 and the QBO signal. Chipperfield et al. (1994) studied the QBO signal in SAGE II O_3 and NO_2 between 1984 and 1991 with the help of a 2-D radiative-dynamical-chemistry model. They observed a change of sign in SAGE II O_3 signal at 28 km, in agreement with our results. The change of sign was explained by the transition from dynamical control below to chemical control above this altitude and was reproduced by the model. The anti-correlation between O_3 and temperature is due to the strong temperature dependence of the ozone loss reaction in the Chapman cycle (Brasseur and Solomon, 2005):

$$O_3 + O \longrightarrow 2\,O_2 \quad (2)$$

Chipperfield et al. (1994) explained the anti-correlation between O_3 and NO_2 in the chemical region by the catalytic destruction of O_3 by NO_x. However this interpretation is still in discussion and Butchart et al. (2003) were able to reproduce the O_3 QBO

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signal in their 3-D chemistry-transport model without QBO induced variations in NO_y. Tian et al. (2006) also showed the change near 28 km from dynamical to chemical control of O_3 by using a fully coupled chemistry-climate model. The phase change for NO_y was near 25 km, 3 km lower than for O_3. Our results confirm the anti-correlation between QBO signals in O_3 and NO_2 up to 36 km and the transition around 28 km between the dynamical control of O_3 below and its photochemical control above.

Above 36 km we observe a second transition with O_3 and NO_2 in phase and anti-correlated with temperature. Most observational studies are limited to altitude below 40 km and are not able to capture this feature. We investigate two possible explanations for this observed O_3-NO_2 correlation:

(1) The first hypothesis is the temperature dependence of chemical reactions involved in O_3 and NO_y photochemistry in the upper stratosphere. The mechanism involved for the O_3 anti-correlation with temperature is the same than between 28 and 36 km. The transformation of NO_2 into reservoir species, N_2O_5 and HNO_3, increases with decreasing temperature (Brasseur and Solomon, 2005) due to the temperature dependence of the chemical reactions:

$$NO_2 + O_3 \longrightarrow NO_3 + O_2 \quad (3)$$

for the formation of N_2O_5 and:

$$NO_2 + OH + M \longrightarrow HNO_3 + M \quad (4)$$

for the formation of HNO_3. This hypothesis is in agreement with the model results of Tian et al. (2006), presented in their Fig. 9, showing an anti-correlation in the QBO response of NO_y and NO_x at 45 km. However, more than 90% of NO_y is under the form of NO_x (NO_2 during night) at altitudes higher than 40 km according to Brasseur and Solomon (2005) and this mechanism is probably not very efficient.

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(2) The second hypothesis is the modulation of N_2O ascent in the upper stratosphere due to a modulation the QBO and the modulation of the Brewer-Dobson circulation. In the equatorial region there is still a significant amount of N_2O up to at 45–50 km. Jin et al. (2009) reported a 35 ppbv N_2O mixing ratio at 2 hPa (about 45 km) using Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) data in agreement with the Canadian Middle Atmosphere Model (CMAM) simulations. An increase of the ascending motion will cause an adiabatic cooling and will bring more N_2O in the upper stratosphere, providing an increase of the source of NO_x from N_2O oxidation by atomic oxygen. This mechanism would explain the observed anti-correlation between NO_2 and temperature in the upper stratosphere. Further observational and model studies are needed to test our hypothesis and to better understand the mechanisms involved in the O_3/NO_2/temperature relationship above 38 km.

7 Conclusions

We analysed more than 6 years of night-time GOMOS O_3 and NO_2 and NO_3 observations in the tropical region to study the inter-annual variability and extract the QBO signal. For a given altitude, latitude and date, the year to year variations in the local concentration can be larger than 10% for O_3 and larger than 25% for NO_2. These variations are mainly due the modulation of the vertical transport and the temperature by the QBO induced circulation.

QBO signals can be found in the evolution of the three constituents up to at least 45 km. We found that NO_3 is positively correlated with temperature up to 40 km in the region where it is in chemical equilibrium with O_3. Above 40 km, NO_3 is no more in equilibrium during night and its concentration is correlated with both O_3 and NO_2.

For NO_2 and O_3, our results confirm the existence of a transition from a dynamical control of O_3 below 28 km, with O_3 correlated with NO_2 and temperature, and a chemical/temperature control of O_3 between 28 and 38 km, with O_3 anti-correlated with NO_2

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Acknowledgements. This study was supported by the European Commission within the SCOUT-O_3 project (contract 505390-GOCECT-2004) and GEOMON project (contract number GOCE-CT-2007-036677) and by the European Space Agency and by the French funding agencies: CNRS-INSU and CNES.

The publication of this article is financed by CNRS-INSU.

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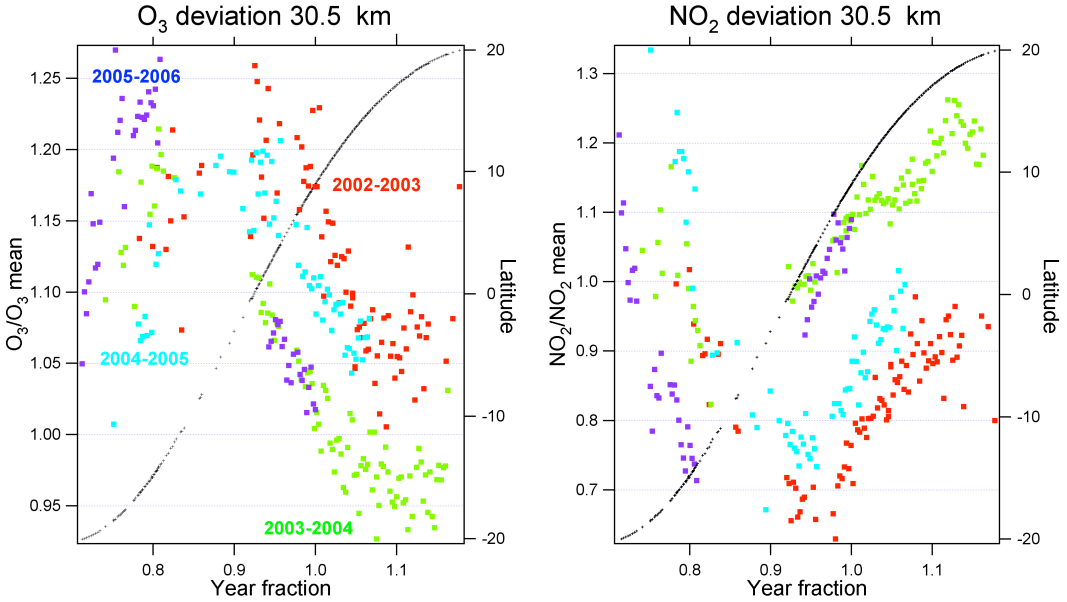

Fig. 1. Evolution of NO_2 and O_3 mixing ratios at 30.5 km obtained from star S029 (Beta Carinae) occultations as a function of the fraction of the year (numbers greater than 1 indicates date after 1 January). Concentrations have been divided by the mean value at 30.5 km for the full period.

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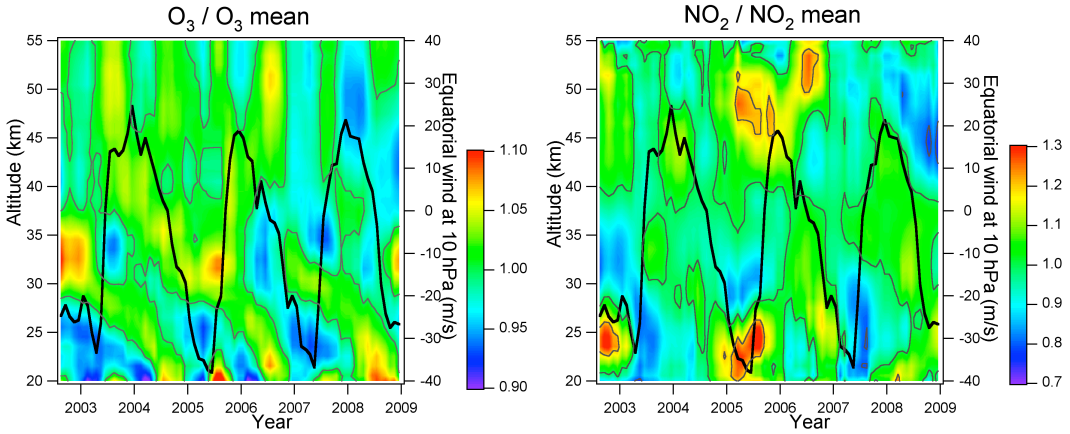

Fig. 2. Evolution of O_3 and NO_2 mixing ratios in the latitude band 15° S–15° N normalized by their seasonal mean evolution. The equatorial wind at 10 hPa (thick black line) is indicated with the right scale.

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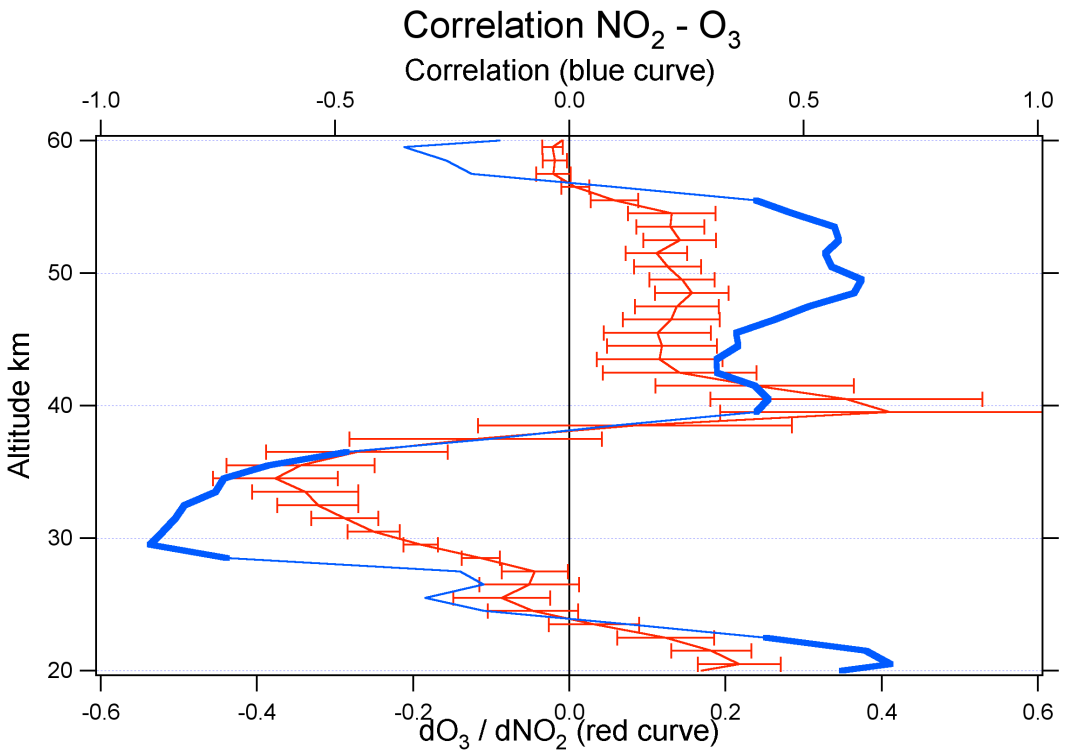

Fig. 3. Correlation between the deviations of NO_2 and O_3 from their mean seasonal value at the same altitude (blue curve, values larger than ±1 standard deviation uncertainty are represented with a thick line) and relative variation of O_3 per relative unit variation of NO_2 (red curve, the 1-σ uncertainty is plotted).

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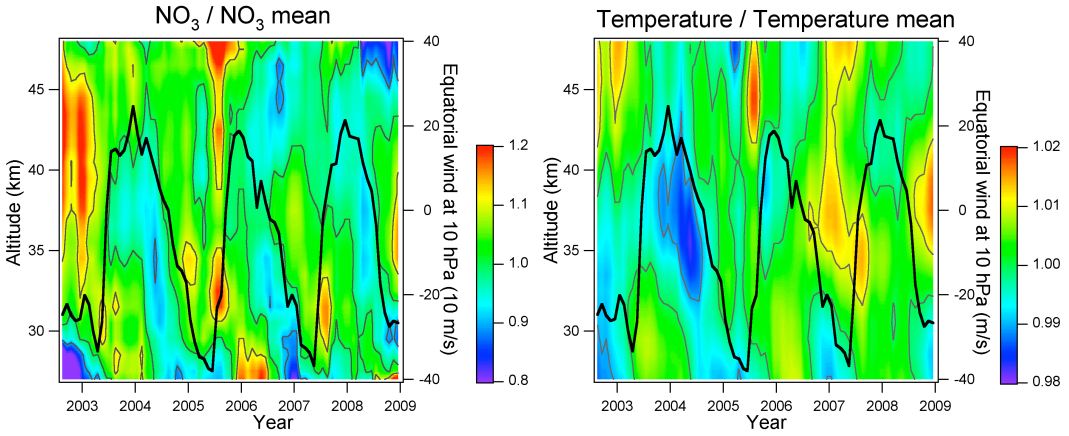

Fig. 4. Evolution of NO_3 mixing ratio and ECMWF temperature in the latitude band 15° S–15° N normalized by their seasonal mean evolution. The equatorial wind at 10 hPa is indicated (right scale). The plot has been limited to altitudes (from 27 km to 48 km) with good signal to noise ratio in NO_3 mixing ratio.

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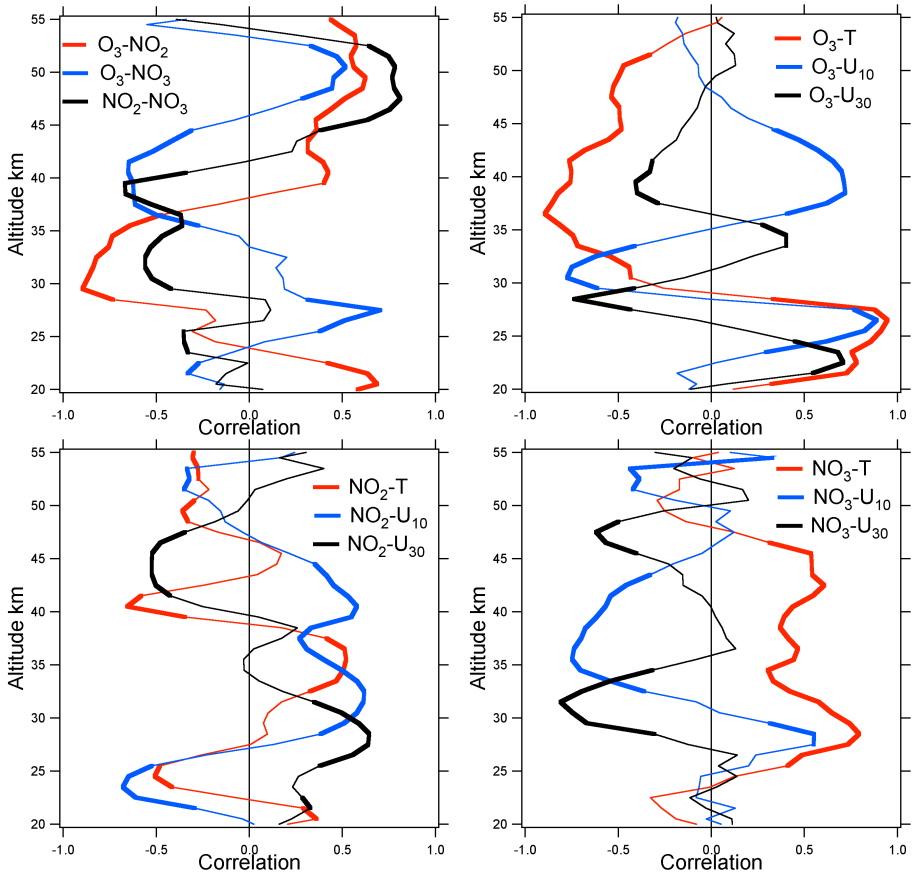

Fig. 5. Correlation coefficients between the deviation from their seasonal evolution of O_3, NO_2 and NO_3 mixing ratios (top left), O_3, temperature and equatorial zonal wind at 10 hPa and 30 hPa (top right), NO_2, temperature and equatorial zonal wind (bottom left), NO_3, temperature and equatorial zonal wind (bottom right). Values larger than ±1 standard deviation uncertainty are represented with thick lines.

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