Replies to Referee #1 on the manuscript 'Retrieval of ozone profiles from OMPS limb scattering observations' by C. Arosio et al.

We thank the reviewer for the time she/he spent reading the manuscript and constructively commenting on the paper. In the text below, we address the comments from the Referee #1. Referee's comments are shown in italicized font and authors' responses are highlighted in blue.

1 Short resume The construction of long-term ozone profile data records for trend studies has been a lively line of research these past few years. The objective is simple, yet has proven hard to realise: obtain a climate data record which is sufficiently stable (better than a few % per decade) over multiple decades and which ideally covers (most of) the globe at high spatial resolution. Arosio et al. explore two established, complementary merging methods to combine measurements by two dense limb samplers SCIAMACHY (2003-2012) and OMPS-LP (2012-now) using a third limb sensor as transfer standard, Aura MLS (2005-now). Contrary to most earlier efforts by other groups, the authors attempt to preserve longitudinal information. The resulting data record is then analysed for trends over the 2003-2018 period using a widely used regression model. The authors discuss the spatial structure of the trends and they claim that longitudinal patterns are discerned that are indicative of changes in the Brewer-Dobson circulation. The paper concludes by extending the SCIAMACHY-OMPS data record to earlier decades using similar techniques and SAGE II measurements. Zonally averaged trends from the longer record confirm earlier findings for the 1985-1997 and 1998-2018 periods.

2 Recommendation

This paper fits the scope of AMT and would be suitable for ACP as well, since equal shares of the manuscript are devoted to merging methods and trend analysis results. I would recommend publication as long as the authors are willing to improve the discussion of several topics.

We thank the reviewer for the positive evaluation, we addressed each comment at our best.

3 Major comments

3.1 Demonstrate that the longitudinal structures are realistic

My main criticism on this paper is that there is no substantial proof of the robustness of the reported longitudinal structure of the time series and derived trends. A much more profound discussion is needed about the validity of the longitudinally-resolved results, especially since this is one of the central points of the paper. Constructing a lon-resolved data record is one thing, but the authors need to show that the longitudinal information in the data record is reliable and stable. This should have been the cornerstone of this paper, but it is entirely missing from the paper. As an illustration (p.12, l.2): "A plot of the longitude-resolved drift values is shown in the Supplements, Fig. S1". But no discussion of key results follows: is there longitude structure in the drift field, or not? Another check would be to compare lon-resolved maps of trend results at neighbouring vertical levels to demonstrate their stability in the vertical domain. Once this validation step is over with, you could gain additional confidence by discussing how the derived trend fields compare to what is expected.

We agree with the reviewer that our discussion of the longitudinally-resolved drift and longitudinal structures observed in the trends was insufficient. We followed the reviewer's suggestions and analyzed in more details the longitudinal structure of the drift with respect to MLS and the vertical consistency of the trend patterns found in Fig. 8.

In the supplements we added the longitudinally-resolved plot of the drift at 41.3 km (Fig. S2, lower panel), which can be compared with Fig. 8, showing ozone trends at the same altitude.

Looking at the drift at this altitude, we see a longitudinal structure: although values are generally non-significant and within $\pm 2\%$, negative values are found in the [0°, 80°] longitude band, whereas positive drift, yet mostly non-significant, are detected within [100°, 260°] longitude. These patterns do not explain the features found in the trends map, even thought they possibly enhance them. The summary of the results has been added to the paper.

To better evaluate the vertical stability of the longitudinal patterns found in Fig. 8 we added in the paper a second panel to this figure, showing the cross section at 60° N of the trend values. In the Supplements, similar plots at 60° S and in the tropics are provided (Fig. S6). We notice that in the northern hemisphere the positive trends over Canada have statistically significant values over a vertically coherent region of the stratosphere, around 40-45 km; at the same time over the Siberian sector values are statistically non-significant at almost all altitudes. In the southern hemisphere the structure is also vertically consistent without large changes as a function of longitude.

To our knowledge no extensive studies of ozone trends as a function of longitude have been published. However, several studies addressed the longitudinal variations of the BDC, using both satellite measurements and atmospheric models. Kozubeck et al., 2015, found not only a two-core structure of opposite meridional winds in the upper stratosphere at northern midlatitudes, but also significant positive trends in this structure, studying the last 20 years. Bari et al., 2013, studied the 3D structure of the BDC comparing a general circulation model and MLS observations. The authors found zonal asymmetries in the meridional mass transport, affecting also the ozone and water vapor distribution, especially in the northern middle winter stratosphere. The results of these studies are discussed in the revised manuscript in Sect. 4.2.

3.2 Elaborate discussion of merging technique

The authors present two merging methods and the resulting difference time series with respect to Aura MLS (Figs. 3-4). Unfortunately, they miss the opportunity to discuss merits and weaknesses of each of the methods and in what way one or the other method can correct for specific issues. I feel such a discussion in Sect. 3 would improve the paper a lot. In the end, readers of this paper will be interested in what you recommend as merging approach: plain-debiasing or anomalies? The answer to this naive question may depend on the use case, of course, but this should be part of the discussion. For instance, Fig. 4 shows a discontinuity of in the anomalymerged time series between 10 N-10 S at 31.5 and 34.8 km. What is the cause of the feature and why is it not present in the plain-debiased time series (Fig. 3)? The trends in the tropics (Fig. 6) are, surprisingly, not very different using both data records. How can this be? On the other hand, p.14, l.14 claims "The general picture in the two panels is very similar, even though trend values in panel (b) are slightly larger". Can this observation be linked to the merging strategy?

Also the reviewer #2 raised a similar question and the answer to it is not straightforward: as the referee already suggests in the comment, it depends indeed on the use of the data set. The subtraction of the seasonal cycle before merging is generally considered a good way to take into consideration the different geometry of observation of several satellite instruments, when merging them. In our case, since SCIAMACHY and OMPS-LP have a similar sampling and geometry (in terms of scattering angle) we performed also a direct merging, removing just the bias between the two time series, with the advantage of providing a long-term time series directly in terms of number density and preserving the original seasonal cycle. On the other hand, at some latitudes, especially towards the polar region, the differences between the seasonal cycles of the two instruments become important and a direct merging of the two time series in terms of number density may introduce artifacts. The different approaches use the same procedure to compute trends (except for the harmonic terms) and, as we showed in the paper, do not significantly affect the results of long-term ozone changes. So, in this example of merging two data sets from similar instruments we don't see strong advantages for trend studies using one of the 2 methods. However, depending on the use of the data set, one approach may be more convenient than the other. For example, for data assimilation and model studies, a time series in terms of number density or VMR is more valuable than ozone anomalies, to which a reconstructed seasonal cycle have to be added before the use. We added a short discussion of this issue in the conclusions and extended the description of Fig. 4 in the paper.

The reviewer is right about the discontinuity visible in Fig. 4 in the tropics particularly at 34.8 km. The reason for this feature is that data were plotted without debiasing the anomalies. Indeed, after removing the seasonal cycle from each instrument time series, OMPS-LP anomalies are debiased with respect to SCIAMACHY, using the overlapping period with MLS. This step was missing in the plotted data. The bias is particularly large in the middle tropical stratosphere, where MLS seasonal cycle shows the largest change over the considered period. The plot has been updated and now Fig. 3 and Fig. 4 look, in this respect, much more consistent.

The slightly larger values in the trends using the plain-debiasing approach are most likely related to the different merging strategy, in particular to the way of computing relative trends. In the anomalies approach, the absolute anomalies are divided by the seasonal cycle to obtain relative anomalies, which are directly used to compute ozone trends in % per decade. In the plain-debiased approach, the trends are computed using ozone number density time series and then normalized to the averaged time series at each altitude, latitude and longitude to obtain the values in terms of % per decade. This is now mentioned in the manuscript right after the sentence mentioned by the reviewer (lines 19-22, p.16 of the revised version).

3.3 Absolute vs relative offset corrections

The adopted plain-debiasing method (Eqs. 1-2) removes additive biases but not the multiplicative biases. And vice versa, the adopted anomalies method (Eqs. 3-4) removes multiplicative biases but not the additive biases. Can the authors clarify the statement on p.8, l.8-9: "Through the merging process biases will be subtracted, whereas the discrepancies in the shape of the seasonal cycle are accounted for when calculating anomalies (subtraction of the SC)."?

The reviewer is right, the additive bias are removed only in the plain-debiasing approach, whereas in the calculation of relative anomalies, the additive bias remains at the denominator. We changed the sentence in the paper accordingly: 'Through the merging process additive biases are subtracted via the plain-debiasing procedure, whereas the multiplicative bias and the discrepancies related to the different shape of the seasonal cycle are accounted for when calculating anomalies.'

We computed, in addition, trends using absolute anomalies, i.e. without dividing by the seasonal cycle as in Eq.4. In this way the additive bias is removed. Trends values computed using absolute anomalies are then divided by the averaged merged ozone time series at each altitude, latitude and longitude. The results are displayed here in Fig. 1 panel (a) and are very similar to the one showed in the paper (Fig. 6 panel (a)), stressing the consistency of the different merging strategies. In particular, we present in panel (b) also the difference map between the zonal trends computed using relative and absolute anomalies: values are extensively within $\pm 1\%$.

3.4 Substantiate claim about stability MLS seasonal cycle

p.8, l.3-4: "In addition, we notice that MLS SC may vary within the instrument life time, as shown at 34.8 km in the tropics with change of up to 5-7 % between the two periods." This is quite a bold statement which may worry the users of Aura MLS data. But this claim is not



Figure 1: In panel (a) zonal trends obtained using absolute merged anomalies over 2003-2018, as a function of altitude and latitude. In panel (b) difference map between trends shown in Fig. 6 panel (a) of the paper, i.e. following the relative anomalies approach, and trends from panel (a) of this picture.

really substantiated by the authors. Should a reader be really worried about the stability of MLS data, while you mentioned earlier on that it is stable? When looking at Fig. 2 only one panel indicates that MLS 2005-2012 deviates clearly from MLS 2012-2016. I would like to see more proof/discussion if you want to keep the statement that MLS SC varies over its life time.

We agree with the reviewer that our statement could be confusing for the reader. The seasonal cycle is affected by the atmospheric natural variability and may change depending on the considered period of time. We did not mean to relate the changing MLS seasonal cycle to instrumental problems. We clarified the sentence in the paper, rephrasing it as follows: 'In addition, the natural variability of the atmosphere plays an important role, with the seasonal cycle that naturally evolves with time: we notice, for example, that the seasonal cycle measured by MLS varies between the two considered periods, in particular at 34.8 km in the tropics, where a change of up to 5–7 % occurs.'

3.5 Collapse of longitude dimension

p.7, l.15: "In this paper we only describe the analysis of the longitudinally resolved ozone profile product". If you do not consider the zonally averaged data in this Section, why mention the binning at all? This is confusing as most of the plots in Sect. 3 are latitudinal cross sections. More importantly, the authors do not clarify in what order and how the different dimensions are collapsed from the underlying alt-lat-lon-time resolved data, perhaps because it has no importance but -in that case- it should be stated somewhere. For e.g. Figs. 3 and 4, did you collapse longitude dimension before computing the difference to MLS, or, first compute difference to MLS then average over longitude?

Several binning possibilities were introduced to show that different products with a higher spatial resolution (in terms of latitude and/or longitude) or a higher temporal resolution are available using data from SCIAMACHY and OMPS-LP. We chose then to consider throughout the paper the following product: monthly mean profiles, spatially gridded every 5° latitude and 20° longitude. However, we did not always show in the manuscript longitudinally resolved results. Indeed, in some cases the longitude-resolved information is not possible to be shown, as it would need too much space (for example for Fig. 3 and 4), in other cases we want to show and compare zonally averaged results from previous studies (for example Fig. 6), or simply we considered as not relevant to show the longitude-resolved plots (for example the correlation

between the merged data set and MLS). We clarified this point in the paper: 'In this paper we consider the longitudinally resolved ozone profile product, i.e. monthly averaged profiles every 5° latitude and 20° longitude. In some cases however we don't show the longitudinally resolved results, either for lack of space or because the zonal averages are directly comparable with previous studies. In this case, the average over longitudes is performed on the level 3 data.'

We thank the referee also for the second point regarding the way zonal mean were calculated. The procedure we followed involves first the computation of ozone zonal mean profiles and then the comparison with MLS data. We included this clarification in the paper before the introduction of Fig. 1.

3.6 Diurnal variation

p.9, l.24-25 reads "Furthermore, at these altitudes diurnal variation of ozone have to be accounted for (Sakazaki et al., 2013), which was not done in this study". This message is repeated on p.15, l.5-6. The correction scheme Eq. 1-2 removes (additive) biases between data records, irrespective of the nature of the bias. Biases due to diurnal variation are part of the total bias. Hence, I infer that diurnal variations are accounted for contrary to what the authors claim. Can the authors respond to this reasoning, and incorporate their answer in the manuscript?

We see the point of the referee and we generally agree with his/her reasoning: diurnal variations are largely removed when the debiasing is performed. This however holds only if they don't change in time between the two instruments. Analyzing diurnal variations for different months above 45 km, we found not only a variability in shape and absolute values over the year, but also that the difference in ozone between the overpass time of SCIAMACHY and OMPS-LP has a different seasonal cycle with respect to the ozone seasonal cycle at the same altitude and latitude. In addition since above 45 km the solar influence gets more important and complex, we believe that a plain debiasing cannot fully account for this. However we deleted the reference to diurnal variations when describing the zonal trends on pg.15, 1.5-6. The Reviewer #2 also raised some concern the about diurnal variation: the reply can be found at p.2 of the answers to the comments.

We included the following sentence in the paper: ' [...] the expected systematic bias between the two instruments is largely removed by the debiasing procedure, even though not completely, because variations with time of this systematic discrepancy may not be accounted for by a 'plain-debiasing'

3.7 Impact of using ERA-Interim data to convert Aura MLS data?

The authors mention that the Aura MLS data record is stable (p.6, l.18-19). However, it is not clear from the paper whether this holds for converted Aura MLS data as well. Please elaborate on how the ERA-Interim data may impact the converted Aura MLS data. Can it induce the change in seasonal cycle reported on p.7, l.6-7? Can it lead to the drift above 50 km reported in p.9, l.22-23?

Also the referee #2 raised some concern about MLS conversion into number density: we performed the conversion of MLS data using pressure profiles from MERRA-2 instead of ECMWF ERA-Interim. The results are shown in the appendix of the paper and demonstrate that the effect on the ozone trends is negligible. However, relative differences between MLS profiles converted using the two different reanalysis were found to reach 3-5% above 55 km.

We don't expect this conversion to cause the change over time of the MLS seasonal cycle reported in the paper, which is mostly related to the natural variability, since it is particularly evident only in the middle tropical stratosphere. The observed drifts in Figs. 3 and 4 above 50 km are too large to be caused by the MLS conversion. We show in Fig. 2 (see below) the drift of the difference between MLS converted using ECMWF ERA-Interim and using MERRA. The drift is in terms of % per decade and computed over the 2005-2016 period. We can notice that above 50 km the drift is around 1–1.5%, smaller than the short term drift found in Figs. 3 and 4. In addition, we did not find any change of sign in this drift over SCIAMACHY and OMPS time, as displayed in the picture in the paper.



Figure 2: Drift of differences between MLS converted using ECMWF ERA-Interim and using MERRA, as a function of altitude and latitude. The drift is in terms of % per decade, computed over the 2005-2016 period.

3.8 Correlation between solar and trend term

The MLR regression model contains a term for the 11 year solar cycle and a linear trend term (p.12, Eq.6). The analysis period (2003-2018) contains one and a half solar cycle, which triggers the question as to how independent the two said low-frequency terms are. Can the authors elaborate on this? Could the change in derived trend for different starting times (p.15, l.7-10) be related to interference between the solar and trend term, this is exactly the region where solar influence should be large. This concern may even be more important for the results shown in Figs. 7 and 9 where even shorter periods are regressed. Perhaps in these cases the non-trend terms were regressed over the entire time period?

The reviewer is right about the issue and we are aware of the problem. That's why we specified in the paper to take the trends over short time periods with caution. We are also aware that more robust results should come from a record covering 2 full sun cycles, but for that we have to wait some more years.

As suggested by the reviewer at this point and in a couple of minor comments, we also tried a different way to regress the trends over the shorter periods (2004–2011 and 2012–2018). First we fit all the proxies excluding the linear terms over the longer time period (2003-2018) and then we perform a linear fit for shorter periods using the residuals after the first step (differences between time series and fit). As an example we show here in Fig. 3 the comparison between the zonal trends over 2004-2011, computed using both methods: in panel (a) the same as the plot presented in the paper and in panel (b) using the wider time range to fit all the non-trend terms. We put this figure in the Supplements (Fig. S7). As we notice, the differences are rather small, even though the bipolar pattern found in the tropics and southern mid-latitudes is less pronounced using the second strategy. However, this method can be applied for the 2 considered sub-periods but we don't see how it could be applied for the 2003-2018 SCIAMACHY-OMPS merged data sets, in which case there are no measurements to be fitted before 2003.



Figure 3: In panel (a) the trends are computed fitting all the terms over the 2004-2011 period (as done in the paper), in panel (b) all the non-trend terms were fitted over the 2003-2018 period. In panel (c), we present the differences of zonal trends over the period 2003-2018, with and without considering the solar proxy.

In order to evaluate the magnitude of the effects of the solar proxy on long-term ozone variations, we computed trends over 2003–2018 without considering the solar proxy in the fit: panel (c) of this figure, shows the difference between the results of this calculation and panel (b) of Fig. 6 in the paper. The differences are within $\pm 1\%$ at most altitudes and latitudes, so that we expect that the interference between the solar and trend term pointed out by the reviewer is smaller than this threshold.

4 Minor comments

p.1, l.11: Be specific about what you mean with "remarkable variability".

We find it difficult to be specific in the abstract, however we added: 'with variations of up to 3-5 % per decade at altitudes around 40 km.'

p.2: Very nice and concise overview of ozone-related processes.

Thanks.

p.3, l.19: Identify "MLS" as "Aura MLS" here and throughout the rest of the paper. You don't want to confuse with the first MLS instrument which was flown in the 1990s-2000s on the UARS satellite.

Thanks for the observation, we specified that we mean the MLS instrument onboard the Aura

satellite to avoid confusion the first time we introduce the acronym, but we kept the use of MLS to indicate Aura MLS, in the rest of the paper.

p.3, l.19-21: You should introduce SAGE II over here, instead of two instruments (ACE FTS and SAGE III/ISS) that are not mentioned in the rest of the manuscript.

We agree with the reviewer to mention at this point the SAGE II instrument but we also left the sentence about ACE FTS, MAESTRO and SAGE III as currently operating solar occultation instruments.

p.3, l.25: Please rephrase. Harris et al (2015) did not merge these data sets, but use them to derive trends.

Thanks for this comment, also the Reviewer #2 highlighted this point: we rephrased as: '... the authors considered several existing merged satellite data sets and examined separately the time spans before and after the peak in ODSs concentration at the end of '90s. The authors combined trends from the different data sets and ...'

p.3, l.34: Remove "applying a multilinear regression analysis". This information is evident and not different from the other analyses you refer to.

True, we deleted this information.

p.4, l.1: Vague statement "Ball et al. (2018) applied a method independent from the ozone turnaround point". The subsequent clause "showed for the first time some evidence of a negative trend in lower stratospheric ozone" seems to imply that the different regression method is leading to this discovery. I am not sure that is what Ball et al. claimed.

We improved the sentence as follows: 'The authors analyzed a longer period of time, together with improved merged time series and considered the lower stratospheric column instead of the ozone profile. With these adjustments, they showed for the first time ...'

p.4, l.4-5: Hanging statement: "This analysis has recently been challenged by Chipperfield et al. (2018)". In what way?

We added, 'who showed that the apparent downward trend in the lower stratosphere (ending in 2017) is a result of longer term variability in atmospheric dynamics.'

p.4, l.6: Clarify what a "pointing drift" is. A general reader will not have a clue what pointing means in this context. Consider vertical pointing, altitude registration, ...

Thanks, we replaced with 'altitude registration', which is more clear: '..., after OSIRIS data were corrected for a drift in the tangent altitude registration of the instrument.'

p.4, l.15: I am not sure LOTUS is "homogenizing" the merging procedures, please double check this with one of the LOTUS participants.

We replaced it with '... studying robust methods to merge data sets ...'.

p.5, l.12: "performing measurements at 3 viewing angles, which differ horizontally by 4.25 deg.

Yes, we reformulated the sentence as suggested by the reviewer.

p.5, Tab. 1: Extend this table to SAGE II and Aura MLS.

We added the information for MLS and for SAGE II.

p.5, Tab. 1: Add level 2 versions in this table as well. This will make life easier for readers in 5 years from now.

Yes, it is indeed a good idea.

p.5, Tab. 1: I advise to show the analysis time period for both instruments. Right now, different information is conveyed in "data time series": SCIA (full mission period) and OMPS (analysis time period). Please use one or the other, but do not mix up.

Thanks, we restricted to the time periods used in our analysis.

p.5, Tab. 1: Align values of spectral coverage and spectral resolution with what is in the main text.

Yes, for SCIAMACHY there was a discrepancy.

p.5, l.20: Add the version of SCIATRAN.

We included the respective versions: v3 for SCIAMACHY and v4 for OMPS.

p.6, l.1: Add the version of the SCIAMACHY L1 data, as was done for OMPS-LP.

We included the following sentence: 'In particular, v8 L1 SCIAMACHY and v2.5 L1 OMPS-LP data were processed.'

p.6, l.9: Clarify "pointing knowledge issues", see also my earlier comment. E.g. "[...] when the issues related to the vertical pointing of the instrument, currently under [...]"

The sentence has been modified as follows: 'data from the lateral slits are planned to be used when the issues related to the tangent altitude registration of the instrument, currently under investigation by NASA, are solved.'

p.6, l.9: Refer to Moy, AMT 2017.

Done

p.6, l.13: Replace "scientific measurements" by a better description or simply drop "scientific".

We replaced it with 'atmospheric observations'.

p.6, l.18: You mention Hubert et al. (2016) for Aura MLS drift. But any trend paper should refer to published drift results for all instruments involved. I.e. add those for SCIAMACHY (Rahpoe-2015, Hubert-2016, LOTUS-2018, ...?) and OMPS-LP (Kramarova-2018) as well, un-

less the L1-L2 versions have changed sufficiently to question the validity of those values.

Both Raphoe 2015 and Hubert 2016 refer to the old version of SCIAMACHY L2 data, while the LOTUS second report still has to be published. The only work to our knowledge which addressed the drift with respect to other satellite data sets of SCIAMACHY v3.5 ozone, is Sofieva et al., 2017. The authors stated that evaluating and inter-comparing the anomalies of the considered instruments, among which SCIAMACHY, they did not find statistically significant drifts with respect to the median anomaly. This was summarized in the paper. We also added a reference to Kramarova et al., 2018, as suggested, addressing OMPS-LP drift with respect to MLS and OSIRIS.

p.6, l.18: Find a better phrasing for "For technical reasons" as it suggests an instrument malfunction. The observations by SAGE II are sparse due to the chosen measurement geometry and is unrelated to the instrument itself.

We agree with the reviewer: we changed the sentence as 'Due to the occultation viewing geometry, ...'.

p.6, l.31: "Aura MLS", see earlier comment.

We specified at the beginning that with MLS we mean Aura MLS.

p.6, l.33: "taking only the latitude covered daily by OMPS-LP". You lost me here, what latitudes are not covered by OMPS? Please clarify what you mean in the main text. And why does this resolve an inconsistency? SCIAMACHY measurements are also made during daytime.

Yes, here we need to be more clear: for every day of the year, we considered MLS measurements within the latitude extremes covered by OMPS-LP during that day. They do not coincide with MLS profiles flagged as day-time observations in L2 data. We reformulated the sentence as: 'For each day, we take only MLS measurements which are made within the latitude range covered by OMPS-LP and SCIAMACHY.'

p.7, l.2: "Aura MLS", see earlier comment. Please incorporate this comment in the rest of the manuscript.

As mentioned, we specified at the beginning that with MLS we mean Aura MLS.

p.7, l.3: Add a motivation for not using the 2002 SCIAMACHY data.

Some studies, like Sofieva et al. 2017, reported anomalous values at the beginning of SCIA-MACHY mission with respect to other satellite observations. We specified it in the paper.

p.7, l.11-12: Figure 1 does not confirm the statement "In both cases we find about 100 profiles on average in each bin." for SCIAMACHY. Each monthly 5deg zonal bin has 1000 profiles, which translates to 56 (=1000/18) profiles per bin, not 100. Did I misunderstand? If not, please change the misleading statement.

With 'about 100 profiles' we meant the order of magnitude, aware that for SCIAMACHY the number is lower than for OMPS-LP and that the available observations depend on latitude and time. Instead of 'about 100', we wrote 'on average 50–100'.

p.7, l.14: Clarify what interpolation method was used.

Done, we mentioned that we used a linear interpolation.

p.7, Fig.1: Add 5° at the end of the caption: "[...] in each 5° zonal monthly bin [...]".

Done

p.8, Fig.2: Is the SCIAMACHY time period identical to that of Aura MLS (2005-2012)? Please add the time period for all four lines in the legend, not just for Aura MLS.

Yes, seasonal cycle for OMPS-LP and SCIAMACHY are computed using the same period as for MLS. We included the time periods for all the instruments in the legend of Fig. 2 and updated the description in the paper.

p.8, l.5-6: The phrasing is not clear whether the time period was only adapted for MLS. In other words, did you compare 2005-2012 for both SCIAMACHY and MLS, and 2012-2016 for both OMPS-LP and MLS? See previous comment.

See previous comment

p.9, l.14: Add a short phrase that the unit of the plain-debiasing data set is ozone number density.

We added this information in the same sentence: 'The merging is then achieved by concatenating the two data sets, in terms of ozone number density, ...'

p.9, l.16: "[...] differences between the merged data set [...]". What merged data set? The zonal one? The longitudinally resolved one?

In the sentence before this one we spoke about the merging of the 2 time series, so we are here directly referring to the plain-debiased longitudinally resolved data set. The picture shows however zonally averaged differences: this has been done for technical reason, it is already a pretty busy figure and we did not find an easy way to show also the longitudinal dimension, which in any case would not add much information. We explained this in the paper when we introduce the binning of the data sets. The collapse of the longitudinal dimension is done again on the Level 3 data: MLS, OMPS and SCIAMACHY data sets are firstly zonally averaged and then the processing (computing, bias, anomalies etc...) is performed.

p.9, l.16-18: What is the sign of the relative difference? (SCIAOMPS - MLS) / MLS or the other way around?

It is (SCIA or OMPS - MLS) / MLS. We added this information in the paper and it is valid for the whole paper.

p.9, l.28-29: Are the larger relative difference values at 15 km truly due to lower data quality or due to the smaller number densities in the UTLS region?

We think it is for both reasons, for sure the smaller number density in the UTLS region espe-

cially in the tropics leads to larger relative differences. We added at p.12, l.19-20 of the revised version that: '... low values of ozone number density, especially in the tropics, amplify the relative differences.'

p.9, l.30-31: Replace by "[...] deseasonalized relative anomalies from [...]" to clarify that you are not working with absolute anomalies.

Done

p.9, l.30-31: What is the motivation behind debiasing the deseasonalized relative anomalies? By computing the anomaly any multiplicative biases are removed by definition.

SCIAMACHY, MLS and OMPS-LP time series are deseasonalized over different periods and have then a zero mean values over their respective periods. So that an additional debiasing using MLS is required, which consists in bringing OMPS-LP anomalies to the average values of MLS anomalies over 2012–2016 and then to SCIAMACHY anomalies over 2005—2012.

p.9, l.31: Replace by "[...] month of the year, m, the (relative) anomalies, [...]".

Done

p.10, Fig. 3 and p.11, Fig. 4: Add in the colour scale the exact sign of the difference: (merged - MLS) / MLS or (MLS - merged) / merged ?

We referred in the caption of Figs. 3 and 4 to Eqs. 3 and 7, which explicitly express how the differences were computed.

p.10, Fig. 3 and p.11, Fig. 4: Add in the caption that MLS data has been offset to SCIA prior to the comparison.

Done, using 'before the comparison' instead of 'prior to the comparison'.

p.10, l.4: Eq. 5 is not really used in the rest of the paper (p.12, l.7-8). I would therefore suggest to drop it, also because (a) you do not explain how the uncertainty for the plain-debiasing data is computed and (b) there is no term for the uncertainty in the seasonal cycle.

We agree with the referee, the equation was removed.

p.10, l.9: Replace by "Figure 4 shows the absolute differences [...]" to clarify that these are absolute differences of relative anomalies.

Done

p.11, l.1: "whereas below 20 km the pattern becomes rather chaotic".

The comment is not complete and thus cannot be addressed.

p.11, Fig.4: Larger differences are found around 35 km in 10S-10N during the OMPS-LP period. What is the cause of this? Does the different MLS SC in the two periods play a role?

We updated this picture, as mentioned in the main comments: now the differences are smaller, therefore we dropped the sentence.

p.11, l.4-5: "The drift is computed as the linear change of the differences between the merged time series and MLS data [...]". Are these relative differences for plain-biased merged data and absolute differences of anomaly-merged data? Clarify this in the text. And add the unit of the drift: % per decade/year/....

We specified that the differences are either relative Eq. 3 or absolute Eq. 7 for the 'plaindebiased' data set and anomalies respectively'. The unit of the drift was added.

p.12, l.1-2: "Very similar results for the drift are obtained using anomalies time series". The timeseries in Figs. 3-4 look fairly different in places, and I am surprised the drift results are very similar for the anomaly time series. This plot has to be in the main paper, also since it may be the basis of an interesting discussion on what merging technique led to most stable results for this particular case. (See also one of my major comments.)

Since the plots of the drifts computed using the two strategies are very similar we decided to keep the other plot in the Supplements (Fig. S1) and we refer to it in the paper.

p.12, Fig. 5: Add in the colour scale the full unit (% per decade/year/...) and the exact sign of the difference: (merged - MLS) / MLS or (MLS - merged) / merged ?

We added the full unit and we refer to the equation expressing the sign of the difference.

p.12, l.6: What are the units of O3 in Eq. 6? The plain-debiased time series are in molec cm^{-3} , the anomaly-merged time series are in %?

We specified the units also at this point in the paper.

p.12, l.14-15: The phrase "The t-th row of the X matrix contains the values of the fit terms for the selected t." does not add information. It could easily be dropped.

We see the point of the referee, we dropped the sentence.

p.13, l.1: The equivalence of the 2σ rule to 95% confidence level is introductory statistics, hence the reference to (Tiao et al., 1990) is not needed.

Done, we agree with the reviewer.

p.13, l.3: How are the plain-debiased time series in molec cm^{-3} regressed to obtain % per decade?

The time series are regressed in terms of molec cm^{-3} , then the obtained trend values are divided by the mean ozone over the time series at each altitude-lat-lon bin. We added this information in the paper.

p.13, l.6-9: Do I understand you correctly that the EHF term is used instead of the harmonic terms, below 25 km and only for the 50-60°N band? Why not for 50-60°S as well, or at other latitudes? Please add that this modified regression model is not applied to the analysis of

anomaly-merged time series.

We refer here to the work of Gebhardt et al. 2014, where it was found that at these latitudes in the northern hemisphere the ozone annual cycle has a larger amplitude with respect to the southern hemisphere, due to the stronger wave activity at northern mid-latitude, which influence the ozone distribution in the lower stratosphere in this region. For the same reason, the annual cycle is also characterized by a strong interannual variability which can lead to an insufficient modeling of the seasonal cycle if considering simple harmonic terms. That's why we used EHF integrated starting from October of each year. The explanation has been improved in the paper.

In addition, EHF are used to regress both the ozone number density time series and anomalies.

p.13, l.23: Please cite more recent work, at least Maycock et al. (2016), perhaps others as well (Ball et al., 2016; Damadeo et al., 2018; ...).

We agree with the referee, the paragraph was revised citing the studies of Soukharev and Hood (2006) and of Maycock et al. (2016).

p.14, l.1: Add the source of the El Nino 3.4 index data, as you did for the other proxy data sets.

Done: 'The data time series is available at: http://www.esrl.noaa.gov/psd/gcos_wgsp/Timeseries/Nino34/.'

p.14, l.6: Add that N34 represents the El Nino 3.4 index anomaly data.

We think that this is already addressed, since we say on p.6 l.1-2 (p.16, l.4-5 of the new version) that '... [El Nino 3.4 index] is based on sea surface temperature anomalies averaged from 5° S–5° N and 170°–120° W.'

p.14, l.11-12 and Fig. 6: Are the trends in Fig. 6 regressed directly from the zonally averaged merged time series, or are these trend results regressed from lat-lon resolved merged time series then averaged over the latitude bands? In the first case, this contradicts an earlier statement that only analysis of lon-resolved data would be described (p.7, l.15). In the latter case, how do you propagate the uncertainties?

The reviewer is right. Our initial statement was probably not clear: as already addressed in the major comment 3.5, we refer to the lon-resolved data set meaning that throughout the paper we considered monthly profiles binned every 5° latitude and 20° longitude. However some of the presented plots show zonally averaged results, due to lack of space to show the lon-resolved fields or for the relevance of the zonal averages. In this case, the zonally averaged L3 time series were considered and regressed to compute the trends. The explanation was added at the beginning of Sect. 3.

p.15, l.2: You may consider adding the LOTUS report and replace the WMO reference in this phrase (WMO, 2018).

We updated the WMO reference, while LOTUS report is not yet published to our knowledge.

p.15, l.9: Perhaps the cause is not instrumental, but related to the interference of the solar and

trend term? See my major comment above.

This is also a possibility; we addressed this issue at the major comment 3.8.

p.15, l.35: Are all terms (seasonal, QBO, solar, ENSO, ...) constrained by the 2003-2011 period or just the linear trend term? This shorter period potentially makes the interference between solar and trend terms even larger. Have you looked into this? The trend results may be more stable/robust when you constrain all non-trend terms (especially solar) to the larger 2003-2018 period.

The plots presented in the paper show trends computed fitting all the proxies in each respective period, in this case 2004-2011. We addressed this issue replying to the point 3.8 of the referee's Major comments.

p.16, Fig. 7: Add complete unit to y-axis label: molec cm^{-3} .

We added the unit to the caption.

p.16, Fig. 7: Adding the anomaly-merged time series and fits would make a fine illustration of how the merging strategy can overcome some of the issues in the data sets as mentioned e.g. in p.16, l.4-7.

We realize that the sentence, which the reviewer refers to, may be misinterpreted: we do not explicitly mean here that the plain-debiasing approach is better, in this regard, with respect to the anomalies strategy. We only meant that at these altitudes and latitudes (tropics, mid-stratosphere), due to the change in time of the MLS seasonal cycle already pointed out, MLS anomalies are sensitive to the period over which the seasonal cycle is computed. As a consequence, also the merged SCIAMACHY/OMPS anomalies data set is affected by MLS deseasonalization. This is not a disadvantage of the anomalies approach by itself but a consequence of the merging of two data sets without an extensive common period.

In order to address the reviewer's request, i.e. add the anomaly-merged time series + fits, another self-standing plot would be needed, since the vertical scale is different. We think that this issue is too technical to be fully discussed in the paper.

p.16, l.1: There is the switch to OMPS-LP in 2012. Can this be a viable alternative explanation to the "discontinuity"? The fits themselves will, in addition, likely be impacted by the solar-trend interference as well.

The change in the instrument plays most probably a role, but not to the extend of explaining the jump. This would be the case only if both instruments had a drift of different sign, which has not been so far identified. In addition, we are currently comparing our results with a run of the TOMCAT CTM and we found similar results between 30 and 35 km in the tropics, which gives us more solidity for such a conclusion. Anyway, we added in the paper that: 'In addition, the switch between SCIAMACHY and OMPS-LP time series and the interference between the solar proxy and the trend-terms may enhance this discontinuity in the long-term changes.'

p.16, l.11-12: Please substantiate why the longitudinal trend results are reliable? A figure like Fig. 8 for neighbouring levels z=41 and 44 km will help to demonstrate the stability of the results in the vertical domain, especially in the US where trends are mostly significant (also in other studies).

At this aim, we included in Fig. 8 a panel (b) illustrating the vertical cross section of the trends as a function of longitude at 60° N. This shows how the positive significant values over the Canadian sector are vertically consistent between 38 and 45 km.

p.17, Fig. 8: Add to the caption what merged data set was used: plain-debiased or anomaly-based?

Done

p.17, Fig. 8: Remove the results in the latitude-range that you mentioned earlier on was not reliable (60° for plain-debiased and 70 for anomaly-based).

We used the anomalies data set, so we keep the results up to 70° .

p.17, l.10-11: Move this discussion to previous paragraph and elaborate on how stable results are in vertical domain.

We left this sentence here, because the study of Kozubeck et al. refers to the upper stratosphere. However we added here a short description of the plots in the Supplements.

p.17, l.15-16: Motivate why you use the anomaly approach.

This was done to take into consideration the different geometry and sampling of the three instruments (particularly, the low density of SAGE II measurements). We included this in the paper.

p.18, l.6-7: Add brief explanation why the harmonic terms are not included (deseasonalized anomaly time series).

As done for the SCIAMACHY-OMPS merged anomalies, we don't need the harmonics here indeed because the seasonal cycle is naturally subtracted when calculating anomalies.

p.18, l.6-7: Slightly confusing, since the trend model is very different from that in previous section. Please clarify whether it is an independent trend (ILT) or a piece-wise trend (PWLT).

The same trend model is applied to the merged SCIAMACHY-OMPS anomalies data set and to both individual data sets. We specified in the paper that independent trends over the two periods have been computed.

p.18, l.13: Add correct unit : "[...] about -2% per decade is detected [...]".

Done

p.18, l.13: As asked before (p.15, l.35), what time period was used to constrain the non-trend terms? And how robust are -especially- the 2012-2018 trend results given the low frequency of the 11 year solar cycle proxy?

This point was already addressed in the reply to the referee's major comment 3.8.

p.18, l.19: You mention 2010-2018 here, while the figure caption says 2012-2018. Which one

is correct?

Thanks, it is 2012-2018, the text was wrong.

p.18, l.32: What do you mean with "up to" the polar regions?

'Including' is indeed better.

p.19, Fig. 9: You mention 2012-2018 in the caption, while the main body text says 2010-2018. Which one is correct?

Same as two comments before.

p.19, Fig. 9: Add the time period to each panel, in addition to (a), (b), ...

We agree, it improves the readability.

p.19, l.6-7: Strong claim that needs demonstration: how reliable is the observed lon-resolved structure?

We softened it by saying 'This is an indication of a possible change in the BDC as a function of longitudes in the northern hemisphere'.

Supplement, Fig. S1: Add in the caption which merged time series are shown: plain-debiased or anomaly?

Done

Supplement, Figs. S1 and S2: Add sign and correct unit (% per decade?) to colour scale or in the caption.

Done

Supplement, Figs. S1 and S2: Each subpanel represents one longitude-bin, all together they convey information about longitude structure of drift of SCIA OMPS wrt MLS. However, the longitude structure would be much more obvious if you would have shown one latitude-bin per subpanel, and then plot drift vs altitude vs longitude. Can you add this to the supplement?

It is a good suggestion, we inserted in the Supplements a panel with the drift as a function of latitude and longitude at 41.3 km. In addition cross sections of trends, i.e. altitude vs longitude trends, have been added to the paper in Fig. 8 and in the Supplements Fig. S6.

Supplement, Figs. S3 and S4: Add to the caption what merged data set was used: plain-debiased or anomaly-based?

Done, these plots are now Figs. S4 and S5.

Supplement, Figs. S3 and S4: Remove lat-range with data that you claimed earlier in the paper are unreliable (poleward of 60° or 70° latitude).

The plots are now up to 70° latitude.

5 Technical corrections

p.1, l.4-5: Replace by "[...] is performed by the processor of the University [...]"

We reformulated the sentence as follows: 'The retrieval of ozone profiles from SCIAMACHY and OMPS-LP is performed using an inversion algorithm developed at the University of Bremen.'

p.1, l.10: Replace "high" horizontal sampling by "dense" horizontal sampling.

Done

p.1, l.24: Remove either "important" or "key". Important implies key and vice versa.

We removed that part of the sentence, leaving: 'The continuous monitoring of the stratospheric ozone layer is required to assess the impact of anthropogenic and natural processes.'

p.2, l.7: Replace by "during the 1990s".

This sentence was changed: 'The adoption of the Montreal Protocol and its amendments regulated the industrial production of chlorine and bromine compounds: in particular, the London amendment in 1990 called for a complete phase out of CFCs production by the year 2000, leading to a decrease of their concentration in the stratosphere starting from the end of the 20th century.'

p.2, l.10: As non-native speaker I expected "GHGs such as", but perhaps "such" is not needed.

We added 'such'.

p.3, l.27: Add a "," in "[...] before 1998, and a positive trend of [...]".

Done

p.3, l.29: Replace by "[...] with uncertainty estimates. [...]".

Done

p.3, l.30: Add "-", replace by "[...] satellite and ground-based data sets [...]".

Done

```
p.3, l.35: Replace by "[...] significant trends [...]".
```

Done

p.3, l.35: Replace by "[...] in the upper stratosphere at mid-latitudes [...]".

Done

p.5, l.4: Replace "[...] in-flight direction [...]" by "in flight direction" or "in the direction of flight".

We used 'in flight direction'.

p.5, l.13: Remove the first "charged" in "charged charged-coupled device".

Done, thanks.

p.5, l.22: Replace "application of SCIAMACHY retrieval scheme" by "application of SCIA-MACHY's retrieval scheme".

Done

p.6, l.3: Replace "we take into account in addition" by "we also take into account".

Done

p.7, l.23: Replace by "[...] the SC of all single instrument data sets [...]".

Done

p.8, l.3: Replace by "[...] the three ozone profile data records in number density [...]".

Done

p.13, l.16: Replace by "[...] and the in-phase at mid-latitudes [...]".

Done

p.13, l.32: Drop "a" in "[...] leading to longitudinally dependent modifications of ozone [..."].

Done

p.14, l.17: Replace by "Bourassa et al. (2018)".

Done

p.15, l.11: Replace "detected" by "found" or "observed". In my view, "detected" implies that the result is significant which is not the case.

We replaced it with 'observed'.

p.15, l.33-34: Remove newline after "[...] panel (a).".

Done

p.17, l.13: Add "s" to "[...] SAGE II occultation observations [...]".

Done

p.18, l.4-5: Replace by "[...] the mean SAGE II latitude plus or minus its standard deviation $[\dots$ "].

Done

p.18, l.7: Replace by "[...] 60° latitude [...]".

We removed the $\pm.$

p.18, l.29: Replace by "[...] is first removed [...]".

Done

p.18, l.30: Remove "one" from "[...] MLS one [...]".

Done

p.18, l.32: Replace by "[...] with respect to the MLS time series [...]".

Done

p.19, caption: Replace by "[...] and in panel (d) over 2012–2018 [...]".

Done

p.19, l.4: Replace by "[...] from 2003 until early 2018 [...]".

Done

p.19, l.5: Replace "detected" by "found" or "observed". In my view, "detected" implies that the result is significant which is not the case.

We replaced it with 'found'.

p.19, l.10: Replace by "[...] has vanished when adding [...]".

Done

Replies to Referee #2 on the manuscript 'Retrieval of ozone profiles from OMPS limb scattering observations' by C. Arosio et al.

We thank the reviewer for the time she/he spent reading the manuscript and constructively commenting on the paper. In the text below, we address the comments from the Referee #2. Referee's comments are shown in italicized font and authors' responses are highlighted in blue.

Major comments The manuscript presents two interesting new merged satellite data sets of ozone vertical distribution, based on SAGE II, SCIAMACHY and OMPS observations. Two different methods are used for merging the data sets. The first one uses MLS data as a transfer function to evaluate the bias between SCIAMACHY and OMPS data sets, which overlap for only 2.5 months, while the second merges deseasonalized anomalies. Both data sets have the advantage of being longitudinally resolved, which is generally not the case for similar merged records except for the SWOOSH data set. Yet, the deseasonalized anomalies record when extended with SAGE II observations is zonally averaged. Ozone trends are then computed from the merged data sets using classical multilinear regression over the 2003 – 2018 and 1985 – 2018 periods for the SCIAMACHY – OMPS and SAGE II – SCIAMACHY – OMPS records respectively. The paper is well written and reference to previous work is adequate. It is suitable for publication in AMT provided that following important comments and recommendations are taken into account.

General Comment

1. The paper is lacking an assessment by the authors themselves of which SCIAMACHY – OMPS merged record they think is best suited for their initial objective of ozone trend evaluation. Comparisons are displayed with MLS data in Figure 3 and 4 of the article, but this record is used as transfer function in both records. What is the advantage for potential users to of using one record over the other one?

There are two points raised by the reviewer.

The first one, regarding which SCIAMACHY/ OMPS-LP merged record is best suited for trend studies, was also addressed by Reviewer #1 and doesn't have a simple answer: we showed that both methods give similar results in terms of ozone trends. The advantage of the plain-debiasing approach is that it maintains the original data sets as they are, so that the resulting merged time series is expressed in terms of ozone number density and preserves the original seasonal cycle. This can be useful for example for data assimilation and model studies, for which a time series in terms of number density or VMR is more valuable than ozone anomalies. The second method involves the subtraction of the seasonal cycle and it is more suitable at altitudes and latitudes (like polar region) where the seasonality of the instruments differ more strongly. The subtraction of the seasonal cycle is a common procedure before the merging, especially when considering several satellite data sets with different observation geometry and latitude coverage. In our case SCIAMACHY and OMPS-LP observe the atmospheric scenes at a very similar scattering angle, the latitude coverage is comparable (as reported in Table 1) and the overpass time differs by 3.5 h. As a consequence we believe that the plain-debiasing method is in this case also suitable and reliable. A sentence was added in the conclusions about this point: 'The anomaly approach is a standard procedure followed in many studies when merging several data sets; in this case, since SCIAMACHY and OMPS-LP observe the atmosphere with a very similar sampling and geometry (in terms of scattering angle), we showed that the plain-debiasing approach is also valid, with the advantage of providing a merged time series expressed in terms of ozone number density and preserving the original seasonal cycle.

The second point relates to the use of MLS as transfer function and for comparison/validation: since MLS data set was not included in the merged record and no drift correction has been

applied, we think that it can be considered an 'independent' data set for validation. The MLS data in the merging procedure is only used to remove the systematic mean bias between the instruments. After the debiasing procedure, the mean levels of the time series are the same for the 3 instruments (at each altitude, latitude and longitude) but the ozone variability and the seasonal structure remain independent. That's why the computation of correlation and relative differences between MLS and the merged time series is justified.

2. An assessment of both records could be provided by comparing them to other independent merged records that have been produced recently, e.g. GOZCARDS, SWOOSH, and others.

Several recent merged data sets such as GOZCARDS and SWOOSH for the period after 2005 are determined by MLS observations. As a consequence, we consider the usage of these data sets for further validation not relevant, since the comparison would lead to very similar results as the ones we got using MLS only. Previous works like Harris et al., 2015 and Steinbrecht et al., 2017 already compared trends from several merged satellite data sets and illustrated the differences.

3. The issue of diurnal variation of ozone deserves some more attention in the article. It is mentioned in page 9 that diurnal ozone variation has to be accounted for above 50 km. However, Sakazaki et al (2013) found significant diurnal variation of ozone well below 50 km and down to 30 km in some latitude ranges.

The reviewer is right, Sakazaki et al., (2012) showed that also at 30–40 km daily variations of ozone play a non-marginal role. However, we did not take them into account for the following reasons. First of all, the equatorial overpass times of SCIAMACHY and OMPS are both around the noon, namely at 10:00 and 13:30 respectively. This was not clearly stated in the manuscript and this information has been added in Table 1 and remarked in a sentence in Sect. 3: 'This considering diurnal variations was not done in our study, because the equatorial crossing time of the two instruments is around noon and differs by only 3.5 h: this would lead to a systematic discrepancy in ozone that we estimate to be about 1-2 % at 30-40 km. Furthermore, the expected systematic bias between the two instruments is largely removed by the debiasing procedure, even though not completely, because variations with time of this systematic discrepancy may not be accounted for by a 'plain-debiasing' procedure.' Indeed, according to the mentioned paper, at altitudes between 30–40 km the ozone concentration has a minimum in the morning after dawn and a maximum in the afternoon, with an amplitude in terms of VMR of 0.15 ppmv, which corresponds to 2-3 % of the ozone at these altitudes. So, considering the satellite equatorial crossing times we expect a difference in the order of 1-2 %. Anyway, the important point to be stressed is that the effect of such a diurnal variation leads to a systematic effect on the whole time series, since the overpass time remains constant; such an offset is then largely removed by debiasing the data sets. We therefore believe that the debiasing procedure to a large extent eliminates this systematic difference between the two instruments, as long as it remains constant with time.

4. More precision is needed on the use of MLS as a transfer function for both records. What is the processing of MLS data in equations 1 and 2? Are they interpolated to the location of SCIAMACHY and OMPS observations? Similarly, not enough attention is given to differences in vertical resolution between the various data sets. Could it be an issue for the merging? In addition, ERA-Interim is used for the MLS data conversion to number density versus altitude. Did the authors test the sensitivity to other reanalyses such as MERRA2?

In Eq. 1 and 2 MLS time series as well as SCIAMACHY and OMPS-LP ones are already con-

sidered as monthly mean profiles, binned into the regularly spaced grid in terms of latitude and longitudes. Before computing monthly averages and binning the data, each single MLS profile has been converted into number density vs. altitude and interpolated onto the common vertical grid (equally spaced every 3.3 km). A sentence was added in the paper to clarify this point: 'In these and following equations, ozone profiles from each instrument are considered as binned monthly averages, interpolated to a common altitude grid.'

The issue related to the vertical resolution is interesting but difficult to remove. It has to be noted that the vertical resolution of the three sensors is similar: about 3 km for all instruments. The vertical sampling is however higher for OMPS-LP and MLS, whose ozone profiles are provided every 1 km, and is equal to 3.3 km for SCIAMACHY. This information has been added to the paper. We interpolated all single profiles from each instrument in the same vertical grid using a linear interpolation scheme. This procedure may indeed introduce artificial discrepancies between OMPS-LP and SCIAMACHY, especially at altitudes where the seasonal cycle significantly changes. To minimize this problem, we chose to perform the interpolation of the 1 km-spaced OMPS-LP and MLS profiles onto the SCIAMACHY lower vertically-resolved grid and not vice versa.

The reviewer raises another interesting point here. It was indeed not properly explained in Sect. 3 that for the conversion from MLS VMR vs. pressure profiles to number density vs. altitude, only pressure profiles from ECMWF are considered, while the temperature profiles are taken from MLS retrievals. We updated the description: 'Volume mixing ratio ozone profiles from MLS on a pressure grid are converted to geometric altitude vs. number density using collocated pressure information from the ECMWF ERA-Interim database and temperature profiles retrieved by MLS.' No issues are known by the authors about ERA-Interim pressure, so that we think that the usage of a different reanalysis product would lead to non-relevant changes. However, a comparison in terms of relative differences between MLS ozone profiles converted using ECMWF and MERRA-2 has been performed and the results have been included in the paper, in Appendix A, Fig. A1. Computing the relative difference between number density MLS zonally averaged ozone distributions over 2016 as a function of altitude and latitude, computed using the two reanalysis, we see (panel (a) of the picture) that the discrepancy increases with altitude, up to 3-5 % above 55 km, while in the lower stratosphere differences are within 1 %. In addition, the sensitivity of the ozone trends to the MLS conversion have been studied as well: the right panel of Fig. A1 in the paper reports the differences in terms of % per decade between the trends computed from the merged data set (plain-debiased approach) when using ECMWF or MERRA-2 for conversion. The differences are small even though not always negligible in the upper stratosphere: values are within -0.25 and +0.5 % at most altitudes and latitudes, approaching +1 % above 45 km at some latitudes.

Specific comments

P2-17: The CFCs have been banned by 2010 in Article 5 developing countries.

The sentence have been reformulated as: 'the London amendment in 1990 called for a complete phase out of CFCs production by the year 2000, ...'

P2-l23: Sentence starting with N2O is a long-life GHG needs to be rephrased.

The sentence has been split in two parts for better readability.

P3-113: What is the reference for the NASA LORE/SOLSE instrument?

Thanks, a reference was included also for LORE/SOLSE: McPeters et al. 2000.

P3-l20: Mention instruments on board SCISAT that use the solar occultation technique.

Thanks, instead of mentioning the SCISAT satellite we directly referred to the ACE-FTS and MAESTRO instruments, as we mention SAGE III instrument in the same sentence.

P3-l24: It is an improper description of the Harris et al. (2015) paper. In this paper, merged satellite records are used for trend studies but the merging is not made by the authors. Intercomparison of the merged records is made in Tummon et al. (2015).

We thank the reviewer for this note. The description of Harris et al. (2015) was changed from 'the authors considered several satellite data sets, merged them over the period 1979–2012 and examined separately...' to 'the authors considered several existing merged satellite data sets and examined separately...'

P3-l26-27: In general, provide trend results from published studies with error bars.

A better characterization of the values has been provided for each cited work, except for Harris et. al (2015), for which several uncertainties were discussed and it was difficult to summarize the results in few sentences.

P3-l30: Ozone-CCI is not the name of a record but the name of a project. More generally in this paragraph it would be better to distinguish articles that describe merged records with those retrieving ozone trends from those records.

Thanks, we changed the terminology regarding Ozone-CCI: from 'merged measurements from SAGE II with Ozone-cci and OMPS satellite data sets' to 'merged measurements from SAGE II with several other data sets homogenized within the Ozone-CCI project including OMPS-LP'. We also improved the distinction between papers merging data sets from articles retrieving trends from the merged data sets.

P4-l1: Mention the name of the method used in Ball et al. (2018).

Yes, the name dynamic linear method has been included.

P4-l23: The SWOOSH record is resolved in longitude.

Thanks for the note, "except for SWOOSH" was added.

P4-l24: Sentence starting with 'In addition': Explain why it is better not to extract the seasonal cycle.

Generally speaking it is better to extract the seasonal cycle to remove discrepancies between data sets related to the sampling, geometry, seasonality and overpassing time; in this case we performed also this approach since we consider only 2 instruments with similar characteristics in terms of geometry and sampling. No further explanation has been added at this point but we expanded the discussion of the two approaches in Sect. 3.

P5- Table1: typo on the unit of the spectral resolution. The latitude coverage could be added in the table as additional information.

The table was corrected and the information about the latitude coverage for both instruments included.

P6-l10: A short summary of validation results of OMPS and SCIAMACHY should be added here.

We agree with the reviewer. We included at this point of the paper a couple of sentences about the results of SCIAMACHY validation against ozonesondes and IUP-OMPS validation against MLS and sondes.

P7-l15: Figure 1 does not include altitude information.

Yes, it doesn't include altitude information because it just shows the number of available (retrieved) profiles from the two instruments as a function of time, without considering it as a function of altitude.

P8-Figure 2: at 28.3 km in the 40°SS-20°SS latitude range, the SCIAMACHY seasonal cycle looks very different. Can the authors comment on this discrepancy?

We studied more carefully SCIAMACHY seasonal cycle in this region and the main discrepancies are found at $[40^{\circ}S, 30^{\circ}S]$ latitude, where its seasonal cycle is pretty flat after the maximum in February-March, in comparison with MLS seasonal cycle in the same period. We don't know the reason for this difference.

P9-18-9: Equations 1 and 2 should include indices linked to latitude, longitude and altitude.

We included the indexes '(lat, lon, z)' also in equation 1.

P13-16: Sentence starting with 'For the 50-60' SN latitude': please clarify. Why is seasonal variation handled differently in this latitude range?

The usage of heat fluxes in this latitude range was done following Gebhardt et al. 2014. At these latitudes the seasonal cycle is found to have a strong inter-annual variability that can be insufficiently modeled employing the harmonic terms. Eddy heat fluxes are related to the wave forcing influencing in turn the BDC. The explanation was added to the paper.

P13-l14: Several studies are mentioned but only one (Park et al., 2017) is cited.

Yes, we started the sentence with Park et al., avoiding the inconsistency.

P13-l21: The solar cycle is also used as a proxy in MLR regression of total ozone for trend retrieval in various studies including that from Weber et al. (2018). The solar activity has thus an impact on ozone also in the lower stratosphere. This is worth mentioning.

The discussion of this proxy has been extended as requested, including the studies of Soukharev and Hood (2006) and of Maycock et al. (2016), investigating the ozone response to the 11-year solar cycle as a function of altitude and latitude from several satellite data sets.

P17-Fig. 8: Mention for which merged data set are the trends displayed. Trend results should be restricted to the range of validity of the data.

Thanks, we specified that we are using the anomalies data set and plot the trends up to \pm 70° latitude, in agreement with the specified range of validity.

Merging of ozone profiles from SCIAMACHY, OMPS and SAGE II observations to study stratospheric ozone changes

Carlo Arosio¹, Alexei Rozanov¹, Elizaveta Malinina¹, Mark Weber¹, and John P. Burrows¹ ¹Institute of Environmental Physics, University of Bremen, Bremen *Correspondence to:* carloarosio@iup.physik.uni-bremen.de

Abstract. This paper presents vertically and zonally resolved merged ozone time series from limb measurements of the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) and the Ozone Mapping and Profiler Suite (OMPS) Limb Profiler (LP). In addition, we present the merging of the latter two data sets with zonally averaged profiles from the Stratospheric Aerosol and Gas Experiment (SAGE) II. The retrieval of ozone profiles from SCIAMACHY and OMPS

- 5 is performed OMPS-LP is performed using an inversion algorithm developed at the University of Bremen. Within the merging procedure To optimize the merging of these two time series, we use data from the Microwave Limb Sounder (MLS) as a transfer function and we follow two approaches: (1) a standard conventional method involving the calculation of deseasonalized anomalies and (2) a 'plain-debiasing' approach, generally not considered in previous similar studies, which preserves the seasonal cycles of each instrument. We find a good correlation and no significant drifts between the merged and MLS time series.
- 10 Using the merged data set from both approaches, we apply a multivariate regression analysis to study ozone changes over the 2003–2018 period in the 20–50 km vertical range. Exploiting the high-dense horizontal sampling of the instruments, we investigate not only the zonally averaged field, but also the longitudinally resolved long-term ozone variations, finding a remarkable an unexpected and large variability, especially at mid- and high-latitudes, with variations of up to 3–5 % per decade at altitudes around 40 km. Significant positive linear trends of about 2–4 % per decade were identified in the upper stratosphere between
- 15 <u>altitudes of</u> 38 and 45 km at mid-latitudes. This is in agreement with the predicted recovery of upper stratospheric ozone, which is attributed both to the adoption of measures to limit the release of halogen-containing ozone-depleting substances included in the Montreal protocol(Montreal protocol) and to the decrease in stratospheric temperature resulting from the increasing concentration of greenhouse gases. In the tropical stratosphere below 25 km negative but non-significant trends were found. We compare our results with similar previous studies and with short-term trends calculated over the SCIAMACHY period:
- 20 while a general. While generally a good agreement is found, some discrepancies are seen in the tropical mid-stratosphere. Regarding the merging of SAGE II with SCIAMACHY and OMPSOMPS-LP, zonal mean anomalies are taken into consideration and ozone trends after and before before and after 1997 are showncalculated. Negative trends above 30 km are found for the 1985–1997 period, with a peak of -6 % per decade at mid-latitudes, in agreement with previous studies. The increase of ozone concentration in the upper stratosphere is confirmed considering over the 1998–2018 period. Trends in the middle and
- 25 lower tropical stratosphere are found to be non-significant tropical stratosphere at 30–35 km show an interesting behavior: over the 1998–2018 period a negligible trend is found. However between 2004 and 2011 a negative long-term change is detected

followed by a positive change between 2012 and 2018. We attribute this behavior to dynamical changes in the tropical middle stratosphere.

1 Introduction

The continuous monitoring of the stratospheric ozone layer is considered by the scientific community an important and key activity, required to assess the impact of anthropogenic and natural processes (WMO, 2018). Variations of ozone concentration in time at different altitudes and latitudes respond to and are intertwined coupled with several dynamical and chemistry-related processes in the atmosphere.

Two important chemical forcings that have influenced globally the amount and distribution of stratospheric ozone over the last decades are the loadings of the so-called halogen-containing ozone-depleting substances (ODSs), that is halogen source

- 10 gases released by human activities as chlorofluorocarbons (CFCs), and of greenhouse gases (GHGs) (WMO, 2018). The adoption of the Montreal Protocol and its amendments regulated the industrial production of chlorine and bromine compounds: species like the CFCs have been banned during '90 in most countries particular, the London amendment in 1990 called for a complete phase out of CFCs production by the year 2000, leading to a decrease of their concentration in the stratosphere starting from the beginning of the end of the 20th century (WMO, 2014). This decrease is expected to lead to a recovery of the
- 15 ozone layer globally and in particular over the Antarctic region, which is affected by the spring-time ozone hole. On the other hand, the increasing concentration of GHGs such as CO_2 and CH_4 in the troposphere, is causing a cooling of the stratosphere, through radiative transfer feedbacks. This cooling leads to ozone increases due to the reactions R1 and R2 :

$$O + O_2 + M \to O_3 + M \tag{R1}$$

$$20 \quad O + O_3 \rightarrow O_2 + O_2 \tag{R2}$$

which have a strong temperature dependence (Groves et al., 1978; Groves and Tuck, 1979)

(first predicted by Groves et al., 1978; Groves and Tuck, 1979). Cooling the stratosphere results in increased production and slower loss of ozone: a so called super recovery is thus expected (WMO, 2014). Models suggest that the combined effect of decreasing ODSs and increasing GHGs is going to lead to an increase in stratospheric ozone in the current and in the next

25 decades, depending. The magnitude of the recovery depends on the chosen scenario of anthropogenic emissions and on the actual decrease of ODSs (Waugh et al., 2009; Morgenstern et al., 2018).

Another important species determining stratospheric ozone concentration belongs to the NO_x family (NO, NO_2). The increasing tropospheric emissions of N_2O or its longer residence time is causing a rise of NO concentration in the stratosphere and a more efficient ozone destruction via the temperature-dependent NO_x catalytic cycle. N_2O is a long-life-long-lived

30 GHG and it is expected to play a central role in the ozone recovery process over the next decades (Ravishankara et al., 2009)and. According to (Portmann et al., 2012) it is rapidly becoming the most important ODS emitted by human activities (Portmann et al., 2012). In addition, increasing emissions of CH_4 at the surface result in increasing CH_4 in the stratosphere

and thus also of HO_x (H, OH, HO_2). However the overall impact of increasing CH_4 is complex in the stratosphere: the ozone depletion by the HO_x catalytic destruction cycles occurs in the upper stratosphere, whereas the catalytic production of ozone is favoured by increasing HO_x and sufficient NO_x in the lower stratosphere.

Changes in stratospheric dynamics also affect the latitudinal and altitudinal distributions of ozone. In particular, the speed
of the tropical upwelling, i.e. the strength of the upward branch of the Brewer–Dobson circulation (BDC), is directly related to changes in the ozone distribution in the tropical lower and middle stratosphere. An acceleration of the stratospheric mean mass transport has been predicted by several model studies (Garcia and Randel, 2008), but strong inter-annual variations prevents prevent a significant recognition of this trend from observations. From monthly up to decadal time scale, ozone concentration is also influenced by many well known phenomena such as the 11-year solar activity cycle and solar proton events, the Quasi-10 Biennial Oscilation (OBO), El Niño Southern oscillation (ENSO), and volcanic eruptions.

Interactions of all these chemistry- and dynamics-related contributions are therefore expected to result in a complex spatial pattern, depending on altitude, latitude and longitude. Therefore, to study long-term variations of the ozone field, there is a need for long-term consistent time series with a good temporal and spatial coverage of the whole globe.

- Passive satellite instruments are able to provide good continuous global coverage and can be classified as nadir-viewing and limb-viewing (including occultation) sounders (Hassler et al., 2014). For stratospheric studies the limb geometry is the preferred choice: as it provides a relatively high vertical resolution. Several limb techniques have been developed over the last decades: in this paper we use data from retrieved from measurements of limb scattering, limb emission and solar occultation instruments. The first type A limb scattering sensor collects solar light scattered into the field of view of the instrument, the second one whereas a limb emission instrument measures radiance emitted by atmospheric compounds in the infrared (IR)
- 20 and or microwave spectral region, whereas the latter one looks into the <u>Solar occultation sensors observe the</u> solar disk and measures measure radiance attenuated along the ray-path through the atmosphere. The latter technique enables measurements of atmospheric trace gases profiles with a higher precision with respect to the other two but with a sparser spatial sampling, since because the observations are only made at sunset and sunrise. The use of shortwave limb scatter technique was for the first time first successfully exploited by the NASA LORE/SOLSE (Limb Ozone Retrieval Experiment/Shuttle Ozone Limb Sound-
- 25 ing Experiment) instrument launched in 1997. The 1997 (McPeters et al., 2000). Two instruments soon followed: the Optical Spectrograph and Infrared Imager System (OSIRIS), launched in February 2001 (Llewellyn et al., 1997), and the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY), launched in March 2002 (Burrows et al., 1995; Gottwald and Bovensmann, 2010)followed on. At the end of 2011, a few months before the end of SCIAMACHY lifetimeENVISAT (Environmental Satellite) mission, the Ozone Mapping and Profiler Suite (OMPS) instrument was launched
- 30 and it is still operational (Flynn et al., 2014). Stratospheric ozone profile is currently monitored by limb sounders like the aging OSIRIS and the Microwave Limb Sounder (MLS onaboard the Aura satellite (Aura MLS, in the following referred to as MLS). In addition, solar occultation observations are currently done by the Canadian SCISAT (SCIence SATellite) ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer) and MAESTRO (Measurement of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation) instruments, launched in 2004 on board the SCISAT satellite,
- and the Stratospheric Aerosol and Gas Experiment (SAGE) III on the international space station, which was launched in 2017.

The latter mission follows the successful SAGE II and SAGE III Meteor-3M instruments, which performed solar occultation observations from 1984 to 2005 and from 2002 to 2005, respectively.

In order to study the long-term changes in ozone vertical profiles and understand the impact of natural phenomena and anthropogenic activities on atmospheric ozone, single instrument time series are too short; several methodologies to consistently

- 5 merge satellite data sets have been developed in the last years. In Harris et al. (2015), the authors considered several existing merged satellite data sets , merged them over the period 1979–2012 and examined separately the time spans before and after the peak in ODSs concentration at the end of '90s. The authors reported negative trends combined trends from the different data sets and reported negative values in the upper stratosphere of -5 % to -10 % per decade before 1998, and a positive trend after 1998 of 2 % after 1998 at mid-latitudes and 3 % in the tropics. Three different ways to compute uncertainties are also
- 10 presented. They also stress different features visible in each single data set and the difficulty to establish the significance of trends in the latter period, requesting longer observational records, improvements in the consistency of single data sets, and more accurate data merging with uncertainties uncertainty estimates. Steinbrecht et al. (2017) updated this work, using several available merged satellite and ground based data sets and focusing on ground-based data records, and computing an average ozone trend profile focusing on the 2000–2016 trendsperiod. A significant increase of ozone in the upper stratosphere was
- 15 reported, with values of 2–21.6–2.5 %±1.1% (1σ) per decade at mid-latitudes and 1.5%-1.6±0.6% (1σ)% in the tropics. Sofieva et al. (2017) merged measurements from SAGE II with Ozone-cei and OMPS satellite data sets using several other data sets homogenized within the Ozone-CCI (Climate Change Initiative) project including OMPS limb observations. The authors used deseasonalized anomalies of zonal monthly mean time series to study trends over the 1980–2016 period, applying a multilinear regression analysis. Before 1997 strong negative trends in the range from -4 % to -8%±1.5% (2σ) per decade
- 20 were confirmed in the upper stratosphere. After 1997, the authors showed statistically significant trend at upper stratospheric significant trends in the upper stratosphere at mid-latitudes reaching up to 2 % per decade \pm 0.8 % (2 σ) per decade in the northern hemisphere. Ball et al. (2018) applied a method independent from the ozone turnaround point, called dynamic linear method, to compute trends and from several existing merged data sets. The authors analyzed a longer period of time, together with improved merged time series and considered the lower stratospheric column instead of the ozone profile. With these
- 25 adjustments, they showed for the first time some evidence of a negative trend in lower stratospheric ozone below 60° latitude. The authors claimed that the lower stratospheric decrease offsets the observed recovery in the upper stratosphere, leading to an overall decline of the stratospheric ozone column. This analysis has recently been challenged by Chipperfield et al. (2018), who showed that the apparent downward trend in the lower stratosphere (ending in 2017) is a result of longer term variability in atmospheric dynamics. Bourassa et al. (2018) presented an updated trend analysis merging SAGE II with OSIRIS time series
- 30 till 2017, after a pointing drift in OSIRIS data was accounted for OSIRIS data were corrected for a drift in the tangent altitude registration of the instrument. The authors identified positive ozone trends post-1997 of about 1–3 % per decade above 25 km especially at mid-latitudes. In the lower stratosphere negative trends were found at all latitudes with significant values generally below 20 km.

Two other projects dealing with merging of satellite observations of several trace gas species are SWOOSH (Stratospheric 35 Water and OzOne Satellite Homogenized) (Davis et al., 2016) and GOZCARDS (Global OZone Chemistry And Related

trace gas Data records for the Stratosphere) (Froidevaux et al., 2015). The first study brought together satellite limb observations, providing several products such as water vapor and ozone mixing ratio profiles using different griddings on pressure levels starting from 1980. The second created time series of zonal monthly mean values of several trace gas species gases using NASA satellites. The LOTUS (Long-term Ozone Trends and Uncertainties in the Stratosphere, see http://www.sparc-

5 climate.org/activities/ozone-trends/) project is focused on investigating uncertainties in ozone trends, studying robust methods to merge data sets and homogenizing the merging procedures and the trend evaluations.

This paper describes a merged ozone data set created using limb measurements from SCIAMACHY and OMPS. The two data sets were generated at the University of Bremen by applying a retrieval algorithm, which uses the same radiative transfer model and spectroscopic databases and was individually optimized for SCIAMACHY and OMPS. The overarching scientific

- objective was to derive consistent ozone data sets that could be merged with the help of a transfer function; the latter being 10 necessary because of the limited overlap period of the two instruments (2.5 months). The merged data set comprises monthly averaged ozone profiles. One of the highlights of this merged data set, in comparison with those reported in several previous studies, except for SWOOSH, is that it is longitudinally resolved in steps of 5° latitude and 20° longitude. This enables us to investigate long-term ozone changes as a function of altitude, latitude, and longitude over the past 15 years (2003 to
- 2018). In addition, we perform a merging of the two time series also in terms of ozone number density values, without sub-15 tracting the seasonal cycle from each data set. In order to investigate ozone trends over even longer periods, we also include merged our new data sets with sparser ozone profiles retrieved from occultation measurements made by SAGE II. This SAGE-II/SCIAMACHY/OMPS merged data set is limited to zonal monthly mean anomalies. Section 2 of the paper describes the instruments, data sets, and methods to retrieve ozone profiles used in this study. Section 3 introduces the merging of SCIA-
- MACHY and OMPS limb data sets using two approaches. Section 4 reports about the long-term ozone changes, both zonally 20 averaged and longitudinally resolved as derived from the SCIAMACHY/OMPS merged data. Results are discussed and compared with previous studies in Sect. 4. Section 5 introduces the merging of SCIAMACHY and OMPS zonal mean anomalies with SAGE II and discusses long-term ozone trends over the pre- and post-1997 periods.

Instruments and data sets 2

25 The SCIAMACHY instrument was launched in 2002 on board the ENVISAT satellite platform and made scientific measurements from August 2002 until April 2012, when a failure in the platform-to-ground communication occurred. In the limb mode, SCIAMACHY observed the atmosphere in-flight in flight direction and scanned horizontally, covering 960km km across-track in four steps, and vertically every 3.3 km. The instrument had a wide spectral coverage, collecting radiances in 8 channels spanning from 240 to 2380 nm, with a spectral resolution varying from 0.22 to 1.48 nm depending on the channel (for a detailed description of the instrument see Burrows et al., 1995; Gottwald and Bovensmann, 2010).

30

The OMPS instrument was launched at the end of 2011 on board the Suomi-NPP satellite platform (Flynn et al., 2014). The suite is composed of three instruments, only data from the Limb Profiler (LP) is taken into consideration for this workused for this study (in the following referred to as OMPS-LP). The instrument looks backwards with respect to the flight velocity vector. It observes the whole atmospheric range simultaneously without scanningthrough, via three vertical slits, the central one . The central slit is aligned with the satellite ground track and the other two sideways, performing measurements horizontally separated are sideways, so that the instrument performs measurements at three viewing angles, which differ horizontally by 4.25° at the tangent point. The instrument. The sensor collects spectral radiance on a two-dimensional charged-charged-coupled

5 device (CCD) through two apertures and at two integration times, to account for the wide dynamic range of the scattered radiance. The CCD pixels are then sampled to get obtain a single picture of the atmospheric state and interpolated to obtain derive level 1 gridded data (L1G). OMPS-LP has a spectral coverage from 280 to 1000 nm with a spectral resolution increasing from 1 nm in the Ultraviolet ultraviolet (UV) region to 30 nm in the near-IR.

In Table 1 some details of the SCIAMACHY instrument are reported together with information about the OMPS-LP instrument 10 for direct comparison, MLS and SAGE II instruments are reported.

	SCIAMACHY	OMPS-LP	MLS	SAGE II
Data time series	08.2002-04 01.2003-03.2012*	02.2012 03.2012-06.2018*	01.2005-12.2016*	01.1985-08.2005*
Spectral coverage mm-	240-2300 240-2380 nm	280–1000 <u>nm</u>	<u>118 GHz – 2.5 THz</u>	<u>385–1020 nm</u>
Spectral resolution nm	0.2 0.22–1 .4 .48 nm	1–30 <u>nm</u>	**	<u>1–2 nm</u>
Instantaneous field of view [km]	2.6	1.5	1.5-3	0.5
Number of observations per orbit	~ 120	180 (each slit)	~ 120 (day-side)	2_{\sim}
Latitude coverage	<u>83.5° S-83.5° N</u>	81.3° S-81.3° N	<u>81.8° S–81.8° N</u>	$\underbrace{80.0^\circ \text{ S}-80.0^\circ \text{ N}}_{\text{C}}$
Equatorial crossing time	10:00	13:30	13:45	~
Level 2 data version	3.5	2.6	4.2	7.0

Table 1. Main characteristics of SCIAMACHYand, OMPS-LP, MLS and SAGE II instruments.

* used in this paper

** see details in Waters et al. (2006)

In this study we consider-use version 3.5 of SCIAMACHY ozone profile retrieval and OMPS-LP version 2.02.6: both products were created at the University of Bremen using the SCIATRAN software package (v3 for SCIAMACHY and v4 for OMPS-LP) which includes a radiative transfer model and a retrieval algorithm (Rozanov et al., 2014). In particular, v8 L1 SCIAMACHY and v2.5 L1 OMPS-LP data were processed. As discussed above and listed in Table 1, differences in terms of

- 15 spectral coverage and resolution, observation method and radiance collection prevented a direct application of SCIAMACHY's retrieval scheme to OMPS-LP. However for the retrieval of both data sets we used the same spectroscopic databases and the same initialization for atmospheric composition and optical parameters. Both algorithms are based on a Tikhonov regularization scheme and use spectral windows in the UV Hartley-Huggins and in the visible Chappuis ozone bandbands. The SCIAMACHY ozone profile retrieval algorithm exploits the sun-normalized limb radiance measurements for Huggins and Chappuis bands.
- 20 while measurements in the Hartley band are normalized to an upper-altitude tangent height. For OMPS-LP, measurements of the solar spectral irradiance are not directly reported in $\frac{\sqrt{2}}{\sqrt{2}}$.5 L1G data, so we normalize the radiance in all absorption bands using upper-altitude tangent heights. In both cases we also take into account in addition the absorption of NO₂ and O₄, using

the same cross sections but convolved to the respective resolution of the instruments. The weighting functions of the surface albedo reflectance are included in the fit procedure. The presence of a cloud in the instrument field of view is detected following the color index approach (Eichmann et al., 2016). Aerosol extinction profiles are retrieved for OMPS-LP using the methodology described in Rieger et al. (2018), whereas for SCIAMACHY climatological profiles are considered used. SCIAMACHY profiles

- 5 are reported from 8 to 64 km with a vertical sampling of 3.3 km and a vertical resolution of 2.6 km, OMPS-LP profiles span from 12 to 60 km with typical vertical resolution of 3 km and a sampling every 1 km. Only measurements from the central slit of the OMPS-LP instrument are used in this study; data from the lateral slits are planned to be used when the pointing knowledge issues issues related to the tangent altitude registration of the instrument, currently under investigation by NASA (Moy et al., 2017), are solved.
- For more details about the University of Bremen OMPS-LP retrieval algorithm , implementation and validation readers are referred to Arosio et al. (2018); for a description of SCIAMACHY retrieval and the validation of the ozone profiles to Jia et al. (2015). Briefly, in Arosio et al. (2018) it has been shown that the retrieved OMPS-LP profiles averaged on a yearly basis agree with MLS within 5–10 % between 20 and 50 km, while below 20 km discrepancies are larger especially in the tropical upper troposphere and lower stratosphere. Also the validation with ozone sondes showed an agreement within ± 7 % between
- 15 20 and 30 km in five chosen latitude bands, with a larger overestimation of the retrieved profiles in the tropics below 22 km. The validation of SCIAMACHY v3.5 against single ozonesondes stations performed in Jia et al. (2015) showed an agreement between the two data sets within 10 % between 20 and 30 km, with discrepancies in the tropics generally below 5 % above 22 km.

The first study which addressed a possible drift of SCIAMACHY v3.5 with respect to other ozone satellite data sets, is

20 Sofieva et al. (2017). The authors stated that evaluating and inter-comparing the anomalies of the considered instruments, among which SCIAMACHY starting from August 2003, they did not find statistically significant drifts with respect to the median anomaly. Kramarova et al. (2018) reported an estimation of v2.5 NASA OMPS-LP ozone profiles drift with respect to MLS, finding positive values up to 0.5-1.0 % yr⁻¹ above 35 km.

The MLS instrument was launched on board the Aura satellite and started scientific measurements atmospheric observations in July 2004, observing the thermal emission from atmospheric trace gases in the millimeter/sub-millimeter spectral range. It scans the Earth limb 240 times per orbit providing retrievals of day- and nighttime profiles of several gases including ozone. For a detailed description of the MLS instrument readers are referred to Waters et al. (2006). In this paper, the version 4.2

25

of MLS level 2 (L2) data is used as a transfer function in the SCIAMACHY/OMPS-LP merging procedure. Quality flags and recommendations reported in Livesey et al. (2017) are taken into consideration throughout used in the study. Several

30 studies, among which Hubert et al. (2016), Hubert et al. (2016) investigated the stability of MLS ozone data set and found no significant drifts over the entire stratosphere.

SAGE II was launched in October 1984 on board the Earth Radiation Budget Satellite (ERBS) and operated until August 2005. The instrument had a sunphotometer collecting solar radiance attenuated by the atmosphere in seven wavelength ranges using the occultation geometry. For technical reasonstechnique. Due to the occultation viewing geometry, the observations of

35 SAGE II are sparse in comparison to the other limb instruments: it could perform that from limb instruments. It performed

measurements only twice per orbit, resulting in 30 observations per day. The occultation geometry, however, yields a higher signal to noise ratio and the ozone profiles are provided with a vertical resolution of 0.5 km from cloud top to 60 km. For a more detailed overview of the instrument, readers are referred to McCormick (1987). In this study we use version 7.0 of SAGE II L2 data (Damadeo et al., 2013).

5 3 Merging the data sets

When merging different data sets, calibration discrepancies between the instruments as well as eventual drifts and jumps in the time series must be accounted for (Hubert et al., 2016). Since As the overlap period of SCIAMACHY and OMPS missions is only about 2.5 months, i.e. too short for a reliable bias correction, we select a reference satellite data set to be used as an external transfer function. For this purpose, MLS was chosen because of the stability and reliability of its measurements,

- 10 the extensive overlapping period with both instruments, its broad latitude coverage, and its dense sampling. In particular, we consider use daytime MLS data from January 2005 until December 2016, taking only the latitude covered daily 2016. For each day, we take only MLS measurements which are made within the latitude range covered by OMPS-LP, to avoid inconsistency between day and night measurements and SCIAMACHY. The presence of the so-called South Atlantic Anomaly (SAA) is taken into consideration filtered using for MLS and OMPS-LP the SAA flag provided in their respective L2 data and applying
- 15 for SCIAMACHY a rectangular exclusion mask over the [-70°, -20°] latitude and [270°, 360°] longitude range. SCIAMACHY data set covers in this study from January 2003 till March 2012 are included (April 2012, April 2018 is excluded because data for the first 8 days only are available), whereas 2002 data are excluded because of the large discrepancies of SCIAMACHY anomalies with respect to other satellites identified by Sofieva et al. (2017). OMPS-LP data from February-March 2012 until June 2018 are used for merging. All profiles are provided in units of ozone number density on a geometric altitude grid. Volume
- 20 mixing ratio (VMR) ozone profiles from MLS on a pressure grid, are converted to geometric altitude vs. number density using pressures and temperatures collocated pressure information from the ECMWF (European Centre for Medium-Range Weather Forecasts) ERA-Interim database and temperature profiles retrieved by MLS. In Appendix A we show the sensitivity of the MLS average ozone distribution and of the computed ozone trends to a change of the reanalysis database. In particular, we use data from ECMWF ERA-Interim - and MERRA-2 (Modern-Era Retrospective analysis for Research and Applications, version
- 25 2, Gelaro et al. (2017)). The effect on ozone trends of using different reanalysis databases providing pressure information is negligible, within -0.25 and +0.5 % at most of the altitudes.

Different ways to bin the satellite data have been studied in order to find an optimal tradeoff between sufficient sufficiently high spatial and temporal resolution of the merged product and the number of measurements in each bin, for the values to be representative. Two optimal sets of values are identified: a longitudinally resolved product, with monthly mean values on

30 a 5° latitude and 20° longitude grid or and a zonally averaged product with a temporal resolution of 10 days and a latitude resolution of 2.5°. In both cases we find about 100 profiles on average on average 50–100 profiles in each bin. The vertical grid used for the merged profiles has evenly spaced steps of 3.3 km, which corresponds to the typical SCIAMACHY vertical sampling: MLS and OMPS-LP profiles with higher denser vertical sampling are linearly interpolated to this common grid.

In this paper we only describe the analysis of the consider the longitudinally resolved ozone profile product., i.e. monthly averaged profiles every 5° latitude and 20° longitude. In some cases however we don't show the longitudinally resolved results, either for lack of space or because the zonal averages are directly comparable with previous studies. In this case, the average over longitudes is performed on the level 3 data. Figure 1 shows the number of measurements available for SCIAMACHY

5 and OMPS-LP in each altitude and latitude bin as a function of time. These values have to be divided by 18, the number of longitudinal bins, to determine the number of measurements that contribute to each longitudinally resolved monthly mean value. The density of measurements increases in 2012, because OMPS-LP has a higher sampling per orbit than SCIAMACHY, as reported in Table 1.



Figure 1. Number of SCIAMACHY and OMPS-LP observations as a function of time and latitude in each 5°_{2} zonal monthly bin.

- Two approaches are used to merge the SCIAMACHY and OMPS-LP data. In the first one, the so-called 'plain-debiasing'
 approach, the seasonal cycle (SC) of each instrument is kept: one data set is shifted with respect to the other, with the help of the transfer function, to remove the offset between the two. In the second one, the so-called 'anomalies' approach, which is similar to that used by Sofieva et al. (2017), the SC of the each single instrument data set is determined and anomalies are calculated independently for each data setbefore subtracting. Then the offset between SCIAMACHY and OMPS-LP is subtracted using the MLS anomalies as a transfer function. We study the SCs of the three instruments to asses how well they agree and whether
 they need to be subtracted before merging. Figure 2 shows the SCs of the three instruments ozone profiles ozone profile data
- 15 they need to be subtracted before merging. Figure 2 shows the SCs of the three instruments ozone profiles ozone profile data records in number density [molec cm⁻³] at different altitudes and latitudes.

The SCIAMACHY ozone SC is compared to that from MLS profiles for , computing it for both instruments over the period 2005–2011, whereas the period 2005 to 2012, and the OMPS-LP ozone SC is compared to that from MLS profiles for the period 2012 to 2016. 2012–2016. At first glance, there is generally good agreement; however, discrepancies are visible in

20 terms of the biasand the additive bias, multiplicative bias (different amplitude of SC) and shape of the seasonal cycle SC between the instruments in some cases. Through the merging process biases will be subtracted additive biases are subtracted via the 'plain-debiasing' procedure, whereas the discrepancies in the multiplicative bias and the discrepancies related to the



Figure 2. Seasonal cycle (SC) for the three instruments as a function of latitude and altitude, in terms of ozone number density [molec cm⁻³]. MLS SC is plotted for the overlapping period with SCIAMACHY ($\frac{2005-2012}{2005-2011}$) and with OMPS-LP (2012-2016).

<u>different</u> shape of the SC are accounted for when calculating anomalies (subtraction of the SC). Two clear examples for these types of discrepancies are seen in the latitude band $[-40^\circ, -20^\circ]$ at two altitudes (see Fig. 2):

- 1. at 34.8 km the <u>SC SCs</u> of the three instruments show the same shape but different absolute values;
- 2. at 28.3 km SCIAMACHY SC has a significantly smaller amplitude with respect to the MLS and OMPS-LP.
- 5 Differences in the amplitudes are caused by the different vertical resolutions sampling of the instruments and by the interpolation procedure we adopted; they are more pronounced at latitudes and altitudes where the transition between semi-annual to annual cycle occurs. In addition, we notice that MLS SC may vary within the instrument life time, as shown the natural variability of the atmosphere plays an important role, with the SC that naturally evolves with time: we notice, for example, that the SC measured by MLS varies between the two considered periods, in particular at 34.8 km in the tropicswith, where a
- 10 change of up to 5–7 % between the two periodsoccurs.

Since the SCs of the three instruments As SCIAMACHY and OMPS-LP have a very similar geometry of observation, a comparable latitude coverage and their SCs do not differ significantly except for few latitudes and altitudes, the first approach

for merging SCIAMACHY and OMPS-LP the two time series consists in a plain debiasing of the two 'plain debiasing' of the data sets with respect to MLS. The bias is defined for each latitude, longitude and altitude as follows:

$$BIAS_{SCIAMACHY}(\underline{lat, lon, z}) = mean(SCIAMACHY_{2005-2012}(\underline{lat, lon, z})) - mean(MLS_{2005-2012}(\underline{lat, lon, z}))$$
(1)
$$BIAS_{OMPS}(\underline{lat, lon, z}) = mean(OMPS_{2012-2016}(\underline{lat, lon, z})) - mean(MLS_{2012-2016}(\underline{lat, lon, z}))$$
(1)

5 In these and following equations, ozone profiles from each instrument are considered as binned monthly averages, interpolated to a common altitude grid. These biases are then applied to the OMPS-LP time series in such a way to conventionally keep the SCIAMACHY mean level as absolute reference as follows:

$$OMPS_{deb}(lat, lon, z) = OMPS(lat, lon, z) - BIAS_{OMPS}(lat, lon, z) + BIAS_{SCIA}(lat, lon, z)$$
(2)

In this way, the any offset between SCIAMACHY and OMPS-LP is accounted for with the help of MLS as a transfer standard. The merging is then done-achieved by concatenating the two data sets, in terms of ozone number density, and computing average values from SCIAMACHY and OMPS-LP over the two months of overlap, i.e. February–March 2012. We exclude all bins where the number of observations is lower than 10 or where the measurements from one of the instruments are not available. Figure 3 shows as a function of latitude for several altitudes relative differences between the merged data set and MLS time series (after the subtraction of its bias with respect to SCIAMACHY) - Relative differences are as a function of

15 latitude for several altitudes.

Relative differences for the 'plain-debiased' merged time series are computed as follows:

$Rel \ Diff(lat, lon, z) = (Merged(lat, lon, z) - MLS(lat, lon, z))/(Merged(lat, lon, z) + MLS(lat, lon, z)) * 200 \tag{3}$

<u>Differences are</u> within ± 10 % between 20 and 50 km and between 50° S and 50° N. Dashed vertical lines indicate the transitions between the two instruments. Over the SCIAMACHY measurement period, a small SC signature is observed,

- 20 especially at 30–35 km at mid-latitudes and at 40–45 km at higher latitudes; these differences are already visible in Fig. 2. In the second half of the time series, less pronounced SC signatures are seen, particularly between 35 and 45 km. Below 20 km the differences increase rapidly showing strong seasonal pattern. Above 50 km, we notice a variation of the relative differences with time, suggesting the presence of drifts with respect to MLS within the time span of each instrument. Caution is therefore required in interpreting the computed trends above 50 km. Furthermore, at At these altitudes diurnal variation of ozone have
- 25 to be accounted for(Sakazaki et al., 2013), which, as showed by Sakazaki et al. (2013). This was not done in this study, our study, because the equatorial crossing time of the two instruments is around noon and differs by only 3.5 h: this would lead to a systematic discrepancy in ozone that we estimate to be about 1-2 % at 30-40 km. Furthermore, the expected systematic bias between the two instruments is largely removed by the debiasing procedure, even though not completely, because variations with time of this systematic discrepancy may not be accounted for by a 'plain-debiasing'. In addition, a technical change in the
- 30 L1 processing of OMPS-LP UV data at the beginning of 2014 affects the OMPS-LP UV retrieval and leads to a jump above 50 km between the 2012–2013 period and the last three years of observations. Towards the polar regions, we notice increasing



Figure 3. Relative differences of the debiased merged time series ('plain-debiasing' approach) with respect to MLS as a function of latitude for several altitudes, <u>computed according to Eq. 3</u>. The vertical dashed lines indicate the transition between SCIAMACHY and OMPS-LP data sets. <u>MLS data has been offset to SCIAMACHY before the comparison</u>.

relative differences with respect to MLS, particularly above 40 km and below 25 km. In summary, we recommend the use of the 'plain-debiased' time series only within $\pm 60^{\circ}$ latitudes and the 20–50 km altitude range.

The second approach to merge data follows that from Sofieva et al. (2017) and comprises computing the deseasonalized relative anomalies from each data set and then debiasing them using MLS data. This is a common procedure when merging several data records, in order to account for the different geometry and atmospheric sampling by each sensor. The SC for each month of the year, m, the and the (relative) anomalies, Δ , and the respective uncertainties, $\sigma(t_m)$, are defined as:

5

$$SC_m = \frac{1}{N_m} \sum_{j=1}^{N_m} O_3(t_j)$$

$$O_2(t_j) = SC$$
(4)

$$\Delta(t_m) = \frac{O_3(t_m) - SO_m}{SC_m} \tag{5}$$

$$\underline{\sigma^2(t_m)} = \frac{1}{N_m^2} \sum_{j=1}^{N_m} \sigma_{O_3}^2(t_j)$$
(6)

for SCIAMACHY, OMPS-LP and MLS, where N_m is the number of available monthly mean values $O_3(t_j)$ for the month of the year m in each time series and $\sigma_{O_3}(t_j)$ is the standard error of the mean for each monthly value respectively. The SC is computed for each instrument considering their complete time series. Then, the anomalies $\Delta(t_m)$ of SCIAMACHY and OMPS-LP are debiased using MLS anomalies as a transfer function as described by Eqs.(1) and (2). The merging is performed

5 in the same way as done for the first approach. Figure 4 shows the differences of time series of absolute differences between the merged anomalies with respect to the MLS anomaly time series and MLS anomalies as a function of latitude for several altitudes..., in percentage, computed as follows:

Diff(lat, lon, z) = (Merged(lat, lon, z) - MLS(lat, lon, z)) * 100(7)

The differences are generally within ± 5 % also towards the polar regions between 20 and 50 $\rm km$ for both SCIAMACHY

- 10 and OMPS-LP periods.—, showing a smaller magnitude and a better consistency over the whole time series with respect to Fig. 3. Above 50 km, the presence of a drift within the single data sets is again observed, whereas below the jump observed in Fig. 3 between the first two years of OMPS-LP lifetime and the rest of the time series is strongly reduced. Below 20 km the pattern becomes rather chaotic in this case as well, also due to the fact that low values of ozone number density, especially in the tropics, amplify the relative differences. We recommend the use of this data product within ±70° latitudes and over the
- 15 20–50 km altitude range.

To check the consistency of the SCIAMACHY/OMPS-LP merged data set with respect to MLS, we compute the correlation coefficient and the drift for each latitude-altitude bin with respect to MLS over the period 2005–2016. The drift is computed as the linear change of the differences (either relative Eq. 3 or absolute Eq. 7 for the 'plain-debiased' data set and anomalies respectively) between the merged time series and MLS data, accounting also for seasonal variations as a sum of harmonic

- 20 terms with periods of 6 and 12 months in the fit. Figure 5 shows in panel (a) the Pearson correlation coefficient as a function of altitude and latitude for the zonally averaged merged data set with respect to MLS, for the 'plain-debiased' merged data set (first approach). The correlation coefficient is high being typically above 0.8 between 20 and 50km km and within ±70° latitudes. A very similar result is obtained for the deseasonalized anomalies -(see the Supplements, Fig. S1). Pearson correlation coefficient values are in that case slightly lower because the strong SC removed in the anomalies contributes largely to the correlation.
- 25 Panel (b) of Fig. 5 shows the drift of the merged data set with respect to MLS, in terms of % per decade; dashed areas in this and the following figures indicate non-significant values, using a 95 % confidence level. The drift is positive only in the tropical lower stratosphere and negative above 45 above 40 km towards the polar regions but values are generally non-significant between 20 and 50km km: this means that the three debiased data sets (MLS and debiased SCIAMACHY and OMPS-LP) are consistent with each other over the 11 years of comparison and the long-term ozone changes from the merged data set can be
- 30 computed with high degree of confidence. Very similar results for the drift are obtained using anomalies time series, whose respective plot can be found in the Supplements (Fig. S1). A plot of the longitude-resolved drift values is also shown in the Supplements, Fig. S1S2: we notice in this plot a longitudinal structure: even though the drift is mostly non-significant, negative values are found in the [0°, 80°] longitude band, whereas positive values are detected within [100°, 260°] longitude and close to zero values elsewhere.



Figure 4. Differences of the merged relative anomaly time series with respect to MLS anomalies as a function of latitude at selected altitudes, computed according to Eq. 7. The vertical dashed lines indicate the transition between SCIAMACHY and OMPS-LP data sets. MLS data has been offset to SCIAMACHY before the comparison.

4 Trend analysis

4.1 Multivariate linear regression terms

To study recent long-term ozone variations with the new merged data sets, we have selected the period January 2003–June 2018, consisting of 186 months. We follow a standard approach, applying an unweighted multilinear regression (MLR) model,

2018, consisting of 186 months. We follow a standard approach, applying an unweighted multilinear regression (MLR) model, accounting for several factors affecting ozone variability in the stratosphere. The weighting of each value by using the reciprocal of its corresponding squared standard deviation, i.e. $\sigma^2(t_m)$ in Eq. (6), has been tested but does not affect significantly the results. The autocorrelation of the data set with 1-one month lag is accounted for, assuming the noise, N, to be an autoregressive



Figure 5. Panel (a): Pearson correlation coefficient of the merged debiased data set with respect to MLS time series over 2005–2016. Panel (b): drift of the merged debiased time series with respect to MLS in % per decade, (differences computed according to Eq. 3); dashed areas identify regions where the drift is not statistically significant.

process of the first order (Weatherhead et al., 1998). The following terms are considered in the MLR (Gebhardt et al., 2014):

$$O_{3}(t) = c_{0} + c_{1}t + \sum_{j=1}^{2} \left(c_{2j} sin(\frac{2\pi jt}{12}) + c_{3j} cos(\frac{2\pi jt}{12}) \right) + QBO(t) + Solar(t) + ENSO(t) + N$$

$$or$$

$$O_{3}(t)O_{3}(t) = X\beta X\beta + \underline{N}N$$

$$(8)$$

5 where t is the time in months and c_i are the regression coefficients, contained in the β vector. The t-th row of the X matrix contains the values of the fit terms for the selected tozone time series can be either in terms of number density [molec cm⁻³] or relative anomalies (multiplied by 100) [%]. The trend uncertainty and thus the significance of the linear trend values are computed from the covariance matrix of the regression coefficients; the trend is significant at the 95 % significance level if the following condition is fulfilled(Tiao et al., 1990):

$$10 \quad \left|\frac{c_1}{\sigma_{c_1}}\right| \gtrsim = 2 \tag{9}$$

All trends shown here are expressed in % per decade: the 'plain-debiased' time series are regressed in terms of [molec cm^{-3}] and the obtained trend values are divided by the averaged ozone series in each bin.

The linear term determined from Eq. (8) is the ozone trend at a given altitude, latitude and longitude. The harmonic terms with a period of 6 and 12 months are considered only for the 'plain-debiased' merged data set to approximate the seasonal

15 behavior. For the 50–60° N latitude band, the seasonal variability of ozone below 25 km is approximated by adding using a term containing the eddy heat flux time series instead of harmonic terms. The eddy heat flux is used as a proxy for the strength of the BDC (Weber et al., 2011). Instead of the harmonic terms the 2-Indeed in this latitude band, the strong inter-annual

variability related to the wave forcing might be insufficiently modeled when using harmonic terms only. As a consequences, the two months lagged eddy heat flux at 50 hPa from ERA-Interim is integrated over each year starting from October and used as a fit proxy (Gebhardt et al., 2014).

The <u>Quasi Biennial Oscillation (OBO)</u> OBO is a quasi-periodic variation of the tropical wind direction in the tropical

- 5 stratosphere: easterly and westerly wind regimes propagate downward with a variable period of approximately 28 months at a given altitude level. Even though it is a tropical phenomenon, the effects of this variable wind pattern on ozone are not confined to the tropical region: they extend to mid- and high-latitudes and are associated with the secondary meridional circulation (Baldwin et al., 2001). Several studies (Park et al., 2017) Park et al. (2017) illustrated the effects of the QBO on ozone profiles as a function of altitude with two peaks in the ozone changes found at 20–27 km and at 30–38 km, showing opposite phase
- 10 in the tropics and the in phase in-phase at mid-latitudes. In this study the influence of QBO is accounted for by considering the monthly average of the zonal wind components measured at 10 and 30 hPa by sondes launched at Singapore station (available at http://www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo/index.html) as a fit proxy. This combination of tropical zonal winds is used at all altitudes and latitudes as follows:

$$QBO(t) = c_{4a}QBO_{10_{hPa}}(t) + c_{4b}QBO_{30_{hPa}}(t)$$
⁽¹⁰⁾

- The solar activity has a noticeable impact on ozone especially in the upper stratosphere as a consequence of e.g. the 11 year cycle and associated strong solar proton events. Several studies based on satellite data sets showed the presence of an in-phase solar cyclevariation of 2-4 % in upper stratospheric ozone (e.g. Remsberg and Lingenfelser, 2010). Soukharev and Hood (2006) studied a 25-years period and found statistically significant ozone variation between the maximum of the solar cycle and its minimum in the upper and in the lower stratosphere. The main contribution to the total ozone column response to the
- 20 <u>11-years solar cycle is found to come mainly from altitudes below 25 km</u>. The correlation is found to be positive and without time lag. More recently Maycock et al. (2016) compared the solar-ozone response from several recently updated satellite time series. In particular, they used the updated v7.0 of SAGE II data, finding a reduced variations in ozone in the tropical upper stratosphere (1 %) due to the solar cycle. This is in agreement with their analysis of Solar Backscatter Ultraviolet Instrument (SBUV) v8.6 data set. As a proxy for the solar activity we consider use Mg II index, which is the core-to-wing
- 25 ratio derived from the Mg II doublet that is known to be highly correlated to solar irradiance variability from the UV to the extreme-UV (Snow et al., 2014). The composite Mg II data set we use was derived at the University of Bremen from the Global Ozone Monitoring Experiment (GOME), SCIAMACHY, GOME-2A and GOME-2B data (and available at http://www.iup.unibremen.de/UVSAT/Datasets/mgii). The solar proxy is then-applied at all latitudes and altitudes, given by:

$$Solar(t) = c_5 MgII(t) \tag{11}$$

30 A further dynamical process impacting stratospheric ozone is the ENSO. This ocean-atmosphere coupled oscillation over the tropical eastern Pacific Ocean has been shown to impact the BDC and is responsible for temperature anomalies in the upper troposphere and lower stratosphere, leading to a longitudinally dependent modifications of ozone in this region (Randel et al., 2009). We include the El Nino 3.4 index as a fit proxy for ozone variations in the lower stratosphere, which is based on sea surface temperature anomalies averaged from 5° S–5° N and $170^{\circ}-120^{\circ}$ W. The data time series is available at *http://www.esrl.noaa.gov/psd/gcos_wgsp/Timeseries/Nino34/*. In particular, we considered a proxy based on a combination of El Nino 3.4 index anomalies and its derivative, in order to account for the time lag between the ENSO proxy and its signature in the ozone time series, as follows:

5
$$ENSO(t) = c_6[N_{34} + \frac{dN_{34}}{dt}\Delta(t)]$$
 (12)

where Δ indicates the time lag in months. An iterative procedure is used to assess Δ . Starting from a time lag of $\frac{2}{2}$ two months the MLR is repeated, updating the time lag at each iteration until it approaches a fraction of a month. The final time lag is allowed to vary between 0 and 12 months. If it does not converge within 10 iteration or exceeds this range, the ENSO proxy is not used in the regression. ENSO is taken into consideration only in the tropical regions (20° S–20° N) and up to below 25 km.

10 4.2 Zonally and longitudinally resolved long-term ozone variations

Figure 6 shows long-term ozone changes of zonally averaged ozone as a function of latitude and altitude calculated using the MLR model applied to the two versions of SCIAMACHY/OMPS-LP merged data sets. In panel (a) considering the anomalies data set and in panel (b) following the 'plain-debiasing' approach. The longitudinally resolved trends are reported in the Supplements, Fig. <u>\$2\$3</u>. The general picture in the two panels is very similar, even though similar, noting that trend values in



Figure 6. Zonal mean linear long-term ozone changes over the 2003–2018 period derived from the SCIAMACHY/OMPS-LP merged data sets: panel (a) shows the results using anomalies, panel (b) shows the results the 'plain-debiased' data set. Dashed areas indicate non-significant trends.

15 panel (b) are slightly larger compared to those in panel (a). This fact may be related to the method use to compute trends: in the anomalies strategy, the absolute anomalies are divided by the SC and then directly used to compute trends in % per decade. In the 'plain-debiasing' approach, trends are computed using the time series in number density and then normalized to the average ozone values at each altitude, latitude and longitude, to obtain values in terms of % per decade. Long-term changes are only statistically significant at mid-latitudes in the upper stratosphere. In this region the long term change is about 3–4 % per decade. This increase shows an asymmetry between the two hemispheres, with higher values at northern high-latitudes, also seen in other studies as (Bourassa et al., 2018) such as Bourassa et al. (2018). As discussed in Sect. 1, a recovery of upper

- 5 stratospheric ozone as a consequence of decreasing ODSs and increasing GHG GHGs emissions is expected and in agreement with recent studies (e.g. WMO (2014)WMO (2018)). This is because at these altitudes the production of ozone results from the photolysis of ground molecular oxygen, O_2 (${}^3\sum_{-}^{g}$) and the subsequent three body reaction of ground state oxygen atoms, O(3P), with $O_2({}^1\sum_{-}^{g})$ whereas ozone is lost by temperature-dependent catalytic odd oxygen cycles involving ClO_x , BrO_x , HO_x and NO_x . Above 48 km in the tropics the negative trends appear significant. As discussed in Sect. 3, these values have
- 10 to be taken with cautionas variations in the diurnal cycle are not accounted for. In addition, we tested the robustness of the trends by changing the starting point of the time series. When the time series starts from mid-2003 or beginning of 2004, the negative trend between 45 and 50 km is strongly reduced and is not significant anymore, whereas the positive trends get larger at mid-latitudes (of about 1%). This is a hint that this may be related to some instrumental issues implies that an unresolved instrumental issue at the beginning of the SCIAMACHY data set at these altitudes exists or that the starting point of the
- 15 time series is not optimal to draw conclusions on long-term trends. In comparison with Ball et al. (2018), we do not identify extensively negative trends below 25 km in the tropics: at these altitudes in the [-30°, 30°] latitude range negative trends are detected observed but are generally not significant. As can be seen in the Supplements, Fig. S2S3, only around 18–20 km in some longitude bins a statistically significant ozone decrease is detected. We also performed the merging procedure and the trend computation using absolute instead of relative anomalies: the results, here not presented, show a discrepancies with respect to panel (a) of Fig. 6 within ±1 % at all altitudes and latitudes.
- Gebhardt et al. (2014) applied a MLR analysis to an older version of SCIAMACHY data and found over the 2002–2012 period positive trends in the upper stratosphere at mid-latitudes as well as in the tropics but also a strong negative trend in the tropics between 30 and 38 km up to -10 to -15 % per decadefrom a MLR analysis applied to an older version of SCIAMACHY data. Other studies have shown similar negative ozone changes in this altitude region. Kyrölä et al. (2013) found
- 25 an ozone decrease of -2 to -4 % for 1997–2011 using merged data from SAGE II and GOMOS (Global Ozone Monitoring by Occultation of Stars), Eckert et al. (2014) found similar changes in 2002–2012 using MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) observations and Nedoluha et al. (2015) showed a decrease of -0.06 ppmv yr⁻¹ using HALOE (Halogen Occultation Experiment) and MLS measurements. Stiller et al. (2017) attributed the changes in the stratospheric age of air (AoA) in the 2002–2012 period to the shift of sub-tropical mixing barriers, which also affects the calculation of long-term
- 30 changes of stratospheric trace gases. Recently, Galytska et al. (2019) have studied this altitude range in the tropics using SCIAMACHY observations and TOMCAT a run of the Toulouse Off-line Model of Chemistry And Transport (TOMCAT) chemical transport model (CTM). Model simulations reproduced the observed behavior in the period 2004–2012. They found anti-correlated changes in ozone and NO₂ from both SCIAMACHY observations and CTM calculations. They showed that these chemical changes are dynamically controlled by seasonal variations in the age of air (AoA). AoA and thus in the vertical
- 35 velocity of the BDC. In particular, the CTM showed a slow-down of the vertical transport during autumn months followed by a

speed-up during winter months, causing changes in the residence time of N_2O and as a consequence in NO_2 and ozone profiles. When averaged over the whole year, AoA trends of different signs cancel out, resulting in no significant annual mean change, whereas the responses of N_2O and as a consequence NO_2 and ozone remain, due to a non-linear relation between chemistry and transport (Galytska et al., 2019). This explains the annual mean trends in the SCIAMACHY ozone profiles observed in the tropical middle stratosphere until 2012. The model Model studies for the 2004–2018 period are ongoing.

- Comparing the results presented in the above mentioned studies with Fig. 6 we notice that the negative trends found in the tropics around 35 km are not detected anymoreand it is worth to have a look at the time series of ozone in this region, shown in. To investigate this behavior, ozone time series are displayed in Fig. 7, panel (a).
 - In this plot, the. The debiased time series is are plotted along with the full regression fit and the linear trend term, considering
- 10 both the whole terms, for the entire time series and the 2003–2011 period. At 34.8 km, panel (a), we notice a decline in ozone until 2010–2012 (with a value close to -10 % per decade), whereas after 2012 the ozone amount in this region leveled up returned to values recorded in mid-2000, resulting in nearly no change in ozone. This fact is enhanced by the anomalous QBO event that occurred in 2015–2016 (Newman et al., 2016), which led to higher ozone in the tropical region during 2016 (Tweedy et al., 2017). In addition, the switch between SCIAMACHY and OMPS-LP time series and the interference between the solar
- 15 proxy and the trend-terms may enhance this discontinuity in the long-term changes. We have to notice that in this region MLS the SC, as shown-reported in Fig. 2, shows a particularly strong variation between SCIAMACHY and OMPS-LP periods: as a consequence, we found a strong sensitivity of the merging procedure for anomalies to the period over which MLS SC is computed.

In the lower tropical stratosphere, panel (b) of Fig. 7, the trend over 2003–2018 is also close to zero. However, looking at the period before and after 2011, we can notice that over the SCIAMACHY time a positive trend is present (7 % per decade), which was already reported by Gebhardt et al. (2014). Over the OMPS-LP period the tendency becomes flat or slightly negative, reducing the overall value of the trend.

Focusing on the altitudes where the ozone recovery is identified, <u>panel (a) of</u> Fig. 8 illustrates the latitudinal and longitudinal structure of the long-term ozone changes at $\frac{3841.3}{1.3}$ km, <u>using the anomalies data set within $\pm 70^{\circ}$ latitude</u>. The longitudinal

- 25 variability is remarkablelarge, especially in the extra-tropical regions. For example, at northern mid- and high-latitudes ozone changes peak at above 56 % per decade over Canada but are near zero non-significant and around 1–2 % over Siberia. Above Antarctica the trend is also positive, but a dedicated study focusing on ozone distribution during Antarctic spring is needed to assess the on-going ozone recovery in this region. The longitudinal patterns found in the drift plots and discussed at the end of Sect. 3 do not explain the variability seen in Fig. 8. In order to study the vertical consistency of these longitudinal structures.
- 30 we show in panel (b) of Fig. 8 the cross section of the trends field at 60° N. We notice that the significant positive values found especially between 180° W and 20° W are vertically homogeneous over three grid levels, from 38 till 44 km. At eastern longitudes the values are consistently non-significant over the whole profile. In the Supplements, Fig. S6 shows in panel (b) the same cross section at 60° S, where the longitudinal structure is less pronounced, as can be seen also from Fig. 8 panel (a), but it still displays a good vertical consistency. Panel (a) of the same picture shows the cross section in the tropics, where values
- 35 are mostly non-significant and the longitudinal variability is within 1-2 %.

5



Figure 7. Merged number density [molec cm⁻³] time series of SCIAMACHY/OMPS-LP merged ozone ('plain debiased') from 2003 to 2018 (in black) with MLR fit (in red) and linear trend term superimposed in blue (the dashed lines refer to the SCIAMACHY period until early 2012); in panel (a) at 34.8 km and in panel (b) at 21.7 km in the tropics, i.e. [-5° , 5°] latitude bin.

Kozubek et al. (2015) presented the structure of the BDC as a function of longitude and its impact on the ozone distribution, using reanalysis data. At 10 hPa a two-core structure of opposite meridional winds have been clearly identified by the authors at higher northern mid-latitudes, one centered over the Canadian and the other over the Asian sectors. Investigating trends in meridional wind at these heights, they found significant trends in these region, showing a weakening of the two-core structure

- 5 after the ODS ODSs turn-around point in 1997. These changes in the dynamics of the stratosphere impact the ozone distribution in this region as well. This illustrates the limitations of the zonal mean approach to describe stratospheric dynamics and related ozone trends. Bari et al. (2013), studied the 3D structure of the BDC comparing a general circulation model and MLS observations. The authors found zonal asymmetries in the meridional mass transport, affecting also the ozone and water vapor distribution, particularly in the northern middle winter stratosphere.
- Similar maps showing the longitudinally resolved ozone field at 21 km and at 35 km are reported in the Supplements (Fig. S3 and S4 and S5, respectively). In the lower stratosphere, we find the already described negative trends in the tropics and irregularly positive values at mid- and high-latitudes, in both cases mostly non-significant. This is a possible indication of the speed-up of the BDC, which transports more efficiently ozone towards higher latitudes. At 35 km we recognize a similar



Figure 8. Longitudinally Panel (a) : longitudinally resolved ozone trends at 3841.3 km in % per decade, computed over the 2003–2018 period from the SCIAMACHY/OMPS-LP anomalies merged data. Dashed areas indicate non-significant trends and the gray polygon labeled as SAA indicate the location of the South Atlantic Anomaly. In panel (b), cross section of the longitudinally resolved trends at 60° N.

distributions of the values as at 38–42 km, but with significant positive trends only in the southern hemisphere. At northern mid- and high-latitudes a kind of two-cell structure is found, featuring positive values over Europe and Canada and negative over Russia, even though extensively non-significant.

5 Merging with SAGE II data set

The merging of SCIAMACHY and OMPS-LP data sets with SAGE II occultation observation observations is carried out considering zonal averaged monthly values, gridded every 10° latitude. This approach was followed in order to account for the different geometry and sampling of the three instruments. This enables us to extend the SCIAMACHY/OMPS-LP data

- 5 record back to 1984. The sparseness of SAGE II data prevents longitudinally resolved consideration or a finer latitude grid. The merging approach is based on anomalies: SAGE II, SCIAMACHY, MLS and OMPS-LP data sets are deseasonalised using their own SCs and then the offset with respect to SCIAMACHY is removed as done in Sect. 3. The debiasing of SAGE II time series with respect to SCIAMACHY is done using the overlapping period between August 2002 and August 2005. In the merging procedure we reject altitude–latitude bins in two cases:
- 10 if less then 10 measurements are available in the bin;
 - if the distribution of SAGE II latitudes inside the bin is not representative for the latitude range, i.e. the mean SAGE II latitude and ± its standard deviation do not include the center latitude of the bin.

The same MLR model as discussed in Sect. 3 (without harmonic terms) is applied over four periods and the resulting trends are shown in Fig. 9, between 20 and 48 km and within $\pm 60^{\circ}$ latitudes. In the two-particular, the trends over the two periods

- 15 are computed independently and not as a piece-wise linear trend. In the two upper panels of the figure the periods 1985–1997 and 1998–2018 are considered, assuming that in 1997 ODSs concentration peaked in the stratosphere. In agreement with the results presented for example by Sofieva et al. (2017) and Steinbrecht et al. (2017), we find negative trends above 30 km before 1997, reaching up to -6 % in the upper stratosphere at mid-latitudes. After 1998 the trends become positive and are significant at mid-latitudes above 35 km. We don't see in panel (b) negative trends in the tropics at 35 km. This results from the inclusion
- of the last 18 months of data. When we consider the time series until 2015 or 2016, a significant negative trend of about -2 % per decade is detected in this region. In the two lower panels of Fig. 9 the focus is brought to the SCIAMACHY and OMPS-LP observation periods to see how short-term ozone changes depend on the periods selected in the MLR. In particular, the January 2004–December 2011 and February 2012–June 2018 periods are considered, as shown in panels (c) and (d), covering approximately an integer number of QBO cycles. Results in panel (c) can be compared with the trends reported in Gebhardt
- et al. (2014) and Galytska et al. (2019). Consistent with previous studies, we notice strong negative trends in the tropical middle stratosphere and positive significant trends in the southern lower stratosphere and in the upper stratosphere at northern mid-latitudes. The trends shown in panel (d) over the 2010–2018-2012-2018 period show an opposite picture with respect to panel (c) in the middle and lower stratosphere: positive changes in the tropics around 35 km and negative changes at southern mid-latitudes. Above 35 km extensively significant positive trends are found at all latitudes. These last two panels show how
- 30 that the long-term changes computed over the last 15 years are the result of complex changes in stratospheric dynamics, which occurred over shorter time scales, and the difficulty to disentangle atmospheric variability from long-term trends. To investigate the possible interference between the solar and the trend terms, we calculated long-term ozone changes for the shorter periods (panel c and d) regressing all non-term terms over the longer period 2003–2018 and then performing a linear trend over the

2004–2011 and 2012–2018 periods. The results for the 2004–2011 ozone changes are shown in the Supplements, Fig. S7: the trend pattern is the same in both cases but differences are visible in terms of absolute values, with smaller trends when the non-trend terms are regressed over the longer period (panel b).



Figure 9. Zonal <u>mean</u> trends computed using the merged SAGE II, SCIAMACHY and OMPS-LP data set, in panel (a) over the 1985–1997 period, in panel (b) from 1997 to 2018, in panel (c) over 2002–2012 (SCIAMACHY observation period) and in panel (ed) over 2012–2018 (OMPS-LP observation period). Dashed areas show non-significant trends.

6 Conclusions

- 5 In this paper we described the approach and results of merging SCIAMACHY limb ozone profiles with OMPS-LP measurements. Monthly averaged data have been considered, binned every 5° latitude and 20° longitude, from January 2003 until June 2018. The merging has been achieved using MLS ozone profiles as a transfer function by following two approaches: in the first one ozone number density profiles are directly merged accounting for the bias between the two instruments independently at every latitude, longitude and altitude; in the second the SC of each instrument is firstly first removed and debiased anomalies
- 10 are then merged. The latter approach is a standard procedure followed in many studies when merging several data sets; in this case, since SCIAMACHY and OMPS-LP observe the atmosphere with a very similar sampling and geometry (in terms of scattering angle), we showed that the 'plain-debiasing' approach is also valid, with the advantage of providing a merged time series expressed in terms of ozone number density and preserving the original SC. Comparing the merged time series

with MLSone, we found residual seasonal features using the first approach, preventing reliable results at high-latitudes. In the second approach, the merged and MLS anomalies showed discrepancies within ± 5 % up to including the polar regions. A correlation coefficient above 0.8 with respect to the MLS time series and no significant drift between 20 and 50 km and between -70° and 70° latitude pointed out a good consistency of the merged data set.

- A MLR model has been applied to the merged data set to study long-term changes in the ozone profile, accounting for several factors affecting stratospheric ozone. Zonal mean trends showed a positive recovery of ozone at mid- and high-latitudes above 35 km, with significant positive changes of about 2–3 % per decade from 2013-2003 until early 2018. Negative but non-significant trends were detected found in the lower tropical stratosphere. Exploiting the high-spatial resolution of the data set, we also studied longitudinally resolved ozone changes, finding in the middle and upper stratosphere a remarkable trend
- 10 patternthat are indicative of changes. This is an indication of a possible change in the BDC varying with longitudes at northern mid-latitudes as a function of longitudes in the northern hemisphere. A comparison of our results with ozone long-term zonal trends reported in previous studies showed a general consistency with regard to the apparent ozone recovery in the upper stratosphere. However, a change in the sign of the trends in the tropical region over the last 15 years was detected: the strong decrease around 35 km found for example by Gebhardt et al. (2014) over the SCIAMACHY period has vanished when adding
- 15 nearly five years of data. This is a consequence of several facts, such as the anomalous QBO event in 2015–2016, which led to positive ozone concentration anomalies in the tropics, and a possible change in the stratospheric dynamics with respect to the last decade. The merging Galytska et al. (2019) has recently explained the feature observed over 2004–2011 in terms of a slow down of the BDC during autumn months. At this stage, we hypothesize that the BDC has increased and compensated the previous loss during recent years. Although we have now identified this fluctuation we have not yet unambiguously found its
- 20 dynamical origin. A model study is planned to improve our understanding of the dynamical impact on the ozone trends in the tropical region over the last 15 years and the variations in ozone concentration over the SCIAMACHY and OMPS-LP periods. We showed that the differences in terms of trends using the two merging approaches are generally negligible, even though the merging procedure may affect the trends especially in regions where the SC of the instruments showed significant changes over the considered period, for example around 35. As a consequence, we don't see in our case strong advantages using one of
- 25 the two merging strategies in terms of ozone trend results. Users needs should guide the choice of one of the two merged data sets.

We also studied the impact of MLS conversion from VMR to number density profiles on the computed ozone trends, using pressure profiles from ECMWF ERA-Interim and from MERRA-2 data sets. The results shown in the Appendix, suggested that no significant impact on long-term ozone changes are related to this conversion of the transfer function, with differences

30 within -0.25 and +0.5 % at most altitudes and latitudes. However, the change of the reanalysis database have a non-negligible effect on the MLS profiles themselves, with difference up to 3–5 % above 55 in the tropics. km.

The merging of monthly zonal mean anomalies of SAGE II with SCIAMACHY and OMPS-LP data sets was performed to facilitate the study of zonal trends in particular over the periods 1985–1997 and 1998–2018. We obtained results in agreement with previous studies: decreasing trends up to -6 % per decade at mid-latitudes in the upper stratosphere before the ODSs

35 turnaround point and an upper stratospheric recovery of about 3 % per decade after 1997, as a result of the implementation of

measures agreed in the Montreal Protocol and its Amendments. A model study is planned to improve our understanding of the dynamical impact on the ozone trends in the tropical region over the last 15 years and the variations in ozone concentration over the SCIAMACHY and OMPS-LP periods. amendments.

7 Data availability

5 Our results and data sets are available upon request to the first two authors.

8 Appendix A

We discuss here the impact of using different reanalysis databases to convert MLS ozone profiles from VMR vs. pressure into number density vs. altitude, as requires for the merging of SCIAMACHY and OMPS-LP time series. In particular, we use two reanalysis databases which provide pressure information needed for the conversion: ECMWF ERA-Interim and MERRA-2.

10 In both cases temperature profiles are taken from MLS observations. In Fig. A1 the effects of this different conversion are displayed. In panel (a) we plot the relative differences as a function of latitude and altitude between the zonally averaged MLS ozone profiles over 2016, converted using the two databases. In detail, the quantity MLS_{Bdiff} is shown:

$$MLS_{Rdiff}(lat, alt) = \frac{MLS_{ecmwf}(lat, alt) - MLS_{merra}(lat, alt)}{MLS_{ecmwf}(lat, alt) + MLS_{merra}(lat, alt)} * 200$$
(13)

where MLS_{ecrnwf} and MLS_{merra} stand for the yearly zonally averaged MLS ozone distributions converted into number
density vs. altitude using ERA-Interim and MERRA-2, respectively. Using pressure profiles from ECMWF leads to slightly higher ozone values going towards the upper stratosphere, with a systematic bias up to 3–5 % above 55 km.

We are also interested in the impact that such a conversion has on ozone trends. Indeed, MLS time series converted using the two different reanalysis data sets serves as a transfer function for the merging of the SCIAMACHY and OMPS-LP, so that eventual drifts in one or both reanalysis would lead to some artificial trend. Panel (b) of Fig. A1 shows the differences as a

20 function of altitude and latitude between the 2003-2018 trends computed from the merged 'plain-debiased' SCIAMACHY/OMPS-LP data set, when the MLS time series is converted using the two different reanalysis. As expected the differences in the trends are small but not negligible in the upper stratosphere: values are within -0.25 and +0.5 % at most of the altitudes and latitudes but approach +1 % above 45 km at some latitudes.



Figure A1. Panel (a): relative differences between MLS ozone profiles zonally averaged over 2016 in case the pressure profiles for the conversion from VMR vs. pressure to number density vs altitude are taken from ECMWF or from MERRA-2 reanalysis. Panel (b) effect of using MERRA-2 pressure profiles instead of ECMWF to convert MLS in terms of ozone trends over 2003-2018.

Author contributions. CA provided OMPS-LP data set, performed the merging of the time series, developed the procedure to compute trends and wrote the paper. AR supervised and guided the merging and the computation of the trends, provided SCIAMACHY data set and the tools for the MLR model and reviewed the manuscript. EM provided stratospheric aerosol data for OMPS-LP retrieval and revised the manuscript. MW contributed with the fit proxies for the MLR, with the revision of the manuscript and guided SAGE II data handling. JPB initiated and proposed the research and led the project, he contributed to the data analysis, the establishment of the scientific outcomes and the preparation of the manuscript.

Competing interests. The authors declare that they have no conflict of interest

5

Acknowledgements. This work was partially funded by ESA within the Ozone CCI Ozone-CCI project and was supported by the University and State of Bremen. We would like to acknowledge the NASA team for providing OMPS-LP LIG and Aura MLS data and support. Part of

10 the data processing has been done at the German HLRN (High Performance Computer Center North).

References

20

- Arosio, C., Rozanov, A., Malinina, E., Eichmann, K.-U., von Clarmann, T., and Burrows, J. P.: Retrieval of ozone profiles from OMPS limb scattering observations, Atmospheric Measurement Techniques, 11, 2135–2149, doi:10.5194/amt-11-2135-2018, https: //www.atmos-meas-tech.net/11/2135/2018/, 2018.
- 5 Baldwin, M., Gray, L., Dunkerton, T., Hamilton, K., Haynes, P., Randel, W., Holton, J., Alexander, M., Hirota, I., Horinouchi, T., et al.: The quasi-biennial oscillation, Reviews of Geophysics, 39, 179–229, 2001.
 - Ball, W. T., Alsing, J., Mortlock, D. J., Staehelin, J., Haigh, J. D., Peter, T., Tummon, F., Stübi, R., Stenke, A., Anderson, J., et al.: Evidence for a continuous decline in lower stratospheric ozone offsetting ozone layer recovery, Atmospheric Chemistry and Physics, 18, 1379–1394, 2018.
- 10 Bari, D. D., Gabriel, A., Körnich, H., and Peters, D.: The effect of zonal asymmetries in the Brewer-Dobson circulation on ozone and water vapor distributions in the northern middle atmosphere, Journal of Geophysical Research: Atmospheres, 118, 3447–3466, 2013.
 - Bourassa, A. E., Roth, C. Z., Zawada, D. J., Rieger, L. A., McLinden, C. A., and Degenstein, D. A.: Drift-corrected Odin-OSIRIS ozone product: algorithm and updated stratospheric ozone trends, Atmospheric Measurement Techniques, 11, 489–498, doi:10.5194/amt-11-489-2018, https://www.atmos-meas-tech.net/11/489/2018/, 2018.
- 15 Burrows, J., Hölzle, E., Goede, A., Visser, H., and Fricke, W.: SCIAMACHY—Scanning imaging absorption spectrometer for atmospheric chartography, Acta Astronautica, 35, 445–451, 1995.
 - Chipperfield, M. P., Dhomse, S., Hossaini, R., Feng, W., Santee, M. L., Weber, M., Burrows, J. P., Wild, J. D., Loyola, D., and Coldewey-Egbers, M.: On the Cause of Recent Variations in Lower Stratospheric Ozone, Geophysical Research Letters, 45, 5718–5726, 2018.
 - Damadeo, R., Zawodny, J., Thomason, L., and Iyer, N.: SAGE version 7.0 algorithm: application to SAGE II, Atmospheric Measurement Techniques, 6, 3539–3561, 2013.
- Davis, S. M., Rosenlof, K. H., Hassler, B., Hurst, D. F., Read, W. G., Vömel, H., Selkirk, H., Fujiwara, M., and Damadeo, R.: The Stratospheric Water and Ozone Satellite Homogenized (SWOOSH) database: A long-term database for climate studies, Earth System Science Data, 8, 461, 2016.

Eckert, E., Clarmann, T. v., Kiefer, M., Stiller, G., Lossow, S., Glatthor, N., Degenstein, D., Froidevaux, L., Godin-Beekmann, S., Leblanc,

- 25 T., et al.: Drift-corrected trends and periodic variations in MIPAS IMK/IAA ozone measurements, Atmospheric Chemistry and Physics, 14, 2571–2589, 2014.
 - Eichmann, K.-U., Lelli, L., von Savigny, C., Sembhi, H., and Burrows, J. P.: Global cloud top height retrieval using SCIAMACHY limb spectra: model studies and first results, Atmospheric Measurement Techniques, 9, 793–815, doi:10.5194/amt-9-793-2016, https://www.atmos-meas-tech.net/9/793/2016/, 2016.
- 30 Flynn, L., Long, C., Wu, X., Evans, R., Beck, C., Petropavlovskikh, I., McConville, G., Yu, W., Zhang, Z., Niu, J., et al.: Performance of the ozone mapping and profiler suite (OMPS) products, Journal of Geophysical Research: Atmospheres, 119, 6181–6195, 2014.
- Froidevaux, L., Anderson, J., Wang, H.-J., Fuller, R. A., Schwartz, M. J., Santee, M. L., Livesey, N. J., Pumphrey, H. C., Bernath, P. F., Russell III, J. M., and McCormick, M. P.: Global OZone Chemistry And Related trace gas Data records for the Stratosphere (GOZ-CARDS): methodology and sample results with a focus on HCl, H₂O, and O₃, Atmospheric Chemistry and Physics, 15, 10471–10507, doi:10.5194/acp-15-10471-2015, https://www.atmos-chem-phys.net/15/10471/2015/, 2015.

- Galytska, E., Rozanov, A., Chipperfield, M. P., Dhomse, Weber, M., Arosio, C., Feng, W., and Burrows, J. P.: Dynamically controlled ozone decline in the tropical mid-stratosphere observed by SCIAMACHY, Atmospheric Chemistry and Physics, 19, 767–783, doi:10.5194/acp-19-767-2019, https://www.atmos-chem-phys.net/19/767/2019/, 2019.
- Garcia, R. R. and Randel, W. J.: Acceleration of the Brewer–Dobson circulation due to increases in greenhouse gases, Journal of the Atmospheric Sciences, 65, 2731–2739, 2008.
- Gebhardt, C., Rozanov, A., Hommel, R., Weber, M., Bovensmann, H., Burrows, J., Degenstein, D., Froidevaux, L., and Thompson, A.: Stratospheric ozone trends and variability as seen by SCIAMACHY from 2002 to 2012, Atmospheric Chemistry and Physics, 14, 831– 846, 2014.
- Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A., Darmenov, A., Bosilovich, M. G., Reichle, R.,
- et al.: The modern-era retrospective analysis for research and applications, version 2 (MERRA-2), Journal of Climate, 30, 5419–5454, 2017.
 - Gottwald, M. and Bovensmann, H.: SCIAMACHY-Exploring the changing Earth's Atmosphere, Springer Science & Business Media, 2010. Groves, K. and Tuck, A.: Simultaneous effects of CO2 and chlorofluoromethanes on stratospheric ozone, Nature, 280, 127, 1979. Groves, K., Mattingly, S., and Tuck, A.: Increased atmospheric carbon dioxide and stratospheric ozone, Nature, 273, 711, 1978.
- 15 Harris, N. R. P., Hassler, B., Tummon, F., Bodeker, G. E., Hubert, D., Petropavlovskikh, I., Steinbrecht, W., Anderson, J., Bhartia, P. K., Boone, C. D., Bourassa, A., Davis, S. M., Degenstein, D., Delcloo, A., Frith, S. M., Froidevaux, L., Godin-Beekmann, S., Jones, N., Kurylo, M. J., Kyrölä, E., Laine, M., Leblanc, S. T., Lambert, J.-C., Liley, B., Mahieu, E., Maycock, A., de Mazière, M., Parrish, A., Querel, R., Rosenlof, K. H., Roth, C., Sioris, C., Staehelin, J., Stolarski, R. S., Stübi, R., Tamminen, J., Vigouroux, C., Walker, K. A., Wang, H. J., Wild, J., and Zawodny, J. M.: Past changes in the vertical distribution of ozone – Part 3: Analysis and interpretation of trends,
- 20 Atmospheric Chemistry and Physics, 15, 9965–9982, doi:10.5194/acp-15-9965-2015, https://www.atmos-chem-phys.net/15/9965/2015/, 2015.
 - Hassler, B., Petropavlovskikh, I., Staehelin, J., August, T., Bhartia, P., Clerbaux, C., Degenstein, D., Mazière, M. D., Dinelli, B., Dudhia, A., et al.: Past Changes in the Vertical Distribution of Ozone Part 1: Measurement Techniques, Uncertainties and Availability, 2014.

Hubert, D., Lambert, J.-C., Verhoelst, T., Granville, J., Keppens, A., Baray, J.-L., Cortesi, U., Degenstein, D., Froidevaux, L., Godin-

- 25 Beekmann, S., et al.: Ground-based assessment of the bias and long-term stability of fourteen limb and occultation ozone profile data records, Atmospheric measurement techniques, 9, 2497, 2016.
 - Jia, J., Rozanov, A., Ladstätter-Weißenmayer, A., and Burrows, J.: Global validation of SCIAMACHY limb ozone data (versions 2.9 and 3.0, IUP Bremen) using ozonesonde measurements, Atmospheric Measurement Techniques, 8, 3369–3383, 2015.

Kozubek, M., Krizan, P., and Lastovicka, J.: Northern Hemisphere stratospheric winds in higher midlatitudes: longitudinal distribution and long-term trends, Atmospheric Chemistry and Physics, 15, 2203–2213, doi:10.5194/acp-15-2203-2015, https://www.atmos-chem-phys.

- net/15/2203/2015/, 2015. Kramarova, N. A., Bhartia, P. K., Jaross, G., Moy, L., Xu, P., Chen, Z., DeLand, M., Froidevaux, L., Livesey, N., Degenstein, D., et al.: Validation of ozone profile retrievals derived from the OMPS LP version 2.5 algorithm against correlative satellite measurements, Atmospheric
- Measurement Techniques, 11, 2837–2861, 2018.

5

30

35 Kyrölä, E., Laine, M., Sofieva, V., Tamminen, J., Päivärinta, S.-M., Tukiainen, S., Zawodny, J., and Thomason, L.: Combined SAGE II– GOMOS ozone profile data set for 1984–2011 and trend analysis of the vertical distribution of ozone, Atmospheric Chemistry and Physics, 13, 10645–10658, 2013.

- Livesey, N., Read, W., Wagner, P., Froidevaux, L., Lambert, A., Manney, G. L., Millan Valle, L. F., Pumphrey, H. C., Santee, M. L., Schwartz, M. J., Wang, S., Fuller, R. A., Jarnot, R. F., Knosp, B. W., and Martinez, E.: Version 4.2x Level 2 data quality and description document, 2017.
- Llewellyn, E., Degenstein, D., McDade, I., Gattinger, R., King, R., Buckingham, R., Richardson, E., Murtagh, D., Evans, W., Solheim, B.,
- 5 et al.: Osiris—An Application of Tomography for Absorbed Emissions in Remote Sensing, in: Applications of Photonic Technology 2, pp. 627–632, Springer, 1997.
 - Maycock, A., Matthes, K., Tegtmeier, S., Thiéblemont, R., and Hood, L.: The representation of solar cycle signals in stratospheric ozone–Part 1: A comparison of recently updated satellite observations, Atmospheric Chemistry and Physics, 16, 10021–10043, 2016.

McCormick, M.: SAGE II: an overview, Advances in Space Research, 7, 219-226, 1987.

- 10 McPeters, R. D., Janz, S. J., Hilsenrath, E., Brown, T. L., Flittner, D. E., and Heath, D. F.: The retrieval of O3 profiles from limb scatter measurements: Results from the Shuttle Ozone Limb Sounding Experiment, Geophysical research letters, 27, 2597–2600, 2000.
 - Morgenstern, O., Stone, K. A., Schofield, R., Akiyoshi, H., Yamashita, Y., Kinnison, D. E., Garcia, R. R., Sudo, K., Plummer, D. A., Scinocca, J., Oman, L. D., Manyin, M. E., Zeng, G., Rozanov, E., Stenke, A., Revell, L. E., Pitari, G., Mancini, E., Di Genova, G., Visioni, D., Dhomse, S. S., and Chipperfield, M. P.: Ozone sensitivity to varying greenhouse gases and ozone-depleting substances in CCMI-1
- 15 simulations, Atmospheric Chemistry and Physics, 18, 1091–1114, doi:10.5194/acp-18-1091-2018, https://www.atmos-chem-phys.net/18/ 1091/2018/, 2018.
 - Moy, L., Bhartia, P. K., Jaross, G., Loughman, R., Kramarova, N., Chen, Z., Taha, G., Chen, G., and Xu, P.: Altitude registration of limbscattered radiation, Atmospheric Measurement Techniques, 10, 167–178, 2017.
- Nedoluha, G., Siskind, D., Lambert, A., and Boone, C.: The decrease in mid-stratospheric tropical ozone since 1991, Atmospheric Chemistry
 and Physics, 15, 4215–4224, 2015.
 - Newman, P., Coy, L., Pawson, S., and Lait, L.: The anomalous change in the QBO in 2015–2016, Geophysical Research Letters, 43, 8791– 8797, 2016.
 - Park, M., Randel, W., Kinnison, D., Bourassa, A., Degenstein, D., Roth, C., McLinden, C., Sioris, C., Livesey, N., and Santee, M.: Variability of Stratospheric Reactive Nitrogen and Ozone Related to the QBO, Journal of Geophysical Research: Atmospheres, 122, 2017.
- 25 Portmann, R., Daniel, J., and Ravishankara, A.: Stratospheric ozone depletion due to nitrous oxide: influences of other gases, Phil. Trans. R. Soc. B, 367, 1256–1264, 2012.
 - Randel, W. J., Garcia, R. R., Calvo, N., and Marsh, D.: ENSO influence on zonal mean temperature and ozone in the tropical lower stratosphere, Geophysical Research Letters, 36, 2009.

Ravishankara, A., Daniel, J. S., and Portmann, R. W.: Nitrous oxide (N2O): the dominant ozone-depleting substance emitted in the 21st

30 century, science, 326, 123–125, 2009.

35

Remsberg, E. and Lingenfelser, G.: Analysis of SAGE II ozone of the middle and upper stratosphere for its response to a decadal-scale forcing, Atmospheric Chemistry and Physics, 10, 11779–11790, 2010.

Rieger, L. A., Malinina, E. P., Rozanov, A. V., Burrows, J. P., Bourassa, A. E., and Degenstein, D. A.: A study of the approaches used to retrieve aerosol extinction, as applied to limb observations made by OSIRIS and SCIAMACHY, Atmospheric Measurement Techniques,

Rozanov, V., Rozanov, A., Kokhanovsky, A., and Burrows, J.: Radiative transfer through terrestrial atmosphere and ocean: software package SCIATRAN, Journal of Quantitative Spectroscopy and Radiative Transfer, 133, 13–71, 2014.

11, 3433–3445, doi:10.5194/amt-11-3433-2018, https://www.atmos-meas-tech.net/11/3433/2018/, 2018.

Sakazaki, T., Fujiwara, M., Mitsuda, C., Imai, K., Manago, N., Naito, Y., Nakamura, T., Akiyoshi, H., Kinnison, D., Sano, T., et al.: Diurnal ozone variations in the stratosphere revealed in observations from the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) on board the International Space Station (ISS), Journal of Geophysical Research: Atmospheres, 118, 2991–3006, 2013.

Snow, M., Weber, M., Machol, J., Viereck, R., and Richard, E.: Comparison of Magnesium II core-to-wing ratio observations during solar minimum 23/24, Journal of Space Weather and Space Climate, 4, A04, 2014.

Sofieva, V. F., Kyrölä, E., Laine, M., Tamminen, J., Degenstein, D., Bourassa, A., Roth, C., Zawada, D., Weber, M., Rozanov, A., et al.: Merged SAGE II, Ozone_cci and OMPS ozone profile dataset and evaluation of ozone trends in the stratosphere, Atmospheric Chemistry and Physics, 17, 12 533–12 552, 2017.

Soukharev, B. and Hood, L.: Solar cycle variation of stratospheric ozone: Multiple regression analysis of long-term satellite data sets and comparisons with models, Journal of Geophysical Research: Atmospheres, 111, 2006.

- Steinbrecht, W., Froidevaux, L., Fuller, R., Wang, R., Anderson, J., Roth, C., Bourassa, A., Degenstein, D., Damadeo, R., Zawodny, J., Frith, S., McPeters, R., Bhartia, P., Wild, J., Long, C., Davis, S., Rosenlof, K., Sofieva, V., Walker, K., Rahpoe, N., Rozanov, A., Weber, M., Laeng, A., von Clarmann, T., Stiller, G., Kramarova, N., Godin-Beekmann, S., Leblanc, T., Querel, R., Swart, D., Boyd, I., Hocke, K., Kämpfer, N., Maillard Barras, E., Moreira, L., Nedoluha, G., Vigouroux, C., Blumenstock, T., Schneider, M., García, O., Jones, N.,
- 15 Mahieu, E., Smale, D., Kotkamp, M., Robinson, J., Petropavlovskikh, I., Harris, N., Hassler, B., Hubert, D., and Tummon, F.: An update on ozone profile trends for the period 2000 to 2016, Atmospheric Chemistry and Physics, 17, 10675–10690, doi:10.5194/acp-17-10675-2017, https://www.atmos-chem-phys.net/17/10675/2017/, 2017.
 - Stiller, G. P., Fierli, F., Ploeger, F., Cagnazzo, C., Funke, B., Haenel, F. J., Reddmann, T., Riese, M., and Clarmann, T. v.: Shift of subtropical transport barriers explains observed hemispheric asymmetry of decadal trends of age of air, Atmospheric Chemistry and Physics, 17, 11, 177, 11, 102, 2017.

20 11 177–11 192, 2017.

25

5

- Tiao, G., Reinsel, G., Xu, D., Pedrick, J., Zhu, X., Miller, A., DeLuisi, J., Mateer, C., and Wuebbles, D.: Effects of autocorrelation and temporal sampling schemes on estimates of trend and spatial correlation, Journal of Geophysical Research: Atmospheres, 95, 20507– 20517, 1990.
- Tweedy, O. V., Kramarova, N. A., Strahan, S. E., Newman, P. A., Coy, L., Randel, W. J., Park, M., Waugh, D. W., and Frith, S. M.: Response of trace gases to the disrupted 2015–2016 quasi-biennial oscillation, Atmospheric Chemistry and Physics, 17, 6813–6823, 2017.
- Waters, J. W., Froidevaux, L., Harwood, R. S., Jarnot, R. F., Pickett, H. M., Read, W. G., Siegel, P. H., Cofield, R. E., Filipiak, M. J., Flower, D. A., et al.: The earth observing system microwave limb sounder (EOS MLS) on the Aura satellite, IEEE Transactions on Geoscience and Remote Sensing, 44, 1075–1092, 2006.

30 spheric ozone recovery, Geophysical Research Letters, 36, 2009.

Weatherhead, E. C., Reinsel, G. C., Tiao, G. C., Meng, X.-L., Choi, D., Cheang, W.-K., Keller, T., DeLuisi, J., Wuebbles, D. J., Kerr, J. B., et al.: Factors affecting the detection of trends: Statistical considerations and applications to environmental data, Journal of Geophysical Research: Atmospheres, 103, 17149–17161, 1998.

Weber, M., Dikty, S., Burrows, J. P., Garny, H., Dameris, M., Kubin, A., Abalichin, J., and Langematz, U.: The Brewer-Dobson circulation
 and total ozone from seasonal to decadal time scales, Atmospheric Chemistry and Physics, 11, 11 221–11 235, 2011.

WMO: Scientific Assessment of Ozone Depletion 2014, Global Ozone Research and Monitoring Project Report 55, World Meterological Organization, https://www.esrl.noaa.gov/csd/assessments/ozone/2014/chapters/2014OzoneAssessment.pdf, 2014.

Waugh, D., Oman, L., Kawa, S., Stolarski, R., Pawson, S., Douglass, A., Newman, P., and Nielsen, J.: Impacts of climate change on strato-

WMO: Scientific Assessment of Ozone Depletion 2018, Global Ozone Research and Monitoring Project Report 58, World Meterological Organization, https://www.esrl.noaa.gov/csd/assessments/ozone/2018/chapters/2018OzoneAssessment.pdf, 2018.