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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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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 40 km. We found that NO_3 is positively correlated with temperature up to 45 km in the region where it is in chemical equilibrium with 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 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 different regime is found with O_3 and NO_2 correlated with each other and anti-correlated with temperature. For the NO_2/temperature anti-correlation in the upper stratosphere, our proposed explanation is the modulation of the N_2O ascent by the QBO up to 45 km. 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.

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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 blowing alternately from east and from west with a mean period of about 28 months (Fig. 1) and with a downward propagation of regions of westerly and easterly winds. Reed (1964) was the first to propose an empirical model for the QBO, explaining the relation observed between zonal wind and temperature. The region with negative vertical gradient of equatorial zonal wind (region situated between westerly winds below and easterly winds above) is associated with an upwelling at the equator inducing an adiabatic cooling. The opposite is found when the vertical gradient of zonal wind is positive. This feature is limited to the 15° S to 15° N latitude band. The subtropical region (15° to 30° latitude) exhibits wind and temperature variations in opposite phase with the equatorial region as a result of the residual cell circulation induced by QBO with upwelling (downwelling) at the Equator and downwelling (upwelling) in the sub-tropics. Lindzen

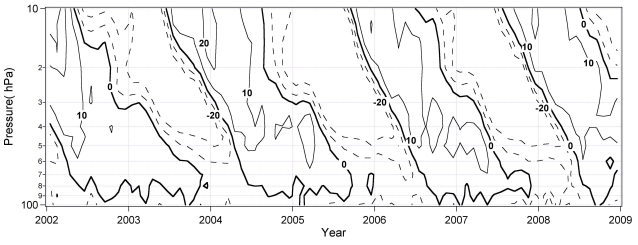

Fig. 1. Time-height section of the equatorial QBO wind updated from Naujokat (1986). Easterly wind contours (negative values) are plotted with dashed lines.

and Holton (1968) showed in a 2-D model that the downward propagating QBO structure could be driven by a spectrum of vertically propagating gravity waves with phase speeds in both eastward and westward directions, each wave accelerating the wind in the direction of its phase propagation at the altitude where its momentum flux is transferred to the zonal flow. The QBO induces variations in the vertical and meridional transport of constituents and in the rate of temperature dependent chemical reactions, leading to changes in the atmospheric composition. 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 $\pm$4% modulation in the ozone equatorial column 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. The 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.

2 The GOMOS instrument

GOMOS, one of ten instruments on ESA's ENVISAT mission, is a stellar occultation spectrometer aimed at building 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 stellar spectrum absorbed and scattered by the atmosphere and the reference stellar 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 gives an excellent 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 the measurement of O_2 and H_2O and two fast photometers (1 kHz sampling rate) are used to correct stellar 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 stellar brightness and temperature. Data acquired on dark limb (night-time) are of better quality than for the bright limb (day-time) because of a smaller perturbation by background light. Hot stars have a strong UV emission and allow obtaining ozone profiles to be obtained up to the lower thermosphere using UV absorption bands. Cool-star ozone profiles are limited to the stratosphere using the 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 at al., 2005).

3 Data selection and processing

One difficulty with the stellar occultation technique is the sensitivity of the data quality to stellar 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

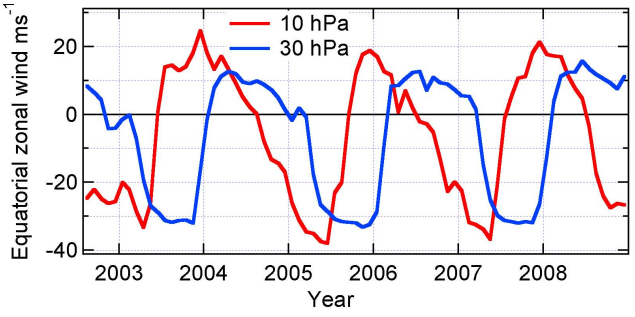

Fig. 2. Evolution of the equatorial QBO wind at 10 and 30 hPa updated from Naujokat (1986).

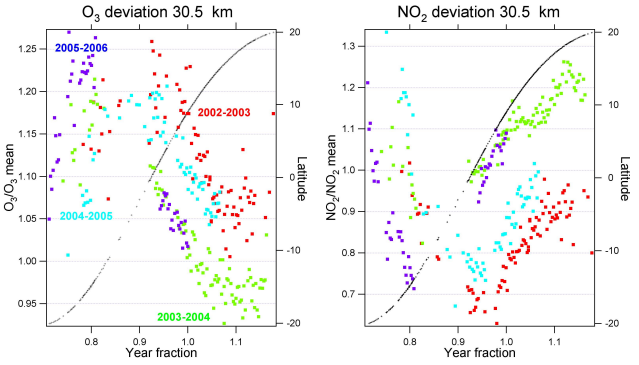

Fig. 3. Evolution of NO_2 and O_3 mixing ratios at 30.5 km obtained from star S029 (Beta Carinae) occultations as a function of the fraction of the year (numbers greater than 1 indicates dates after 1st January). Concentrations have been divided by the mean value at 30.5 km for the full period.

(illumination flag "bright limb") to eliminate noisy O_3, NO_2 and NO_3 profiles. Concentration profiles are converted to mixing ratio profiles using a combined atmospheric density profile from ECMWF analyses below 1 hPa (about 48 km) and from the 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 selected from the 14–15 available. 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 the correlation coefficient between each pair of parameters for O_3, NO_2, NO_3, ECMWF temperature and QBO winds at 30 hPa and at 10 hPa is computed. Deviations from their mean value for the full period at the given altitude are used to compute the correlation coefficient. QBO winds are Singapore (1° N) zonal wind radiosonde data compiled by the Berlin Free University updated from Naujokat (1986). Data are downloaded from the web site:

http://www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo.

QBO winds at 30 hPa and 10 hPa are chosen because they are almost orthogonal (correlation coefficient 0.10) and the wind evolution at 30 hPa follows that at 10 hPa with a delay of about a quarter of the QBO period as seen in Fig. 2.

4 Interannual variability

For a given star, it is possible to obtain one profile per orbit with the latitude of the tangent point varying slowly in time and the longitude shifted by 25° westward from one orbit to the next. 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 azimuths since 2005, we are not able to follow all stars between 2002 and 2008 in a periodic manner. The star S029 in the GOMOS catalogue (Beta Carinae) has been chosen because it is always occulted in the tropical region. Figure 3 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 year 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 most likely linked with the QBO phase. This QBO link will be investigated in detail in the next section. Our analysis concentrates on the response of stratospheric constituents to the QBO that appears to be the dominant contributor to our results. We are aware that there may be some influence of other inter-annual variability, in particular ENSO (El Nino-Sothern Oscillation) but also solar variability and the increase of greenhouse gases. However it is not the goal of this paper to study these other terms due to the limited length of the data series (6 years and 5 months).

5 Relationship with QBO and temperature

Figure 4 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 (∼31 km). We consider the QBO wind at 10 hPa as a good proxy because this altitude 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

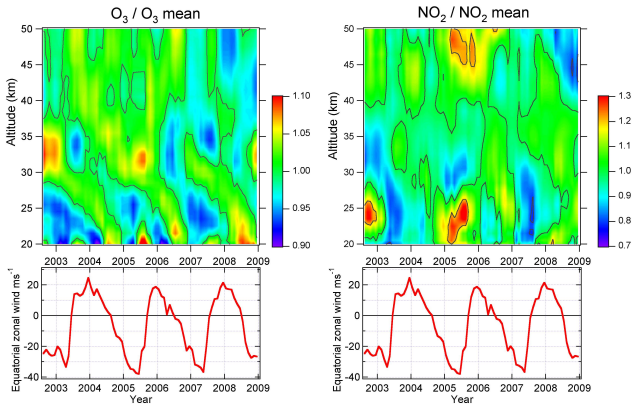

Fig. 4. Top) Evolution of O_3 and NO_2 mixing ratios in the latitude band 15° S–15° N divided by their mean value at the given altitude for the full period; Bottom) Equatorial zonal wind at 10 hPa updated from Naujokat (1986).

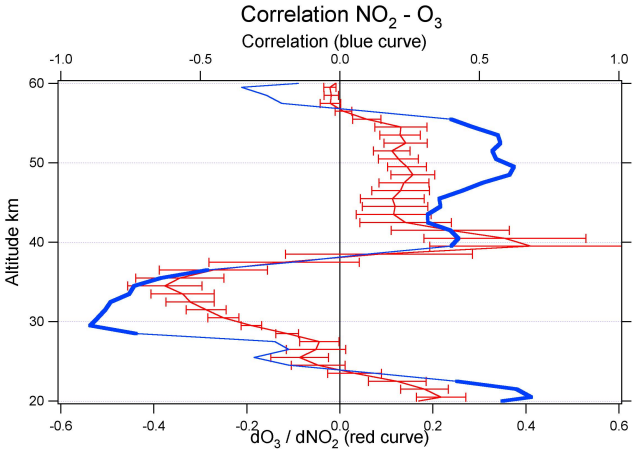

Fig. 5. Correlation between the deviations of NO_2 and O_3 from their mean value for the full period at the given altitude (blue curve, values larger than ± 1 standard deviation uncertainty are represented with a thick line) and relative deviation of O_3 per unit of relative deviation of NO_2 (red curve, the 1-sigma uncertainty is plotted).

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. Fig. 4 shows clearly that the variation of O_3 and NO_2 are correlated or anti-correlated depending on the altitude. This O_3 - NO_2 correlation (Fig. 5) 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 to the location of observations (Fig. 6) between 25 and 45 km. The

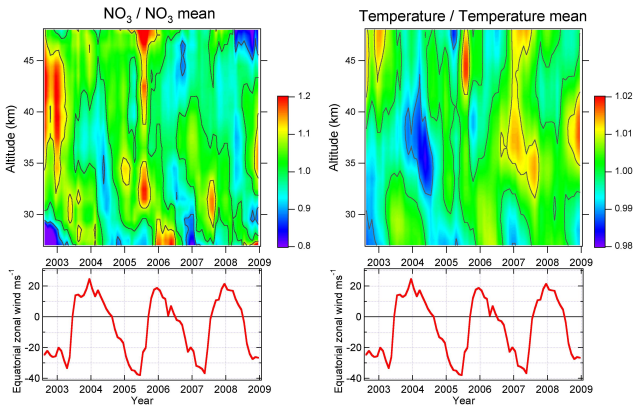

Fig. 6. Top) Evolution of NO_3 mixing ratio and ECMWF temperature in the latitude band 15° S–15° N their mean value at the given altitude for the full period. The plot is limited to altitude range 27–48 km to exclude regions where the signal to noise ratio in NO_3 mixing ratio is to poor; Bottom) Equatorial zonal wind at 10 hPa updated from Naujokat (1986).

limitation of the good correlation to this altitude range is not due to physical or chemical processes but to the increase of NO_3 concentration uncertainty outside this range preventing the detection of any significant correlation (Hauchecorne et al., 2005). The NO_3-temperature correlation is due to the strongly positive temperature dependence of the chemical reaction forming NO_3:

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad \text{(R1)}$$

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.

The evolution of O_3, NO_2, NO_3 and temperature seems to be dominated by the QBO up to about 40 km and the downward propagation of the QBO signal can be followed from 40 km down to 27 km for NO_3 and to 22 km for O_3 and NO_2. Above 40 km the QBO signature is less clear and the interpretation of the observed correlations has to be made with caution. However we consider it interesting to look at the sign of the correlation with temperature even if the causes of these temperature changes are not fully understood.

In order to identify all phase relationships between variables, Fig. 7 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. 5. As expected T is alternately correlated and anti-correlated with U_{10} and with U_{30} between 20 and 45 km with a downward shift in altitude from U_{10} to U_{30}. This feature is consistent with the fact that the maximum of warming occurs where the vertical gradient of zonal wind is maximum, a few km below the altitude of wind maximum, at 26 and 21 km for the correlation with U respectively at 10 hPa ($\sim$31 km) and 30 hPa ($\sim$24 km). For O_3/T, the correlation is positive between 20 and 28 km and negative between 30 and 50 km related to 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

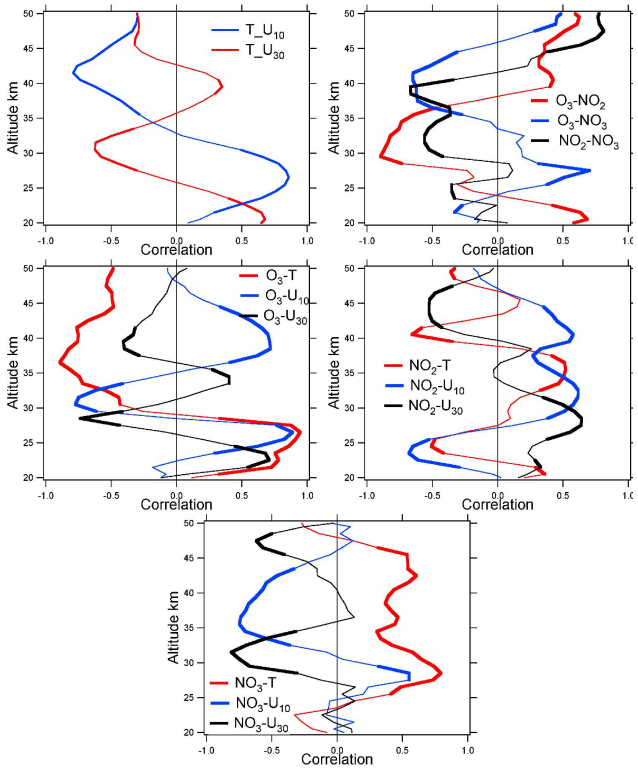

Fig. 7. Correlation coefficients between the deviation from their mean value for the full period at the given altitude of; Top left) Temperature (T) and equatorial wind at wind at 10 hPa and 30 hPa (U_{10} and U_{30}); Top right) O_3, NO_2 and NO_3 mixing ratios (top left); Middle left) O_3 and T, U_{10} and U_{30}; Middle right) NO2 and T, U_{10} and U_{30}; Bottom) NO_3 and T, U_{10} and U30. Values larger than ± 1 standard deviation uncertainty are represented with thick lines.

positive and negative regions with a downward shift in altitude between U_{10} to U_{30} corresponding to the altitude difference between the levels as for the correlation with temperature. For NO_2/T the correlation is negative between 23 and 26 km, positive between 28 and 38 km and negative again around 40 km and at 48–50 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.

6 Discussion

There are only a 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 2D 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):

$$O_3 + O \rightarrow 2O_2 \quad \text{(R2)}$$

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 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 38 km and the transition around 28 km between the dynamical control of O_3 below and its photochemical control above.

Above 38 km we observe a second transition with O_3 and NO_2 correlated with each other and anti-correlated with temperature. Most observational studies are limited to altitudes 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 between 28 and 36 km. The transformation of NO_2 into the 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:

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \quad \text{(R3)}$$

$$NO_2 + NO_3 + M \rightarrow N_2O_5 + M \quad \text{(R4)}$$

for the formation of N_2O_5 and:

$$NO_2 + OH + M \rightarrow HNO_3 + M \quad \text{(R5)}$$

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 in 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.

2) The second hypothesis is the modulation of N_2O ascent in the upper stratosphere due to a modulation the QBO at least up to 45 km and by any other dynamical process, for instance 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 Conclusion

We analysed more than 6 years of night-time GOMOS O_3, 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 40 km. We found that NO_3 is positively correlated with temperature up to 45 km in the region where it is in chemical equilibrium with O_3.

For O_3, our results confirm the existence of a transition from a dynamical control of O_3 below 28 km, with O_3 correlated temperature, and a chemical/temperature control of O_3 between 28 and 38 km, with O_3 anti-correlated with NO_2 and temperature. Above 38 km and up to 50 km a different regime is found with O_3 and NO_2 correlated with each other and 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 at least up to 45 km and by any other dynamical process above 45 km, for instance 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 available for NO_y formation.

Finally, due to the limitation of the length of the GOMOS data series (6 years and 5 months), our analysis concentrates on the QBO that appears to be the dominant contributor to ozone and nitrogen oxide inter-annual variability in the equatorial stratosphere. However it is not excluded that other phenomena (ENSO, solar variability, increase of greenhouse gases) may be have some influence on this variability. We plan to examine this point in further studies.

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