Answer to Referee #1

We thank the referee for his constructive comments and suggestions.

Regarding major concerns: i) we hope that the scope of the paper is now better defined, oriented towards the construction of optimum merged satellite data series for further trend analyses, through a thorough evaluation of satellites observations in the tropics by comparison with the two SAOZ, and the title has been changed accordingly; ii) within this scope, the paper has been reorganized and all sections carefully revised, meeting hopefully now better the standard of scientific publications.

Indeed, although several validations of the satellites measurements used here are available, they hardly apply to the tropics where significant differences between them and with ground-based observations are still present, originating mainly from the masking of the troposphere by cirrus clouds and thunderstorms anvils, particularly over land areas in the summer.

Detailed comments

P4853 The introduction is short and does not properly introduce the NDACC network, the SAOZ instrument and measurement principal. Add references.

The introduction has been re-written according to the revised scope of the paper and appropriate references added.

P4853/L14-18: The current knowledge (i.e. results from previous studies) on the NO2 and O3 distribution at Bauru and Reunion (e.g. from SOAZ, other GB measurements or models) should be discussed in more detail. Add references.

The description of the current performances of the SAOZ measurements and vertical column retrievals, their advantages and disadvantages, the impact of clouds and the way the cloudy data are removed has been added. Although, the SAOZ tropical data have been (e.g. Celarier et al., 2008) and are currently used within several validation programs (ESA CCI, EU NORS projects) the results of these studies are not yet published and this is precisely the objective of the paper.

P4853 It should be clearly described how the comparisons of the SOAZ data and the satellite measurements can be used to check the reliability of the SOAZ data: The absolute accuracy of the SOAZ measurements has not been analysed here, however, the comparisons with satellite measurements do provide information about the robustness 2and consistency of the SOAZ time series.

As explained in the introduction, the goal is not to validate the SAOZ using satellite information, but in the opposite to understand the origin of differences between satellites and SAOZ and moreover between satellites which are quite large in the tropics. The accuracy of SAOZ slant column measurements is not the problem, as has been evaluated during the many NDACC instrument intercomparison campaigns quoted in the paper. As explained by Hendrick et al. 2011, the main issue in the SAOZ accuracy is the Air Mass Factor used to convert slant into vertical columns. In the case of ozone this is done by using daily AMF derived from the TOMSv8 zonal profile climatology, For NO_2 this is not yet possible and a yearly mean profile is used derived from HALOE and balloon measurements using a SAOZ balloon version in Bauru. Fortunately, the stratospheric O_3 and NO_2 profiles are not varying too much in the tropics (in contrast to mid- and high latitudes) and do not introduce additional significant uncertainty. All details on this issue can be found in Hendrick et al and are not repeated here.

Section 2.1

P4854 The SOAZ measurements should be described in more detail. Especially, the sensitivity of the SAOZ measurements to the vertical distribution of ozone and NO2 in the troposphere and stratosphere should be discussed (averaging kernels).

As explained above, the procedure, including averaging Kernels, is fully described in Hendrick et al., 2011

P4854/L13: Add reference to DOAS technique.

Added: Platt, U., Differential Optical Absorption Spectroscopy (DOAS), in Air Monitoring by Spectroscopic Techniques, Chem. Anal. Ser., vol. 127, edited by M. W. Sigrist, pp. 27– 76, Wiley-Interscience, Hoboken, N. J.,1994

P4854/L19 Add references to TOMS V8 ozone profile climatology.

Reference to TOMS V8 ozone profile climatology added (Mc Peters et al, 2007)

P4854/L21 The description of the SOAZ NO2 retrieval is insufficient. What is the reference for the accuracy estimate? (the estimate mentioned here seems different from the one in Ionov et al., 2008)

Information regarding the NO_2 retrieval have been added as follows:

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 in Bauru (detail of the retrieval can be found in Ionov et al., 2008).

The accuracy given here was that given in M. Pastel thesis. Since unpublished, it has been replaced by the figures given by Ionov et al: 11% precision, 21% accuracy.

Section 2.2

P4855 Two different error/accuracy estimates are given for the TOMS V8 ozone columns. Is the estimate mentioned on L13 only valid for 2002? Is the larger error related to the empirical correction? This is confusing.

The sentence has been rephrased as follows:
The estimated accuracy is about 3 to 5 %. Since 2000, EP-TOMS experienced instrumental problems introducing a 3% bias in total ozone (Bramstedt et al., 2003). In the version 8 used in this study, an empirical correction was applied by NASA to overcome this bias (McPeters et al., 2007).

Section 2.3

P4856/L1: Add reference to GDP4 O3 validation paper (Balis et al., JGR, 2007), internet link is wrong.

Reference added and Internet link changed for (http://wdc.dlr.de/sensors/gome/)

Reference Balis at al., 2007 added in the introduction.

P4856/L3: add reference to GDP4 and IUP GOME NO2 products.

References added:

For NO_2, two retrievals are available, from the Institut für Umweltphysik (IUP) of the University of Bremen (Richter et al., 2005) and from the ESA GOME Data Processor GDP4 (Lambert et al., 2004).

Ref IUP: Richter, A., F. Wittrock, M. Weber, S. Beirle, S. Kühl, U. Platt, T. Wagner, W. Wilms-Grabe, and J. P. Burrows, GOME observations of stratospheric trace gas distributions during the splitting vortex event in the Antarctic winter 2002 Part I: Measurements, *J. Atmos. Sci.*, 62 (3), 778-785, 2005

P4856/L5-18: The description of the two GOME NO2 products is confusing. From the algorithm description available at DLR and IUP, it follows that both products provide a total NO2 column based on a stratospheric AMF (underestimating the tropospheric NO2 contribution in case of polluted conditions). The main difference between the two products (i.e. the correction applied in the IUP product, which is related to the effect of spectral artifacts in the GOME solar spectrum due to the GOME diffuser problems) should be explained in more detail, since this has a large impact on the GOME IUP NO2 column time series for Bauru and Reunion. Both internet links are wrong.

This section has been changed as follows:

For NO_2, two retrievals are used, from the Institut für Umweltphysik (IUP) of the University of Bremen (Richter et al., 2005) version 1 and from the ESA GOME Data Processor GDP version 4 (Lambert et al., 2004). Both are using the same cross-sections (Burrows et al., 1998) at 221K and the DOAS spectral analysis method between 425-450 nm. The Data Processor GDP version 4(GDP4) retrieval is based, on the GDOAS algorithm created by BIRA-IASB (Van Roozendael et al., 2006). The difference between the two versions is coming from the different AMF calculation and the correction applied for the diffuser offset.

Both versions provide total column based on a stratospheric AMF. For IUP, the columns are derived from the SCIATRAN model (Rozanov et al., 2005) using a US standard atmosphere profile with the troposphere set to 0 (Richter et al., 2005). For the ESA product, the columns are derived from AMFs calculated calculated by the multiple scattering radiative transfer model LIDORT (Linearized Discrete Ordinate Radiative Transfer) (Spurr et al., 2001), using a NO_2 profile climatology (Lambert et al., 2004).

To compensate the effect of spectral artifacts in GOME solar spectrum due to the diffuser offset, the IUP applies a normalization on each slant column from a slant column selected in a clean region in the Pacific, assuming in this area a total column of $2*10^{15}$ molec/cm^2 independent of the season. The resulting columns are assumed a good estimation of stratospheric columns in clean areas but they may be relatively higher than the reality in polluted regions (Richter et al., 2005). The accuracy is estimated to 5-10%. The data used here are available on http://www.doas-bremen.de/gome_no2_data_quilt.htm.

On the other hand, an optimal calibration on the post process data has been applied on the GDP4 product. Details of the calibration can by found in Lambert et al., 2004.

The accuracy is estimated between 5% and 10%, in unpolluted regions. The data used here are available at: http://wdc.dlr.de/data_products/TRACEGASES/.

The internet addresses of both products have been changed.

Section 2.4

P4857/L3 is this the correct reference?

The internet link has been changed for: http://wdc.dlr.de/data_products/TRACEGASES

P4857/L9 To my understanding, the IUP NO2 column (GOME and SCHIAMACHY are total NO2 columns)

Change has been made all over the paper. NO_2 column is now referred as total column

P4857/L11 An ESA SCIAMACHY NO2 product (total NO2 column based on a stratospheric AMF) is available since the beginning of the SCIAMACHY mission.

The SCIAMACHY SGP version 5.02 data were made available after the submission of the paper which is now used. Section 2 has been revised as follows:

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 local time 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 - 790nm) with a spectral resolution of 0.2 to 1.5 nm and a spatial resolution of 30 x 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 (http://wdc.dlr.de/data_products/TRACEGASES/). This product has been developed based on GOME GDP4.0 (Bracher et al, 2005). The estimated accuracy is about 5% for SZA lower than 60°.

As for GOME, two NO_2 retrievals are used in this study,from the Institut für Umweltphysik (IUP) of the University of Bremen (Richter et al., 2004) version 2 and from the ESA off-line Processor SGP version 5.02 (Lichtenberg et al.,2010). Both versions are using the same cross-sections (Bogumil et al., 2003) at 243K and a DOAS spectral analysis method between 425-450 nm. The data processor version 5.02 is based, for the trace gas slant column retrieval, on the SDOAS algorithm created by BIRA-IASB. The SDOAS algorithm is similar to the GDOAS algorithm used for GOME GDP. Both products are using the same AMF as described in the GOME versions. Except for SGP where HALOE profiles are used instead of climatology (Lambert et al, 2000).

The estimated accuracy for the IUP version is about 5 to 10% (Richter et al., 2004) and the data can be found at http://www.iup.uni-bremen.de/doas/scia_no2_data_acve.htm.

The estimated accuracy of the SGP version ranges from 5% to 20% at polar latitudes and in the Northern Hemisphere. The NO_2 columns are low biased by about $5 \cdot 10^{14}$ molec/cm^2 at low and middle latitudes of the Southern Hemisphere. This low bias exhibits a seasonal cycle (larger in summer) and a latitudinal dependence. The NO_2 columns are affected by larger errors over polluted areas (large underestimation) and in the South Atlantic Anomaly (noisier data). The data can be ordered via the ESA EO Principal Investigator Portal.

NO_2 SCIAMACHY SGP5.02 reference added:

Lichtenberg, G., Bovensmann, H., Van Roozendael, M., Doicu, A., Eichmann, K.-U., Hess, M., Hrechanyy, S., Kokhanovsky, A., Lerot, C., Noel, S., Richter, A., Rozanov, A., Schreier, F. and Tilstra, L.G., SCIAMACHY Offline Level 1b-2 Processor ATBD (ENV-ATB-QWG-SCIA-0085, issue 1A), 2010

Section 2.5

P4857/L18 Which advantages of GOME and SCIAMACHY are combined?

The idea was that OMI is providing a better spatial resolution than GOME and a better overpass frequency than SCIAMACHY.

The description of OMI has been changed for:

OMI is a nadir-viewing UV/Visible spectrometer with a spectral resolution about 0.63 nm for the visible channel (349–504 nm) and about 0.42 nm for the UV channel (307–383 nm). It measures the solar light scattered by the atmosphere in the 270–500 nm wavelength range with a spatial resolution at nadir of 13km×24 km. The satellite crosses the equator during its ascending orbit at 13:42 local time.

P4858 The OMI NO_2 ***description is confusing. Here, the retrieval of the total NO2 column, including a correction for tropospheric NO2 is described. However, in Section3.2.2 it is mentioned that a stratospheric OMI NO2 product is used. This should be clarified and the version number of the OMI NO2 data product used in this study should be mentioned as well.***

The OMI NO_2 version was omitted, and changed as follows: For NO_2, the OMI data version 3 used here is that developed by NASA, using the cross-sections of Vandaele et al., (1998) at 220K and the DOAS technique in the wavelength interval 405-465nm nm (Boersma et al., 2002).

It was assumed that the total column corrected for tropospheric NO_2 is a stratospheric column, which is wrong. This assumption led to a misinterpretation and was corrected in all of the paper.

Section 3.1

P4859 Please add a plot of the seasonal cycle in the O3 columns at the two sites to fig1 (in addition to the plot of the seasonal cycle of the difference). The seasonal cycle at the two sites (and the difference between the two) could then be discussed in more detail here, including the contribution of tropospheric ozone.

Figure 1 has been modified as shown below. The figure shows on the left the monthly mean time series of SAOZ total O_3 in Bauru and Reunion. On the top right: the O_3 seasonal variation in Bauru and Reunion and on the bottom right: the difference between the two stations.

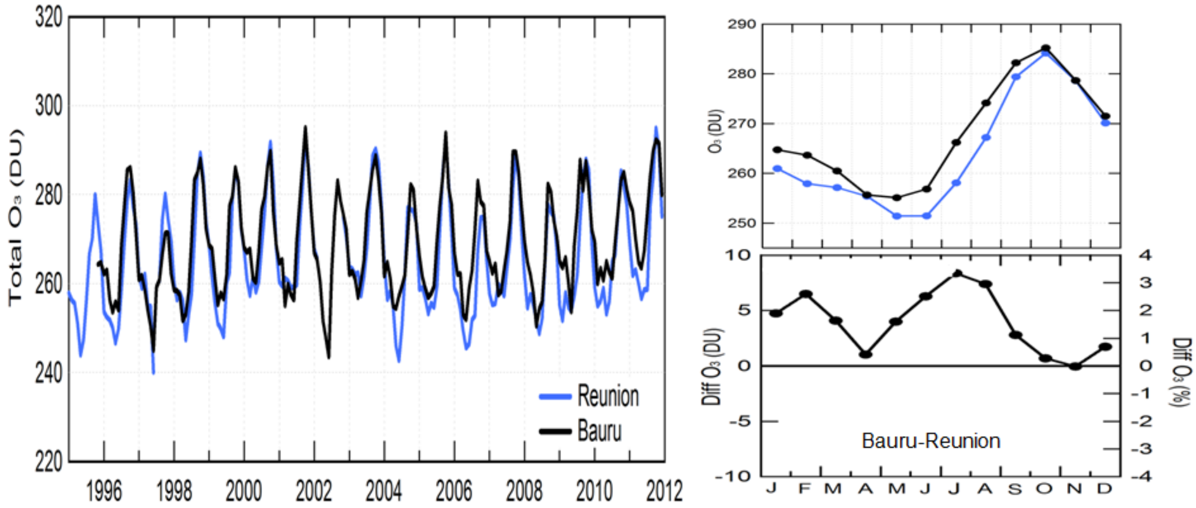

Figure 1. Left: Monthly mean time series of SAOZ total O_3 in Bauru (black) and Reunion (blue). Top right, seasonal variation in Bauru and Reunion. Bottom: difference between the two stations.

The description of the figure has been re-written, including a discussion of the origin of the O_3 winter maximum over Bauru demonstrated to come from the lower stratosphere and not the troposphere as said in the first version of the paper.

P4859/L11-14: Please include a more precise and complete trend analysis. Trend in O3 and NO2 column from both the SAOZ and satellite measurements should be calculated using a standard linear regression method and discussed in the paper

The trend study and that of the sources (solar QBO, ENSO, etc) of interannual variability is available, but is far too heavy for the current paper. The idea is to submit these results to the more appropriate ACP as soon as the current AMT paper is finished.

Table 1 What is exactly meant with 'Bauru – Reunion' number?

This was the difference between the difference of amplitude between the seasonal cycles at the two stations for each satellites. But we admit that it was confusing. The data have been removed for the table, replaced by figures showing the seasonal cycle of satellites-SAOZ differences (as asked below).

Fig 2,3 Plots showing the seasonal cycle of the satellite minus GB difference should be added for the different satellite products. For some satellite measurements there might be a systematic seasonal cycle in the difference, but this is not obvious from Fig. 2 for all satellite measurements

Figures and discussion were added in the paper.

Table 2 How are the numbers in table 2 computed?

The amplitude in DU (%) of the seasonal cycle of the difference between satellites and SAOZ now calculated. Data removed from the Table, replaced by a full discussion of the figures..

P4860/L1 It is unclear what is meant with the 5.8 DU difference.

This was an error. The sentence has been corrected as follows:

“Systematic biases are observed between satellites and SAOZ. 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 satellites are lower on average by 5.5 DU (-2.04%) in Bauru and larger by 0.5 DU (0.20%) in Reunion.

P4860/L9 It is not clear from Fig 2 that the satellite observed seasonal cycle is smaller than measured by SOAZ.

We agree that it was not clear. The subject is now fully treated by adequate plots and discussion.

P4860/L14 What is meant with sharper seasonal bias?

The author meant that the seasonal variation of the difference between SAOZ and satellites is more pronounced with OMI TOMS. The sentence has been rephrased..

P4860/L15-16 What is meant with “only EP TOMS columns are similar” ? Correct is that EP TOMS shows the smallest differences with the SOAZ measurements.

The comment of the referee is correct, the sentence was reformulated.

P4860/L20 What is meant with “the two versions of OMI are anticorrelated in 2005 and 2006”?

The two versions of OMI are showing anticorrelated variations in 2005 and 2006.

P4860/L20-24 It is mentioned that the largest dependencies on the stratospheric temperature and SZA are found for EP-TOMS and OMI-TOMS, but the comparisons with the SAOZ measurements show the smallest seasonality in the difference for these two satellite products. This apparent inconsistency should be further discussed and clarified.

The referee is right, the largest dependencies on the stratospheric temperature and SZA are found for EP-TOMS and half with OMI-TOMS. Nevertheless, these dependences are very small in the tropics because of the limited seasonality of those parameters. As shown by all the satellites bias drop in Jan-Mar in Reunion (Fig. 6) and in Oct-Mar in Bauru (Fig 4) and the larger noise there, the most important contributor to uncertainties is the cloud cover. As clouds are masking tropospheric ozone, this effect is corrected by a so called ghost column taken from climatology using cloud top and cloud fraction estimations procedures which are highly variable in the retrieval procedures.

P4862/L3 Should be Table 1?

The referee is right. The Table 1 is mentioned.

P4862/L7-8 After the bias correction of the satellite measurements one would expect that the average bias between the satellite and SOAZ measurements is close to zero. What do the (large) intercept values mean? Why are the satellite observations higher at Reunion after the bias correction?

The new merged data set presents a correlation of 96% and 97% with SAOZ data at both stations. The difference between the data is less than 1%. Note that the correction applied is only a normalization by addition of the satellite’s respective biases with SAOZ. Therefore their respective variations have not been changed. This is why a large intercept is seen on the comparison in Reunion. Satellites observed variations, which is not seen by the SAOZ.

P4862/L8 A more detailed discussion on the seasonal cycles itself at the two sites (before focusing on the differences between the seasonal cycles) is missing in the manuscript (including the impact of tropospheric ozone).

Done.

P4862/L26-29 The explanation for the difference between the satellites and SOAZ O3 columns for Oct-Dec is not convincing. The results from Thompson et al., 2003 indicate that the enhanced tropical tropospheric ozone concentrations at the two longitude regions of Bauru and La Reunion are found at similar altitudes in the middle and upper troposphere. So one would expect a similar underestimation of the ozone column by SOAZ at the two sites.

The interpretation has been revised.

"The only difference between the two data sets is the larger column (5 DU) in Reunion in Oct-Nov. The explanation of this difference is the advection over the Indian Ocean of a biomass-burning plume from Africa, resulting in an increase of ozone concentration in the middle troposphere over Reunion between 5-12 km (Randriambelo et al., 2000). The smaller sensitivity of SAOZ compared to the one of the satellites when considering 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. "

Fig 6 Please add a plot of the seasonal cycle in the NO2 column at the two sites toFig.6 (in addition to the plot of the seasonal cycle of the difference).

Done.

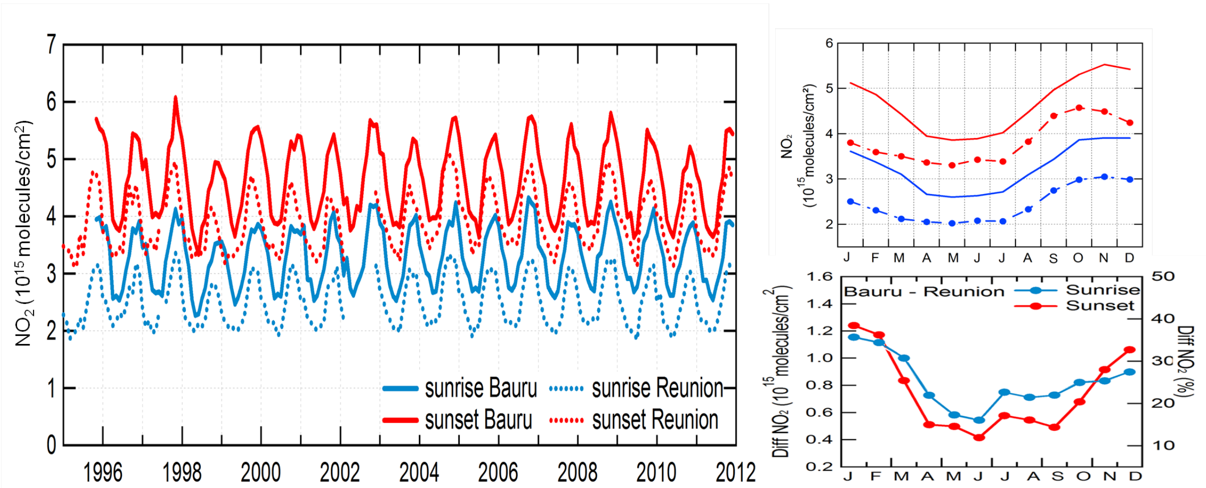

Figure 9: Left: Monthly mean time series of SAOZ total NO 2 columns at sunrise (blue) and sunset (red) in Bauru (solid lines) and Reunion (dotted). Top right, seasonal variation in Bauru and Reunion. Bottom: difference between the two stations.

P4864/L1 If the larger noise at Bauru is only visible in the satellite measurements and not in the SOAZ measurements than this could be an indication of variability in tropospheric NO2.

The referee is right. The figure does not display the SAOZ data but the satellites data only. Indeed the measurements are performed at various local time and SAOZ at sunrise and sunset. A correction for NO_2 diurnal variation has to be applied first for comparing those data.

P4864/L23-24 It is not clear why SOAZ NO2 column measurements are used to adjust the satellite measurements for the diurnal variation in NO2.

The scope of this paper is to define a new data set of satellite database. The SAOZ data are considered as reference. Since satellite measurements are closer to SAOZ sunrise observations, for comparison with SAOZ, satellite measurements have been corrected to match SAOZ sunrise.

P4865/L21-24 This is unclear. As explained in Section 2.3, the main difference between the IUP and ESA GOME NO2 columns is the normalization over the Pacific in the IUP product. Both product provide a total NO2 column based on a stratospheric AMF (but underestimating tropospheric NO2 in case of polluted conditions).

Rewritten. Differences between ESA and IUP versions due to the different AMF calculations and the correction applied for the diffuser offset

P4866/L15 This is unclear. Is a tropospheric contribution subtracted in the retrieval? I suppose you mean how the tropospheric contribution is taken into account?

The referee is correct; the authors meant the tropospheric contribution is taken into account

P4866/L17-19 This is unclear. Which OMI NO2 column is used, the stratospheric or the total column? The OMI stratospheric column is not determined as described here (see Bucsela et al., 2006).

Following the previous remarks of the referee, regarding the confusing use of total and stratospheric column, the word stratospheric column has been replaced total column everywhere.

P4867/L13 This is a strange formulation. Tropospheric NO2 has a larger effect on the total column that O3, but the previous section shows the importance of tropospheric O3 variability for the total column as well.

Correct, the authors wanted to say that the tropospheric impact on NO2 is more visible on the data than the O_3.

P4867/L13-21 In general, one would expect that the satellite NO2 retrieval is more sensitive to NO2 in the lower and middle troposphere than SAOZ (at least for the ESA GOME and the SCIA NO2 products). So in case of enhanced tropospheric NO2 in Bauru, one would expect that to be visible in the satellite vs. SOAZ comparisons. But this is not the case in Fig 9, 10 / Table 3. Is that because of the dominating contribution of lightning NOx emissions at 10-15 km in Bauru (for which both SAOZ and sat measurements are sensitive)? Please discuss

From the figure of the seasonal cycle difference between SAOZ and the merged data set one can see that satellites are observing a slightly higher seasonal cycle than SAOZ but the variations are within the error bars. The NO_2 increase during the austral summer is caused by the production of NOx by Lightning in the upper troposphere.