

Harmonisation and trends of 20-years tropical tropospheric ozone data

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Abstract. Using a convective clouds differential (CCD) method, developed in house and applied to retrievals of total ozone and cloud data from three European satellite instruments (viz. GOME/ERS-2 (1995–2003), SCIAMACHY/Envisat (2002–2012), and GOME-2/MetOp-A (2007–2015)) *Authors: monthly mean* tropical tropospheric columns of ozone (TTCO) have been retrieved, which are in good agreement with *Authors: in-situ measurements ozone sondes* *RI: (biases less than 6 DU)*. As small differences in TTCO between the individual instruments were evident, it was necessary to develop a scheme to harmonise the three datasets into one consistent time-series starting from 1996 until 2015. Correction offsets (bias) between the instruments using SCIAMACHY as intermediate reference have been calculated and six different harmonisation /merging scenarios have been evaluated. Depending on the *Authors: choice of harmonisation merging approach*, the magnitude, pattern, and uncertainty of the trends strongly vary. The harmonisation/merging represents an additional source of uncertainty in the *Authors: merged dataset and derived trend estimates trends* *Authors: (2 DU/decade on average, exceeding in most of the cases the uncertainty from the regression)*. *Authors: For the preferred harmonised dataset, the trend ranges between -4 and 4 DU decade⁻¹.* *Authors: For studying further details on tropospheric ozone trends on various spatial scales in the tropics we stick with one preferred merged dataset that shows best agreement with ozone sondes. In this merged dataset no correction was applied for GOME and mean biases with respect to SCIAMACHY in the overlapping period (2007–2012) was calculated and applied for GOME-2 in each grid-box (2.5° × 5°). In contrast with other studies we found that the tropospheric trend averaged over the tropics (15°S–15°N) is not statistically significant. The* *Authors: trend of the tropically averaged mean* tropospheric ozone trend equals -0.2 ± 0.6 DU decade⁻¹ (2σ). Regionally, tropospheric ozone has a statistically significant increase of ~ 3 DU decade⁻¹ over southern Africa (~ 1.5 % year⁻¹), the southern tropical Atlantic (~ 1.5 % year⁻¹), southeastern tropical Pacific Ocean (~ 1 % year⁻¹), and central Oceania (~ 2 % year⁻¹) *RI: Additionally, and by* ~ 2 DU decade⁻¹ over central Africa (2 – 2.5 % year⁻¹) and south India (~ 1.5 % year⁻¹). *Authors: These regional positive tropospheric ozone trends may be linked to anthropogenic activities, such as emissions in mega-cities or biomass burning in combination with changes in meteorology or/and long range transport of precursor emissions.* On the other hand, tropospheric O₃ decreases by ~ 3 DU decade⁻¹ over the Caribbean sea and parts of the North Pacific Ocean (~ -2 % year⁻¹), and by less than -2 DU decade⁻¹ over some regions of the southern Pacific and Indian Ocean (~ -0.5 – -1 % year⁻¹). *Authors: Possible reasons for these decreases are changes in dynamical processes, convection,*

~~STE, and precipitation. The comparison of the calculated trends from the current study with tropospheric ozone trends from Heue et al., 2016 and Ebojie et al., 2016 in ten selected mega-cities showed that they agree within 2σ of the trend uncertainty.~~

1 Introduction

~~Authors: The massive global population growth and urbanization of our societies inevitably leads to increased energy consumption activities related to industry, transportation, and food production. As is well known since the industrial revolution the earth's population and its standard of living have grown dramatically. At the same time the urban population has grown. Since 2011, more than 50% of the world's population live in urban areas and the population has now passed 7.5 Billion. In the past two decades, the population has grown by more than 2 Billion. An increasing population and standard of living inevitably leads to increased energy consumption, which is used in industry, transportation, and food production.~~ These human activities release a large number of atmospheric pollutants which can be harmful to public health and/or vegetation and modify the terrestrial climate (Crutzen, 2002). Climate change may also impact air pollution events^{RI}:~~since they are both interdependent~~ (WMO/IGAC, 2012). Tropospheric ozone (O_3) is regarded as one of the most important surface pollutants. ~~Authors: This is because~~ it oxidizes the biological tissues causing respiratory problems or even death (WHO, 2006), acts as a greenhouse gas (IPCC, 2007), and controls the oxidizing capacity of the troposphere (Jacob, 2000). O_3 in the troposphere is expected to increase by 60 to 80% by 2050 in Southeast Asia, India and Central America under the A2 IPCC (2013) scenario. However, the effects of climate change, especially the increased tropospheric temperatures and water vapor, may offset this increase by 10% to 17% (Stevenson et al., 2000; Grewe et al., 2001; Hauglustaine et al., 2005; IPCC, 2013).

Ozone is not directly emitted in the troposphere but it is a byproduct of the oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x) and sunlight (Crutzen, 1970; Chameides and Walker, 1973). Young et al. (2013) estimated that 4877 ± 1706 (2σ) Tg of O_3 are chemically produced every year. Additionally, 477 ± 392 Tg $\cdot yr^{-1}$ are transported from the stratosphere to the troposphere via the stratosphere to troposphere exchange (STE) (Holton and Lelieveld, 1996; Young et al., 2013). Tropospheric ozone loss is controlled by deposition to the Earth's surface and chemical destruction, mainly by photolysis to atomic oxygen ($O(^1D)$), followed by the reaction of $O(^1D)$ with water (H_2O) to produce two hydroxyl radicals ($2OH$) (Levy, 1972). The net chemical production (production minus loss) is estimated at 618 ± 550 Tg $\cdot yr^{-1}$ (2σ) (Young et al., 2013; IPCC, 2013). The mean tropospheric ozone burden is 337 ± 46 Tg (2σ) today, which is about 30% more than in 1850 (Young et al., 2013).

The sources of ozone precursors (VOCs and NO_x) can be both of anthropogenic and natural origin. ~~Authors: Anthropogenic sources of NO_x and VOCs emissions could be fossil fuel combustion, transport, electricity production and industrial processes, agriculture, solvent use and chemical manufacturing. The dominant natural sources of NO_x are lightning, biomass burning, and soil and of VOCs released by several kinds of terrestrial vegetation, mainly forests and shrubs Jacob2000. The spatio-temporal formation of low level ozone is non-linear and depends on the ratio between NO_x and VOC concentrations which determines the regime of ozone production sensitivity. In a NO_x limited regime (low NO_x and high VOCs), ozone production is insensitive to hydrocarbons and increases when NO_x concentration increases. In a VOCs limited regime (low VOCs and high NO_x) a decrease in NO_x results in an increase of ozone concentration whereas a decrease in VOCs decreases ozone Jacob2000, Seinfeld2006. VOCs limited regime is more likely to exist over~~

urban or industrial regions, while NO_x limited regime is more likely to occur in rural areas downwind of pollution sources Duncan2010. For this reason, a successful strategy to mitigate tropospheric ozone pollution effectively requires the knowledge of chemical ozone production regime in order to determine which precursor emissions (NO_x or VOCs) should be controlled. Various efforts towards reducing NO_x and VOC emissions have been taken in developed countries, particularly in Europe and North America, leading to negative surface ozone trends on a local scale (Derwent et al., 2003; Cooper et al., 2014; Parrish et al., 2014). Nevertheless, tropospheric ozone pollution is a matter of global concern, because ozone and its precursors are transported from polluted areas to clean regions over continental distances and into the free troposphere through atmospheric dynamics, increasing the tropospheric ozone abundances over remote areas. For example, air masses originated from eastern China have increased ozone abundance over Japan and North America's West Coast, despite the US legislation of reducing NO_x emissions (Parrish et al., 2009; Cooper et al., 2010; Oltmans et al., 2013; Verstraeten et al., 2016). Additionally, the high tropospheric ozone amounts noticed over the south Atlantic ocean, the so-called "tropical Atlantic paradox", arise from ozone precursor emissions by biomass burning taking place in south America and Africa (Thompson et al., 2000; Diab et al., 2003).

The long-term evolution of tropospheric ozone is complex and depends upon the evolution of precursor emissions and climate change. As the predicted increase of trace gases emissions for the next years is mainly located over low latitudes (Grenfell et al., 2003), long term observations of tropospheric ozone in the tropics should receive particular attention. Various studies have been performed in urban and rural sites using in situ data in order to estimate tropical tropospheric ozone trends. Lelieveld et al. (2004) noticed an increase in surface ozone in the order of 0.4 ppbv year⁻¹ over the northeastern tropical Atlantic, 0.4 ppbv year⁻¹ over the southeastern tropical Atlantic, and a smaller trend of 0.1 ppbv decade⁻¹ over the southwestern tropical Atlantic Ocean, based on ship-borne measurements (1977–2002). Oltmans et al. (2013) *Authors: using surface and ozonesonde observations* observed an increase of 3.8 % decade⁻¹ (0.16 ppbv year⁻¹) *Authors: in surface ozone* in Mauna Loa, Hawaii (19.5°N) in the North Pacific since 1974 and a smaller insignificant trend in the order of 0.7 % decade⁻¹ (0.01 ppbv year⁻¹) in American Samoa (14.5°S) after 1976. Additionally, Cooper et al. (2014) report a significant increase of 0.19 ppbv year⁻¹ in the subtropical site of Cape Point in South Africa from 1983 to 2011. Thompson et al. (2014) using ozonesonde data from the SHADOZ stations in Irene (25.9°S, 28.2°W) and Réunion (21.1°S, 55.5°W) noticed statistically significant trends in the middle and upper troposphere of ~ 25 % decade⁻¹ (1 ppbv year⁻¹) and ~35–45 % decade⁻¹ (2 ppbv year⁻¹) respectively during winter (June-August). Smaller positive trends appear, close to the tropopause in summer.

Satellite remote sensing is required to perform trend analysis up to global scale. *Authors: One key challenge to retrieve tropospheric ozone column amounts from the measurements of satellite remote sensing instrumentation is the accurate subtraction of stratospheric ozone from the total column ozone. This requires accurate knowledge of the pressure/ altitude level at which the tropopause is located. However in the tropics, where the the tropopause is not strongly modulated by frontal systems, the retrieval uncertainties due to the day to day variability of the tropopause can be reduced using monthly averages (Jensen et al., 2012). Most of the methods of estimating tropospheric ozone columns from space in the tropics derive from the residual approach (TOR) of Fishman and Larsen (1987) and Fishman et al. (1991). Later, more methods were developed such us the scan angle method from Kim et al. (1996), a modified residual method from Thompson and Hudson (1999), the convective clouds differential (CCD) from Ziemke et al. (1999), the cloud slicing (CS) technique from Ziemke et al. (2001), a modified*

trajectory enhanced tropospheric ozone residual method (TTOR) from Schoeberl et al. (2007) and Doughty et al. (2011), and the limb nadir matching (LNM) from Ebojie et al. (2014).

-These methods have provided valuable datasets with which tropospheric ozone trends have been derived in the tropics. For example, Ziemke et al. (2005) using the *Authors: Convective Cloud Differential* (CCD) method on Total Ozone Mapping Spectrometer (TOMS) version 8 data from 1979 to 2003, found a statistically significant positive linear trend in the mid-latitudes but not in the tropics. Beig and Singh (2007) using the same data found an increasing trend of 7–9 % decade⁻¹ over some parts of south Asia, 4–6 % decade⁻¹ over the Bay of Bengal and 2–3 % decade⁻¹ over the central Atlantic ocean and central Africa up to 2005. Kulkarni et al. (2010) using Tropospheric Ozone Residual (TOR) data from TOMS, SAGE and SBUV instruments, calculated statistically significant trends over three Indian mega-cities during 1979–2005. They showed that ozone increased by 3.4 % decade⁻¹ in Delhi during monsoon period, while it increased by 3.4–4.7 % decade⁻¹ in Hyderabad and 5–7.8 % decade⁻¹ in Bangalore during pre-monsoon and post-monsoon, respectively. *Authors: One objectives of the SCIAMACHY proposal in 1988 (Burrows et al., 1995 and references therein) was the retrieval of tropospheric ozone by making limb and nadir observation in the back scattered and reflected solar radiation.* Ebojie et al. (2016) using the full record of SCIAMACHY limb-nadir matching data (2002-2011) retrieved regional and global tropospheric ozone trends. An insignificant positive trend in the order of 0.5 DU decade⁻¹ was noticed for the northern tropics (0-20°N) and in the order of 0.3 DU decade⁻¹ in the southern tropics (0–20°S). Regionally, they reported statistically significant trends of -1.6 % year⁻¹ over Northern South America (0–10°S, 75-45°W), of 1.6 % year⁻¹ in Southern Africa (5-15°S, 25-35°E), of 1.9 % year⁻¹ over Southeast Asia (15-35°N, 80-115°E), and a trend of 1.2 % year⁻¹ over Northern Australia (20-10°S, 100-130°E). Most recently, Heue et al. (2016) published a study about tropical tropospheric ozone trends using the CCD method on a harmonised dataset consisting of data retrieved from GOME, SCIAMACHY, GOME-2 and OMI satellite instruments from July 1995–December 2015 which are based upon different total ozone and cloud retrievals as well as merging approaches. *Authors: The main differences between our CCD algorithm and the one developed by Heue et al. (2016) originate from the corrections that we have applied in the above cloud column calculation of GOME and GOME-2 data and handling of the outlier data (Leventidou et al., 2016).* The mean tropical tropospheric ozone trend that they found is 0.7 DU decade⁻¹ and regionally the trend reaches 1.8 DU decade⁻¹ near the African Atlantic coast, and -0.8 DU decade⁻¹ over the western Pacific. Seasonally, they found that the trend over the South African coast maximises in summer, whereas the negative trend over the southwest Pacific ocean maximises during autumn. *Authors: As discussed earlier, the trend results from the various studies vary significantly, and in some cases they do not agree with each other, even though the same dataset was used.*

Authors: The purpose of this study is to harmonise three individual tropical troposphere ozone (TTCO) datasets retrieved using the Leventidou et al., 2016, and to estimate the tropical tropospheric ozone trend from the merged datasets between 1996 and 2015. This paper is organised as follows: Sect. 2 presents various scenarios used in order to harmonise the three separate datasets into merged time series. Sect. 3 describes the regression model used to derive trends and presents the trend results (on global, regional, and local scale), along with their sensitivity to different harmonisation approaches used, and finally Sect.4 summarizes and discusses the findings of this study.

Authors: The main goal of this study is to derive long-term trends from our merged CCD tropical tropospheric ozone datasets. In a first step the three satellite data are merged into a consistent long-term dataset. Six possible approaches for merging the data

are considered and evaluated by comparisons to SHADOZ ozone sondes and by trend evaluations (Section 2). The comparisons to ozone sondes is used to identify the preferred merging scenario. The trend evaluation will allow us to roughly estimate the impact from the merging on trend uncertainties. In Section 3 the multiple linear regression model is briefly described. Detailed trend results for the tropics 15S -15N as well as for selected regions, and separated by seasons are presented in Section 4 for the preferred merged dataset. This paper ends with a summary and discussion (Section 5).

2 Harmonisation/merging of the TCO datasets

2.1 Tropical tropospheric O₃ data

Monthly mean TCO data have been retrieved ~~Authors: in a previous work of~~ as reported by Leventidou et al. (2016) using the Convective Clouds Differential (CCD) method on GOME (Burrows et al., 1999), SCIAMACHY (Burrows et al., 1995; Bovensmann et al., 1999), and GOME-2 (Callies et al., 2000) total ozone and cloud data from 1996 to 2015. These instruments have different properties such as spatial resolution, cloud algorithms, overpass time, etc. The individual TCO datasets have been created taking into account these specific characteristics. The individual TCO datasets have been separately validated with integrated (until 200 hPa) tropospheric ozone columns by ozone sondes from the SHADOZ network (Thompson et al., 2003) (see: Leventidou et al. (2016)). The biases between them have been found to be within ~~Authors: 1.6–6.46~~ DU ~~Authors: and the root~~ mean-square (RMS) deviation less than 13 DU for all the instruments which is mostly within the uncertainties of the mean biases of 6 DU (1 sigma). One large source of uncertainties in these comparisons are the low sampling of the sondes (typically less than five launches in a month) and the fact that CCD ozone is only derived as monthly means covering rather large areas (grid box). . The uncertainty of the tropospheric ozone column retrieval with the CCD method is in the order of 3 DU (~ 10%). For most of the stations, the bias with the ozone sondes is within the retrieval uncertainty, with the exception of GOME-2 TCO which is in the order of 5 DU. Finally, the CCD TCO from SCIAMACHY data have been compared with the Limb-Nadir-Matching (LNM) tropospheric O₃ columns up to 200 hPa altitude from the same satellite instrument, showing that the bias and the RMS values are within the ones calculated for the comparison with ozone sondes.

2.2 Correction offsets between GOME and GOME-2 with respect to SCIAMACHY TCO

~~Authors:~~ For trend calculations the existence of a constant bias (in clouds and ozone) between the instruments, caused by the spatial and temporal differences of the individual instruments, can be removed by using a suitable merging approach as will be shown here. Correction offsets have been calculated in order to create one consistent tropical tropospheric columns dataset from the CCD method for the whole timespan of the ~~R1: European satellites operation~~ operation of the European satellites (1996–2015). SCIAMACHY TCO were used as the reference ~~for the correction offset calculation~~, because ~~R1:~~ it is the only instrument that overlaps (2002–2012) both with GOME and GOME-2 and has the smallest bias with respect to the ozone sondes (< 2 DU). The average difference (bias) for each grid-box during the common years of the instruments operation (2002 for SCIAMACHY–GOME and 2007–2012 for SCIAMACHY–GOME-2) was computed and applied (added) to GOME and GOME-2 TCO data. The mean

biases, shown in Fig. 1, range between -6 and 6 DU for GOME, with positive differences (3–6 DU) located mainly over land. There are also two ^{Authors:} ~~stripes~~ regions with positive biases appearing north of 7.5°N until 20°N, and between -5 and -7.5°S. For GOME-2, the bias ranges between -8 and 0 DU, with the biases being smaller over land, especially over south America and north/central Africa. Possible reasons for the biases are the different cloud algorithms used for each instrument (SACURA for
5 SCIAMACHY and FRESCO for GOME and GOME-2) and the small biases noticed in the total ozone columns (e.g. ~ -2.5 DU between SCIAMACHY and GOME-2). ^{R2:} Differences in spatial resolution and overpass time of the instruments have also minor contributions in the biases.

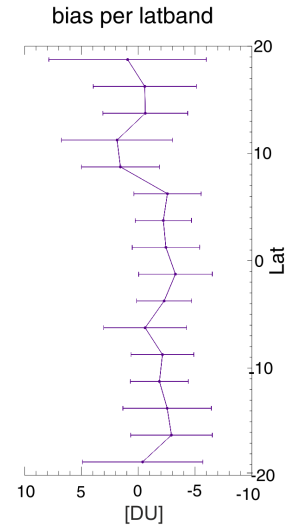
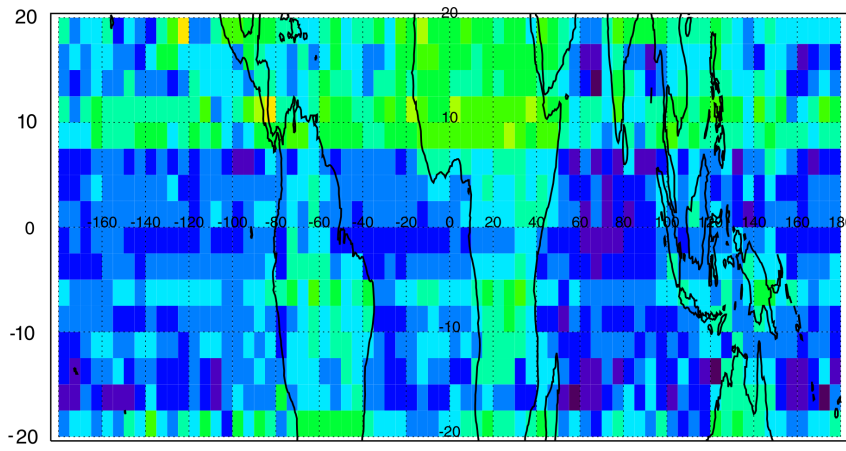
The latitudinal dependence of the mean bias is shown at the bottom of Fig. 1. The average differences between GOME and GOME-2 with SCIAMACHY are generally negative (less than 5 DU) in all latitude bands with the exception of the northern
10 tropical latitudes, where GOME mean biases are positive (0–2 DU). GOME mean biases have stronger latitudinal variability than those of GOME-2. This behavior may be explained by the short time of common operation (Jan. 2002–Jun. 2003) between GOME and SCIAMACHY instruments. The 1σ standard deviation (uncertainty bars) of the mean bias per latitude band is comparable to the magnitude of the biases, ranging from less than 5 DU close to the equator to 7 DU for latitude bands close to the tropical borders. For the case of GOME, the mean correction offset is -1.2 DU, whereas for GOME-2, it is -5.7 DU.
15 The mean offset of GOME-2 is almost twice the CCD retrieval uncertainty (~3 DU). For this reason and because of the large biases with the ozonesonde data, ^{R1:} it seems reasonable to apply a correction for the GOME-2 TCO dataset.

The drift on the average differences (bias), β , has been estimated using a simple linear regression model such as: $Y = \alpha + \beta \cdot X_t$, where Y is the time-series of the biases, X_t is the time variable in months, and α is the offset. The drift between SCIAMACHY and GOME-2 is shown in Fig. 2. There are not enough overlapping years to calculate a trend in the GOME-
20 SCIAMACHY difference time-series. The drift is generally less than ~0.4 DU per year and is statistically not significant ($\beta/\sigma_\beta < 2$ (Weatherhead et al., 1998; Wilks, 2011)) for nearly all grid boxes, with the exception of the 17.5–20 °N latitude band, where it is statistically significant and exceeds 1 DU year⁻¹. During local winter months at the tropical borders, there are often missing TCO data owing to the movement of the ITCZ and the inability to retrieve a reliable stratospheric O₃ column. For this reason, calculated drifts for these latitudes are not reliable, in spite of the fact that they might appear to be statistically
25 significant. ^{R1:} ~~They do not cover the entire studied period (2007–2012) but only a subset of them.~~ Consequently, the trend of the correction offsets is ^{Authors:} ~~not considered here~~ considered to be negligible.

2.3 Six Harmonisation scenarios

The creation of a consistent tropical tropospheric ozone column dataset from multiple satellite instruments demands a careful selection of the optimal harmonisation approach, since it introduces additional uncertainty in the merged dataset. ^{Authors:} ~~For this~~
30 ~~purpose,~~ Six harmonisation scenarios have been tested. They all use the SCIAMACHY TCO dataset as a reference, which is in the middle of the time period, as follows:

SCIA - GOME Trop. O3 column bias



SCIA - GOME-2 Trop. O3 column bias

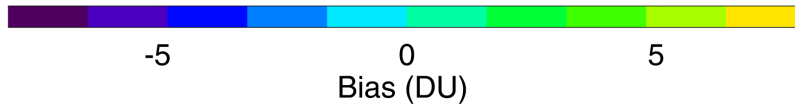
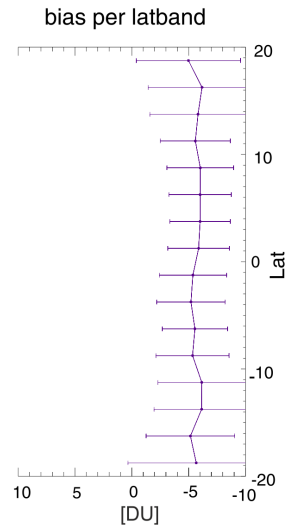
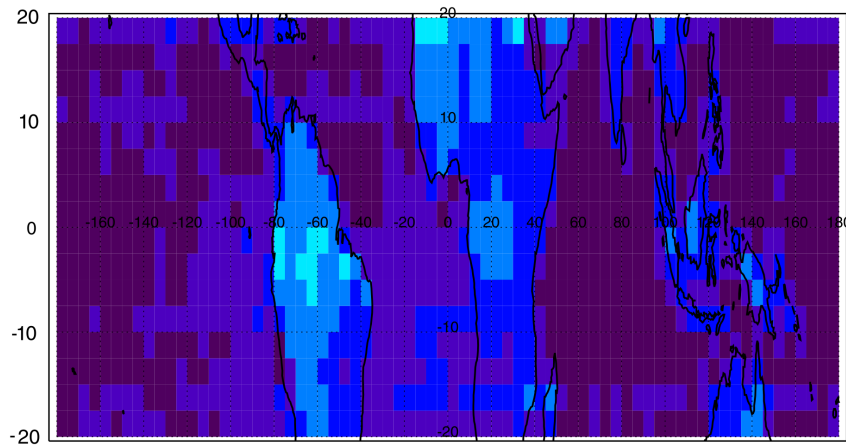


Figure 1. Correction offsets using SCIAMACHY TCO as reference. (up) Correction offset for GOME: average difference of GOME from SCIAMACHY TCO for the years 2002-2003. (down) Correction offset for GOME-2: average difference of GOME-2 from SCIAMACHY TCO (in DU) for the years 2007-2012. On the right are shown the biases per latitude band. The error bars denote the 1σ standard deviations of the latitudinally averaged biases.

SCIA - GOME2 Trop. O3 column Bias drift

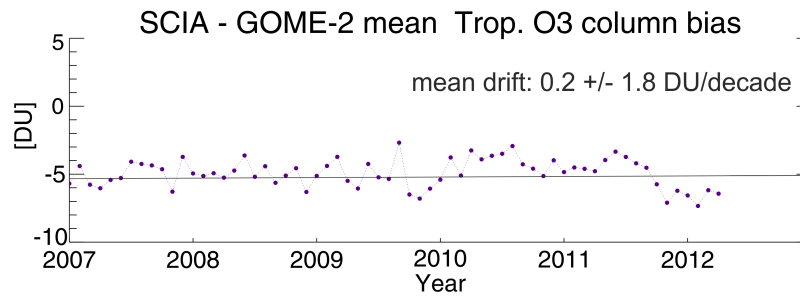
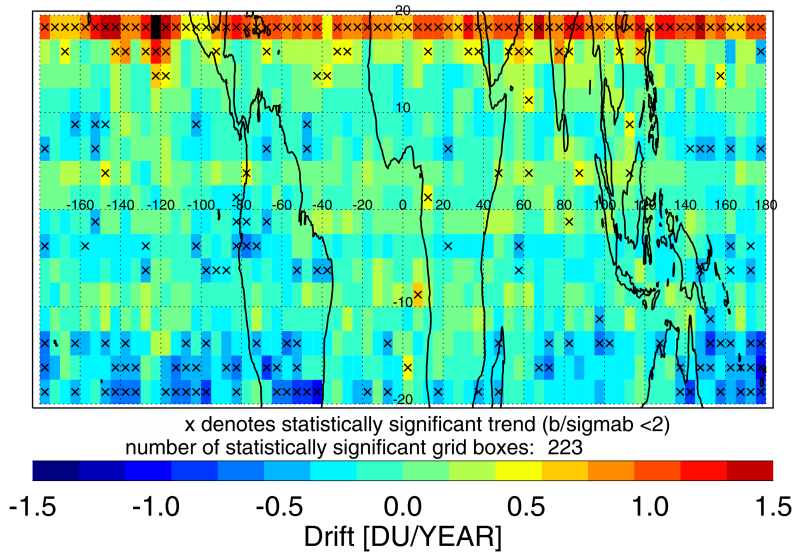


Figure 2. Top: drift in the correction offset for GOME-2. Black "x" denotes statistically significant trend. Bottom: average difference and drift in the correction offset for GOME-2 between 2007 and 2012.

5

- **Scenario 1:** No correction applied to GOME data (which maybe justified by the very short overlap period), while GOME-2 is corrected using for each grid-box the mean bias with respect SCIAMACHY for the common years of operation (2007–2012 for GOME-2).
- **Scenario 2:** No correction applied to GOME data and the average bias (-5.7 DU) with respect SCIAMACHY is added to all GOME-2 TCO data.
- **Scenario 3:** GOME and GOME-2 have been corrected using for each grid-box the mean bias with respect to SCIAMACHY for the common years of operation.

- **Scenario 4:** The average bias with respect to SCIAMACHY (-1.2 DU) is added to all GOME TTCO data, whereas GOME-2 TTCO has been corrected using for each grid-box the mean bias with respect to SCIAMACHY for the common years of operation (2002 for GOME and 2007-2012 for GOME-2).
- **Scenario 5:** The average bias with respect to SCIAMACHY (-1.2 DU) for GOME and for GOME-2 (-5.7 DU) is added to all GOME and GOME-2 TTCO data respectively.
- **Scenario 6:** No correction applied to GOME, whereas for GOME-2 both the bias and the drift is included in the correction of GOME-2 TTCO in each grid-box.

After the correction terms for all scenarios have been applied to the original data, the "corrected" GOME (1996-2002) and GOME-2 (2007-2015) TTCO were averaged with the ones from SCIAMACHY (2003-2012) for the overlapping months (Jan. 2002–Jun. 2003 and Jan. 2007–Dec. 2012, respectively).

Table 1. *RI:* Mean differences (in DU) between the harmonised TTCO datasets using six different harmonisation scenarios with integrated ozone columns until 200 hPa from nine ozonesonde stations. The regions where the harmonisation scenarios have the smallest biases with ozone sondes are marked bold Mean differences (in DU) between merged TTCO data, retrieved with the CCD method using six possible harmonisation scenarios, with integrated ozone columns up to 200 hPa from nine SHADOZ stations. The stations marked with asterisk present data from the newest reprocessed (V05.1_R) version (Thompson et al., 2007; Witte et al., 2017). The regions where the merged scenarios have the smallest biases with the ozone sondes are marked with bold. Scenario 1 has the smallest mean bias for all the stations.

CCD – Sondes TTCO [DU] /Site	scenario 1	scenario 2	scenario 3	scenario 4	scenario 5	scenario 6
Am. Samoa (14.4S,170.6W)*	-0.89	-0.92	-1.99	-0.61	-0.93	4.59
Ascension (8S,14.4W)	0.03	-0.14	-0.77	-0.42	-0.60	0.03
Java (7.6S,111E)	-0.11	-0.12	-1.12	-0.54	-0.55	-0.11
Kuala Lumpur (2.7N,101.7E)	-1.81	-2.12	-2.12	-2.14	-2.48	-1.78
Nairobi (1.3S,36.8E)	1.81	1.10	1.80	1.48	0.74	1.84
Natal (5.4S,35.4W)	0.56	0.63	-0.21	0.22	0.28	0.57
Paramaribo (5.8N,55.2W)*	-2.98	-2.95	-3.02	-4.11	-4.34	-0.11
Mean bias for all stations	-0.48	-0.64	-1.06	0.87	-1.13	0.72

In order to decide which is the most suitable harmonisation scenario, the various merged datasets were compared with integrated ozone columns up to 200 hPa altitude from nine ozonesonde stations: (a) Ascension (8°S, 14.4°W), b) Paramaribo (5.8°N, 55.2°W), c) Java (7.6°S, 111°E), d) Natal (5.4°S, 35.4°W), e) Samoa (14.4°S, 170.6°W), f) Nairobi (1.4°S, 36.8°E), and g) Kuala Lumpur (2.7°S, 101.7°E). *Authors:* Fiji (18.1S, 178.4E) station is not included in the comparison because it is highly influenced by air coming in from mid-latitudes and the upper troposphere (Thompson et al., 2017). *Authors:* Hilo (19.4N, 155.4W) is influenced by volcanic out-gassing with high SO₂ emissions, resulting in negligible ozone concentrations at the boundary layer. Therefore, this station is also not included. As seen in Table 1, the mean bias between the six harmonised

TTCO datasets and the ozone sondes range between R^1 : ~~2.5 and 1.8-1.1 and 0.9~~ DU which is well within the retrieval uncertainty R^1 : showing that for most scenarios the spatio-temporal offsets with respect to ozonesondes are minimised. However, the biases of each scenario with ozone sondes are very close to each other for every station. The same R^1 ; occurs for the correlation between the harmonised TTCO datasets and the ozone sondes (not shown here). Although the comparison between the TTCO

5 from the individual harmonised scenarios and the ozonesonde data does not favor clearly any harmonisation scenario, R^1 : ~~the scenarios that can be confidently rejected according to this comparison are scenarios 3, 4 and 5, which have the biggest bias with the ozone sondes. Scenario 6 presents smaller bias at four out of nine ozonesonde stations, whereas scenario 1 at three out of nine ozonesonde stations. Nevertheless, scenario 6 has larger biases with respect to ozone sondes compared to scenario 1 (with the exception of one station).~~ the scenarios that can be confidently rejected are scenarios 3, 4 and 5 where GOME data are corrected with respect to SCIAMACHY since the overlap period between GOME

10 and SCIAMACHY is very short (10 months, 8/2002-6/2003). Scenario 6 can also be rejected due to the fact that the drift in GOME-2 correction offset at 81% of the grid-boxes is statistically non significant. Lack of significant drifts in the comparison between GOME-2 and SCIAMACHY over the overlapping period shows that the data records are quite stable. Finally, scenario 1 (no drift corrections and bias correction for GOME-2) has the smallest mean bias with the ozone sondes (-0.4 DU). For these reasons, scenario 1 has been selected to be the preferred harmonisation scenario for merging the TTCO datasets.

15 ~~Authors: All results (without explicit indication of the harmonisation scenario used) presented here are based on harmonisation scenario 1.~~ Before we discuss in details tropical tropospheric trends using the preferred scenario (Section 5), we try to estimate the potential contribution of the merging approaches to trend uncertainties in tropical tropospheric ozone.

2.4 Sensitivity of the trend to the merging approach

~~Authors: The statistical trend uncertainty derived from a single dataset usually does not account for uncertainties due to the merging approach applied. Here we will provide a rough estimate on how large the trend uncertainties may be. We applied the multivariate linear regression model (see Section 3, Eq. 1 for details on the regression) Authors: and correction for noise have been applied to six individual harmonised CCD tropical tropospheric ozone columns datasets (see: Section 4) from 1996 to 2015. Authors: to derive trends from all six merged datasets.~~ The tropospheric O_3 trends from all scenarios range between ~ -4 and 4 DU decade $^{-1}$, with mean values between 0 and 0.8 DU decade $^{-1}$, without any of them being statistically significant ~~Authors: for the global tropics~~ (see Fig. S1 in te supplement). The maximum trend difference among all six harmonisation scenarios is on average 2 DU decade $^{-1}$ exceeding the $2\sigma_\beta$ uncertainty of the trends which is ~ 1.2 DU decade $^{-1}$ (see Fig. S2 in te supplement). These differences in the trends among the differently harmonised datasets reveal the additional uncertainty which results from the harmonisation procedure of multiple TTCO datasets.

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~~R^2 : The maximum absolute differences ($>3-6$ DU decade $^{-1}$) are found mainly over land and more specifically over south America, and northern Africa, while the minimum absolute differences are over the Oceans with the exception of the Indian and the southern Pacific Ocean. Nevertheless, all scenarios agree that there is a positive trend of tropospheric ozone over the south tropical Atlantic Ocean, and some parts of central Africa and India, while a negative trend appears over the Caribbean sea, the north and south Pacific Ocean. Scenarios 1, 4, and 6 have a similar pattern with each other which is caused by the absence of correction of the GOME TTCO dataset. Nevertheless, the range of the trends is different, with scenario 4 showing higher positive trends ($2-4$ DU decade $^{-1}$), mainly over Africa, south America and the southern tropical borders. Scenarios 2 and 5 have also similar pattern with each other, driven~~

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by the average offset applied to GOME-2 data. The pattern of these scenarios consist of a characteristic decrease in tropospheric ozone (-2 DU decade⁻¹) over central-south America and over the Indonesian peninsula. The tropospheric O₃ trends calculated with scenario 3 repeat the meridional pattern of GOME correction offsets (see: Fig. 1), which appears as an artifact in the trend results.

3 The multi-linear regression trend model

5 Changes in ozone precursor emissions due to urbanization and land use, along with changes in the atmospheric ^{Authors: oscillations} dynamics which ^{Authors: affect processes that modulate the impact} tropical upwelling or the horizontal ozone transport, may cause long-term changes in the tropospheric ozone burden. ^{Authors: and influence} This in turn impacts the photochemical ozone production and loss in the troposphere (Ziemke and Chandra, 2003; Solomon et al., 2007; Chandra et al., 2009; Voulgarakis et al., 2010; WMO, 2011; Neu et al., 2014; Monks et al., 2015). Some of these factors can be represented by periodic seasonal proxies, such as the El Niño Southern Oscillation (ENSO), the quasi-biennial oscillation (QBO), and the solar cycle (SC). These indexes are embodied in the trend model described here. ^{Authors: The seasonal variation of the linear trend is also included using harmonic functions, which represent the annual, semi-annual and quarterly harmonic oscillations.}

The time series of the monthly mean tropical tropospheric ozone columns Y_t at a specific latitude and longitude (i, j) (running every 2.5° and 5°, respectively) ^{Authors: can be generally} is described by the following trend model:

$$15 \quad Y_t(i, j) = \alpha(i, j) + \beta(i, j) \cdot X_t + S_t(i, j) + R_t(i, j) + N_t(i, j), \quad (1)$$

where a is the offset ^{RI: TFCO} for the first month $t=1$, β the linear trend in DU month⁻¹, X the time variable (months running from zero to 239) covering the years 1996–2015, S_t is the seasonal variation, R_t are the terms with the various proxies (ENSO, QBO, solar cycle) and N_t is the noise of the time series, representing the unexplained portion of the variability in the fit. The seasonal cycle is modeled by a Fourier series (see Eq. 2), with $\gamma_{11}, \gamma_{21}, \gamma_{12}, \gamma_{22}, \gamma_{13}, \gamma_{23}$ being the regression coefficients for 12-, 6- and 4-month periodicities, with sine and cosine terms for each periodicity, respectively:

$$20 \quad S_t(i, j) = \sum_{n=1}^3 (\gamma_{1n} \cdot \sin(\frac{2 \cdot \pi \cdot n \cdot t}{12}) + \gamma_{2n} \cdot \cos(\frac{2 \cdot \pi \cdot n \cdot t}{12})). \quad (2)$$

R_t , represents the time dependent regression coefficients for the ENSO, QBO, and solar cycle proxies which can be expressed as:

$$R_t = \delta \cdot ENSO_t + \varepsilon \cdot QBO_{30t} + \zeta \cdot QBO_{50t} + \eta \cdot SC_t. \quad (3)$$

25 Because the tropospheric ozone lifetime approaches a month, the pattern of tropospheric ozone for a month has the tendency to ^{RI: recur on the next month} persist into the next month. Even after removing ^{Authors: to the largest extent} the seasonal and other effects in the time series shown in Eq. 1, there is still a month-to-month correlation (ϕ) in residuals. This phenomena is called persistence (Wilks, 2011) and is quantified by the degree of autocorrelation of a parameter, shifted by p time steps (lag p). Therefore, the ^{Authors: first order} autocorrelation of the noise ^{Authors: (AR[1])} is included in the model, as explained by Weatherhead et al. (1998).

4 Tropical tropospheric ozone trends

For the rest of the discussion about tropical tropospheric ozone, the trend refers to the preferred harmonisation scenario (scenario 1).

4.1 Tropical distribution of tropospheric O3 trends and mean tropical trend

5 Figure 3 summarises the tropical tropospheric ozone trends calculated in a $2.5^\circ \times 5^\circ$ grid as derived from the preferred merged CCD TCO dataset *Authors: using the multivariate regression model* (Eq. 1) between 1996 and 2015. As shown in 3a, the trend varies between -3.2 and 3.7 DU decade⁻¹, and the average trend for the period 1996–2015 is statistically not-significant and equal to -0.1 ± 1.2 DU decade⁻¹ (2σ). *Authors: The noise is random (white noise) following very well a Gaussian distribution.* Fig. 3b shows the 2σ of the trend, which is in the order of $\sim 0-4$ DU decade⁻¹ (mean: 1.2 DU decade⁻¹), with higher values at the tropical
10 borders and values close to zero along the equator. Fig. 3c shows the correlation between the model and the time-series. The correlation coefficient reaches 1 over the north and central-east Pacific and the southern Atlantic Ocean. The regions of smaller correlations are mostly over the west Pacific, the Caribbean sea, the south-east Asia, and over the central African continent. The main reason for the low correlation is the weak seasonal cycle observed in these regions. Fig. 3d shows the RMS between the time-series and the model fit. The RMS is less than 3 DU close to equator and reaches 7 DU at the tropical borders. Fig.
15 3e presents only those grid boxes where the trend is statistically significant and exceeds the maximum difference of the trends calculated from all six scenarios. This additional criterion (to exceed the differences between harmonisation scenarios) allows us to identify grid boxes that have significant trends with higher confidence. Using this stricter criterion, tropospheric ozone trends are positive over some parts of central Africa (~ 2 DU decade⁻¹), southern Africa and Atlantic Ocean ($\sim 2-3$ DU decade⁻¹), India (~ 2 DU decade⁻¹) and Oceania ($\sim 3-4$ DU decade⁻¹) but are negative over the Caribbean sea and parts of
20 North Pacific Ocean ($\sim -2-3$ DU decade⁻¹), as well as over some regions of the southern Pacific Ocean (~ -2 DU decade⁻¹) seem to be relevant, however, for all other grid boxes trends are highly uncertain and mainly dependent on the choice of the harmonisation scenario. The negative trends appearing *Authors: as a stripe at a region at the* northern latitudes (Caribbean sea and northern Pacific) may *RI: still* be an artifact of the data-set (low sampling of data, *RI: 54 out of 240 months of data*). Finally, Fig. 3f shows the tropical tropospheric ozone trends in per cent per year ($\% \text{ year}^{-1}$) that are statistically significant *for the*
25 *harmonised TCO data according to scenario 1 (S1)*. Here the maximum increase is observed over central Africa, $\sim 3\% \text{ year}^{-1}$, over southern Africa, south tropical Atlantic and Oceania $\sim 1.5\% \text{ year}^{-1}$, and finally over India and south-east Asia $\sim 1\% \text{ year}^{-1}$. The maximum tropospheric ozone decreasing trend is observed over the Caribbean sea and the north-east tropical Pacific, about $\sim -2\% \text{ year}^{-1}$, followed by the central-south Pacific and Indian Ocean, $\sim -1\% \text{ year}^{-1}$.

Authors: The southern and northern boundary of the tropics (15–20S and 15–20N) is strongly influenced by stratospheric intrusions via tropopause foldings and air masses being transported from the mid-latitudes and the upper troposphere (Pickering et al., 2001; Thompson et al., 2017). *Authors: Therefore, in order to estimate a more reliable mean trend for the tropics the multivariate regression model (Eq. 1) has been applied to the mean tropical time-series between 15S to 15N*. The fit results are shown in Fig. 4. The mean ("global") tropical trend equals -0.2 ± 0.6 DU decade⁻¹ (2 sigma). This means that there is no
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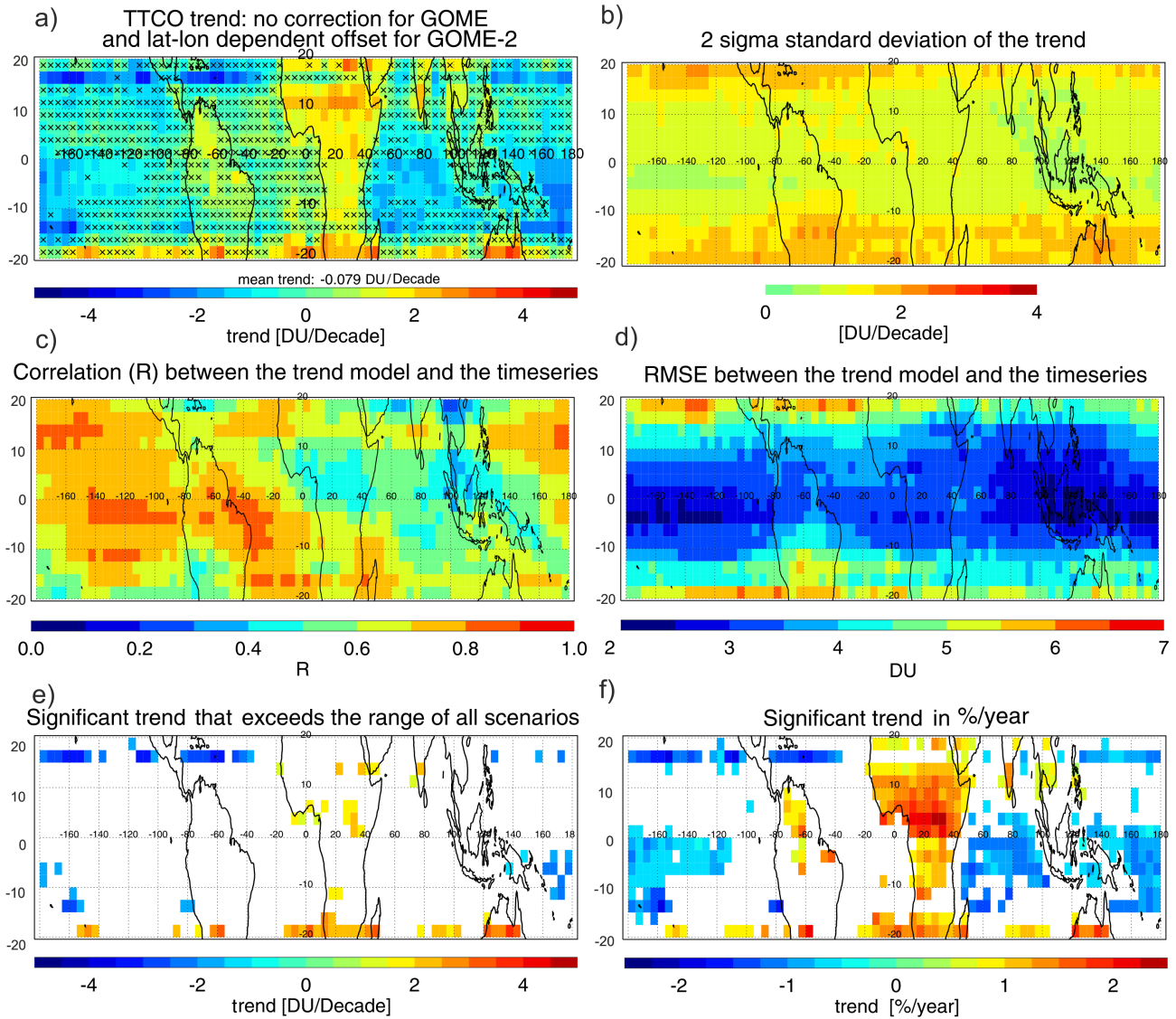


Figure 3. (a) Tropical tropospheric ozone trends using a linear multivariate first order auto-regression model for the selected harmonised scenario 1 in DU decade^{-1} . Grid-boxes marked with "x" are statistically non-significant at the 95% confidence level ($b > 2\sigma_\beta$). b) 2σ standard deviation of the trend. c) The correlation coefficient, R, between the multi-linear trend model fit and the original time-series. d) The RMS error between the trend model and the time-series. e) The statistically significant trend that exceeds the maximum absolute difference of the trends calculated for all six scenarios. f) The significant tropical tropospheric ozone trend in $\% \text{ year}^{-1}$.

significant trend for tropospheric ozone in the tropics. ^{RI:}The mean tropospheric ozone trend is in agreement with Ziemke et al. (2005) ^{RI:}(using solar backscatter ultraviolet (SBUV) and Total Ozone Mapping Spectrometer (TOMS) version 8 data from 1979 to 2003) and Ebojje et al. (2016) ^{RI:}(using SCIAMACHY limb-nadir-matching (LNM) observations during the period 2003–2011) who also indicated insignificant and near zero global trends in the tropics, although their analysis was based on different datasets and covered shorter time periods. ^{Authors:}Nevertheless, Heue et al.(2016), using a similar CCD method on the same period and satellite instruments, reported a significant average increase of 0.7 ± 0.1 DU decade⁻¹. The tropical mean

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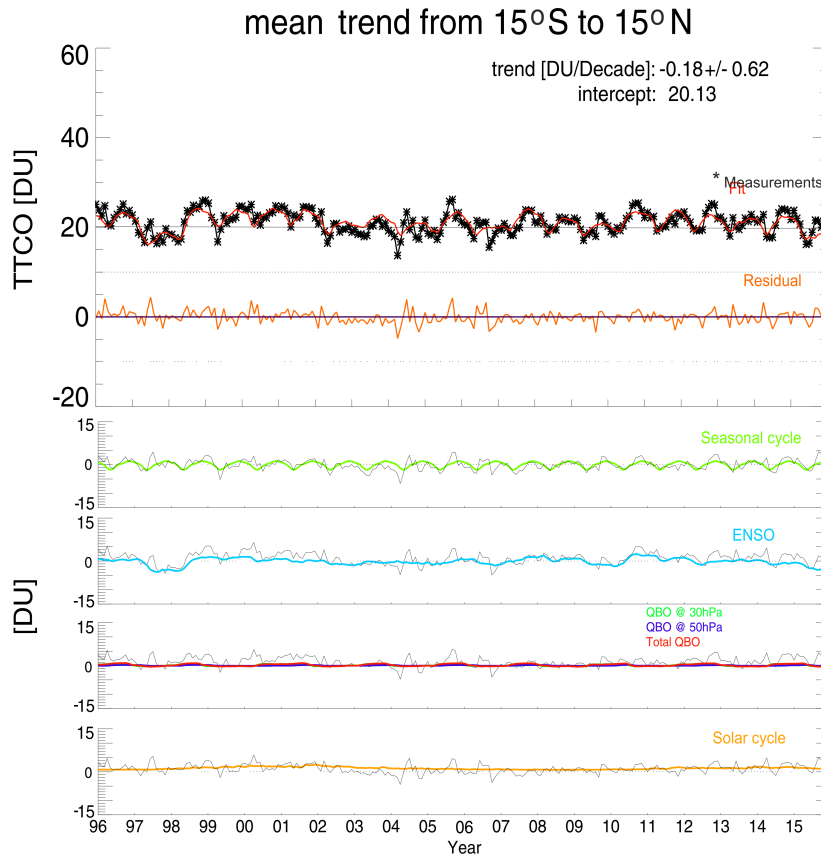


Figure 4. Mean tropical tropospheric ozone trends between 15°S – 15°N for the period 1996 to 2015. Top: The multivariate linear trend (black), the fit (red) and the residual (orange) are over-plotted. The mean tropical tropospheric ozone trend is equal to -0.18 and the 2σ uncertainty of the trend is ± 0.62 DU decade⁻¹. The next panels show the harmonic functions (green), ENSO (light blue), QBO (red), solar (orange). Overlaid in black for all proxies are the time series with all fit terms removed except the particular fit parameter.

tropospheric ozone time-series (black stars) shows a seasonal cycle with higher values in ~~late summer-autumn~~ Jul.–Oct.. The time-series are well followed by the regressed tropospheric ozone (red line) and the residual (orange line in upper panel) is less than ^{Authors:}35 DU. The seasonal cycle contributes the most to the TCO variability in the tropics by about ± 32 DU. Tropical tropospheric ozone is reduced by ~~3-4~~ DU during El Niño years (1997-98, 2006-07, 2009-10, 2015) and slightly increases

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by 1 – 2 DU during strong La Niña years (1999-00, 2007-08, 2010-11). QBO and the solar cycle, do not contribute to the inter-annual mean tropical tropospheric ozone variability. Overlaid in black for all proxies are the time series with all fit terms removed except the particular fit parameter. This allows us to relate the magnitude of changes due to a selected process to the observed residuals (or unexplained variations).

5 4.2 *Authors: Regional and Mega-cities* **Regionally averaged tropical tropospheric ozone trends**

We also studied regional trends focusing on the regions where the trends are statistically significant. The TTCO have been regionally averaged for eight regions and the regression analysis applied to them. The regions are: A: Caribbean Sea (15° – 17.5°, -85° – -45°), B: India (10° – 20°, 70° – 85°), C: north-south America (0° – 10°, -75° – -60°), D: North Africa (5° – 15°, -17.5° – 50°), E: east Pacific Ocean (0° – 7.5°, -180° – -110°), F: Indian Ocean (0° – 7.5°, 50° – 100°), G: west Pacific Ocean (0° – 7.5°, 160° – 180°), and H: southern Africa (-20° – -12.5°, 10° – 50°).

Table 2. Regional tropospheric ozone trends in 8 tropical regions. Bold are the regions where the trend is greater than three times the standard deviation of the trend (3σ).

Area	Tropospheric O ₃ trend $\pm 2\sigma$ in DU decade ⁻¹
A) Caribbean sea	-1.59 \pm 1.30
B) India	1.10 \pm 0.86
C) North South America	0.99 \pm 0.94
D) North Africa	1.54 \pm 1.09
E) East Pacific Ocean	-1.21 \pm 0.65
F) Indian Ocean	-1.61 \pm 0.83
G) West Pacific Ocean	-1.87 \pm 0.72
H) South Africa	1.44 \pm 1.28

As shown in Figure 5 and Table 2, regions B, C, D and H show significant increase in the order of 1–1.5 DU decade⁻¹ and regions A, E, F, and G a significant ozone decrease in the order of -1.2 – 1.9 DU decade⁻¹. The observed significant positive changes in tropospheric O₃ over north Africa and parts of the Arabian sea (D), south Africa and the southern African outflow (H), parts of India (B), and north south America (C) agree well with results of Lelieveld et al. (2004), Beig and Singh (2007), Kulkarni et al. (2010), Ebojie et al. (2016) and Heue et al. (2016) who also observed an increasing ozone trend over these regions. *Authors: Although, Ebojie et al. (2016) observe a decreasing trend of -0.5 DU decade-1 in tropospheric ozone over north-east Africa (D).* *RI: They can be attributed to changes in anthropogenic NO_x and other tropospheric O₃ precursors, due to population and energy consumption increases, which are transported to these areas (Hilboll2013, Kulkarni2010, Dahlmann2011, Schneider2015, Cooper2014, Duncan2016,*

Trop. O3 column trend

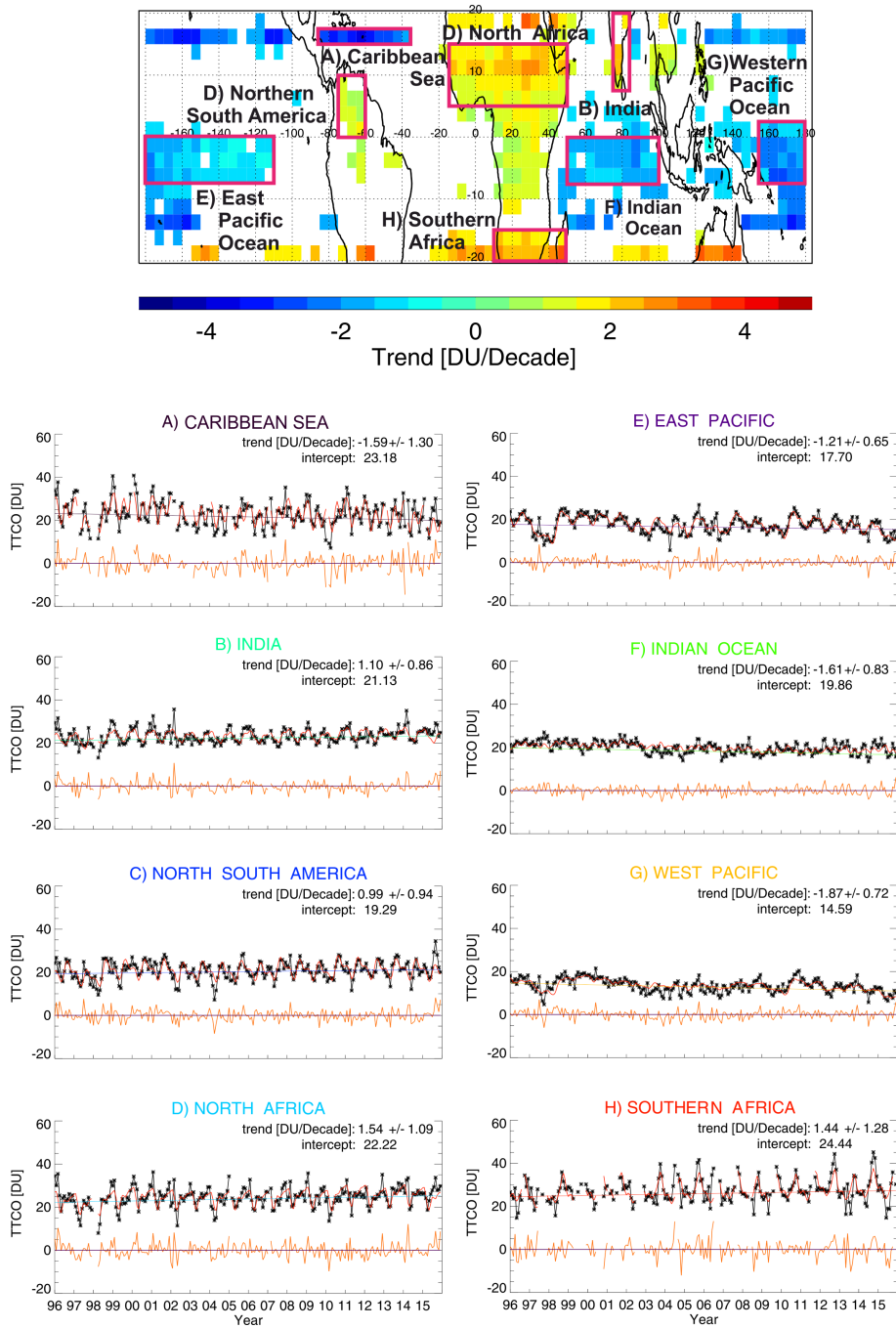


Figure 5. Tropical tropospheric ozone trend in A) central America, B) India, C) east Pacific Ocean, D) South America, E) central Atlantic Ocean, F) Indian Ocean, G) south Atlantic Ocean, and H) southern Africa.

Hilboll2017). Biomass burning may also have an impact on tropospheric ozone changes. For example, the burned area in southern tropical Africa increased by 1.8 %/yr during the period 2000 to 2011 (Giglio2013). (Ziemke2009b) and (Wai2014) estimated that biomass burning can contribute to an increase in tropospheric ozone column by $\sim 20\%$. Additionally, changes in meteorology, convection, and dynamical oscillations, such as the MJO, stratospheric intrusions (STE) and shorter timescale atmospheric dynamics or cyclones may have influence the transport of pollutants and contribute locally to observed tropo-
5 spheric ozone changes (Ziemke2009b, Beig2007, Parrish2009, Ebojie2016, Chandra2004, Oltmans2004, Sauvage2007). Another factor that may influence tropospheric ozone are the changes in stratospheric ozone column. For example, an increase in the tropical upwelling caused by a stronger Brewer-Dobson circulation is expected to reduce both lower stratospheric and the total column ozone in the tropics, increasing the UV-B radiation reaching the troposphere (WMO2014). This could result in an enhance of tropospheric ozone photolysis (photochemical ozone sink). However, the increase of UV-B radiation at the surface would also lead to increased concentrations of OH (hydroxyl radicals) and subsequently increased concentrations of HO₂ and RO₂ radicals, which
10 may enhance the production of ozone if NO_x are available (e.g. in mega-cities) (UNEP1998). Consequently, there are multiple feedbacks from these changes that could either increase or decrease ozone in the troposphere.

The negative changes in TCO over the Caribbean sea (A) are in agreement with the results of Ebojie et al. (2016). ^{RI:} Although they might be influenced by the decrease in NO_x emissions over the north American continent (Duncan2016, Hilboll2013) or by changes in strato-
spheric intrusions via the tropopause foldings (Hwang2005, Ojha2017) However, the observed trends over the northern and southern tropical
15 latitudes (18° – 20° in SH and NH) should be generally interpreted with caution because they are influenced by low sampling of data ^{RI:} Despite the fact that might appear to be statistically significant, they should be interpreted with caution since they are influenced by gaps in the TCO time-series due to the movement of the ITCZ, which reduces the cloudy data during local winters and makes the above cloud ozone column (ACCO) retrieval difficult, violating in some cases the invariance of the ACCO per latitude band. The decreasing tropospheric ozone trend over the ^{Authors: western} Pacific (^{Authors: E and G}) and Indian (F) Oceans agrees well with Heue
20 et al. (2016) ^{Authors: results, RI:} It might be associated with changes in the burden of organic and inorganic halogens on these areas as well as changes in dissolved organic matter (DOM) photochemistry in surface waters could be an additional source of volatile organic compounds that can contribute to ozone destruction (Dickerson1999, Ebojie2016). Additionally it may be attributed to changes in the humidity burden of the troposphere. For example, (Fontaine2011) indicated that the Outgoing Long-wave Radiation (OLR), which allows to differentiate between clear-sky (high OLR) and deep convective regions (low OLR) has been decreasing over these regions, which can indicate deeper convective clouds appearing over the Caribbean, the west-central Africa in summer and the
25 Indian Ocean in autumn. The increased deep convection is associated with ozone loss due to convective outflow and increased cloudiness and humidity which contribute to photochemical O₃ loss (Morris2010, Wai2014). (Fontaine2011) showed that the location of OLR minima has been shifted northwards which can be associated with a shift on the ITCZ by 0.5 – 0.8° northwards. These changes are subsequently associated with changes in the location of tropical jets, with changes in rainfall amounts and weather systems. All these changes maybe responsible in some degree for the statistically significant tropospheric ozone trends observed close to the location and the branches of the ITCZ (e.g in the Indian and Pacific Oceans), but their contribution remains vague. ^{Authors: On the}
30 other hand, the decreasing trend over the eastern Pacific Ocean (E) is in disagreement with Heue et al., (2016) who reported a significant increase in the order of 0.5 – 1 DU decade⁻¹.

4.3 Seasonal tropospheric O₃ trends

Seasonal tropospheric O₃ trends can be useful for understanding the connection between the factors (e.g. meteorology or emissions) that contribute to tropospheric ozone changes and its distribution. For this reason, the multi-linear regression model

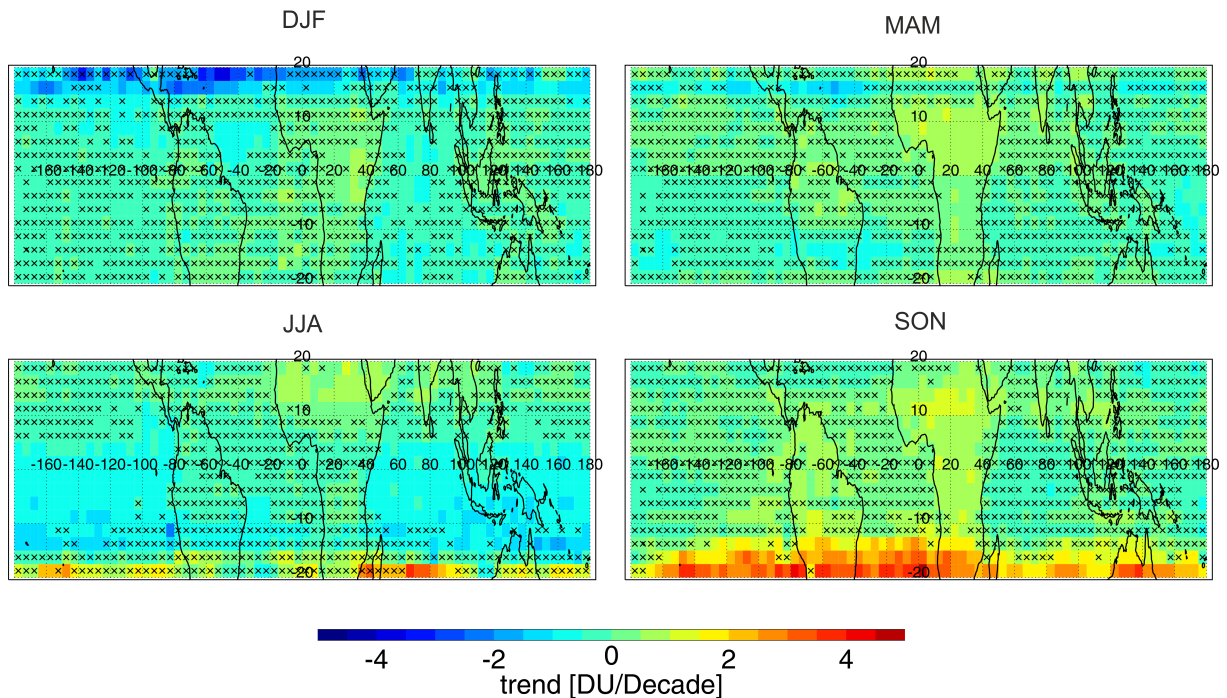


Figure 6. Tropical tropospheric ozone trends for winter (DJF), spring (MAM), summer (JJA), and autumn (SON) for the years 1996 to 2015.

Authors: Black "x" denotes statistically significant trend.

has been applied to *Authors:* monthly time-series containing only the following months, Dec.–Feb., Mar.–May, Jun.–Aug., and Sep.–Nov respectively. *Authors:* TTCO time-series and proxies (ENSO, QBO, solar cycle) in order to calculate TTCO trends for respectively with the only difference that the sine and cosine terms that reflect the seasonal cycle are neglected in the regression. For these time series no seasonal terms are used in the regression. In Fig. 6, the maximum decreasing trends appear during *RI:* winter December to February over the northern tropical Atlantic and Pacific Oceans (~ 4 DU decade⁻¹). These air masses are more affected by changes occurring in the mid-latitudes due to the southward movement of the ITCZ in *RI:* winter these months and the strong westerly air flow over the tropical borders *RI:* in winter (Oltmans et al., 2004). Therefore, it is possible that changes in ozone precursors, such as NO₂ over North America and Europe may have affected the O₃ trends over these tropical latitudes (Logan et al., 2012; Hilboll et al., 2013b). *RI:* The winter This decrease might also be associated with the limited number of TTCO measurements on the northern tropical borders, thus it demands a more careful investigation. The trends are mostly insignificant *RI:* during spring between March and May, with the exception of Africa where ozone is increasing by ~ 1 DU decade⁻¹ and some parts over South America where ozone is decreasing by less than 1 DU decade⁻¹. During *RI:* summer June to August, ozone shows a small statistically significant decrease over the Pacific and Indian Oceans (1-2 DU decade⁻¹). Possible reasons for tropospheric ozone decrease over the oceans may be related to changes in sea surface temperatures (SSTs), which are closely tied to the tropospheric humidity (Trenberth, 2011; IPCC, 2007). As discussed earlier, *Authors:* the production of HOx (OH and OH₂) from water vapor in the troposphere *RI:* consists

of accounts for one of the most important sinks of tropospheric ozone (Jacob, 2000). An increase in vertical convective patterns over the tropical oceans may result in lower ozone mixing ratios in the upper troposphere where the WFDOAS retrieval is more sensitive (Morris et al., 2010; Wai et al., 2014; Fontaine et al., 2011; Ziemke et al., 2008; Coldewey-Egbers et al., 2005). Several studies have shown that the total column of water vapour (TCWV) has increased over the tropics. Mieruch et al. (2014) and Trenberth et al. (2005) found that the TCWV has increased by $\sim 1\text{--}2\%$ decade⁻¹ over the oceans. Chen and Liu (2016) found that also the precipitable water vapor (PWV) increased by $1\text{--}2\%$ in the tropics between 1992–2014. The precipitation increase is about 4% over the ocean, while a decrease of 2% is found over land in the latitude range 25°S to 25°N, between 1979 and 2001 (Adler et al., 2003). The significant positive trend of ozone at the southern tropical Atlantic, southern Africa, South America, and Oceania maximise during ^{R1:} autumn September to November (~ 4 DU decade⁻¹). According to MODIS/TERRA Fire Radiative Power (mW/m²) data (<https://disc.gsfc.nasa.gov/neespi/data-holdings/mod14cm1.shtml>) autumn is the season with the most intense fires over southern Africa and South America. ^{R2:} The burned area in southern tropical Africa increased by 1.8 %/yr during the period 2000 to 2011 (Giglio et al., 2013). Ziemke et al. (2009b) and Wai et al. (2014) estimated that biomass burning can contribute to an increase in tropospheric ozone column by $\sim 20\%$. Hence, it is very likely that biomass burning could be the origin of the observed ozone increase.

15 5 Summary and discussion

The new harmonised dataset of tropical tropospheric ozone columns for the last 20 years between 1996 and 2015, makes it possible to calculate and study long-term tropospheric O₃ variability and trends. Correction offsets have been calculated for GOME and GOME-2 TCO using SCIAMACHY as reference (in the middle of the time-series) in order to reduce the instrumental effects in the long-term time series. Nevertheless, the short overlap period between GOME and SCIAMACHY limits the harmonisation of the GOME dataset. The correction offsets for GOME presented artificial features which are also visible afterwards in the trend (see Fig. S1). In order to identify the best way to merge the CCD data and also to investigate how the harmonisation approach may affect the observed trends, six different harmonisation scenarios have been ^{Authors:} tested/evaluated by comparing with ozone sondes. The ^{Authors:} merging scenario, using no correction for GOME ^{Authors:} (short overlap) and the mean bias correction of GOME-2 with respect to SCIAMACHY ^{Authors:} in each grid box was found to show slightly smaller differences ^{Authors:} comparing to ozone sondes and therefore, was considered to be the preferred scenario. ^{Authors:} From the trend analysis of all merged datasets a rough estimate on the variability of trends due to merging approaches was provided ($\sim \pm 2$ DU decade⁻¹). After the harmonisation, the data obtained from the different instruments agree better with each other and with ozone sondes.

Harmonisation and merging of multi-instrument datasets is one of the largest sources of uncertainty. Most of the trend studies that use multiple satellite data (e.g. Xu et al. (2011), Loyola et al. (2009), Heue et al. (2016) ^{Authors:} , and TOAR) ^{Authors:} underestimate the uncertainty that harmonisation might introduce, and they calculate their results using only one harmonisation approach. do not account for uncertainties related to the merging approach. Therefore, in order to quantify the uncertainty due to harmonisation, multi-linear tropospheric ozone trends using all six harmonised datasets have been derived and the maximum deviation between them has

been calculated. The trends range between about -4 and 4 DU decade⁻¹ and the average difference between the trends from the six scenarios has been found to be ~ 2 DU decade⁻¹, exceeding locally the 2σ of the ^{Authors:}individual trends (0 to 4 DU decade⁻¹). We conclude that ^{RI:} the statistical regression analysis using the $\beta > 2\sigma_\beta$ as criterion to report significant trends in the 95% confidence level is not adequate in order to conclude whether the trends are significant with confidence since the overall uncertainties in the trends are larger than the statistical ones reported.

Despite the fact that ^{Authors:} all the trend results ^{Authors:} from this study using the preferred merged dataset are small ($< \pm 4$ DU decade⁻¹ or 3 % year⁻¹) and mostly uncertain (66 % are statistically insignificant), there are regions such as over southern Africa, the southern tropical Atlantic, south-east tropical Pacific Ocean, and central Oceania where tropospheric O₃ increased significantly by ~ 3 DU decade⁻¹. In central Africa and southern India, tropospheric ozone increased by ~ 2 DU decade⁻¹. Regional positive tropospheric ozone trends of similar magnitude were also observed in other studies (e.g. Lelieveld et al., 2004; Beig and Singh, 2007; Kulkarni et al., 2010; Cooper et al., 2014; Ebojje et al., 2016; Heue et al., 2016). ^{Authors:} They might be linked to anthropogenic activities such as emissions in mega-cities or biomass burning in combination with changes in meteorology or/and long range transport of the precursor emissions Hilboll2013, Wai2014, Giglio2013, Schneider2015, Cooper2014, Duncan2016, Hilboll2017. On the other hand, tropospheric O₃ decreases by ~ 3 DU decade⁻¹ over the Caribbean sea and parts of North Pacific Ocean, and by less than -2 DU decade⁻¹ over some regions of the southern Pacific Ocean. ^{Authors:} Possible reasons for this decrease maybe changes in dynamical processes, changes in STE, convection, humidity or precipitation Morris2010, Wai2014, Fontaine2011, Ebojje2016, Mieruch2014, Trenberth2005, Adler2003, Chen2016, IPCC2007. The most important limitation in interpreting the observed trends over the northern and southern tropical latitudes ^{RI:} (18°–20° in SH and NH) is the low data sampling at these latitudes. Due to the ITCZ movement, cloudy data during local winters are reduced, making the above cloud ozone column (ACCO) retrieval difficult or violating the invariance of the ACCO per latitude band. Therefore, even though they might appear to be statistically significant, they should be referred to with caution.

^{Authors:} Using the preferred merged dataset, The mean tropospheric ozone trend ^{Authors:} has been estimated between 15S and 15N during the period 1996–2015. This restriction has been applied in order to avoid the influence of sub-tropical air masses on tropospheric ozone abundances at the tropical borders (Thompson et al., 2017). The global mean trend is found to be almost equal to zero (^{Authors:} 0.002–0.1 ± 0.3 % year⁻¹) and statistically non significant. This is in agreement with studies of Ziemke et al. (2005) (nearly zero trend) and Ebojje et al. (2016) (^{RI:} 0.2 \pm 0.3 \pm 0.4 % year⁻¹ for the southern tropics and 0.1 \pm 0.5 % year⁻¹ for the northern tropics) who also found no trend or insignificant trends. This is in contrast with the results of Heue et al. (2016) who found ^{Authors:} an averaged a mean increase of 0.7 ± 0.1 DU decade⁻¹ ^{Authors:} for the entire tropics.

Focusing on trends in ten selected mega-cities, a slight tropospheric ozone decrease is observed at the largest cities, such as Jakarta and Mexico (-0.3 DU decade⁻¹), whereas statistically significant increases (-1 DU decade⁻¹) are noticed over Manila, Bangkok, and Kinshasa. It has been shown that tropospheric ozone increase is not linearly related with the size and the population of the selected mega-cities. This is not surprising since tropospheric ozone production from its precursors, NO_x and VOCs, is not linear and occurs downwind of the pollution source. For example, large increase of NO_x or VOCs result in destruction of ozone close to and within mega-cities. It is also broadly recognised that the mechanisms that modulate tropospheric ozone variability are not straightforward according to precursor emission. In addition, meteorological conditions and atmospheric dynamics also play an important role Ziemke2003, Solomon2007, Chandra2009, Voulgarakis2010, WMO2011, Neu2014, Monks2015. Comparing our trend results with those of Heue2016 and Ebojje2016

trend results in these ten mega-cities, we found that they agree slightly better (within the combined uncertainties) with the ones from Heue2016. The most possible reasons for the mismatch with Ebojie2016 trends is the fact that their retrieval reaches up to the tropopause including more upper-tropospheric ozone information, and additionally the fact that they investigated a shorter time period, between 2003 and 2012.

Authors: around the world due to its outstanding importance as a greenhouse gas and hazardous pollutant *Authors:* Comparison of several independent studies conducted on tropospheric ozone trends shows that the trends vary in sign and magnitude for the past few decades in the tropics (Cooper et al., 2014; Ziemke et al., 2005; Monks et al., 2015; Oltmans et al., 2013; Lelieveld et al., 2004; Lin et al., 2014; Beig and Singh, 2007; Kulkarni et al., 2010; Thompson et al., 2014; Heue et al., 2016; Ebojie et al., 2016). *Authors:* This is a significant issue for the scientific community, especially climate modelers who try to use recent past data to evaluate the performance climate and global atmospheric chemistry models for future prediction (Zhang et al., 2016). *Authors:* At the moment, there is a new activity of the International Global Atmospheric Chemistry Project (IGAC), named Tropospheric Ozone Assessment Report (TOAR), which aims to assess our knowledge of the tropospheric ozone distribution, pattern and trends, using the available surface ozone data, ozone sonde, aircraft and satellite observations (currently under review in Elementa: <https://collections.elementascience.org/toar/>).

RI: The accurate interpretation of the trend results is challenging and requires the parallel investigation of changes in numerous factors that impact on ozone production, loss, and transport in the troposphere, including various feedbacks e.g. Cooper et al. (2014) and references therein. *Authors:* For example, tropospheric ozone changes can be linked to changes in anthropogenic NO_x and other tropospheric O₃ precursors from human activities, alterations to the humidity burden of the troposphere and NO_x produced by lightning Morris2010, Hilboll2013, Schneider2015, Cooper2014, Duncan2016, Hilboll2017, Wai2014. Changes in the burden of organic and inorganic halogens on marine areas as well as changes in dissolved organic matter (DOM) photochemistry in surface waters could be an additional source of volatile organic compounds and halogen chemistry that can contribute to ozone destruction Dickerson1999. Meteorology, convection, and dynamical oscillations, such as the MJO, stratospheric intrusions (STE) or the tropical upwelling modulated by the Brewer-Dobson circulation, and in shorter timescale, atmospheric dynamics or cyclones may have influence on the transport of pollutants and contribute locally to the observed tropospheric ozone changes Ziemke2009b, Beig2007, Parrish2009, Chandra2004, Oltmans2004, Sauvage2007. Additionally, changes in stratospheric ozone column may influence the tropospheric ozone production via photolysis Fontaine2011, WMO2014. Finally, the attribution of observed TCO trends in specific regions to the various processes is not possible without the additional use of chemistry-transport models that can potentially disentangle the different contributions to tropospheric ozone variability (dynamics and chemistry)(Grewe et al., 2012; Coates et al., 2015). *Authors:* For example, monthly tagged CTM runs could give insight into tropospheric ozone sources for specific locations (Coates et al., 2015). With this method, the fate of emitted species is followed and their chemical reaction pathways are tracked. Using labeled CTM mechanisms for NO_x and VOCs emissions, and their degradation products, the ozone burden can be attributed to the relevant emission source

Authors: In the future, t The launch of Sentinel 5 precursor(S5p) satellite *Authors:* (fall 2017)in 2017 and the planned launches of three consecutive Sentinel 5 instruments until 2030 will extend the TCO record *Authors:* at least for 7 more years (expected lifetime). It is also expected that the extension of the time series willwhich will likely result in more reliable trends. The grid box size used in this study was relatively coarse (2.5°×5° degrees), due to the instruments spatial resolution (GOME pixel ≈320 km), and in order to remove the residual noise. The high spatial resolution (7×7 km) of the TROPOMI instrument aboard S5p will improve the trend estimates of tropospheric ozone in particular over mega-cities.

6 Data availability

Data used in this publication can be accessed via the IUP website: <http://www.iup.uni-bremen.de/UVSAT/datasets> or by contacting the corresponding author.

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Answers on Anonymous Referee #1
Received and published: 7 November 2017

General Comments:

This study attempts to harmonize a 20-year satellite record including GOME/ERS-2, SCIAMACHY/Envisat, and GOME-2/MetOp-A using SCIAMACHY as a transfer standard. The authors use 6 different schemes in their attempt to harmonize the data and evaluate the relative success of the various approaches via comparisons with in situ measurements (i.e., sondes) when and where possible. The authors suggest that by using 6 different approaches, they are better able to estimate the uncertainty in apparent trends owed to the harmonization itself. Like prior studies, the authors find few areas of significant tropospheric ozone trends. Their analysis of trends over tropical mega cities seems to produce results not too different from prior studies. Frankly, I am really not sure what to take away from this study, and I have several important issues with what is (and is not) presented.

The authors should be aware of a major reprocessing effort of the Southern Hemisphere Additional Ozonesonde (SHADOZ) network data being led by Thompson and Witte. While I realize the papers on their work are just making their way into the literature, their efforts have been ongoing for several years. Since the authors of this study leverage the SHADOZ data, I am surprised that the paper communicates no awareness of the reprocessing effort nor of its potential impact on the results of this study.

At the very least, the authors could have contacted SHADOZ PI Thompson to make sure she was aware of this study and had the opportunity to communicate important updates relevant to the authors. While entirely up to the authors, having Thompson as a co-author would have strengthened the credibility of the sonde results presented in this paper.

Finally, while the paper appears to present different approach to tropical tropospheric ozone trend analyses that recent studies, I do not find the results particularly compelling or worthy of publication in this form at this time.

Recommendation:

I recommend this manuscript be declined for publication in ACP at this time pending major revisions.

Our answer:

Many thanks for the very helpful comments. As proposed by the reviewer, we have contacted Drs. Anne Thompson and Bryan Johnson who are responsible for reprocessing the ozonesondes data from SHADOZ network. Both accepted our invitation to become co-authors of the paper and supported us in revising the paper. We now used reprocessed SHADOZ data (available for American Samoa and Paramaribo). The recent reprocessing mainly focused on improving old sonde data at the stratospheric ozone peak and above. It had very minor impact on our findings. The stations of Hilo and Fiji have been removed from the comparisons. Fiji is affected by air masses originating from the

mid-latitudes and the upper troposphere and Hilo is strongly affected by volcanic outgassing, resulting in negligible ozone concentrations in the boundary layer.

Additionally, Klaus Peter Heue is included as co-author in the paper for granting us access to his tropospheric ozone data which we used for some comparisons.

Since Reviewer #2 suggested to shorten the discussion on the various merging approaches we moved some material into a supplement and discuss mainly the lessons learned from looking at trends from differently merged datasets in Section 2. The estimation of the mean tropical trends is moved to Subsection 4.1 and is now limited between 15°S and 15°N since the tropical borders are strongly influenced by air masses being transported from the mid-latitudes and stratospheric intrusions (Thompson et al., 2017).

Section 3.3.3. about trends in megacities has been removed from the revised version of our paper as also suggested by reviewer #2.

At various places we have expanded on the comparisons to the Heue et al. results. Although similar instruments have been used, the results from this study and Heue et al. are different and are discussed in more detail (see detailed comments).

Our main findings can be summarized as follows:

Tropical tropospheric ozone trends critically depend on the merging/harmonisation approach. This was investigated by investigating six different merging scenarios. The trend of tropical tropospheric ozone is estimated using a multiple linear regression model and for all six scenarios the sensitivity of the derived trends to the harmonisation approach is investigated. Such an approach has not been reported before and may explain why tropospheric ozone trends from different studies do not agree (see e.g. TOAR report). The main conclusion is that the (statistical) trend uncertainties from one scenario may be smaller than the variation of trends from the different merging approaches, which means that the trend uncertainties are in reality larger. At the end we selected the preferred merging scenario by comparing these six merged datasets with ozonesonde data (some of them reprocessed now) from the SHADOZ network.

Detailed Comments:

Page 1

--Line 3: What does “good agreement” mean? Quantify.

We consider the bias between the CCD retrievals and the integrated O₃ profiles from ozone sondes good since they are less than 6 DU which is about the 1sigma uncertainty of the mean station bias (RMS in Table 2 of Leventidou et al., 2016).

We changed in the main text (page 4, line 30) as follows: “The biases between them have been found to be within 6 DU which is mostly within the uncertainties of the mean biases of 6 DU (1 sigma). One large source of uncertainties in these comparisons are the

low sampling of the sondes (less than five launches in a month typically) and the fact that CCD ozone is only derived as monthly means covering rather large areas (grid boxes). In the abstract we only mention the average bias between CCD and sondes”.

Line 14: “Additionally, over central ...” Awkward sentence.

The sentence has been changed to “... and by ~2DU/decade over central ...”

Line 19: “... reasons for these decreases are...”

The sentence has been removed.

Page 2

--Line 2: delete “both”

Deleted

Page 3

--Line 10: “3.8% decade⁻¹ (0.16 ppbv year⁻¹)” What is the difference between these numbers? Unclear. You mention “surface and ozonesonde observations,” so which is it?

The trend in ppbv year⁻¹ represents the change of surface ozone in volume mixing ratio per year. The sentence (page 3, line 5) has been changed to: “Oltmans et al. (2013) observed an increase of 3.8 % decade⁻¹ (0.16 ppbv year⁻¹) in surface ozone in Mauna Loa, Hawaii (19.5°N) in the North Pacific since 1974 and a smaller insignificant trend in the order of 0.7 % decade⁻¹ (0.01 ppbv year⁻¹) in American Samoa (14.5°S) after 1976. ”

Page 4

--Line 4: The reference to Leventidou et al., 2016 may be a recent one for the CCD method, but I think you should be referencing the original paper for this approach, which I believe goes back to Ziemke ..

As the referee mentions, the CCD method was developed by Ziemke et al., 1998 and further improved by Valks et al., 2003. The citation of Ziemke et al. 1998 has been added in the introduction, along with the most significant contributors on tropospheric ozone retrievals from remote sensing in the past.

--Line 10ff: This would be a good place to remind the reader of the specific application of the CCD approach you’re using. To what altitude is tropospheric column ozone being computed? Does it vary scene-to-scene? On what fraction of pixels can it be applied?

The CCD method is described in detail in Leventidou et al., 2016. All tropospheric O3 columns are calculated up to 200 hPa. The main reason is that most clouds do not reach the tropopause.

--Line 14: “overpass time” – is this not a critical element influencing tropospheric ozone, especially in regions near megacities?

For tropospheric trace gases that show diurnal variations, overpass time is important. All satellite data used here are in the morning hours differing at most one hour (9:30 to 10:30). This is believed to have little impact on the results.

The following text has been added (page 9, line8): “As seen in Table 1, the mean bias between the six harmonised TCO datasets and the ozone sondes range between -1.1 and 0.9 DU which is well within the retrieval uncertainty showing that for most scenarios the spatio-temporal offsets with respect to ozonesondes are minimised.”

--Line 25: “ ... whole timespan of the operation of the European satellites...”
changed

--Line 26: “...since it is the only...”
changed

Page 5

--Line 2: “Possible reasons for the biases are...” The paper is filled with these statements. It would be good to know if this is the problem. Could you test your hypothesis by applying the same cloud algorithm to both retrievals? I realize that requires working with the instrument teams, but even a limited test application could prove useful. The differences that appear between the two panels of Figure 1 are striking. To me, this subject is more interesting than the one that is the current main focus of the paper.

It would be desirable to have the same cloud algorithm for all instruments. However, any bias from the cloud algorithm has been removed by the harmonisation process. We believe that the different spatial resolutions of the instruments is more important (GOME :320 x 40 km², SCIAMACHY: 60 x 30 km², and GOME-2: 80 x 40 km²).

--Figure 1. It occurs to me in looking at the upper panels that perhaps it would be worthwhile to separate the lower plots into “over land” and “over water” components. Visually, It would also be helpful to the reader if the plots were rotated 90 deg. so that the latitudes ran up and down the page as they do in the top panel.

In order to keep it simple we leave the figure as is. The land-sea contrast can be clearly seen in the 2D plots. The error bars in Fig. 1 (line graphs) mainly reflect the longitudinal variation possibly due to land-sea contrast. The line graphs have been changed so that the y-axis is the latitude.

--Line 8 – 9: “This behavior may be explained by the short time of common operation...” Another undemonstrated hypothesis. How could you test this assumption? And if it’s true, is not your transfer standard idea (i.e., reference to SCIAMACHY) compromised?

The larger variation in the bias with latitude in GOME data is most likely due to the short overlap period (10 months, from August 2002 to June 2003 (when GOME lost its global coverage)). For GOME-2 the overlap with SCIAMACHY was more than 5 years, making the latitude dependence smoother.

Page 6

--Line 3: "...ozonesonde data, it seems reasonable..."
changed

--Figure 2. What is important about this plot is how little of the area is actually statistically significant. Perhaps you should reverse and only mark with "x" those cells that ARE statistically significant.

Also, remind the reader, what fraction of the cells are statistically significant? This result is key in your argument that you can use a constant offset to "correct" the GOME 2 data, but you do not seem to make much of a point of that in the text.

Figure 2 has been changed, showing with x the statistically significant grid boxes. It is clear from the figure that the vast majority of the grid points are statistically insignificant and there is no need to specify.

Line 14, page 9 on the following modified sentence: "Scenario 6 can also be rejected due to the fact that the drift in GOME-2 correction offset at 81% of the grid-boxes is statistically non significant. "

We have added a line plot to Fig. 2 to show that the drift is not significant. The reviewer is correct that this is the main reason that a drift correction is not needed as already mentioned in the text.

--Line 12: "...but only a subset of them." Which subset? Which years?

.....of the ITCZ (no cloudy data available in the western Pacific)

The sentence has been removed since the explanation is given the sentence before.

--Sect. 2.3: This seems to be an important part of the paper, but frankly, I do not find it well motivated. Why 6 scenarios? Have you exhausted all possibilities. Does the reader need to know the details or simply your recommendation for the best approach to harmonize the data – with a discussion of the other approaches you tried and how they inform your estimate of the component of the calculated trend uncertainties that arise from the harmonization process itself?

We think that this is one of the most important results of this paper. We show here that the hamonisation procedure (merging) is one of the largest error sources of the trends. In the six scenarios we checked different reasonable assumptions on how to handle the differences between the individual instruments. The trends derived from the various merged dataset show larger differences than the statistical uncertainty from the trend regression applied to one of them. This is usually neglected in other studies.

To make this section a bit shorter as also suggested by Reviewer #2, we moved

this figure showing the maximum trend difference among all six merging scenarios to the supplementary material. Fig. S2 shows that the mean differences in trends from all pairs of merged datasets is about 2 DU/decade, exceeding in most cases the uncertainty from the single data regression.

Page 7

--Table 1. Not sure what to do with this table. Why are the stations in the order in which they appear? What is the table communicating? Here's where my earlier comment about SHADOZ reprocessing becomes relevant: are you using the reprocessed sonde data? How would these results change if you did? You are integrating the sondes to 200 hPa. How does that compare to the altitude used for the CCD approach?

Table 1 showed comparisons between integrated ozone columns up to 200hPa from 9 tropical ozonesonde stations from the SHADOZ network (version V05) with 6 different possible merging scenarios of Tropical tropospheric ozone columns from GOME, SCIAMACHY, and GOME-2. The tropospheric ozone columns retrieved with our CCD algorithm are adjusted to 200 hPa using climatological values (Leventidou et al., 2016).

The change in the differences of tropospheric ozone columns (up to 200 hPa) between collocated CCD_results and SHADOZ for the stations of Paramaribo, Am. Samoa, Hilo, and Fiji due to changes from SHADOZ V05 to V05.1R .

<i>Station</i>	<i>Scenario 1</i>	<i>Scenario 2</i>	<i>Scenario 3</i>	<i>Scenario 4</i>	<i>Scenario 5</i>	<i>Scenario 6</i>
<i>Am. Samoa</i>	<i>0.6</i>	<i>1.0</i>	<i>0.6</i>	<i>0.0</i>	<i>0.7</i>	<i>4.3</i>
<i>Paramaribo</i>	<i>1.8</i>	<i>0.7</i>	<i>1.7</i>	<i>2.7</i>	<i>1.8</i>	<i>-1.1</i>

Following the comments of the reviewer the comparison has been updated including the newest version of SHADOZ data (for two stations). The stations of Fiji and Hilo have been removed from the paper as discussed earlier. The following text has been added: "Fiji (18.1S, 178.4E)) station is not included in the comparison because it is highly influenced by air coming in from mid-latitudes and the upper troposphere (Thompson et al., 2017). Hilo (19.4N, 155.4W) is influenced by volcanic out-gassing with high SO2 emissions, resulting in negligible ozone concentrations at the boundary layer. Therefore, this station is also not included."

The order of the stations has changed to alphabetical, and the title and the table has changed as follows:

"Mean differences (in DU) between merged TCO data, retrieved with the CCD method using six possible harmonisation scenarios, with integrated ozone columns up to 200 hPa from nine SHADOZ stations. The stations marked with asterisk present data from the newest reprocessed (V05.1_R) version (Thompson et al., 2007; Witte et al., 2017). The regions where the merged scenarios have the smallest biases with the ozonesondes are marked with bold. Scenario 1 has the smallest mean bias for all the stations."

<i>Station</i>	<i>Scenario 1</i>	<i>Scenario 2</i>	<i>Scenario 3</i>	<i>Scenario 4</i>	<i>Scenario 5</i>	<i>Scenario 6</i>
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<i>Am. Samoa*</i>	<i>-0.89</i>	<i>-0.92</i>	<i>-1.99</i>	<i>-0.61</i>	<i>-0.93</i>	<i>4.59</i>
<i>Ascension</i>	<i>0.03</i>	<i>-0.14</i>	<i>-0.77</i>	<i>-0.42</i>	<i>-0.6</i>	<i>0.03</i>
<i>Java</i>	<i>-0.11</i>	<i>-0.12</i>	<i>-1.12</i>	<i>-0.54</i>	<i>-0.55</i>	<i>-0.11</i>
<i>Kuala Lumpur</i>	<i>-1.81</i>	<i>-2.12</i>	<i>-2.12</i>	<i>-2.14</i>	<i>-2.48</i>	<i>-1.78</i>
<i>Nairobi</i>	<i>1.81</i>	<i>1.10</i>	<i>1.80</i>	<i>1.48</i>	<i>0.74</i>	<i>1.84</i>
<i>Natal</i>	<i>0.56</i>	<i>0.63</i>	<i>-0.21</i>	<i>0.22</i>	<i>0.28</i>	<i>0.57</i>
<i>Paramaribo*</i>	<i>-2.98</i>	<i>-2.95</i>	<i>-3.02</i>	<i>-4.11</i>	<i>-4.34</i>	<i>-0.11</i>
<i>Mean bias for all stations</i>	<i>-0.48</i>	<i>-0.64</i>	<i>-1.06</i>	<i>0.87</i>	<i>-1.13</i>	<i>0.72</i>

The results with the updated ozonesonde data do not differ significantly from our earlier results. Nevertheless, we present our results now using the updated V05_R1 ozonesonde data for the available stations.

Page 8

--Line 7: "The same occurs for the..." delete the comma.
changed

--Line 9: "...the scenarios that can be confidently rejected according to this comparison are..." I'm not sure I have confidence that any scenario can be rejected until I know more about the sonde data you used and the altitudes used for the satellite tropospheric column amount.

The updated ozonesonde data to not change the conclusions. The tropospheric ozone columns from the sonde data were calculated exactly as the satellite data up to 200 hPa.

The text has been changed as follows(page 9, line 14):

" ... Although the comparison between the TCO from the individual harmonised scenarios and the ozonesonde data does not favor clearly any harmonisation scenario, the scenarios that can be confidently rejected are scenarios 3, 4 and 5 where GOME data are corrected with respect to SCIAMACHY since the overlap period between GOME and SCIAMACHY is very short (10 months, 8/2002-6/2003). Scenario 6 can also be rejected due to the fact that the drift in GOME-2 correction offset at 81% of the grid-boxes is statistically non significant. Lack of significant drifts in the comparison between GOME-2 and SCIAMACHY over the overlapping period shows that the data records are quite stable. Finally, scenario 1 (no drift corrections and bias correction for GOME-2) has the smallest mean bias with the ozone sondes (-0.4 DU). For these reasons, scenario 1 has been selected to be the preferred harmonisation scenario for merging the TCO datasets."

Page 9

--Line 1: "where α is the offset ..."
Changed

--Line 12: "...to persist into the next month."

changed

--Line 16: define "AR(1)"

The sentence has been changed as: "Therefore, the first order autocorrelation of the noise (AR[1]) is included in the model, as explained by Weatherhead et al. (1998)."

--Lines 25ff: "Nevertheless, all scenarios shown in Fig. 3 agree that there is a positive trend..." My quick read of Figure 3 is that very little of the map shows statistically significant trends. As to the fact that one appears to exist "over the southern tropical Atlantic Ocean" and a couple of other sites cited by the author, I am not sure what to make of it. The authors provide no explanation for such trends or why they might exist. I would find this more compelling if the authors could simplify the presentation, show the best correction scheme, show the best estimation of uncertainty (including that resulting from the harmonization scheme), and then spent some time in the text discussion what the resulting trend data showed and why. In its current form, I find the presentation more confusing than compelling.

Figure 3 e) shows the regions where the statistically significant trends calculated using the preferred merging scenario exceed the maximum difference of the trends among all six merging scenarios (now Fig S2) and can be reported with the highest confidence.

Page 11

--Line 8: "... following very well a Gaussian distribution." You don't show that in the paper. But I'm not sure what to make of it, either. Are you saying that there is no signal anywhere on the map? What do you mean by "the noise is random?" What is the noise?

This sentence has been removed as it is out of context here.

--Line 10ff: "This result is in agreement with Ziemke et al. (2005) and Ebojie et al. (2016)..." What periods did they examine? What data did they use? Are there any implications from the fact that it does not appear to have changed from their analyses to your analysis? What new have we learned from your analysis?

The refereed sentence (now page 11, line 30) has been modified as follows:

"The mean tropospheric ozone trend is in agreement with Ziemke et al. (2005) (using solar backscatter ultraviolet (SBUV) and Total Ozone Mapping Spectrometer (TOMS) version data from 1979 to 2003) and Ebojie et al. (2016) (using SCIAMACHY limb-nadir-matching (LNM) observations during the period 2003–2011) who also indicated insignificant and near zero global trends in the tropics, although their analysis was based on different datasets and covered shorter time periods. "

Page 12

--Line 10: "Figure 5 summarizes the tropical tropospheric ozone trends ..."

changed

--Line 25: "...may still be an artifact of the data-set." Is there a way to know?

The sentence (now page 11, line 18) has changed as follows: " The negative trends appearing in a region at the northern latitudes (Caribbean sea and northern Pacific) may be an artifact of the data-set (low sampling of data, 54 out of 240 months of data)."

--Line 31ff: Just to be clear, you selected your regions based on where you found statistical significance? That led to larger regions that then had statistically significant trends?

Yes, we selected the regions in order to have large number of grid points with significant trends for highlighting.

--Your Table 2 shows some impressive trend results. You then follow that with a list of possible factors that led to the trends (anthropogenic NO_x, population, energy consumption, biomass burning, changes in meteorology, dynamical oscillations, stratospheric intrusions) and you cite some prior works that have made these suggestions, but you provide no evidence within this paper for the proximal cause (or causes) in each of the regions you list in Table 2, nor is there really any justification for the selection of the boundaries of those regions other than they produce significant trends. If, as the title of this section suggests, mega-cities are responsible, it seems the regions might have been more narrowly defined. It would have been nicer to select regions based on a hypothesis and then identify the existence of significant trends (or not) to accept or reject that hypothesis.

The conversation about the possible reasons for the noticed trends has been moved in the conclusions. This section now summarises the areas where we observe statistically significant TTCO trends and compares these results with other studies.

Page 16

--Line 10: "Despite the fact that might appear to be..." What might appear?

The sentence has changed as follows (page 14, line14): "However, the observed trends over the northern and southern tropical latitudes (18° –20° in SH and NH) should be generally interpreted with caution because they are influenced by low sampling of data due to the movement of the ITCZ, which reduces the cloudy data during local winters and makes the above cloud ozone column (ACCO) retrieval difficult, violating in some cases the invariance of the ACCO per latitude band."

--Lines 22ff: "...cloudiness and humidity which contribute to photochemical O₃ loss..." I think the missing factor identified in the Morris et al., 2010 paper was significant lightning production, which they hypothesized led to NO_x production and O₃ loss in the absence of sunlight. The presentation here is a bit oversimplified. Deep convection alone can loft relatively low O₃ concentrations from near the surface (especially over the sea) to the upper troposphere. Those decreases are not "loss" but reductions resulting from transport.

We removed most of the discussions on possible causes as we can only speculate on it. In the Summary (page 18, line 34) we briefly mention that we cannot attribute the observed changes in tropospheric ozone as numerous factors may contribute the trends (production, loss, transport). Only with the help of modelling data one can disentangle the various factors.

Page 17

--Line 2: “winter” – what does “winter” mean in the tropics? Perhaps it’s better to identify seasons by months rather than such ambiguous names.

Changed

--Line 3: “as NO₂ over North America and Europe may have affected the O₃ trends...” moved the comma.

Changed

--Line 8ff: “Possible reasons...” There’s a whole list of possibilities here with no conclusions or evidence to support any one (or combination) of them.

The speculations about possible reasons that are responsible for the observed trends has been removed from the text. Instead a paragraph has been added in the summary where the complexity of trends' interpretation is discussed.

--Line 10: “...water vapor in the troposphere accounts for one of the most important...”

Changed

--Lines 10 – 12: “An increase in vertical convective patterns over the tropical oceans may result in lower ozone mixing ratios in the upper troposphere...” True if lofting low ozone from the surface. If lightning is present in the convection, however, you might see enhancements.

Thus, the influence is unclear.

See our earlier reply above (has been removed)

Page 18

--Lines 23ff: I would replace all of this text with a table. No need to write it all out.

Section 3.3.3. and the discussion about trends in mega cities has been removed from the paper as also suggested by Reviewer #2.

Page 19

--Table 3. Like previous tables, what is the logic of the order of cities in this table? How do the periods of study for Heue, Ebojie, Schneider, Hillboll, and this work compare? What impacts do differences in study periods have on interpretation of the results? The data in this table appear to have been compiled using 2.5 X 5 deg boxes. That’s roughly an area 250 km X 500 km in the tropics. Can you actually see signatures from megacities spread out over such a large area? For a control, should you also compute trends around cities that have not

grown (perhaps ones that have shrunk) or that have reduced emissions just to see if they behave any differently than the ones you list here?

See previous reply

--Line 3: "The derived tropospheric trends clearly show that tropospheric ozone increase is not proportional to ..." If you're going to make this claim, I think you need to show the population data, perhaps in Table 3, and the proxy you're using for industrial activity as well.

See previous reply

--Lines 8 – 9: "The degree of tropospheric ozone change strongly depends on the NO₂ amount..." As the second half of this sentence correctly relates, it depends on the relative NO₂ and VOC concentrations. I think I would get rid of the word "strongly" in this sentence.

See previous reply

Page 20

--Line 21: "... since the uncertainties in the trends are larger..."

See previous reply

--Line 24: Cite the uncertainties associated with the trends published in Ebojje et al.

See previous reply

--Line 31 – 32: "They might be linked to..." You list a whole bunch of possibilities. Has anyone shown the specific relevant link for your study? If not, how can you test these hypothetical influences?

The speculations about possible reasons that are responsible for the observed trends has been removed from the text. Instead a paragraph has been added in the summary where the complexity of the interpretation of the trends is discussed. It is out of the scope of this paper to attribute the trends to specific processes.

Page 21

--Line 4: "...tropical latitudes (> 18°N and S)..." What range?

The sentence has changed as follows (page 18, line13): "The most important limitation in interpreting the observed trends over the northern and southern tropical latitudes (18°–20° in SH and NH) is the low data sampling at these latitudes. "

--Line 10: "It has been shown that tropospheric ozone increase is not linearly related..." I'm not sure this study has shown that result conclusively or persuasively.

This text has been removed (mega cities)

--Line 18: "...the fact that their retrieval reaches up to the tropopause ..." That seems like an important factor. How different are the retrievals? What impact do the differences have on your results/homogenization scheme?

It is true that we cannot directly compare the trend results but, as mentioned earlier, we use them as indications of the range of the estimated trends.

--Line 28: “...(expected lifetime of the Sentinel 5 precursor satellite).” No need to introduce an abbreviation in the last paragraph.

changed

Anonymous Referee #2

Received and published: 14 December 2017

Review of the paper by Leventidou et al.

The authors present in the paper a detailed trend analysis of tropospheric ozone over the tropics, using a long term homogenized data set based on satellite measurements using the Convective Clouds Differential method. This method and its application on individual satellite sensors has already been presented in various studies including a publication from the same group in AMT (Leventidou et al. 2016). In this paper they homogenize the data from three sensors and examine the variability and the trends over regions and mega cities within the tropics. The paper is well written and structured but there are many significant issues that should be considered before being accepted for publication in ACP.

The content of the paper has many similarities with the paper by Heue et al. 2016 in AMT. Although it is clear that the Heue et al., paper uses a different version of total ozone data it is not clear from the current paper what are the differences between these two data sets concerning the application of the CCD method and the resulting tropospheric ozone estimates. The authors should elaborate more here.

Our reply:

Many thanks for the comments.

The discussion on the various merging approaches is shortened and a part of it is moved into a supplement. Now we discuss mainly the lessons learned from looking at trends from differently merged datasets in Subsection 2.4. The estimation of the mean tropical trends is now limited between 15°S and 15°N since the tropospheric ozone retrievals are questionable (also for the case of Heue et al. (2016)) due to the fact that the tropical borders are strongly influenced by air masses being transported from the mid-latitudes and stratospheric intrusions (Thompson et al., 2017).

At various places we have expanded on the comparisons to the Heue et al. (2016) results. Although similar instruments have been used, the results from this study and Heue et al. (2016) are different and are discussed in more detail. In page 4, line 6 we added: “The main differences between our CCD algorithm and the one developed by Heue et al. (2016) originate from the corrections that we have applied in the above cloud column calculation of GOME and GOME-2 data and handling of the outlier data (Leventidou et al., 2016).”

Our study shows that despite the fact that the same instruments are used, the trends differ. These differences can be attributed to the different harmonization/merging approaches applied in addition to the different ozone and cloud retrievals used. This paper clearly shows that the merging approach is rather a large source of uncertainty in determining tropospheric ozone trends. This is in our opinion is demonstrated for the first time in this paper.

Detailed comments:

Section 2.2

The authors attribute most of the differences between the TCO mostly to the different cloud algorithms involved. Why they exclude eventual biases between the sensors also in the initial total columns?

For trend calculations a constant bias (in clouds and ozone) is not really an issue and can be removed using a suitable merging approach as shown here. In the periods of overlaps both total ozone and tropospheric columns agree well after applying a bias correction. In particular the lack of significant drifts in the comparison between GOME-2A and SCIAMACHY over an extended period show that the data records are quite stable. A time-varying bias (drift), however, may add significantly to trend uncertainties if not properly accounted for.

In the text we mention: “Possible reasons for the biases are the different cloud algorithms used for each instrument (SACURA for SCIAMACHY and FRESCO for GOME and GOME-2) and the small biases noticed in the total ozone columns (e.g. ~ -2.5 DU between SCIAMACHY and GOME-2). Differences in spatial resolution and overpass time of the instruments have also minor contributions in the biases.”

Is there any explanation for the different behavior of GOME-SCIA differences over 10°N shown in Figure 1?

The larger variation in the bias with latitude in GOME data is most likely due to the short overlap period (10 months, from August 2002 to June 2003, when GOME lost its global coverage). For GOME-2 the overlap with SCIAMACHY was more than 5 years, making the latitude dependence smoother. It should be noted that the shift in the bias at 10°N is within the uncertainty of the observed biases at these latitudes.

In the manuscript (page 5, line 22) it is now mentioned that: “GOME mean biases have stronger latitudinal variability than those of GOME-2. This behavior may be explained by the short time of common operation (Jan. 2002–Jun. 2003) between GOME and SCIAMACHY instruments.”

The authors should also provide an explanation for the GOME-2/SCIA drift. Does this originate from a potential drift in the total columns?

There seems to be a positive drift in the GOME-2-SCIAMACHY difference (Fig. 1) which is quite small and statistically not significant. One possible explanation are changes in the instrument response function with time (e.g. De Smedt et al, 2012).

Reference: De Smedt, I., Van Roozendaal, M., Stavrou, T., Müller, J.-F., Lerot, C., Theys, N., Valks, P., Hao, N., and van der A, R.: Improved retrieval of global tropospheric formaldehyde columns from GOME-2/MetOp-A addressing noise reduction

and instrumental degradation issues, Atmos. Meas. Tech., 5, 2933–2949, doi:10.5194/amt-5-2933-2012, 2012.

Section 2.3.

The discussion of six scenarios in the paper is confusing, since they don't differ substantially concerning the outcome. I think the authors should just describe here the chosen approach of harmonization.

We think that this section is one of the most important results of this paper. In the six scenarios we checked different reasonable assumptions on how to handle the differences between the individual instruments. We show here that the harmonisation procedure (merging) is one of the largest error sources of the trends since the trends derived from the various merged dataset show larger differences than the statistical uncertainty of the trend derived from any of the single dataset (one of the six). This is usually neglected in other studies. We, however, shortened that section a bit and moved some of the figures to the supplementary material. The added uncertainty from the merging approach is also discussed in more detail in the summary section.

Section 3.3.1.

The authors present regional trends in this sections. The choice of the regions to my understanding is based only on the significance of the trends and in a sense this looks like a random choice. Do these regions have some special characteristics that have to do either with prevailing dynamic features or emission sources? In general the discussion here should be improved.

Indeed we studied regional trends focusing on the regions where the trends are statistically significant across many grid points. As we are not trying to speculate too much on the possible causes (would require substantial modelling efforts) we leave it as is. In the introduction and summary we discuss some of the possible causes of trends and make it clear that long-range transport of tropospheric ozone can contribute to trends in rather remote areas.

Section 3.3.2.

The authors attribute the positive trends to South Africa and South America to biomass burning. Is there any indication from another source that there is increased biomass burning over the years that can cause such a trend?

The following text has been added (page 17, line 15): “The burned area in southern tropical Africa increased by 1.8 %/yr during the period 2000 to 2011 (Giglio et al., 2013). Ziemke et al. (2009b) and Wai et al. (2014) estimated that biomass burning can contribute to an increase in tropospheric ozone column by ~20%. Hence, it is very likely that biomass burning could be the origin of the observed ozone increase.”

Section 3.3.3.

The authors show trends over mega-cities in the tropics. The authors should provide a comment why they think a grid-point of 2x5degrees can represent the variability of tropospheric ozone caused by a mega city.

This Section (3.3.3) has been removed from the manuscript.

The discussion against NO₂ trends as shown in the paper is also not conclusive. Are there studies (modelling or in-situ ones) to support their findings?

The discussion about NO₂ trends has been removed (megacities).

The authors also compare their results with Heue et al and although the approach is pretty similar there are differences. They should elaborate more here to explain this.

The comparison with Heue et al. (2016) trends for 10 mega cities has been removed from the manuscript.