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Comparison of long term series of total ozone and NO_2 column measurements in the southern tropics by SAOZ/NDACC UV-Vis spectrometers and satellites

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Abstract

Long series of ozone and NO_2 total column measurements at the Southern tropics are available from two ground-based SAOZ (Système d'Analyse par Observation Zenithale) UV-visible spectrometers operated within the NDACC (Network for the Detection of Amtospheric Composition Change) deployed in Bauru in S-E Brazil and Reunion Island in the S-W Indian Ocean in 1995 and 1993 respectively. Although at the same latitude, the data show larger columns of both species above the South American continent than above the Indian Ocean. For verifying the reliability of these data before carrying out trend analysis, they have been compared to satellites observations available during the same period. However, since no single satellite was operating from 1995 until present, the comparison requires the building of a composite, called merged satellites series. As systematic differences exist between the individual data sets because of the many differences between instruments, spectral ranges, absorption cross-sections, and retrieval procedures used, the building of such a composite requires thorough evaluation and normalisation of each. From comparisons with SAOZ, the merged satellite data set build with EP-TOMS from 1995 to 2004 and OMI-TOMS from 2005 to 2012 are found best for ozone in the Southern tropics. After correction for biases with SAOZ, both are confirming the larger ozone columns reported by SAOZ above South America compared to the Indian Ocean shown to origin from ozone production by lightning NO_x (LNO_x) over the continent in the summer and the advection from Africa of ozone produced by biomass burning emissions in the winter. For NO_2, best matching the SAOZ is a combination of GOME GDP4 1996–2003 and SCIAMACHY 2003–2012 products, after correction for the photochemical diurnal change of the concentration of the species between the SAOZ twilight observations and the time of satellites overpasses. The merged data series built from the data of these two satellites fully confirms the larger NO_2 column reported by SAOZ above the South American continent as well as and its seasonality. The 35 % larger column above Brazil in the summer is shown to be due to the NO_x production in the upper troposphere by the frequent lightning during

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the thunderstorm season, whereas the winter maximum is shown to come from the larger exchange of NO_x rich air with mid-latitudes in the lower stratosphere due to the more equatorial latitude of the subtropical jet above South America compared to the Indian Ocean.

1 Introduction

In the frame of the international Network for Detection of Atmospheric Composition Change (NDACC), two ground-based SAOZ (Système d'Analyse par Observation Zenithale) UV-visible spectrometers (Pommereau and Goutail, 1988) have been deployed for monitoring the long-tem evolution of atmospheric total ozone and NO_2 columns in the little instrumented southern tropics in Bauru at 22° S, 49° W in S-E Brazil in 1995 and Reunion Island at 21° S, 55° E in the S-O Indian Ocean in 1993. Those data have been extensively used for validating EP-TOMS, GOME-ER2, SCIAMACHY-ENVISAT and OMI AURA satellite observations (Richter et al., 2000, 2004; Piters et al., 2006; Celarier et al., 2008; Hendrick et al., 2011). But although deployed at the same latitude, the measurements of the two instruments are displaying significant differences between the two stations, Bauru showing larger ozone and NO_2 above the South American continent compared to Reunion in the Indian Ocean, and moreover a large seasonal variation of the difference. For checking the reliability of the ground-based observations before further trend analysis, SAOZ data have been compared to those of the same species measured by the several satellites available since 1995, using a merged satellite composite specifically built for checking the robustness of the results of the trend analysis.

The satellite data available in coincidence with the SAOZ measurements are those of EP-TOMS for ozone (McPeters et al., 1998) between 1996–2005, GOME-ERS2 (Burrows et al., 1999) for both O_3 and NO_2 between 1995–2003 in the Southern Hemisphere, SCIAMACHY-ENVISAT (Bovensmann et al., 1999) in 2002–2012 and OMI-AURA (Levelt et al., 2006) from 2004 until present. After a brief recall of the instruments

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characteristics and the products available from each in Sect. 2, their data are compared to those of the SAOZ and a merged satellite composite is built, first for ozone, then for NO_2. Then, the differences between the two stations seen by the satellites and SAOZ, respectively are examined in Sect. 3, and the results are summarised in Sect. 4.

2 Observational data

The data used in this study are the ozone and NO_2 columns from the ground-based SAOZ/NDACC stations at the two tropical stations Reunion Island and Bauru and EP-TOMS, GOME, SCIAMACHY and OMI products for the overpasses of the satellites above the SAOZ stations.

2.1 Ground-based SAOZ UV-visible total NO_2 and O_3 columns

SAOZ O_3 and NO_2 total columns measurements began in 1995 in Bauru and in 1993 in Reunion. The O_3 and NO_2 columns are retrieved using the DOAS (Differential Optical Absorption Spectroscopy) technique at dawn and dusk between 86–91° Solar Zenith Angle (SZA), in the visible Chappuis bands between 450–550 nm for O_3 and 410–530 nm for NO_2 with a spectral resolution of 0.8 nm. The ozone data available are those of the version 2 retrieval algorithm following the recommendation of the NDACC UV-VIS working group described by Hendrick et al. (2011). The largest change compared to the previous SAOZ version 1 retrieval is the use of a daily air mass factors (AMF) calculated from the TOMS V8 ozone profile climatology for converting slant into vertical columns instead of a yearly mean profile. The random error estimated by Hendrick et al. (2011), is 4.7 % and the total accuracy ~5.9 %. For NO_2, since a similar daily profile procedure is not yet available, a yearly mean profile is used built from HALOE (Halogen Occultation Experiment) solar occultation measurements above 20 km complemented for the lower stratosphere and upper troposphere by SAOZ-balloon profiles measured

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in Bauru (Ionov et al., 2008). The total accuracy on the NO_2 vertical column is estimated to $\sim$ 10–15 %.

2.2 EP-TOMS

EP-TOMS, is a 6 wavelengths nadir viewing instrument on the helio-synchronous Earth Probe (EP) satellite, crossing the equator at 11h local time which operated between July 1996 and the end of 2005 (McPeters et al., 1998). The data used here are those of the version 8 retrieval algorithm available on (ftp://jwocky.gsfc.nasa.gov/pub/eptoms/data/overpass/). The ozone measurements are based on a differential pair method at 317.5 and 331.2 nm for a SZA smaller than 70° always available in the tropics (Wellemeyer et al., 2004). The estimated accuracy is about 3 to 5 %. Since 2000, EP-TOMS experienced instrumental problems inducing a 3 % bias in total ozone (Bramstedt et al., 2003). An empirical correction was performed in version 8 to overcome this bias. In 2002, and depending on the latitude, the estimated error was of 4–10 % (WMO, 2007).

2.3 GOME

The GOME instrument (Global Ozone Monitoring Experiment) was launched on 21 April 1995 aboard the ESA ERS-2 satellite, at 795 km altitude on a sun-synchronous orbit of 98.5° inclination. GOME is a nadir-viewing spectrometer, which observes solar radiation reflected or scattered by the atmosphere and the land surface in the UV-Visible spectral range (240–790 nm) with a spatial resolution of 320 × 40 km (Burrows et al., 1999). The satellite passes over the equator during its ascending orbit at 10:30 LT (local time). The instrument was designed to measure trace gases in the troposphere and the stratosphere, including ozone, and was the first space instrument to measure NO_2 total columns. Global coverage is achieved in 3 days.

The ozone data used here are the GOME version GDP4 ESA products retrieved between 325 and 335 nm with an analysis based on the DOAS method and have an

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accuracy of 2.4 to 3.3 % (Van Roozendael et al., 2006; http://atmos.caf.dlr.de/cgi-bin/gdp4/.

For NO_2 two retrievals are available, one from the Institut für Umweltphysik (IUP) of the University of Bremen and one from the ESA GOME Data Processor GDP4. Both are using the same cross-sections (Burrows et al., 1998) at 221 K and the same DOAS spectral analysis method between 425–450 nm. The main difference is in the vertical column retrieval and a normalisation over the Pacific which is applied in the NO_2 retrieval to account for seasonal changes in the GOME solar irradiance measurements. The IUP data are stratospheric columns using AMF derived from the SCIATRAN model (Rozanov et al., 2005). The data are a good estimate of stratospheric columns in clean areas but they may be relatively higher than the reality in polluted areas (Richter and Burrows, 2002). The accuracy is estimated to 5–10 %. The data used here are available on http://www.iup.uni-bremen.de/doas/gomeno2_dataquilt.htm.

The ESA GDP4 version is a total column calculated using a NO_2 profile climatology. (Lambert et al., 2004). The accuracy is estimated between 5 and 10 %, in unpolluted regions. The data used here are available at: http://atmos.caf.dlr.de/cgi-bin/gdp4/. Following the breakdown of the on-board recorder in 2002, the ERS-2 spatial coverage was reduced and no data are available in the southern tropics after this date.

2.4 SCIAMACHY

The SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) instrument was launched on March 2002 on the ENVISAT platform. The satellite crosses the equator at 10:00 LT on a sun-synchronous orbit at 800 km of 98.5° inclination. SCIAMACHY is a spectrometer that measures solar radiation scattered by the atmosphere at limb and nadir in the spectral UV-Visible range (240–790 nm) with a spectral resolution of 0.2 to 1.5 nm and a spatial resolution of 30 × 60 km (Bovensmann et al., 1999). The high spectral resolution and the use of a wide range of wavelengths allow the detection of several trace gases. Because of the alternate nadir and limb observations, its global coverage is of six days, a factor of two lower than that of GOME. The ozone data used in this study are those of the ESA operational off-line processor version 3.01 (ftp-ops-dp.eo.esa.int). This product has been developed based on GOME GDP4.0 (Bracher et al, 2006). The estimated accuracy is about 5 % for SZA lower than 60°. NO_2 columns are those retrieved by the IUP Bremen (http://www.iup.uni-bremen.de/doas/scia_no2dataacve.htm). The analysis is based on the DOAS technique between 425–450 nm (Richter et al., 2004) using the NO_2 cross section from Bogumil et al. (2003) at 243 K. The retrieval is the same as the one applied to GOME NO_2 version IUP (cf. Sect. 2.3). The SCIAMACHY columns used here are thus stratospheric columns retreived with an AMF derived from the SCIATRAN model (Rozanov et al., 2005). The estimated accuracy is about 5 to 10 % (Richter et al., 2004). Since the GDP4 products were not available at the time of this work, the SCIAMACHY NO_2 data used here ar limited to the IUP version.

2.5 OMI

OMI (Ozone Monitoring Instrument) was launched on 15 July 2004 aboard the EOS Aura sun-synchronous platform at 705 km of 98.2° inclination (Levelt et al., 2006). OMI is a UV-Vis imaging spectrometer that measures solar radiation at nadir reflected and scattered by the atmosphere and the surface. For ozone and NO_2, it combines the advantages of its predecessors (GOME and SCIAMACHY) by measuring the spectrum (270–500 nm) in the UV-visible range at very high spatial resolution of 13–24 km with a spectral resolution of 0.63 nm in the visible. The satellite crosses the equator during its ascending orbit at 13:42 LT. It achieves daily global coverage. In this study, two algorithms are used for total ozone. The first, called OMI-TOMS, is identical to that used for EP-TOMS V8 (Buchard et al., 2008) providing total ozone measurements with a relative accuracy of 2 % at SZA lower than 70°. The second, called OMI-DOAS, developed by KNMI (Koninklijk Nederlands Meteorologisch Instituut) is based on the DOAS method with a relative accuracy of 3 % on cloudy days and 2 % on clear sky (Barthia et al., 2002). The data of the two versions are available from http://avdc.gsfc.nasa.gov/.

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For NO_2, the OMI data used here are those developed by NASA, using the cross-sections of Vandaele et al. (1998) at 220 K and the DOAS technique in the wavelength interval 405–465 nm (Boersma et al., 2002). The vertical columns are calculated with a stratospheric AMF which depends of the viewing geometry, the surface albedo and the shape of the NO_2 vertical profile. As described in Ionov et al. (2008), for each location, the algorithm uses a single mean unpolluted profile derived from a stratospheric model and a geographically gridded set of annual mean polluted profiles obtained from a tropospheric model. An initial estimate of the NO_2 vertical column is obtained by dividing the slant column by an unpolluted AMF, which is then geographically gridded using the data acquired within ±12 h from the target orbit. Areas shown by the model to contain climatologically high tropospheric NO_2 amounts are then masked, and the remaining regions are smoothed in latitude bands to construct a global stratospheric field. Where the initial vertical column exceeds the estimated stratospheric NO_2, the presence of tropospheric NO_2 is inferred and the vertical column is recalculated using an AMF computed from an assumed tropospheric NO_2 profile (Bucsela et al., 2006). The estimated accuracy of the NO_2 vertical column in clear sky and unpolluted conditions is 5 % (0.2×10^{15} mol cm^{-2}) and 20 % (0.8×10^{15} mol cm^{-2}) in polluted cases. In the presence of pollution and clouds, the error can reach 50 % (Celarier et al., 2008; Boersma et al., 2002). The data are available at http://avdc.gsfc.nasa.gov/.

3 Comparison between SAOZ and collocate satellite measurements

Since no single satellite covers the full period of SAOZ observations, the comparison of the long-term evolution requires the building of a composite series called "merged satellite data", built from the best available satellite data. However, because of many differences between instruments, wavelength ranges, absorption cross-sections and retrieval procedures, satellites are showing systematic biases and seasonality compared to SAOZ, which requires evaluation and correction. The satellite data analysed in the following are GOME-GDP4, EP-TOMS, OMI-TOMS and OMI-DOAS for ozone and GOME-IUP V1, GOME-GDP4, SCIAMACHY-IUP V2 and OMI-DOAS V3 for NO_2. However, because of the strong perturbation of both satellite and ground-based measurements by the aerosols injected in the stratosphere by the eruption of the Mount Pinatubo in 1992, data prior to 1995 will be ignored in the following.

3.1 Ozone

3.1.1 SAOZ seasonal and interannual variability

The SAOZ monthly mean O_3 columns in Bauru and Reunion since 1995 are shown in Fig. 1. At both stations the mean column is about 270 DU with a seasonal cycle of 30–40 DU amplitude with a spring maximum. The seasonal and interannual variations are very similar at both stations, displaying a biennial cycle of about 10 DU amplitude, with a maximum during the east phase of the QBO (Quasi Biennial Oscillation). At both stations, 3 periods are noticed: from 1993 to 2002 where a slight increase of 15 DU is observed, then from 2003 to 2007 where O_3 variations are constant and from 2008 to 2011 where an increase of 20 DU (more pronounced at Bauru) is seen. Most remarkable is the difference between the two stations which is displayed in the right part of Fig. 1. Bauru is showing larger ozone than Reunion in the summer (~ 2 %, about 5 DU from January to March) and in the winter (~ 3 % about 6–7 DU from May to August).

3.1.2 Comparison between satellites and SAOZ

The monthly mean GOME GDP4, EP-TOMS, SCIAMACHY-OL3, OMI-TOMS, OMI-DOAS total ozone and the SAOZ sunrise-sunset average ozone columns as well as the various differences satellite-SAOZ are presented in Fig. 2 for Bauru and in Fig. 3 for Reunion. Mean biases and standard deviations with SAOZ are summarised in Table 1 and the mean amplitude of the seasonal cycle of the difference in Table 2.

Systematic biases are observed between satellites and SAOZ, lower by 5.8 DU (2.1 %) in Bauru compared to Reunion with a smaller standard deviation with EP-TOMS and OMI-TOMS than with other satellites. The difference also shows systematic seasonality, smaller with EP-TOMS than with other satellites. With the exception of the beginning of GOME in 1995 and EP-TOMS in late 2002 (immediately after the correction for the instrumental problems), the difference does not exceed ±5 %. However, significant biases appear between the satellites, larger above Bauru than Reunion. At Bauru, a seasonal variation of the difference between SAOZ and satellites is observed. Except for EP-TOMS, the satellite observed seasonal cycle is lower than that measured by SAOZ with smaller amplitude during the austral summer up to 5 % for GOME GDP4, 4 % for SCIAMACHY, 6 % for OMI DOAS and 8 % for OMI TOMS. The two OMI versions present different amounts of O_3, with a difference of −3.4 % (OMI TOMS) and −2.10 % (OMI DOAS) relative to SAOZ columns; however, the columns of OMI TOMS have a sharper seasonal bias than OMI-DOAS. It is also noted that SCIAMACHY columns present a slight decrease (2 %) after 2004. Finally, only the EP TOMS columns are similar to those of SAOZ with a mean bias of −0.59 %.

At Reunion, the differences between satellites and SAOZ columns are lower. Only GOME and SCIAMACHY columns present larger seasonal variation with a minimum in austral winter. EP-TOMS columns are higher than those of SAOZ (1.04 %) with large variations. Finally, the two versions of OMI are anticorrelated in 2005 and 2006, OMI-TOMS columns showing larger variations with peaks of up to −4 %. Nevertheless, OMI-TOMS ozone columns are close to those measured by the SAOZ with a difference of 0.47 % against −0.04 % for OMI DOAS.

The mean biases between the satellites and SAOZ ozone retrievals originate partly from errors in the absorption cross-sections in the various spectral ranges, but more importantly from the way longitudinal ozone variations are treated in the retrievals and the sensitivity of the measurements to tropospheric ozone, as shown by the mean 2 % low bias in Bauru compared to +0.2 % in Reunion and the large differences between the two stations reported by the various instruments. The influence of the instrument

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sensitivity is illustrated by the difference between EP-TOMS and OMI-TOMS, the latter presenting a positive bias of 2.9 % compared to EP-TOMS over Bauru and a low bias of 0.6 % only over Reunion. This difference is fully consistent with that already found by McPeters et al. (2008) from comparisons between the two instruments and ground-based measurements at 74 stations in the Northern Hemisphere. Finally, with the exception of GOME at the very beginning of its operation in 1995, the GOME, EP-TOMS, OMI-TOMS and SCIAMACHY biases remain constant during their full operating periods. The only satellite showing a non-constant bias is OMI-DOAS displaying a 2–3 % jump of unknown origin in 2009 over both stations.

The seasonality, of variable amplitude between the satellites, has been shown by Hendrick et al. 2011) to come from a combination of stratospheric temperature and solar zenith angle dependeces at the satellite overpass over the station. The largest temperature dependences of 0.21 % $^{\circ}C^{-1}$, similar to that observed between the ground-based Dobson and SAOZ, are observed with EP-TOMS and OMI-TOMS performing in the short UV spectral range, while that of GOME of 0.06 % $^{\circ}C^{-1}$ and SCIAMACHY of 0.11 % $^{\circ}C^{-1}$ performing in the mid-UV are of lower amplitude and that with OMI-DOAS insignificant. Since SAOZ is performing in the visible Chappuis bands where the ozone absorption cross-sections are not sensitive to temperature, this dependence must be attributed to the satellites. The stratospheric temperature seasonality in the tropics being lower than 10 °C, the impact of this dependence is $\leq$ 2 % on EP-TOMS and OMI-TOMS, $\leq$ 1 % on GOME and SCIA and inexistent on OMI-DOAS. As shown by Hendrick et al. (2011), the SZA dependence is quadratic. The largest are observed on OMI-DOAS, OMI-TOMS, SCIA and GOME resulting in a seasonality of 1 to 4 % depending on the ephemeris of the orbit.

3.1.3 Merged satellite total ozone series

The best satellite series covering the full SAOZ measurements since 1996 is a combination of EP-TOMS (1995–2004) and OMI-TOMS (2005–2011), both showing the smallest biases and dispersion and the smallest seasonality of the difference with

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SAOZ. A merged satellite composite has then been constructed from the series of these two satellites after normalisation by addition of their respective biases with SAOZ above Bauru and Reunion as indicated in Table 2.

The merged satellite series and the SAOZ ozone columns over Bauru and Reunion since 1996 are displayed in Fig. 4. The two data sets show very high correlation of 0.96 % in Bauru and 0.97 % in Reunion, although of slightly larger slope (0.95 instead of 0.87) and larger intercept (32 DU instead of 12 DU) over Reunion, which indicates that the satellite columns are higher than SAOZ observations at this station. The seasonal variation of the difference between the two stations is represented in Fig. 5. The satellite series confirms the larger ozone in Bauru seen by SAOZ in the summer (January–March) and the winter (June–August), but differs in spring (October–November) when the satellites are reporting a 5.7 DU larger column over Reunion. The 5 DU larger column over Bauru seen by both data sets during the summer convective season is due to photochemical ozone production in the upper troposphere by lightning NO_x (LNO_x) and entrainment of O_3-rich air masses from the upper troposphere-lower stratosphere region (Rivière et al., 2006; Huntrieser et al., 2007) to which both satellite nadir and ground-based zenith sky measurements are sensitive. The larger maximum of 10 DU in the winter displayed by both data sets, consistent with the TOMS total ozone longitudinal variability seen by Fishman and Larsen (1987) and the wave number one distribution of tropospheric ozone seen by the SHADOZ ozone sondes (Thompson et al., 2003a,b), is due to the westward advection across the Atlantic of ozone rich air masses from biomass burning in Africa. The smaller difference between the two stations in October–November is also due to biomass burning in Africa, but in that case rapidly advected to the close Indian Ocean after the reversal of tropospheric wind direction in spring, resulting in an increase of ozone concentration over the close Reunion Island between 5–12 km (Randriambelo et al., 2000). The lesser sensitivity of SAOZ than satellites to this event is due to the low altitude of the ozone rich layer to which ground-based zenith sky twilight measurements are less sensitive than the nadir viewing satellites.

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In conclusion, with the exception of mid-tropospheric ozone increase over Reunion during the African biomass burning season less seen by the ground-based zenith sky instrument, there is full agreement between the satellite merged ozone data series constructed from the EP-TOMS (1995–2004) and OMI-TOMS (2005–2011) products after correction for biases, with the SAOZ measurements performed since 1996 in the Southern tropics. Both are showing larger ozone columns above the South American continent station of Bauru compared to Reunion Island in the Indian Ocean.

3.2 Nitrogen dioxide

3.2.1 SAOZ seasonal and interannual variability

The SAOZ sunrise and sunset monthly mean columns since 1996 in Bauru and Reunion are displayed in Fig. 6. On average, the NO_2 column is larger by $0.70 \times 10^{15}\,\mathrm{mol\,cm^{-2}}$ in Bauru. At both stations, the seasonal variation exhibits a spring maximum but of larger amplitude ($0.5 \times 10^{15}\,\mathrm{mol\,cm^{-2}}$) and delayed by 2 months in Bauru. A drop can be seen in late 1998 and early 1999 during the largest El-Niño event since 1996 and a biennial cycle of $0.5 \times 10^{15}\,\mathrm{mol\,cm^{-2}}$ amplitude with a NO_2 column maximum during the West phase of the Quasi-Biennial Oscillation (QBO). The seasonal cycle of the difference between the two stations (Fig. 6, right panel) shows a 38 % larger column over Bauru in the summer in November–March, a little larger at sunset, and a smaller increase of 20–25 % in July–August, larger at sunrise.

3.2.2 Satellite observations

Figure 7 displays the monthly mean time series of GOME-GDP4 (total) and -IUP, SCIAMACHY-IUP (stratospheric) and OMI-NASA (stratospheric) columns at the time of satellite overpass over the equator around 10:00 LT for SCIAMACHY, 10:30 LT for GOME and 13:00 LT for OMI. The measurements all show a seasonal cycle with a spring maximum, but of variable amplitude depending on the satellite. In addition they

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display larger noise in Bauru, suggesting larger NO_2 column variability there, particularly GOME-GDP4 measurements. GOME-GDP4 exhibits a larger seasonality than GOME-IUP by 2×10^{15} mol cm^{-2} in Bauru and 1.2×10^{15} mol cm^{-2} in Reunion, larger than SCIAMACHY by 0.5×10^{15} mol cm^{-2} in both stations. OMI agrees with SCIAMACHY in Bauru but shows larger columns by $0.5 10^{15}$ mol cm^{-2} in Reunion with less seasonality.

3.2.3 Correction for NO_2 diurnal variation

Since NO_2 displays a strong photochemical diurnal variation and SAOZ and satellite measurements do not coincide in time, a correction is required for comparing their measurements. Figure 8 shows the diurnal variation of the NO_2 column at 20° S in January and June calculated with the SLIMCAT 3-D Chemical Transport Model (Chipperfield et al., 1999; Denis et al., 2005), the time of satellite overpasses and that of the weighted average twilight SAOZ measurements between 86–91° SZA. The column at the time of satellite overpasses is always smaller than that of SAOZ at twilight ($\sim0.2\times10^{15}$ mol cm^{-2} at sunrise and around 1.1×10^{15} mol cm^{-2} at sunset) and different between the satellites. Indeed, Although there is thirty minutes between GOME and SCIAMACHY measurements and 3 h with OMI, the diurnal variation induces a bias of 0.05×10^{15} mol cm^{-2} between the first and 0.3×10^{15} mol cm^{-2} with the last. Moreover, following the seasonal variation, winter columns are greater (0.4×10^{15} mol cm^{-2}) than summer ones. Following these various biases observed between satellites and SAOZ, and the exact time when the satellites overpass the station, a diurnal correction is applied to convert all satellite data at SAOZ sunrise measurement. The diurnal cycle has been simulated with a photochemical box model derived from the SLIMCAT 3-D Chemical-Transport Model (Denis et al., 2005). In order to correct satellite data, a reference column calculated from the weighted average of SAOZ sunrise measurements between 86° and 91° SZA has been defined. The calculation of the diurnal variation of the ratio NO_2 (reference)/NO_2 (model) has been performed for the twelve months of the year. Then, all the satellite measurements have been normalized using this ratio.

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3.2.4 Comparison between satellites and SAOZ

The monthly mean GOME GDP4, GOME IUP, SCIAMACHY, OMI and the SAOZ columns as well as the relative mean difference satellites-SAOZ after applying the photochemical corrections are displayed in Fig. 9 for Bauru and Fig. 10 for Reunion. Mean biases and standard deviations of the difference with SAOZ are given in Table 3 and the amplitude of the seasonal cycle of the difference in Table 4.

Between 1996 and 2003, best agreement between the two GOME retrieval versions with SAOZ over the two stations is the GDP4 data showing smaller bias and seasonality, little difference between the two stations and high correlation with SAOZ (0.91 instead of 0.65 for IUP over Bauru and 0.92 instead of 0.73 for IUP over Reunion). Indeed, at both stations, GOME IUP presents a weaker amplitude of the seasonal cycle with systematically higher values (0.3×10^{15} mol cm^{-2}) during the spring period compared to SAOZ and the ESA versions. On average, IUP columns at Bauru are closer to SAOZ, compared to Reunion where the columns are systematically larger (0.4×10^{15} mol cm^{2}). A seasonal variation of the difference is observed for the IUP version, with a maximum in winter (∼ 20 %) and a minimum from October to February. The mean difference with SAOZ is smaller at Bauru (∼ 2.6 % compared to 19.1 % at Reunion) but has a high standard deviation (12.7 % ∼ 0.5×10^{15} mol cm^{-2}). One the other hand, the ESA version presents a steady difference of −6.5 % at Bauru and 5.8 % at Reunion with no seasonal variation.

The main difference between the two retrievals is the way the tropospheric contribution is treated. GDP4 is providing a total column using a NO_2 profile zonal climatology for the stratospheric part (Lambert et al., 2004), whereas the IUP product is a stratospheric column, underestimating any tropospheric NO_2 present in the measurements.

Finaly, regarding the consistency between GOME and SCIAMACHY data (2003–2012), the gap between them is less pronounced with the ESA version at Bauru and inexistent at Reunion, compare to the IUP version where a mean bias of 20 % is observe between the two satellites. The GDP4 total column product is thus better suited for comparison with SAOZ.

The two satellites following GOME measurements after the breakdown of the on-board recorder are SCIAMACHY and OMI (2004–2012), both representing stratospheric and not total columns. They both agree with SAOZ above Reunion ($r^2 = 0.90$ for OMI and 0.92 for SCIAMACHY) where the NO_2 tropospheric content is limited, although OMI is displaying a larger seasonality of the difference with SAOZ (mostly during winter with an amplitude of the biais 0.5×10^{15} mol cm^{-2}). It is also noticed that SCIAMACHY presents a slight bias during summer (∼ mean bias −4.1 %).

More importantly, both satellite data sets underestimate the columns over Bauru by 19 % for SCIAMACHY and 29 % for OMI and have smaller amplitude of the seasonal cycle than SAOZ. In addition, their maximums are shifted by 1 month. The OMI maximum (0.35×10^{15} mol cm^{-2}) occurs from April to August and the SCIAMACHY maximum from March to September (0.2×10^{15} mol cm^{-2}).

The difference between both satellites depends on how the tropospheric contribution is subtracted in the retrieval: indeed, SCIAMACHY IUP column have ben retrieved with the same method as GOME IUP and OMI stratospheric column with an AMF using a single mean unpolluted profile derived from a stratospheric model and a geographically gridded set of annual mean polluted profiles obtained from a tropospheric model.

In summary, although not representing the full total column, the SCIAMACHY product is better suited for comparison with SAOZ.

3.2.5 Merged satellite NO_2 series

The best satellite series covering the full SAOZ measurements since 1996 is thus a combination of GOME GDP4 (1995–2003) and SCIAMACHY (2003–2011), both showing the smallest biases and dispersion and the smallest seasonality in the difference with SAOZ. A merged satellite composite has then been constructed from the series of these two satellites after normalisation by by addition of their respective biases with SAOZ above Bauru and Reunion as indicated in Table 3.

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The satellite and SAOZ NO_2 column series over Bauru and Reunion and the correlation between them are shown in Fig. 11. The satellite composite series present more variation from 1996 to 2002 (GOME GDP4) compared to SAOZ at both stations. However, despite these differences, the two data sets are highly correlated (92 % in Bauru, 94 % in Reunion) displaying the same seasonality and interannual variability. The seasonal variation of the difference between Bauru and Reunion displayed by the two data sets (Fig. 12) is almost identical, both showing a larger column above Bauru particularly in summer (January–March) where the difference reaches to 1.1×10^{15} mol cm^{-2} that is 35 % and a minimum in winter (April–June) of 18 % (such as 0.6×10^{15} mol cm^{-2}).

Two factors can contribute to the differences between the two stations:

- the longitudinal distribution of stratospheric NO_2 (such as for the O_3)
- the tropospheric NO_2 column.

Unlike for ozone, the contribution of the tropospheric NO_2 to the total column is important. Although SAOZ measurements are less sensitive, the tropospheric NO_2, in particular the part present in the upper troposphere cannot be ignored in our study, especialy when a comparison to satellites is performed.

All sources of NO_x (urban and industrial pollution, biomass burning, production by lightning) are consistently higher over the continents and outside of main shipping routes negligible over the oceans. One year of OMI NO_2 tropospheric measurements has been studied over the region of Bauru, and the average tropospheric NO_2 column is about 3×10^{15} mol cm^{-2}, about 50 % of the total column reported by SAOZ. It should be noted that winter is the season where maximum pollution is detected at the highly polluted city of São Paulo, located 300 km to the east of Bauru. The increase of tropospheric NO_2 in spring (late August, seen Fig. 12)) is related to sugar cane fires, a traditional practice in Brazil, as reported for example by the GOME NO_2 observations (Richter and Burrows, 2002).

The explanation for this large difference in the summer (December to March), seen by both nadir viewing satellites and twilight zenith sky SAOZ, is the high NO_x

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concentration between 10–15 km above S-E Brazil produced by the frequent lightning's during the thunderstorm convective season (Pommereau et al., 2004; Rivière et al., 2006; Huntrieser et al., 2008), as opposed to the Indian ocean and maritime regions in general where lightning is rare (Zipser et al., 2006).

Another contributor to the NO_2 column difference between Bauru and Reunion from autumn to spring is the location of the station relative to the sub-tropical jet in the lower stratosphere which has an impact on the longitudinal distribution of stratospheric NO_2. The equivalent latitude parameter calculated from the potential vorticity (PV) output (from 1995 to 2012) of the high-resolution advection model MIMOSA (modeling Meso-scale isentropic transport of Stratospheric Ozone by advection (Hauchecorne et al., 2002) developed by the LATMOS, have been used to identified the origin of the air mass above the stations. As shown by the seasonal variation of equivalent latitude calculated above the two stations (Fig. 13), at any level, the position of Reunion can be defined as a tropical station with equivalent latitude varying around 22° S ± 2° throughout the year. On the other hand, Bauru's equivalent latitude presents larger variations, and is much more under the influence of mid-latitudes than Reunion from April to December at 450 K (increases up to 33° S) and from July to October (up to 30° S) at 475 K. HALOE V19 profiles (from 1995 to 2005) have been used in order to corrorborate those results (data can be found on http://haloe.gats-inc.com/home/index.php). To get the maximum profiles, the satellite grid observation has been expanded to 200 km radius around the station (Bauru: 22° ± 3° S, 50° ± 15° W, Reunion: 20 ± 3° S, 50 ± 15° E). Despite this extension, only 100 profiles were used at Bauru and 110 at Reunion. Because of the extension of the grid and the time of satellite measurement (between 7 and 9 a.m.), all profiles had to be standardized from a reference profile calculated using a photochemical box model derived from 3D chemistry transport model SLIMCAT (Denis et al., 2005). As shown by the seasonal variation of the HALOE NO_2 profiles above the two stations (Fig. 14), the NO_2 concentration is larger ($\sim 0.2 \times 10^9$ mol cm^{-3}) in the lower stratosphere above Bauru than above Reunion in the winter, although no difference is seen between November–March.

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In conclusion, there is excellent agreement between the merged satellite associating GOME GDP4 products from 1996 to 2003 and SCIAMACHY IUP version from 2003 until present after correction for the diurnal variation of NO_2 and systematic biases, with the long series of SAOZ observations in the southern tropics both showing larger NO_2 column above the South American continental station of Bauru compared to the Indian Ocean Reunion Island.

4 Conclusions

Long series of ozone and NO_2 total column SAOZ-NDACC observations at the Southern tropics are available since 1995 in Bauru on the South American continent and since 1993 in Reunion Island in the Indian Ocean, providing a unique tool for investigating possible trends in the composition of the stratosphere at this latitude. For evaluating the reliability of these data before performing such studies, an essential exercise is to compare them to the measurements performed by the several satellites operating during the same period. Since none of the satellites covers the full period until present, and systematic biases exist between them because of the many differences between instruments, wavelength ranges, absorption cross-sections and retrieval procedures, they require evaluation and bias corrections.

From comparisons with SAOZ, the best satellite data series for ozone in the Southern tropics are shown to be EP-TOMS (1995–2004) and OMI-TOMS (2005–2011) products. Both confirm the larger ozone columns reported by SAOZ above the South American continent station of Bauru compared to the Indian Ocean Reunion Island because of the ozone production by lightning NO_x (LNO_x) over the continent in the summer and the advection from Africa in the winter of ozone produced by biomass burning resulting in the known wave number one distribution of tropospheric ozone reported by the SHADOZ soundings. The only difference between the data sets is the underestimation by SAOZ compared to satellites of the ozone enhancement in the mid- troposphere

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above Reunion following the fast advection of biomass burning from the nearby African continent during the reversal of tropospheric wind direction in Spring.

Best matching the SAOZ NO_2 column measurements is a combination of GOME GDP4 products in 1996–2003 and SCIAMACHY in 2003–2012, both confirming, like ozone, the larger column seen by SAOZ above the South American continent, attributed to LNO_x production by frequent lightning in the summer, and to the larger exchange of NO_x rich air with the mid-latitudes in the winter in the lower stratosphere due to the more equatorial latitude of the subtropical jet in the winter above South America compared to the Indian Ocean.

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Table 1. Mean biases and standard deviation in DU (%) between satellites and SAOZ total ozone.

Satellite	Bauru	Reunion	Bauru-Reunion
EP-TOMS	-1.58 ± 3.1 (-0.59)	2.80 ± 3.6 (1.04)	-4.38 (-1.6)
OMI-DOAS	-6.09 ± 5.2 (2.1)	-0.08 ± 4.5 (-0.04)	-6.17 (-2.28)
OMI-TOMS	-9.47 ± 3.2 (-3.43)	0.31 ± 3.4 (0.47)	-9.16 (-3.39)
GOME-GDP4	-5.49 ± 4.2 (-1.82)	0.44 ± 4.9 (0.15)	-5.05 (-1.87)
SCIAMACHY-OL3	-4.93 ± 4.7 (-1.7)	-0.73 ± 2.9 (-0.24)	-4.2 (-2.9)
Average	-5.51 (-2.04)	0.55 (0.20)	-5.8 (-2.15)

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Table 2. Amplitude in DU (%) of the seasonal cycle of the difference between SAOZ and satellites.

Satellite	Bauru	Reunion
EP-TOMS	4.6 (1.7)	4.4 (1.6)
OMI-DOAS	9.0 (3.3)	7.9 (2.9)
OMI-TOMS	8.8 (3.2)	8.7 (3.2)
GOME-GDP4	11.9 (4.3)	9.3 (3.4)
SCIAMACHY-OL3	10.8 (4.0)	10.5 (3.9)

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Table 3. Mean biases and standard deviation in 10^{15} mol cm^{-2} (%) between satellites and SAOZ NO_2 columns.

Satellite	Bauru	Reunion	Bauru-Reunion
GOME V1.0 IUP	0.04 ± 0.5 (2.6)	0.44 ± 0.3 (19.1)	−0.4 (−19)
GOME GDP4 ESA	−0.23 ± 0.2 (−6.5)	−0.13 ± 0.2 (−5.8)	−0.1 (−5.7)
SCIAMACHY V2.0 IUP	−0.63 ± 0.3 (−18.8)	−0.10 ± 0.1 (−4.1)	−0.53 (−21.2)
OMI DOAS V3	−0.9 ± 0.28 (−28.9)	−0.02 ± 0.2 (0.28)	−0.88 (−29.4)
Average	−0.43 (−2.04)	0.05 (0.20)	−0.47 (−19.2)

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Table 4. Amplitude in 10^{15} mol cm^{-2} (and %) of the seasonal cycle of the difference between SAOZ and satellites.

Satellite	Bauru	Reunion
GOME V1.0 IUP	0.73 (22)	0.52 (13)
GOME GDP4 ESA	0.18 (4.2)	0.17 (4.1)
SCIAMACHY V2.0 IUP	0.51(13)	0.09 (3.7)
OMI DOAS V3	0.97 (14.1)	0.16 (4)

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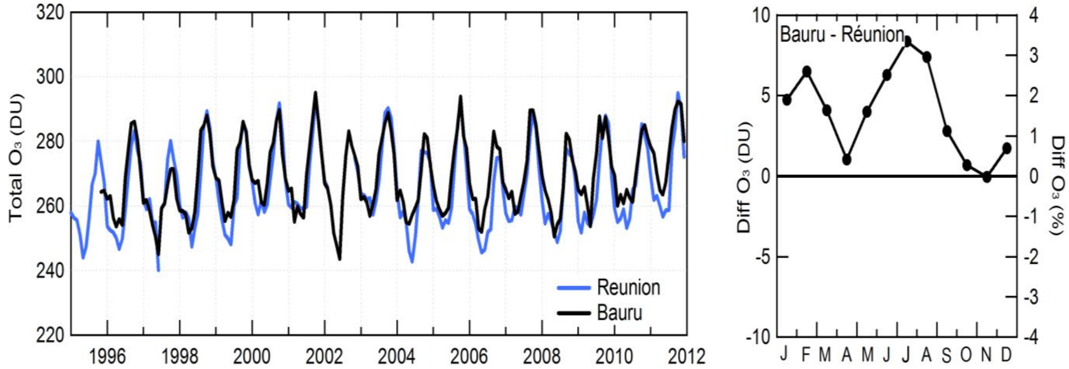

Fig. 1. Left panel: monthly mean SAOZ total O_3 in Bauru (black) and La Reunion (blue). Right panel: seasonal cycle of difference between the two stations.

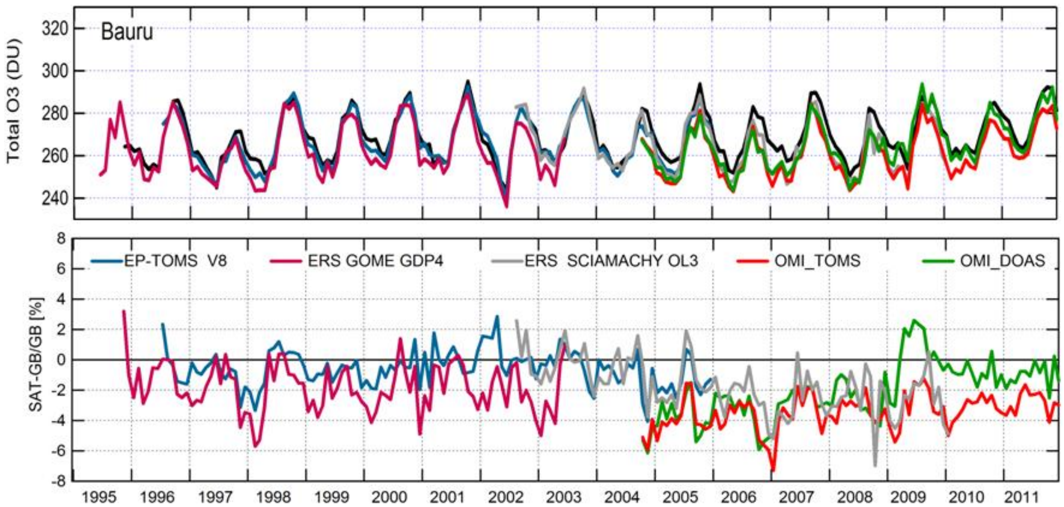

Fig. 2. Total ozone over Bauru. Top panel: monthly mean column GOME GDP4 (pink), EP-TOMS (blue), SCIAMACHY OL3 (grey), OMI-TOMS (red), OMI-DOAS (green) and SAOZ sunrise-sunset average (black); bottom panel: differences with SAOZ.

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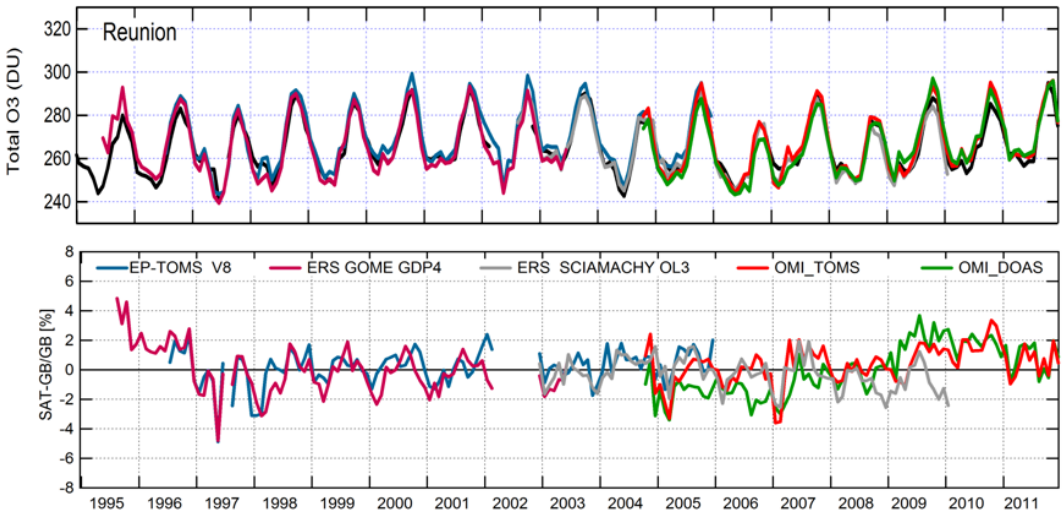

Fig. 3. Same as Fig. 2 for Reunion.

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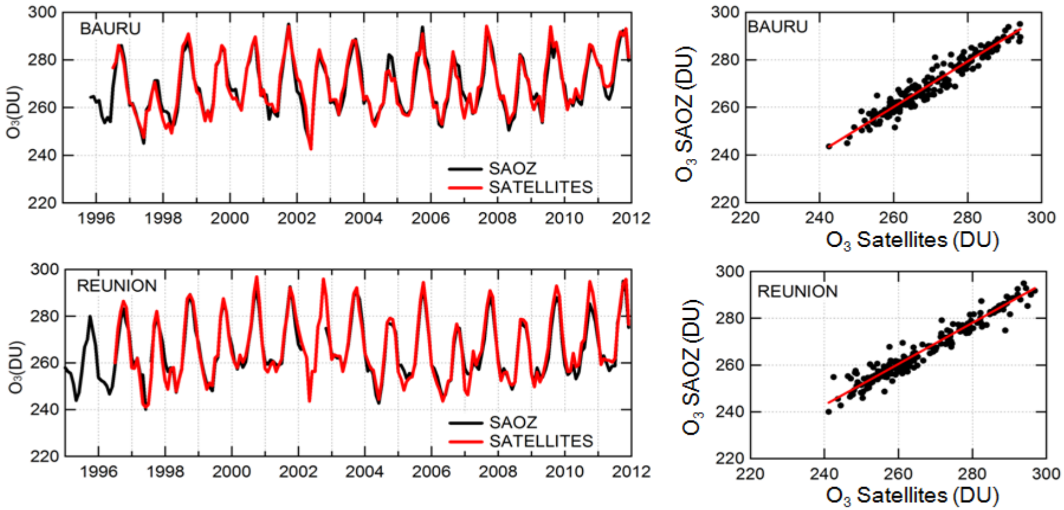

Fig. 4. Left panels: time series of SAOZ (black) and merged satellites (red) total O_3 column (DU) in Bauru (top panels) and Reunion (bottom panels). Right panels: correlation between SAOZ and merged satellites ($r^2 = 0.96$ in Bauru and $r^2 = 0.97$ in Reunion).

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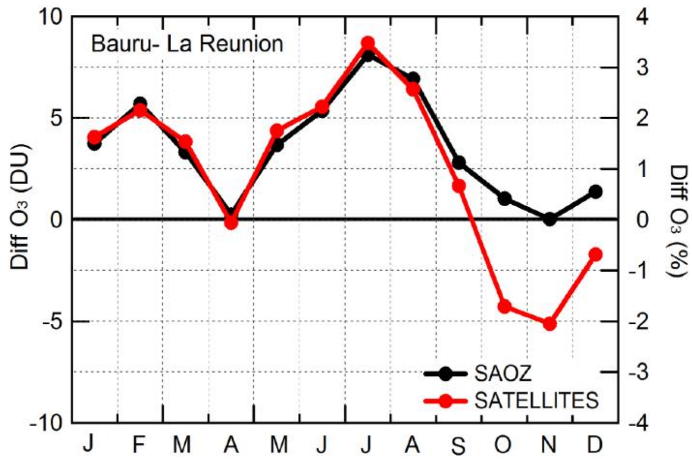

Fig. 5. Seasonal cycle of the difference between Bauru and Reunion for SAOZ (black) and merged satellite composite (red).

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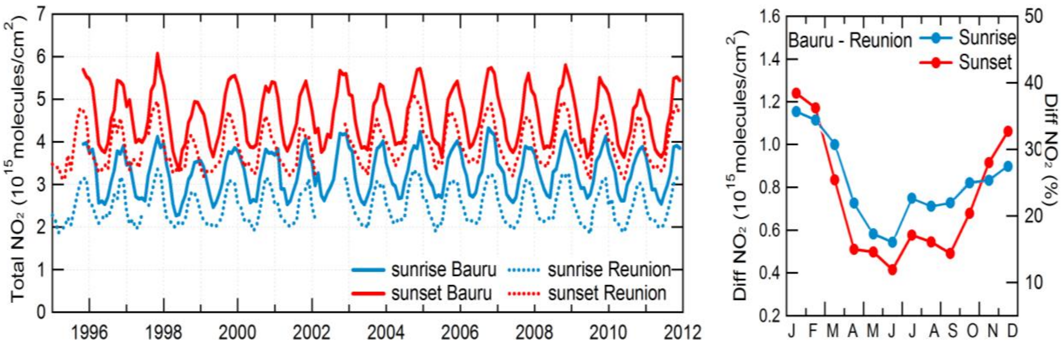

Fig. 6. Left panel: NO_2 sunrise (blue) and sunset (red) columns in Bauru (solid lines) and Reunion (dotted). Right panel: seasonal cycle of the difference between the two stations.

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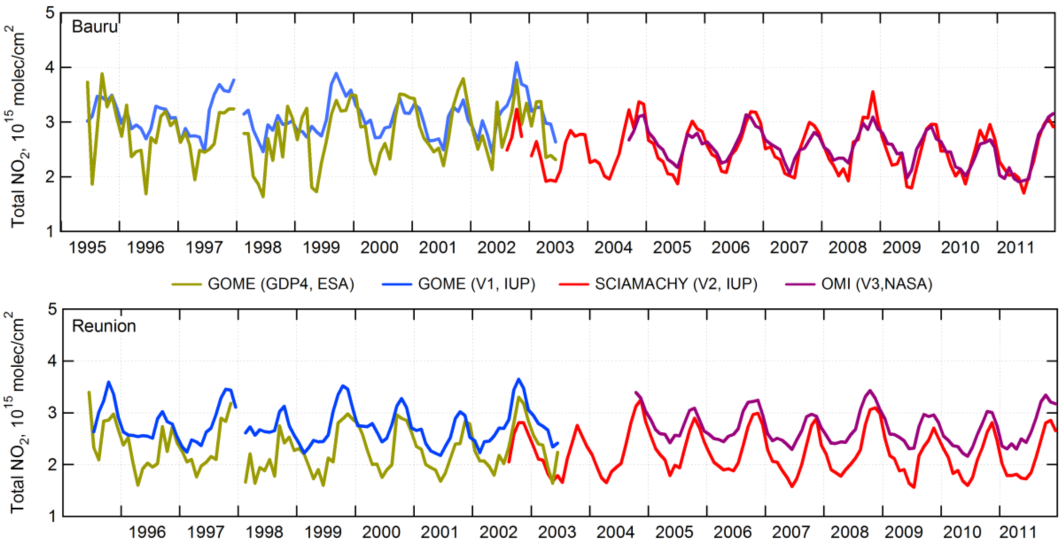

Fig. 7. Monthly mean NO_2 time series of GOME GDP4 (green), GOME IUP V1 (blue), SCIAMACHY IUP V2 (red) and OMI DOAS V3 (purple) total NO_2 over Bauru (top panel) and Reunion (bottom panel).

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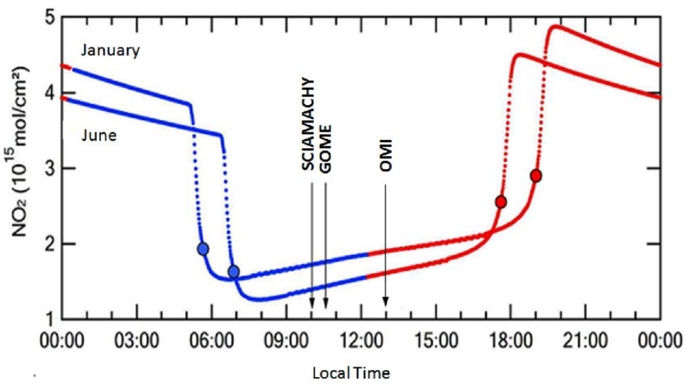

Fig. 8. Simulated NO_2 column diurnal variation in the tropics in January and July. The markers show the time of sunrise (blue) and sunset (red) SAOZ measurements at 90° SZA and the arrows that of ERS-2 GOME, Envisat SCIAMACHY and Aura OMI overpasses.

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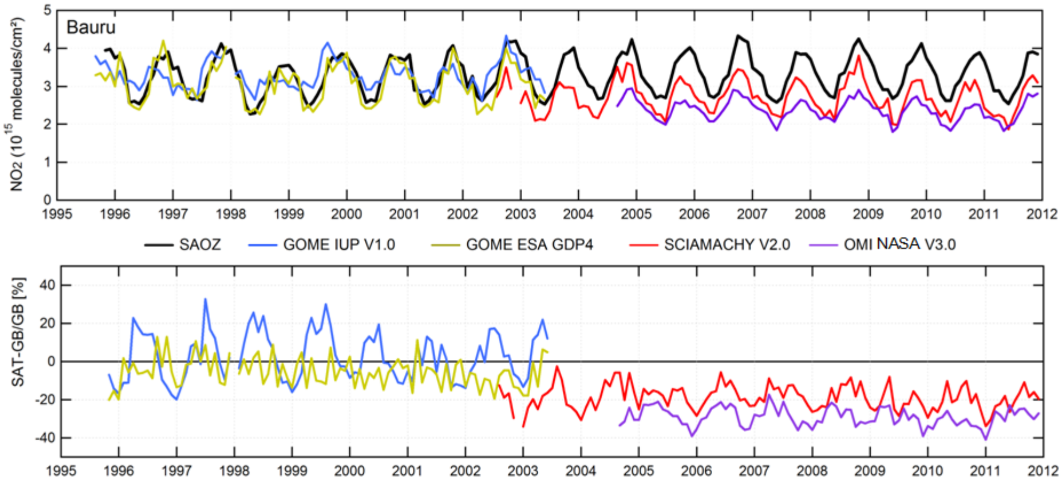

Fig. 9. Monthly mean NO_2 column over Bauru. Top panel: GOME GDP4 (green), GOME IUP V1 (blue), SCIAMACHY IUP V2 (red), OMI DOAS V3 (purple) and SAOZ sunrise (black). Bottom panel: relative mean difference with SAOZ in %.

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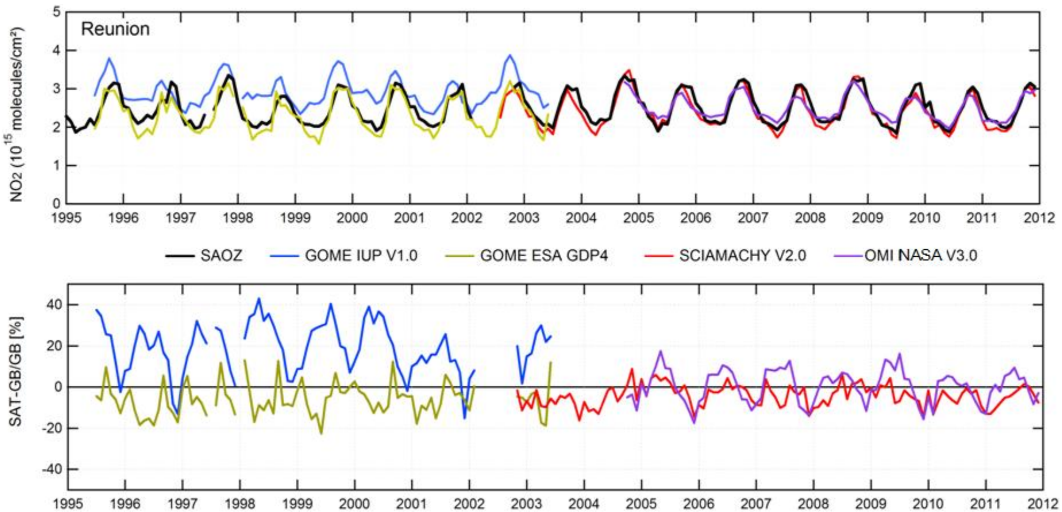

Fig. 10. Same as Fig. 9 for Reunion.

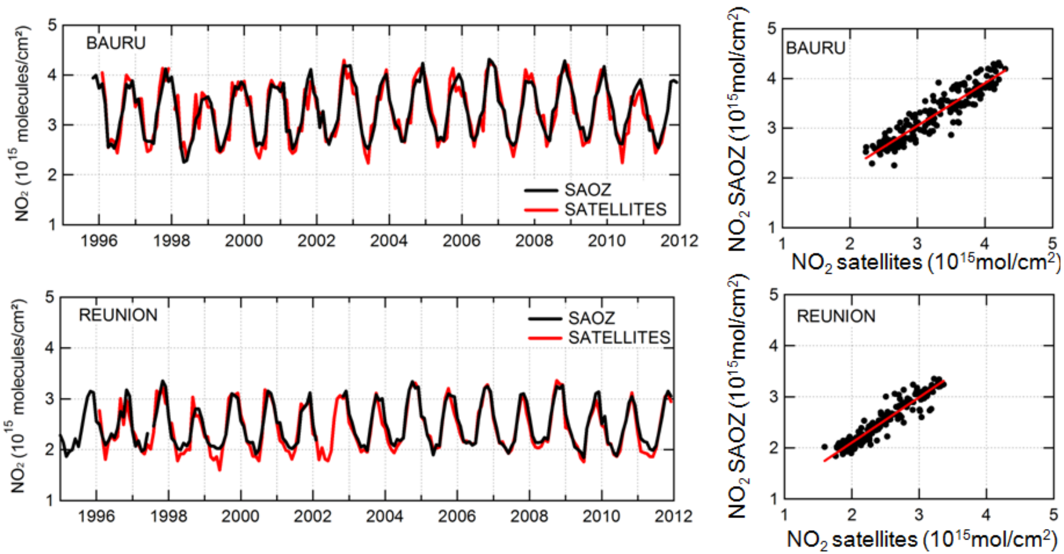

Fig. 11. Left panels: SAOZ (black) and merged satellites (red) NO_2 columns (10^{15} mol cm^{-2}) in Bauru (top panels) and Reunion (bottom panels). Right panels: correlation between SAOZ and merged satellites data ($r^2 = 0.94$ in Bauru and $r^2 = 0.92$ in Reunion).

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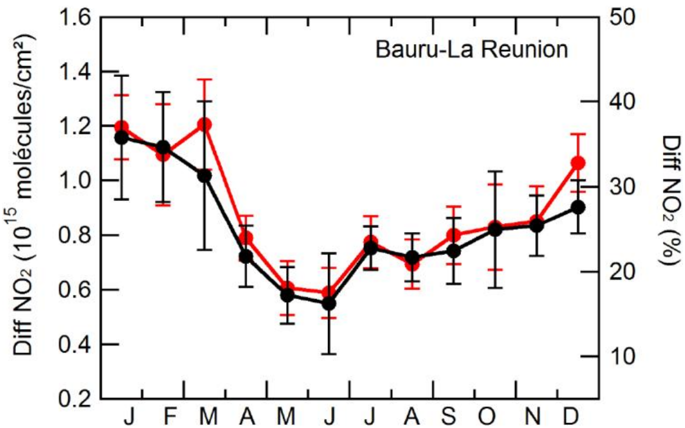

Fig. 12. Seasonal cycle of the difference between Bauru and Reunion from SAOZ (black) and merged satellites (red).

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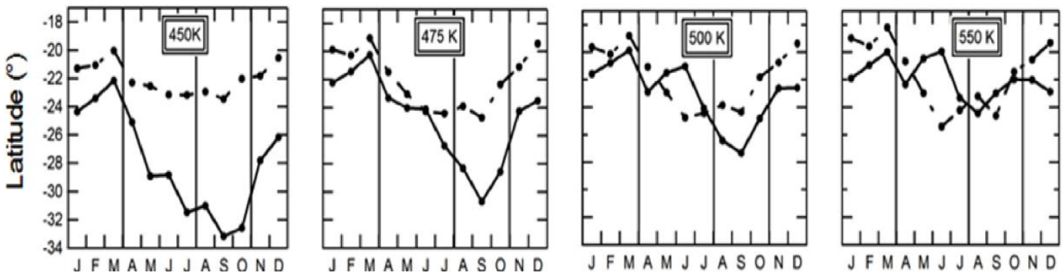

Fig. 13. Seasonal variation of equivalent latitude above Bauru (solid line) and Reunion (dashed) at 450, 475, 500 and 550 K.

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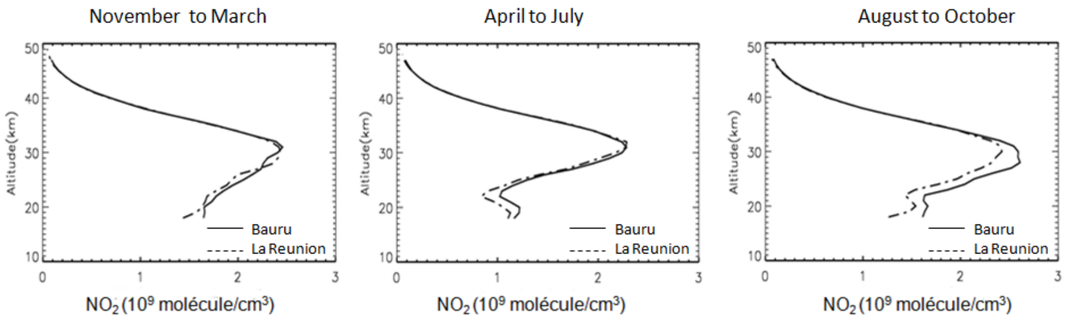

Fig. 14. HALOE (1995–2005) NO_2 mean profiles over Bauru (solid line) and Reunion (dashed) in November–March (left panel), April–July (middle panel) and August–October (right panel).