acp-2017-391: Steinbrecht et al. "An update on ozone profile trends for the period 2000 to 2016"

Response to Reviewers

We thank both reviewers for their positive reception of the manuscript, and for the helpful and constructive comments. The revised version of the manuscript addresses all their comments. Our detailed response to each comment follows below.

The reviewers comments are given in normal typeface, our responses are italicized and in red.

The two major points raised by both reviewers were

- 1.) To improve and expand the discussion of differences and similarities between the present study, WMO (2014), and Harris et al. (2015).
- 2.) To clarify how trend uncertainty was determined for the CCMVal-2 multi-model simulations.

To address these major points we have

- 1.) expanded the discussion of similarities and differences between the approach here, in WMO 2014, and Harris et al. 2015 throughout the paper. Additional text and explanation have been added in many parts, e.g. in abstract, introduction and discussion. We have also added two tables pointing out differences between the underlying data sets. Subsectioning of the text has also been increased to more clearly point out these comparisons.
- 2.) changed Figure 1 and added the ±2 standard deviation range of modelled anomalies, which is used to estimate trend uncertainty of the model simulations. We have also added a few explaining sentences to the beginning of Section 4 "Ozone Profile Trends" and the captions of Figures 2 and 5.

In addition to the changes outlined below, which were mostly minor textual changes, there were also a few other small text changes and changes in references. The description of QBO proxies, which in fact include an annual cycle, was also corrected. The original description was not correct.

Responses to Reviewer 1

General comment:

I recommend to discuss in some more detail the (conceptual) differences between Harris et al., 2015 and WMO, 2014 (2014), see below.

This is also suggested by reviewer 2. Consequently, the discussion of similarities and differences between the approach in this paper and in WMO 2014 and Harris et al. 2015 has been expanded throughout. Additional text and explanation have been added in many parts of the manuscript, e.g. in abstract, introduction and discussion. We have also added two tables pointing out differences between the underlying data sets and the way to determine average trend and its uncertainty.

Specific comments:

Abstract:

1. p. 2, line 10/11: I agree that a "detailed attribution of the observed increases to declining ozone depleting substances and to stratospheric cooling" is required: From a formal point of view a suggest to mention this point in the conclusions as well.

Good point, we have added a few sentences to the conclusions. Also suggested by reviewer 2.

Introduction:

2. p. 2, Line 21/22: I don't believe, that the Montreal protocol was (only) signed because of the ozone hole, this is a too strong oversimplification for me: Indeed the ozone hole was very important to enhance public awareness but in the same period the results of the International Ozone Trend Panel Report were elaborated showing first time significant negative trends in northern mid latitudes which was certainly important for the signature (and gradual strengthening) of the Montreal Protocol too.

We have reworded the paragraph to avoid this oversimplified and incorrect impression.

Ozone profile data records:

3. I suggest to clarify whether additional data were used compared to WMO (2014) and for which data sets important revisions were made.

As also suggested by reviewer 2, the discussion of similarities and differences between the data / approach in this paper and WMO 2014 and Harris et al. 2015 has been expanded throughout. A table, additional text and explanation have been added.

4. I think it would be useful to clarify whether (all) satellite merged series used in this paper were used in Harris et al. (2015) and vice versa: I think in this paper SAGE-GOMOS merged series are not used. Is there a particular reason not to use these data ? please explain

Same as point 3 above.

5. Second last para on page 4: I would have preferred to put the sentence "Table 2 summarizes the ground-based stations used in the present study" at the beginning of the para.

OK. Done.

6. p. 4, line 29: I suggest to extend the paragraph staring on p. 4, line 29 about the comparison of Hubert et al., 2016: Which NDACC measurements were used ?

Good point. We have added the information that only ozone sondes and lidars were used.

7. Figure 1, legend: I am not sure, whether Umkehr measurements belong to "NDACC groundbased stations" – Umkehr measurements at least started earlier than NDACC exists.

OK, while Dobson and Brewer instruments are NDACC-associated for total ozone, their Umkehr profiles are not really NDACC. We have reworded the legend.

8. Figure 1: Is there an explanation why upper stratospheric ozone decrease in extra tropics in the first years of the 1980s seems considerably larger in available measurements than in numerical simulations ?

As far as we know, there is no accepted clear-cut explanation. A number of factors can play a role:

a.) The SAGE I data (before 1982) have very sparse sampling, and have an altitude shift that is not very well known, and might be not well corrected for.

b.) the Umkehr data are also very sparse and have poor sampling. So the only "remaining" record is Nimbus 7 SBUV, which may or may not have a drift, and which may or may not be matched well to the later SBUVs. Another factor is the solar-cycle which is not simulated by all models, and not necessarily modeled well in the remaining models.

With the model envelope now given in the revised Figure 1, the difference in the earlier years is not as striking / clear cut anymore.

Because the early data and the trends before 1997 are not really at the focus of this study, we decided to not add discussion of this feature. As indicated above, we feel such a discussion would be inconclusive and might be more confusing than helpful.

9. Figure 1: what's the reason for missing data in the black curve 1983-1985 in the upper panel (northern midatitude) ? are the Umkehr data missing ?

In the post El-Chichon and Pinatubo years (1982, 1983, 1991 to 1993) the Umkehr data were heavily contaminated by volcanic aerosol and SBUV-NASA data are not available for 1992. Therefore data from these years were not used. We have added a corresponding sentence to the caption of Fig. 1.

10. p. 5, line 7 ff: The reference Eyring et al., 2010 seems rather old. Are no more recent publications available ? The data after 2010 are predictions in Eyring et al., 2010 Ozone profile trends:

Eyring et al. 2010 is still the most comprehensive / relevant reference for the CCMVal2 simulations. The more recent CMIP5 / ACCMIP interactive ozone simulations are only becoming available now, and were not available for the manuscript. For the upper stratosphere region of interest here changes between CCMVal1 and CCMVal2 were minimal, and no significant changes are expected from CMIP5 / ACCMIP. No change in the reference. Technically, the CCMVal2 data after 2010 are "predictions". However, we feel that it is not necessary to introduce this technical term here in the text, and feel that describing them as "simulations" is adequate. No change to text.

11. Fig. 2: How is the significance levels determined for numerical simulations ? Is this (directly) comparable with significant trends in measurements ? Please explain

Trend uncertainty for the simulations is determined using the standard deviation of the simulated anomalies (=uncertainty of each data point). For the observations it is determined from the fit residuals. While not directly comparable, both approaches give similar results, because standard deviation of simulated anomalies and fit residuals are of comparable magnitude. We have added a few sentences to explain this to the discussion in the first paragraph of Section 4 "Ozone Profile Trends". Responding to similar questions by reviewer 2, we have also changed Figure 1 and added the ± 2 standard deviation range of modelled anomalies, which essentially gives the uncertainty of the model simulated trends.

From individual data sets trends to the average trend:

12. p. 7., line 22 ff: The use of weighting with inverse squared uncertainty might be viewed as scientifically arbitrary. I believe, that weighting with inverse squared uncertainty of the individual data series tends to increase magnitude of trends of the ensemble. Please comment

We do not agree with these statements of the reviewer, and we have not changed the text here. Our reasoning is as follows:

a.) Weighted mean was used in WMO 2014 and Harris et al. 2015. We cannot change that, and there is no need to criticize that here. In the current paper, a weighted mean is not used anymore, as is discussed in the same paragraph.

b.) Weighting with inverse squared uncertainty is, in fact, standard procedure and makes perfect sense in many situations:

Assume that n1 and n2 samples are taken from the same random distribution with mean X and standard deviation σ . We can then expect the mean of samples n1 to be X1 with uncertainty of the mean dX1= σ /n1, and the mean of samples n2 to be X2 with uncertainty of that mean dX2= σ /n2. Using both samples together as one will give the mean X3 = (n1*X1+n2*X2)/(n1+n2) with uncertainty of that mean dX3= σ /(n1+n2).

This is precisely what is achieved by weighting X1 and X2 with their inverse squared uncertainties $n1/\sigma^2$ and $n2/\sigma^2$ (σ^2 falls out in the equation for X3). Gaussian error propagation $(dX3^2=(\partial X3/\partial X1)^2dX1^2+(\partial X3/\partial X2)^2dX2^2$; independent samples!) also gives $dX3^2 = (n1/(n1+n2))^2 \sigma^2/n1 + (n2/(n1+n2))^2 \sigma^2/n2 = \sigma^2/(n1+n2)$, or $dX3=\sigma/(n1+n2) - exactly$ what is needed. Therefore, weighting by inverse squared uncertainty makes good sense. In particular it makes sure that more uncertain data points / samples carry less weight.

Why would weighting by inverse squared uncertainty increase the magnitude of the average trend? That would only happen, if larger trends were associated with larger weights, in this case smaller uncertainty. There is no reason why this would be the case here.

We, therefore, disagree with these statements of the reviewer, and we have not changed our text.

13. I suggest to extend the first para of this section, I think this discussion is important for the community

We assume that the referee means the first paragraph of Section 4.2 "from individual trends to average trend". As suggested by reviewer #2, we have modified the manuscript in several places, to give a more coherent discussion of the trends obtained here versus the trends in WMO 2014 and Harris et al. 2015. These text changes and the new additional table, in our opinion, also address this comment.

14. Fig. 5: I am wondering whether it is justified to show the uncertainty of Harris et al., 2015 (yellow shading). The main result of this study seems to me that the uncertainty of Harris et al, 2015 no longer corresponds to the present knowledge which is shown in Table 4

While we agree that the large uncertainty reported in Harris et al. 2015 is probably outdated, we still feel that it makes sense to show it, and to compare it with the new results. One of the results of our study is that the uncertainty of Harris et al. 2015 are conservative and probably too large! Therefore, to us, it makes perfect sense to show these uncertainties in the Figure. No changes.

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Responses to Reviewer 2

General Comment:

... The paper is generally well-written and the analysis is robust. There needs to be, however, a clearer, more focused discussion of the relationship between this paper and Harris et al. [2015] as a thread that one can follow throughout the paper. Instead, I found myself having to re-read Harris et al. to understand the primary difference between their analysis and WMO2014, and then having to read between the lines here to understand whether this was an apples-to apples type update to the Harris et al. analysis or a simple extension of the WMO2014 analysis. The issue is easily fixed by being more clear about the differences between WMO2014 and Harris et al. (2015) early in the paper and by guiding the reader to understand that the analysis here uses the same uncertainty assessment (based on the J-distribution) that led to the larger uncertainties in Harris et al. (2015) relative to WMO2014....

We thank the reviewer for this overall very positive, helpful and detailed review. As already mentioned in the reply to reviewer 1, the discussion of similarities and differences between this paper, WMO 2014 has Harris et al. 2015 has been expanded throughout the revised manuscript. We have also added two tables to clarify this.

Specific Comments: Page 2 Line 2: It would be clearer to say "ground-based data [collected or measured] by four techniques....

OK, we have added "measured".

Lines 6-8: This sentence should be rearranged to clarify that "more years of observations and updated data sets" refers to a comparison to WMO 2014 and Harris et al. 2015. (Suggested: "This study confirms positive trends already reported in. . . using three to four more years. . .."

OK, this has been reworded.

Lines 8-10: Here it would be helpful if the authors were specific regarding the reduction in uncertainty relative to Harris et al. that is the result of the improved datasets, which have a lower inferred drift.

These are all good points, and we have reworded the abstract accordingly.

Line 29: It seems to me that a brief mention of confounding factors is warranted here. We do indeed expect ozone to increase, but it is important to be clear that variability and trends in the circulation, temperature changes, etc, can easily mask those increases and lead to large uncertainties on calculated trends even for ideal data records.

OK. Masking / confounding factors are now mentioned, with references.

Page 3

Lines 4-6: A more in-depth discussion of the differences between Harris et al. and Hubert et al. and the WMO 2014 results is needed here. What drove the larger uncertainties in those studies

relative to WMO? Are the differences in uncertainties something that can be at least partially addressed by longer and / or improved records?

Good point. We have added discussion and have expanded the paragraph.

Section 2: The last paragraph of the introduction mentions "improved and additional datasets". A brief summary of which datasets have been improved or added to the analysis since WMO 2014 would be beneficial in Section 2.

Done. See our response to the first general comment of both reviewers.

Line 13: For people with less familiarity with these stratospheric trend studies, it might not be clear why there is a focus on records starting before 1990 given that your analysis looks at the 2000-2016 period.

Good point. We have added an explaining sentence at the beginning, have reworded the paragraph slightly, and have added some references.

Lines 23-26: This is confusing if one is unaware that there were two SAGE instruments.

Added a few words for clarification.

Line 29: Improved in what way?

By correcting for drift in instrument pointing. Explanation has been added.

Page 4

Lines 1-3: A brief mention of the different assumptions used in SWOOSH and GOZCARDS would be helpful to a reader trying to understand how independent these datasets are.

SWOOSH and GOZCARDS ozone are largely constructed from the same data and in the same way, using the same assumptions. Explaining sentences have been added, and the text has been corrected to reflect the new versions of GOZCARDS (no use of ACE-FTS anymore) and SWOOSH that are actually used.

Lines 17-28: The role of the ground-based data in this study is not quite clear to me. As far as I understand, they are not used in the average trend analysis, but they are also not used to quantitatively evaluate individual satellite records. It would be helpful if the authors could provide the rationale for their inclusion.

Ground-based data provide an independent verification of the satellite results. We think they are important and we have added a short explanation at the beginning of the paragraph (and a reference), and also later in the discussion of the trend profiles.

Lines 25-27: Is the fact that they are coherent over a wide range of altitudes and latitudes based on high vertical resolution profiles from satellites? Or on models? A reference would be useful here, since Harris et al. (2015) did not use ground-based profiles because of concerns about representativeness and the "coherency over latitude" would seem to negate that issue.

This is a good point. Since the statement is not really important, since we did not find a good reference and to avoid unnecessary discussion, we decided to simply remove the sentence.

Lines 29-35 and Page 5, lines 1-3: These drifts, while not statistically significant, are of the same order of magnitude, if not larger than, the trends reported here. It is clear from the abstract that a more detailed analysis of these drifts is being undertaken by LOTUS, but a somewhat deeper discussion of their relevance to results presented here is needed, particularly given that the differences between Harris et al. and these results seems to stem in part from lower drift in the records used here.

This point has been addressed by adding some discussion, here and in other places, and by adding a table comparing the different data sets use.

Page 5

Lines 5-6: What is the rationale for using the 1998-2008 climatology for normalization?

Added text to explain the rationale.

Lines 14-16: It seems this point could be made more clearly by referring to sparser spatial and temporal sampling rather than "sparser sampling" meaning temporal and "geophysical differences" referring to the spatial sampling.

OK, reworded.

Figure 1: Presumably the grey line in Figure 1 refers the multi-model mean of the CCMVal2 models? This point should be clear in the caption and in the text in Section 2, and the authors should consider providing the full envelope of the models, as the range is fairly large.

Added information to caption and text. We have also changed Figure 1 and are now plotting the envelope of the multi-model simulated anomalies.

Line 27: Is there something missing here between "solar cycle" and "Reisel"?

Thanks. Fixed.

Section 3: It is clear from Figure 1 that there are data gaps in at least some of the ground-based records. How are these handled in the trend analysis?

Only the available data can be used in the trend analysis. There was no special treatment for data gaps. Data gaps will, however, add some variability to the trend results. Since we did not do anything specific, and to avoided confusing details, we have not changed the text. However, the caption of Fig. 1 now mentions the gaps in some data records.

Page 6

Lines 18-25: I found this explanation confusing. It is unclear to me from this description how the first 2 regression terms are used. This seems to imply that only the last 4 terms are used and then a linear trend for 2000-2016 is fitted to the remaining residuals – if so then why are the first 2 regression terms included at all?

Thanks for this important comment. The old text is indeed unclear and confusing. It has been reworded.

Lines 20-23: On what years of data was the initial regression step performed?

All years of the entire time series. Added text to clarify.

Page 7

Lines 2-3: The authors might want to refer to the Tegtmeier et al. paper on the SPARC Data Initiative ozone climatologies here.

Thanks. Done.

Figure 3: What uncertainty was used for the CCMVal 2 results? Is it based on the model range for all of the models or just on the ensemble mean? Please specify.

See response to comment #11 by reviewer 1, and also to our response to reviewer 2's comment about Figure 1 above. Explaining sentences have been added to text.

Page 8

Line 3: Why was SBUV only used above 40 hPa?

The lowest SBUV layers have very wide averaging kernel and include stratospheric and tropospheric contributions. True stratospheric profile information is only available for layers with layer centers above 40 hPa. Short explanation has been added.

Figure 5: The caption needs several clarifications. It states that "uncertainty bars and yellow shading" give the +/- 2 x sigma values for all individual trends and seems to refer to the datasets used here (though it is unclear how both the uncertainty bars and yellow shading show the uncertainties for a single dataset), but then states that the yellow lines and shading show results from Harris et al. The WMO trend is apparently shown, but the color is not specified – is it the blue line? Finally, the clarification is again required for the model simulations – is this the ensemble mean? How are the uncertainties derived? I think perhaps things could be clarified if the sentence about uncertainty bars and shading were moved later in the paragraph.

Thanks. The caption has been reorganized, and missing information has been added.

Lines 20-21: Strictly speaking, a drift analysis requires comparison to independent datasets. It is unclear whether the authors are saying here that such an analysis has been performed and that the drift in the upper stratosphere has been determined to be 1-2% rather than the 6% used in Harris et al., or whether they are simply relying on the J-distribution analysis to argue for a small drift estimate. It is also unclear how this estimate relates to the estimates provide in Section 2, bottom of page 4, which describe drift estimates of 2-5% for the individual satellite records that make up the merged datasets.

We agree. There was no real drift analysis. However, the spread of the trends in Fig. 4, and the (joint distribution) trend uncertainty in Fig. 5 indicate that some of the larger drift estimates of the past (e.g. 6% per decade) no longer apply. Text has been reworded (as has been the text in Section 2, bottom of page 4).

Page 9

Lines 16-21: For completeness, a brief discussion of the attribution of trends should be provided here.

This point is also mentioned by reviewer #1 (his point 1.). It has been addressed by adding a few sentences.

An update on ozone profile trends for the period 2000 to 2016

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Abstract. Ozone profile trends over the period 2000 to 2016 from several merged satellite ozone data sets and from groundbased data measured by four techniques at stations of the Network for the Detection of Atmospheric Composition Change indicate significant ozone increases in the upper stratosphere, between 35 and 48 km altitude (5 and 1 hPa). Near 2 hPa (42 km), ozone has been increasing by about 1.5% per decade in the tropics (20°S to 20°N), and by 2 to 2.5% per decade

- in the 35° to 60° latitude bands of both hemispheres. At levels below 35 km (5 hPa), 2000 to 2016 ozone trends are smaller 5 and not statistically significant. The observed trend profiles are consistent with expectations from chemistry climate model simulations. Using three to four more years of observations and updated data sets, this This study confirms positive trends of upper stratospheric ozone already reported, e.g., in the WMO/UNEP Ozone Assessment 2014, or by Harris et al. (2015). The additional years, and Compared to those studies, three to four additional years of observations, updated and improved data
- sets with reduced drift, and the fact that nearly all individual data sets indicate these increases, ozone increase in the upper 10 stratosphere, all give enhanced confidence. Uncertainties have been reduced, for example for the trend near 2 hPa in the 35° to 60° latitude bands from about $\pm 5\%$ (2 σ) in Harris et al. (2015) to less than $\pm 2\%$ (2 σ). Nevertheless, a thorough analysis of possible drifts and differences between various data sources is still required, as is a detailed attribution of the observed increases to declining ozone depleting substances and to stratospheric cooling. Ongoing quality observations from multiple
- independent platforms are key for verifying that recovery of the ozone layer continues as expected. 15

1 Introduction

Depletion of the stratospheric ozone layer by anthropogenic chlorine and bromine from ozone depleting substances (ODS) has been a world-wide concern since the 1970s (Stolarski and Cicerone, 1974; Molina and Rowland, 1974). Initially, studies predicted the largest ozone losses for the upper stratosphere, at about 42 km or 2 hPa (Crutzen, 1974). For the total column

- of ozone only moderate losses were predicted. The Public perception of the situation changed dramatically with the discovery 20 of the Antarctic ozone hole (Chubachi, 1984; Farman et al., 1985). The ozone hole is characterized by large ozone depletion throughout the lower stratosphere, which is due to heterogeneous reactions on the surface of Polar Stratospheric Clouds (Solomon, 1999). These important reactions had not been known or and not been included in the early predictions. Thus, the The large spring-time ozone losses over an entire continent were a huge surprise. The world's nations reacted to mounting
- 25 evidence that ODS were harming the vital ozone layer, first by signing the International Vienna Convention for the Protection of the Ozone layer (Layer in 1985), and, then by implementing the 1987 Montreal Protocol and its later amendments. Thanks to these agreements, the world-wide production and consumption of ODS have been eliminated almost completely since the early 1990s (WMO, 2007).

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The Montreal Protocol has been very successful. The concentration of ODS in the atmosphere has been declining since the mid-1990s in the troposphere, and since the late 1990s also in the stratosphere (WMO, 2011). Scientific assessments of the state of the ozone layer have shown that the ozone layer is responding: The decline of ozone in the upper stratosphere stopped around 2000 (Newchurch et al., 2003; WMO, 2007). Total ozone columns have also stabilized (WMO, 2011, 2014). Given the current slow decline of ODS concentrations, we now expect ozone to increase accordingly in the stratosphere. However, this small increase is not easily separated from concurrent variability and changes in temperature, atmospheric circulation, and solar ultraviolet flux (Jonsson et al., 2004; Reinsel et al., 2005; WMO, 2007, 2011).

- 5 The last WMO/UNEP ozone assessment (WMO, 2014), therefore, concluded that statistically significant increases of ozone had been observed only in the upper stratosphere (around 42 km or 2 hPa), but not at lower levels, and not for total ozone columns. About half of the increase in the upper stratosphere was attributed to declining ODS, the other half to declining temperature. This stratospheric cooling is caused by increasing CO₂ (Jonsson et al., 2004; Randel et al., 2016). Low temperature enhances ozone in the upper stratosphere, by slowing gas-phase destruction cycles and making ozone production more efficient.
- Studies published after WMO (2014) have confirmed this the tendency of ozone increasing in the upper stratosphere, but they also pointed out that instrument drifts and drift uncertainties might be larger and upward trends than the 1 to 2% per decade assumed in WMO (2014). Hubert et al. (2016), for example, reported drifts and drift uncertainties between satellite and ground-based data exceeding 5% per decade for some instruments, and less than 2% per decade only for a few instruments. Harris et al. (2015) found larger differences between trends from some data sets, exceeding 6% per decade. Based on these
- 15 larger differences, and the larger drift uncertainty estimates (Hubert et al., 2016), Harris et al. (2015) concluded that upward trends of upper stratospheric ozone might not be statistically different from zero(Harris et al., 2015; Hubert et al., 2016).

The purpose of the present paper is to follow up on WMO (2014), using the same methodology, these studies, but with three to four more years of data, and with improved and additional data sets. Here we present initial results. For the future, a A more comprehensive investigation of instrumental and merging uncertainties, and of uncertainties for different regression analyses

20 is planned-under way in the "Long-term Ozone Trends and Uncertainties in the Stratosphere" initiative (LOTUS), which has been started by an activity of the Stratosphere-troposphere Processes And their Role in Climate project (SPARC) of the World Climate Research Programme (WCRP), see http://www.sparc-climate.org/activities/ozone-trends/.

2 Ozone Profile Data Records

Only a few-The determination of ozone trends requires homogeneous data records that extend over several decades, because
 not only ozone variations associated with the quasi-biennial oscillation must be quantified well, but also the slow variations associated with the 11-year solar cycle (Newchurch et al., 2003; Steinbrecht et al., 2004). Available long-term records of ozone profile data start before 1990 and extend past 2014 to the present (see also Tegtmeier et al., 2013; Hassler et al., 2014; Tummon et al., 2015). Tables 1 and 2 summarize the merged satellite records and ground-based stations used in the present study.

2.1 Data Sources

30 The nadir viewing Solar Backscatter UltraViolet (SBUV) instruments on NASA and NOAA satellites have measured ozone profiles continuously since late 1978, covering the sunlit part of the globe, but with only coarse altitude resolution of 10 to 15 kilometers (McPeters et al., 2013). Orbit drifts, differences between individual instruments, instrument degradation, and some other problems require careful assessment, when generating a long-term data set from these measurements. Currently two SBUV based data sets (Version 8.60) are available: The merged SBUV MOD (release 6) ozone data set generated by NASA (Frith et al., 2014), termed SBUV-NASA in the following, and the "coherent" SBUV data set generated by NOAA (Wild and Long, 2017), termed SBUV-NOAA in the following. The two data sets rely on the same SBUV instruments, but differ in the approach taken for merging their individual records (see also Frith et al., 2017).

Ozone profiles with higher vertical resolution (about 2 km), but also with sparser coverage, were provided by the satelliteborne Stratospheric Aerosol and Gas Experiments (SAGE I and SAGE II) and the Halogen Occultation Experiment (HALOE). These instruments measured in solar occultation geometry from 1979 to about 1982 (SAGE I), from late 1984 to 2005 (SAGE II), and from 1991 to 2005 , respectively (Damadeo et al., 2013, 2014; Remsberg, 2008). (HALOE), see, e.g., Damadeo et al. (2013, 2014)

- 10 , and Remsberg (2008). Since 2002, the Optical Spectrograph and InfraRed Imaging System (OSIRIS) measures ozone profiles from ultraviolet light scattered in limb geometry (McLinden et al., 2012). SAGE II and OSIRIS ozone profiles have been combined by Bourassa et al. (2014) to produce a long-term data set, which has subsequently been improved by correcting for a tangent altitude drift of the OSIRIS instrument (Bourassa et al., 2017). Optionally, this data set also includes ozone profiles from the limb viewing instrument of the Ozone Mapping Profiler Suite (OMPS), which has operated since early 2012 (e.g.,
- 15 Flynn et al., 2014).

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Using microwave emissions in limb geometry, the Microwave Limb Sounder (MLS) on the Aura satellite has been measuring many stratospheric trace gases since 2004, including ozone profiles with dense spatial sampling and a vertical resolution of 2.5 to 3 km in the stratosphere (Waters et al., 2006). SAGE, HALOE, ACE-FTS (= Atmospheric Chemistry Experiment Fourier Transform Spe and MLS ozone profiles have been combined in the Global OZone Chemistry And Related trace gas Data records for the Strato-

- 20 sphere (GOZCARDS, Froidevaux et al., 2015) (GOZCARDS, Froidevaux et al., 2015, newer version 2.20 used here) and in the Stratospheric Water and OzOne Satellite Homogenized data set (SWOOSH, Davis et al., 2016). GOZCARDS (v2.20) and SWOOSH (v2.6) are very similar (compare Fig. 1). Both rely to a large degree on the ozone records from SAGE II (1984 to 2005, version 7) and Aura-MLS (2004 to present, version 4.2). Both adjust ozone values from other satellites to those from SAGE II. GOZCARDS additionally uses SAGE I (version 5.9_rev) to extend the ozone record back to 1979.
- For the period from August 2002 to April 2012, ozone profiles were <u>also</u> measured by the SCIAMACHY (= SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY), GOMOS (= Global Ozone Monitoring by Occultation of Stars) and MIPAS (= Michelson Interferometer for Passive Atmospheric Sounding) instruments on board the European ENVISAT satellite. Positive ozone trends have been reported in the upper stratosphere for each of these instruments (Gebhardt et al., 2014; Kyrölä et al., 2013; Eckert et al., 2014). Unfortunately, ENVISAT failed in April 2012, and measurements ceased. The ESA
- 30 Climate Change Initiative has generated a harmonized ozone profile data set (Sofieva et al., 2013; Rahpoe et al., 2015) from the ENVISAT instruments, the SMR (= Sub-Millimeter Radiometer) microwave instrument, the OSIRIS instrument, and ACE-FTS (Sofieva et al., 2013; Rahpoe et al., 2015)(= Atmospheric Chemistry Experiment Fourier Transform Spectrometer, see Bernath, 2017). This "ESA CCI" or "Ozone CCI" ozone profile record has recently been updated and extended, with SAGE II ozone profiles back to 1984, and with OMPS ozone profiles (2D retrieval from U. Saskatoon) from 2012 to the present (SAGE + CCI +
- 35 OMPS, see Sofieva et al., 2017). Another new merged data set, following previous work by Laeng et al. (2017), combines the

MIPAS (Fischer et al., 2008) ozone profile record (KIT/IMK processing) with the records from SAGE II and OMPS (NASA v2 retrieval). Because of short or lacking overlap periods, this SAGE + MIPAS + OMPS record relies on ACE-FTS as a transfer standard for matching MIPAS high spectral resolution mode data (07/2002 until 03/2004) to MIPAS low spectral resolution mode data (01/2005 to 04/2012), and for matching the latter to OMPS data (after 02/2012).

- 5 While satellites provide near global coverage, the limited lifetimes of most satellite instruments makes the construction of consistent long-term records difficult, as indicated above. Long-term consistency, therefore, might be more easily achieved by ground-based measurements, albeit at the cost of only local coverage. Ground-based instruments have provided some of the longest available records for ozone trend analysis (e.g., Zanis et al., 2006; Nair et al., 2013). Therefore, the ground-based stations in Table 2 are used as an independent source for ozone trends in the present study. The longest ground-based ozone
- 10 profile records for the upper stratosphere come from Dobson spectrometers operated in "Umkehr" mode (Petropavlovskikh et al., 2005, 2011). Umkehr ozone profiles have coarse altitude resolution, about 10 km. Long-term ground-based measurements of ozone in the upper stratosphere are also available from the Network for the Detection of Atmospheric Composition Change (NDACC, http://www.ndacc.org, Kurylo et al., 2016). These measurements started in the late 1980s and 1990s, using differential absorption lidars, microwave radiometers (Steinbrecht et al., 2009), and Fourier transform infrared spectrometers
- (FTIRs, Vigouroux et al., 2015). FTIR ozone profiles have coarse altitude resolution (8 to 15 km) and resolve only 3 layers 15 in the stratosphere. Altitude resolution for the microwave radiometers is also 8 to 15 km. Lidars provide altitude resolution between 1 km (below 30 km) and 10 km (above 45 km). Note, however, that altitude resolution is not that important for the investigation of long-term trends in the upper stratosphere, which are usually coherent over a wide range of altitudes and latitudes (different, however, for changes around the tropopause). Table 2 summarizes the ground-based stations used in the
- 20 present study.

A comprehensive intercomparison of limb-viewing satellite instruments with ground-based NDACC instruments ozone sondes and lidars by Hubert et al. (2016) indicates that SAGE II and Aura-MLS, the primary instruments in many of the merged records, are very stable. If drifts exist, they are smaller than $\pm 2\%$ per decade in the 20 to 40 km region, and not statistically significant. Below 20 km and above 45 km uncertainties become larger, because of larger geophysical variation

- in the compared altitude ranges, and because of increasing measurement errors, see also Tegtmeier et al. (2013). Note that 25 in Hubert et al. (2016), the OSIRIS V5.07 ozone data did exhibit a significant drift, up to 8% per decade near 40 km, also apparent in Harris et al. (2015). This drift has been corrected in the revised and updated OSIRIS V5.10 data set used here (Bourassa et al., 2017). Drifts of most SBUV instruments are less than 3 to 5% per decade, and are not statistically significant (Kramarova et al., 2013). Similarly, Rahpoe et al. (2015) report that drifts of several limb viewing instruments including
- ACE-FTS, MIPAS, and OSIRIS are typically less than 3% per decade (even for older processing versions), and not statistically 30 significant. For the current study, newer data sets with reduced drifts were available (especially OSIRIS), and older data sets with apparent large drifts were not used (SAGE + GOMOS).

Table 3 compares data sets and trend periods used here, in WMO (2014), and in Harris et al. (2015). In addition to using three more years of data, the main difference between the present study and WMO (2014) is the use of four more satellite data sets: SBUV-NOAA, SWOOSH, SAGE + OSIRIS, and SAGE + MIPAS + OMPS. OSIRIS and MIPAS (as well as

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SCIAMACHY, GOMOS, and SMR) were included in the HARMOZ / Ozone_CCI merged data set used in WMO (2014), which is replaced here by the new SAGE + Ozone_CCI + OMPS data set. The most important differences between this study and Harris et al. (2015) are the different trend periods, the use of the new and improved Ozone_CCI, SAGE + OSIRIS (now drift-corrected) data sets, and the omission of the anomalous SAGE + GOMOS data set. The latter two data sets provided quite different trend estimates from each other, and from other data sets (compare Fig. 6 of Harris et al., 2015).

2.2 **Time Series**

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Figure 1 shows annual mean ozone anomalies from the different satellite and ground-based data sets, averaged over three latitude bands, and for a level near 2 hPa or 42 km. Anomalies are relative to the 1998 to 2008 climatology of each individual data set. 1998 to 2008 was chosen as the reference period because ozone values were fairly constant over this period, and

- many instruments provide data for a substantial fraction of the period: SAGE II and HALOE until late 2005, ENVISAT 10 instruments since late 2002, and Aura-MLS since August 2004. All data sets show clear ozone declines until the late 1990s and show and generally increasing ozone over the 2000 to 2016 period, especially at mid-latitudes. This observed evolution generally confirms expectations from model simulations by Chemistry Climate Models within their Validation-2 initiative (CCMVal-2, Eyring et al., 2010, grey lines in Fig. 1). (CCMVal-2, Eyring et al., 2010). Grey lines and shading in Fig. 1 give
- 15 the CCMVal-2 multi-model mean and ± 2 standard deviation envelope, obtained over all models (except outliers) and a 25 month sliding window. The simulations attribute the ozone decline until the late 1990s to increasing ODS loading, and predict positive ozone trends due to declining ODS loading since around 2000. In the simulations, the ozone increase is enhanced by overall cooling of the stratosphere due to increasing greenhouse gases over the entire 1978 to 2016 period (see also Jonsson et al., 2004; Randel et al., 2016).

All observational data sets show similar fluctuations from year to year, usually within 1 or 2% of each other. They also indi-20 cate similar long-term tendencies, usually within $\pm 2\%$ per decade of each other, and comparable to the CCMVal-2 simulations. Generally, the station data show larger variations than the zonal means from the satellite data. This is not surprising, given the sparser temporal sampling of most ground-based data (lidar, Umkehr, and FTIR all require clear sky). Geophysical differences between a station location and the Also, the low density of stations will generally result in more variability compared to the smoother wide-band zonal means are also possible from the satellite records. 25

3 Multiple Linear Regression

Multiple linear regression (MLR) has become a standard method for deriving ozone trends (Bojkov et al., 1990; Reinsel et al., 2002; Newchurch et al., 2003; Chehade et al., 2014). MLR can be applied to monthly mean ozone anomaly time series $dO_3(i)$ of many months *i*. The anomalies are obtained by referencing the monthly mean $O_3(i)$ to the climatological mean for each calendar month $O_{3,Clim}(i \mod 12)$.

$$dO_3(i) = \frac{O_3(i) - O_{3,Clim}(i \mod 12)}{O_{3,Clim}(i \mod 12)} \tag{1}$$

MLR attempts to reconstruct the observed anomalies as a linear combination of prescribed predictors $P_j(i)$, which account 5 for known ozone variations, and residual noise $\epsilon(i)$.

$$dO_3(i) = c_0 + \sum_{j=1}^n c_j * P_j(i) + \epsilon(i)$$
(2)

Here our set of predictors P_j includes a linear trend, a change of the trend in January 1997 (hockey stick), two, reflecting the increase of ozone depleting substances until the late 1990s, and their decline since), six proxies for the Quasi-Biennial Oscillation (QBO), equatorial zonal winds at 30 and 10 hPa, plus their modulations by sine and cosine with 12 month (=annual)

- 10 period), and a proxy for the 11-year solar cycleReinsel et al. (as in 2002), as in Reinsel et al. (2002). Like WMO (2014) or Harris et al. (2015), the present study also includes a proxy for stratospheric aerosol loading and for El-Nino / La Nina, which is most relevant for the tropical lower stratosphere (e.g., Oman et al., 2013). Table 4 summarizes the proxies used, and their sources. Other studies may include further proxies for weather patterns and meridional ozone transports, such as circulation indices or eddy heat flux (Steinbrecht et al., 2001; Reinsel et al., 2005), but this was not done here. The coefficients c_j are
- obtained by least squares fitting of the residuals, i.e. minimization of $\sum_i \epsilon^2(i)$. Typically, the residuals $\epsilon(i)$ are of the order of 1 to 10%, large enough to cover fit errors and measurement errors for each monthly mean.

If realistic uncertainties $\Delta O_3(i)$ are available for each monthly mean, the anomalies can be weighted by their inverse squared uncertainty (high weight for low uncertainty), and the uncertainties $\Delta O_3(i)$ can be used to estimate the uncertainty Δc_j of the fitted c_j . However, in many cases reliable uncertainties are not available for monthly means, because it is difficult to account

- 20 correctly for all error terms, and for autocorrelation and covariance of the individual measurement errors (e.g., Toohey and von Clarmann, 2013; Damadeo et al., 2014). A time-invariant bias, for example, might be included in the monthly mean uncertainty, but it would be irrelevant for the long-term trend. So in many studies, including Reinsel et al. (2005), WMO (2014), Harris et al. (2015), and this study, the pragmatic approach is to use the standard deviation of the fit residuals $\epsilon(i)$ for estimating the uncertainties Δc_i of the fitted coefficients.
- Strictly, the uncertainties from the MLR assume that the predictors are orthogonal, and that the residuals $\epsilon(i)$ are uncorrelated white noise. In practice, the predictors above are orthogonal enough (cross-correlations less than 0.3 for the long periods considered), and first order auto-correlation in the residuals is small ($|AC| \ll 0.3$). Still, to correct for first order auto-correlation AC (Reinsel et al., 2002), the Δc_j are multiplied here by $\sqrt{(1 + AC)/(1 - AC)}$. Neglecting higher orders of autocorrelation might result in slightly underestimated uncertainties (Vyushin et al., 2007).
- 30 One problem with the "hockey stick" fit is that the slope of the declining trend and the time of the turning point have an influence on the slope of the second part of the "hockey stick" (Reinsel et al., 2002). To reduce this problem, a second step

was introduced in WMO (2014), and this approach is also used here. First, Equation 2 is used to estimate fitted for the entire long time series, e.g., from 1978 to 2016. The QBO, solar cycle, aerosol and El Nino effects over the entire record. These latter effects resulting from this first fit are then subtracted from the ozone anomalies , resulting in $dO_3(i)$. This provides time series of ozone residuals $O_{3,res}(i)$ which include the , which have most of the variability associated with QBO, solar cycle, aerosol

- 5 and El Nino removed, but which still contain the long-term trend component, other substantial remaining variability and the $\epsilon(i)$. The use of the entire 30 to 40 year long time-series in the first step is particularly important for a good estimate of the 11-year solar cycle effect, which cannot be estimated well with shorter records. Then, in a second step, a simple linear trend is fitted to the $O_{3,res}(i)$. This trend can be fitted over any desired period, here in this case the period 2000 to 2016, since QBO, solar cycle and ENSO related variations have already been taken care of 2016. The second fit is not constrained as much by the
- 10 by a "hockey stick" assumption, and has more full freedom to react to the remaining ozone variations over the desired period.

4 Ozone Profile Trends

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4.1 Trends for individual data sets

Figures 2 and 3 present the latitude-pressure cross-sections of 2000 to 2016 ozone trends, TR_{∞} (and uncertainties σ) obtained using the two-step method from the previous section for the satellite-based data sets from Section 2. In addition, the top

- 15 right panel shows corresponding trends for the <u>multi-model mean of the CCMVal-2</u> simulations. For the <u>simulations</u>, trend <u>uncertainty was derived from the standard deviation of individual monthly anomalies from the multi-model mean (shaded envelope in Fig. 1). For the observations, trend uncertainty was derived from the fit residuals, as mentioned in Section 2. Although the two approaches differ, fit residuals for the observations and standard deviation of the simulated monthly anomalies have similar magnitude (compare Fig. 1), and the resulting trend uncertainties come out similar. The magnitude of the all trends</u>
- is represented by the color scale. Grey shading indicates regions where trends are not statistically significant (95% confidence level, $|TR| \le 2\sigma$). All satellite data sets show significant ozone increases in the <u>extra-tropical upper stratosphere</u>, above 10 to 5 hPa (30 to 35 km). <u>Some show significant ozone increases also in the tropical upper stratosphere</u>. At levels between 50 and 10 hPa (20 and 30 km), trends are generally not significant, except for islands of significant trends near 20 hPa or 50 hPa in some data sets, mostly in the southern hemisphere. Most data sets (but not SAGE + MIPAS + OMPS) also show significant
- 25 ozone decline in the tropical lowermost stratosphere below 100 hPa (16 km). However, satellite measurements in this region can have large uncertainties and need very careful consideration, both in tropics and extra-tropics (see also Tegtmeier et al., 2013).

The simulations (in the top right panel of Fig. 2) confirm that significant trends should be expected only in the upper stratosphere, between 10 and 0.5 hPa (30 to 55 km), <u>especially in the extra-tropics</u>. Exactly there, the observed data sets give significant increasing trends. Both magnitude - between 1 and 5% per decade - and latitudinal pattern - smaller increases in the tropics, larger increases at higher latitudes - are consistent between the satellite data sets and the simulations.

Figs. 2 and 3, therefore, provide substantial observational evidence for significant ozone increases in the upper stratosphere, consistent with model simulations based on declining ODS and decreasing temperatures in the upper stratosphere. Comparison of Figs. 2 and 3 with Figure 2-10 of WMO (2014) shows that the addition of three more years of data, as well as improved and

additional data sets, have not changed the overall picture very much. Comparable patterns, but slightly smaller increases are also reported in Fig. 5 of Harris et al. (2015) for the 1998 to 2012 period (but only between 60°S and 60°N). Compared to that Figure, the SAGE + OSIRIS trends here have changed considerably because of improved data (see also Bourassa et al., 2017). The SWOOSH trends have increased in magnitude. The SAGE + GOMOS data, which had shown large, and probably unrealistic ozone decline polewards of 40° latitude in Fig. 5 of Harris et al. (2015) are not used here.

A specific look at zonal mean trends from all satellite and ground-based data sets is given in Fig. 4. The basis for these trend calculations are the zonal band anomaly time series as in Fig. 1. In Fig. 4, almost all individual data sets show increasing ozone between 5 and 1 hPa, with trends between 0 and 4% per decade. For the 5 and 2 hPa levels, the plotted $\pm 2\sigma$ uncertainty bars (from the MLR) indicate that most individual trends are statistically significant (95% confidence level). Between 50 and

10 10 hPa (22 and 30 km), most data sets indicate small and non-significant trends. In the lowermost stratosphere, between 100 and 50 hPa (16 and 22 km), several data sets report ozone decreases, but these are generally not statistically significant. Differences between data sets are larger as well. Overall, Fig. 4 confirms significant ozone increases in the upper stratosphere from nearly all satellite and ground-based data sets, whereas ozone trends at lower levels are generally smaller and not significant.

4.2 From individual data set trends to the average trend

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- 15 To compare with other studies, or with model simulations, it It is useful to obtain an average ozone trend profile from all individual trends. In WMO (2014) this was done by a weighted mean of all individual ground-based and satellite trends TR(i). Each individual trend was weighted with its inverse squared uncertainty $(1/\sigma(i))^2$, so more uncertain trends have less weight. Individual uncertainties $\sigma(i)$ came from the regression (as in Section 3), and also included a 1 or 2% per decade drift uncertainty $(2\sigma, depending on the instrument)$ added in quadrature. This standard weighted mean approach (SWM) was also one of the
- 20 approaches used in Harris et al. (2015), however, with much larger drift uncertainties (4 or 6% per decade, 2σ), resulting. This resulted in larger overall uncertainty and in non-significant trends in Harris et al. (2015) compared to WMO (2014).

One problem with the standard weighted mean is that its uncertainty does not depend on the spread of the individual trends (because of Gaussian error propagation). Therefore, Harris et al. (2015) also considered the joint distribution approach (J). There, the uncertainty of the mean trend is essentially given by the standard deviation of σ between the individual trends

- 25 σ (which includes possible instrument drifts), divided by \sqrt{n} , where *n* is the number of data sets. Strictly, *n* should be the number of statistically independent realizations. Since data sets. However, since most merged data sets use the same SBUV, SAGE, MLS, or OMPS instruments, these data sets are not independent. Also, the trend calculation by multiple linear regression uses the same approach and the same proxies for all data sets, which may further reduce independence between the individual trend estimates.
- 30 Given these caveats, To be compatible with Harris et al. (2015), where standard weighted mean approach with large drift uncertainties and joint distribution approach gave similar average trends (1 to 3% per decade in the upper stratosphere) and similar uncertainties (2 to 6% per decade, 2σ), it was decided to also use the joint distribution approach for the average trend in the present study. Table 5 summarizes the methods used in the different studies to arrive at an average trend and its uncertainty.

Fig. 5 shows the joint distribution <u>average</u> trends (black lines), obtained here by averaging the seven satellite data sets (GOZ-CARDS, SWOOSH, SAGE + OSIRIS, SAGE + CCI + OMPS, SAGE + MIPAS + OMPS, SBUV-NASA, and SBUV-NOAA). All were given the same weight, and but SBUV data were used only at levels above 40 hPa (23 km)-, because the lower SBUV layers mix stratospheric and tropospheric ozone information due to their very wide averaging kernels (see also Kramarova et al., 2013)

5 . Ground-based data were not included in the average trends, because of their sparser sampling (which would require small weights), and also to be compatible with Harris et al. (2015). Nevertheless, the ground-based trends, shown in Fig. 4, provide important independent verification of the satellite-based trends.

The (joint distribution) uncertainty bars give ± 2 standard deviations over all seven data sets Fig. 5 (black error bars) give the full $\pm 2\sigma$ standard deviations between all 7 satellite-based trend estimates. Using these uncertainty bars in the Figure

- 10 assumes only 1 independent realization (n = 1), and should give a very conservative uncertainty estimate for the mean trend, TR. Even with this conservative uncertainty estimate, significant increasing trends (|TR| ≥ 2σ) appear in Fig. 5 for the 2 hPa level in the tropics and at northern mid-latitudes. Table 6 gives the same trend results, but now bold letters indicate trends, TR, that are significant with 95% confidence (|TR| ≥ 2σ/√n), assuming n = 3 independent realizations, or n = 2 below 40 hPa. In this less conservative case, significant increasing trends appear nearly everywhere above 10 hPa (30 km). As mentioned
- 15 above, trends at 70 hPa (and below) differ more between data sets and should be considered with care. See also the large error bars below 50 hPa for the tropical latitudes in Fig. 5.

4.3 Comparison to previous studies

For comparison, the yellow lines and shading in Fig. 5 show average 1998 to 2012 trends and uncertainties from Harris et al. (2015) Harris et al. (2015, joint distribution case), and the blue lines give average 2000 to 2013 trends and error bars from WMO

- 20 (2014). See Tables 3 and 5 for a summary of the different data sets and approaches. Overall, the updated 2000 to 2016 trend profiles (black lines) agree quite well with Harris et al. (2015) and with WMO (2014), especially when the overlapping error bars are considered. One difference is that the previous negative trend around 5-8 hPa in the tropics is not observed any more. The major difference, however, is the substantially larger uncertainty range reported in Harris et al. (2015) for the upper stratosphere. This As mentioned before, it is probably caused by two outlying data sets in Harris et al. (2015): 1.) An
- older version of the SAGE + OSIRIS data set, where the OSIRIS (V5.07) data suffered from a large drift (Hubert et al., 2016) (Hubert et al., 2016; Bourassa et al., 2017). 2.) A now outdated SAGE + GOMOS data set, which exhibited unrealistically low / negative trends at latitudes poleward of 45° (see Fig. 5 of Harris et al., 2015). For levels above 5 hPa (35 km) and levels below 30 hPa (25 km), the new and improved merged satellite data sets, and the additional years, provide substantially smaller trend uncertainties than Harris et al. (2015). In particular, there is no indication that the new a look at Fig. 4 indicates that individual
- 30 trends from the merged satellite data sets differ by more than 1 or and the ground-based instruments used here differ, in most cases, by less than 2 or 3% per decadedue to instrumental drifts, at levels above 50 hPa. This much better agreement indicates that previously large instrumental drifts and drift uncertainties (around 6% per decade for some data sets Harris et al., 2015; Hubert et al., 2 have been reduced substantially since.

Compared to WMO (2014), the current work reports slightly larger uncertainty bars. This is expected, because the standard weighted mean uncertainty used in WMO (2014) did not consider the spread of the individual trends (as mentioned above), and also assumed statistical independence for all the data sets in their the average.

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The updated trend profiles in Fig. 5 also show excellent agreement with the CCMVal-2 simulations, with virtually no difference at levels above 50 hPa (20 km). The fact that all individual data sets in Fig. 4 indicate significant increases in the upper stratosphere, the reduced uncertainty since Harris et al. (2015), the excellent agreement with the CCMVal-2 simulations in Fig. 5, and the good agreement with trend results from WMO (2014), all give enhanced confidence that ozone is indeed increasing in the upper stratosphere, and that at least part of the that increase is due to declining ODS.

5 Conclusions

10 New and improved satellite data sets, and the addition of data after 2013 several years of data until the end of 2016, improve our confidence that ozone in the upper stratosphere, between 5 and 1 hPa (35 to 48 km), has been increasing since 2000. Between 50 and 10 hPa (20 to 30 km) trends are small, and there are no clear indications for increasing (or decreasing) ozone. In the lowermost stratosphere, between 100 and 50 hPa (16 and 20 km), there might be an indication for decreasing ozone in the tropics and at northern mid-latitudes. However, differences between data sets in this region are larger. Instrumental difficulties and large natural variability require more careful analysis of these possible ozone decreases. 15

Overall, the updated ozone profile trends are consistent with previous studies, e.g. with WMO (2014) and Harris et al. (2015)-Using the average 2000 to 2016 trend from seven continuing and improved satellite data sets, and their standard deviation as uncertainty, trend uncertainty, but average trend uncertainty in the upper stratosphere is reduced by a factor of two compared to Harris et al. (2015). Ozone increases at the 2 hPa (42 km) level are statistically significant with more than 90 or 95% confidence over a wide range of latitudes. In addition, the majority of all individual satellite and ground-based data sets also 20

indicates significant ozone increases at levels above 10 hPa.

There are, however, remaining questions, for example regarding the merging of different instrumental records, the quality of the records in the lowermost stratosphere, or on the best methods for trend estimation and their detailed uncertainties. These issues are being addressed in the "Long-term Ozone Trends and Uncertainties in the Stratosphere" (LOTUS) initiative, which

- runs under the Stratosphere-troposphere Processes And their Role in Climate project (SPARC) of the World Climate Research 25 Programme (WCRP), see http://www.sparc-climate.org/activities/ozone-trends/. The goals of LOTUS are to further improve the data sets, to better understand all relevant uncertainties, and to achieve a more complete and more precise picture of trends in the stratospheric ozone profile. What is also missing is a thorough quantification and attribution of the contributions from decreasing ozone depleting substances, from stratospheric cooling (due to increasing CO_2), and from transport changes to the
- observed profile trend. While this has been done in modelling studies (e.g. Jonsson et al., 2004; WMO, 2014), quantification 30 of these factors on the basis of observations has not been done yet.

The update presented here, however, already gives strong indications that ozone in the upper stratosphere has been increasing over the last 15 years, and has begun to recover from man-made ozone depleting substances. Simulations show that this process will take many more decades. In order to verify that ozone recovery continues as expected, reliable long-term observations from multiple independent platforms will remain crucial for many years to come.

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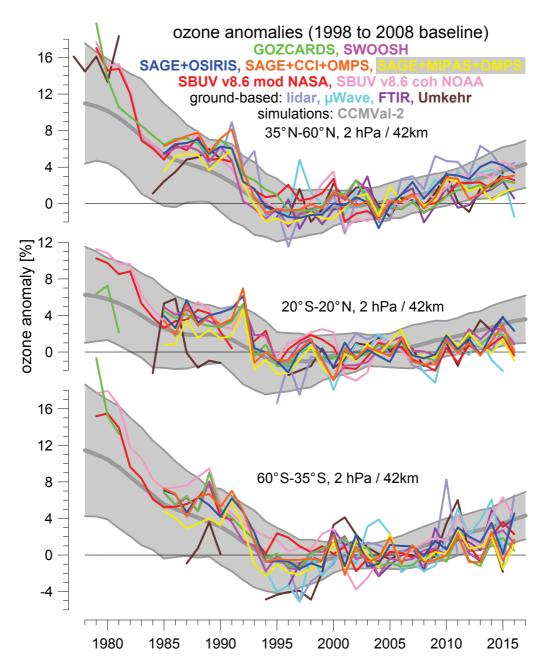


Figure 1. Annual mean ozone anomalies near 2 hPa or 42 km, as recorded by merged satellite data sets and NDACC ground-based stations. Anomalies are referenced to the 1998 to 2008 climatological annual cycle of each individual data set, and are averaged over the indicated zonal bands. NDACC stations Stations close to a zonal band are also included, i.e. NDACC lidar data from Table Mountain at 34.4°N, NDACC FTIR data from Izaña at 28.3°N and Wollongong at 34.4°S, and Umkehr data from Perth at 34.7°S are included in the respective mid-latitude bands. The grey Due to contamination by volcanic aerosol after the eruptions of El Chichon and Mt. Pinatubo, Umkehr data are not used for the years 1982, 1983, and 1991 to 1993, and SBUV-NASA data are not available for 1992. Grey lines show corresponding the multi-model mean ozone anomalies from CCMVal-2 model simulations (Eyring et al., 2010), with the grey shading giving the ±2 standard deviation are taken over all models (except outliers) and within a 25 month sliding window.

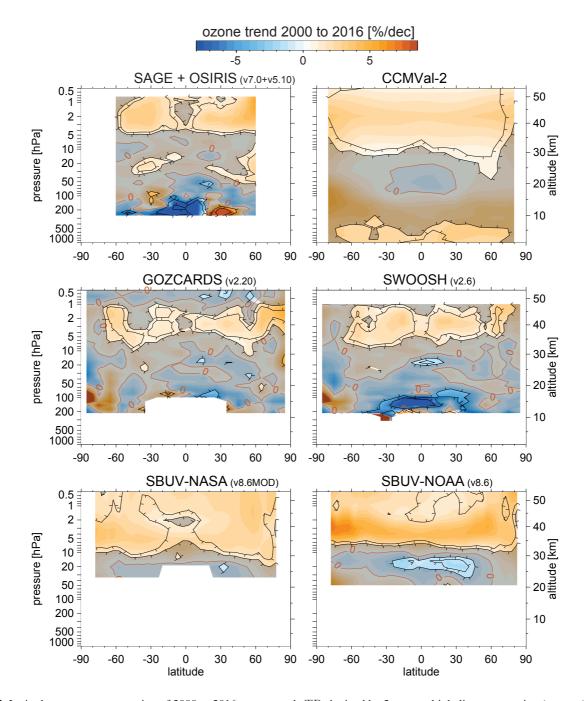


Figure 2. Latitude pressure cross section of 2000 to 2016 ozone trends TR obtained by 2-step multiple linear regression (see text). The top right panel is for model simulations from the CCMVal-2 initiative. The other panels are for merged satellite data sets. The colour scale gives trend magnitude TR. Shading and isolines give the ratio of trend to trend uncertainty, $|TR|/\sigma$. Grey shading, in regions where $|TR| \le 2\sigma$, indicates that trends are not significant at the 95% confidence level. The next isoline is at $|TR|/\sigma = 3$.

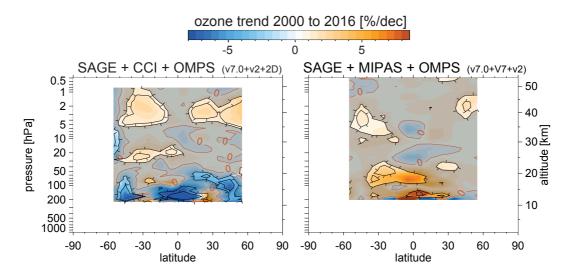


Figure 3. Same as Fig. 2, but showing the 2000 to 2016 ozone trends for the merged SAGE + ESA Ozone CCI + OMPS, and SAGE + MIPAS + OMPS data sets. The SAGE + ESA Ozone CCI + OMPS data set uses the OMPS 2D retrieval from U. Saskatoon. The SAGE + MIPAS + OMPS data set uses the OMPS v2 retrieval from NASA.

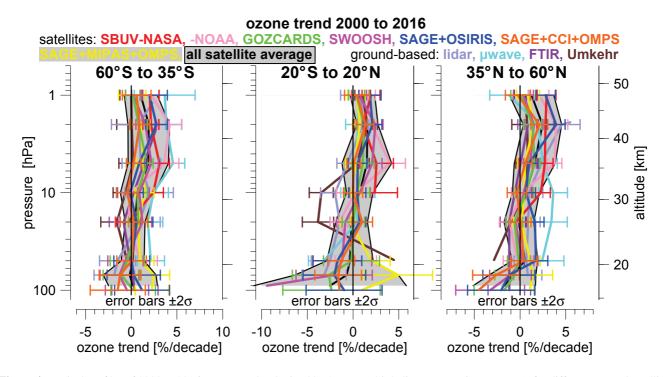


Figure 4. Vertical profiles of 2000 to 2016 ozone trends, obtained by 2-step multiple linear regression (see text), for different merged satellite and ground-based station data sets. Results are for the zonal bands 60° S to 35° S (left), 20° S to 20° N (center) and 35° N to 60° N (right). For the 60° S to 35° S zonal band, FTIR data from Wollongong (34.4° S), and Umkehr data from Perth (34.7° S) are included. For the 35° N to 60° N band, lidar data from Table Mountain (34.4° N) and FTIR data from Izaña (28.3° N) are included. SBUV and Umkehr data are not shown at/ below the 50 hPa level. Black lines and grey shading show the average trend and $\pm 2\sigma$ standard deviations of the 7 satellite based trends (see also Fig. 5 and text).

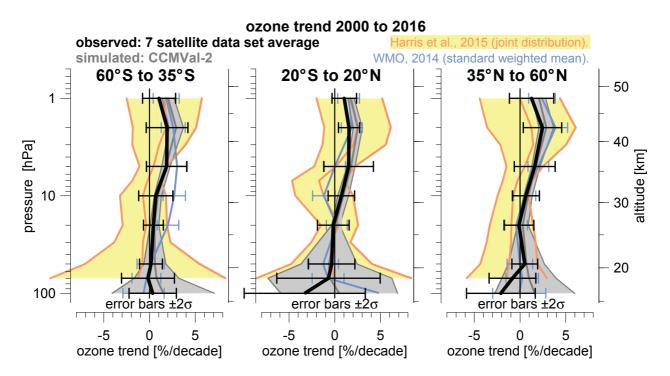


Figure 5. Same as Fig. 4, but giving the average 2000 to 2016 ozone trends (black lines) from seven merged satellite data sets (GOZCARDS, SWOOSH, SAGE + OSIRIS, SAGE + CCI + OMPS(2D), SAGE + MIPAS + OMPS(v2), SBUV-NASA, and SBUV-NOAA). SBUV trends are only used at levels above 40 hPa (23 km). Uncertainty bars and yellow shading give ± 2 times the standard deviation σ of all individual trends in the average. For comparison, the 1998 to 2012 average ozone trend from Harris et al., 2015 (yellow lines and shading), and the 2000 to 2013 average ozone trend from WMO (2014) WMO 2014 (blue lines) are shown as well. In all cases, uncertainty bars or shading give $\pm 2\sigma$ uncertainty. For the 2000 to 2016 satellite average ozone trends (black error bars), and trends from Harris et al., 2015 (yellow shading) the uncertainty is derived from the standard deviation σ between individual trends in the average (joint distribution case). For the WMO 2014 trends (blue error bars), the uncertainty of the standard weighted mean is given (see text for details). Grey lines and shading give corresponding trends the trend and uncertainties $\pm 2\sigma$ uncertainty obtained from multi-model mean and standard deviation of the CCMVal-2 model simulations (see text in Section 4.1).

Table 1. Merged satellite data sets used in the present study. The URLs serve as an entry point only, and do not always provide the newest and most complete data set used here. See text for references.

Name	Version(s)	from	to	URL
SBUV-NASA	v8.60MOD	05/1970 ^a	12/2016	https://acd-ext.gsfc.nasa.gov/Data_services/merged/
SBUV-NOAA	v8.60	11/1978	12/2016	ftp://ftp.cpc.ncep.noaa.gov/SBUV_CDR/
GOZCARDS	v2.20	02/1979 ^b	12/2016	https://gozcards.jpl.nasa.gov/
SWOOSH	v2.6	10/1984	12/2016	https://www.esrl.noaa.gov/csd/groups/csd8/swoosh/
SAGE II + OSIRIS (+ OMPS) c,d	$v7.0 + v5.10 (+ 2D^d)$	10/1984	12/2016	http://osirus.usask.ca/
SAGE II + Ozone_CCI + $OMPS^d$	$v7.0 + v2 + 2D^d$	10/1984	12/2016	http://www.esa-ozone-cci.org/
SAGE II + MIPAS + OMPS ^{e}	$v7.0 + KIT v7 + v2^e$	10/1984 ^f	03/2017	https://www.imk-asf.kit.edu/english/304_2857.php

 a^{a} gap from 05/1976 to 10/1978; b^{b} includes also SAGE I, but gap from 12/1981 to 09/1984, when SAGE II begins; c^{c} the SAGE + OSIRIS data set optionally includes OMPS data. These start in 04/2012, and give very similar trend results. However, to keep more independence between the various data sets, the version with OMPS data is not used here; d^{c} OMPS 2D retrieval from U. Saskatoon; e^{c} OMPS retrieval from NASA; f^{f} MIPAS high resolution data from 07/2002 to 03/2004, reduced resolution data from 01/2005 to 04/2012, gap in between.

of Atmospheric Composition Change (NDACC), and are originally available at http://www.ndacc.org. Umkehr data were provided by I. Petropavlovskikh.

Table 2. Stations and instruments used in the present study. Lidar, microwave and FTIR data are from the Network for the Detection

Name	latitude	longitude	instrument	from	to
Fairbanks	64.8°N	147.9°W	Umkehr	03/1994	09/2015
Hohenpeissenberg	47.8°N	11.0°E	lidar	09/1987	12/2016
Bern	46.9°N	7.5°E	microwave	11/1994	12/2016
Payerne	46.8°N	7.0°E	microwave	01/2000	12/2016
Arosa	46.8°N	9.7°E	Umkehr	01/1956	12/2015
Jungfraujoch	46.6°N	8.0°E	FTIR	05/1995	11/2016
Haute Provence	43.9°N	5.7°E	lidar	07/1985	10/2016
Haute Provence	43.9°N	5.7°E	Umkehr	01/1984	11/2015
Boulder	$40.0^{\circ}N$	$105.3^{\circ}W$	Umkehr	01/1984	12/2015
Table Mountain	34.4°N	$117.7^{\circ}W$	lidar	02/1988	09/2016
Izaña	28.3°N	16.5°W	FTIR	03/1999	10/2016
Mauna Loa	19.5°N	155.6°W	lidar	07/1993	09/2016
Mauna Loa	19.5°N	155.6°W	microwave	07/1995	05/2015
Mauna Loa	19.5°N	155.6°W	Umkehr	01/1984	12/2015
Wollongong	34.4°S	150.9°E	FTIR	05/1996	11/2016
Perth	34.7°S	138.6°E	Umkehr	01/1987	12/2015
Lauder	45.0°S	169.7°E	microwave	10/1992	10/2016
Lauder	45.0°S	169.7°E	lidar	11/1994	12/2016
Lauder	45.0°S	169.7°E	FTIR	10/2001	12/2016
Lauder	45.0°S	169.7°E	Umkehr	02/1987	12/2015

 Table 3. Comparison between principal data sets, trend periods, and regression method used in the present study, in Harris et al. (2015), and in WMO (2014). Here only major changes in data sets are indicated. Boldface indicates the most relevant differences.

	<u>this study</u>	Harris et al. (2015)	<u>WMO (2014)</u>
SBUV-NASA	used	used	used
SBUV-NOAA	used	used	not used
GOZCARDS	used	old version	used
SWOOSH	used	old version	not used
Ozone_CCI	new version	not used	old version ^a
SAGE + OSIRIS	<u>new version</u> ^b	old version ^c	not used
SAGE + GOMOS	not used	old version ^c	not used
SAGE + MIPAS + OMPS	used	not available	not available
ground-based	\underbrace{used}^d	\underbrace{used}^d	$\underbrace{used^e}_{e}$
trend period	2000 to 2016	<u>1998 to 2012</u>	2000 to 2013
regression method	two steps	hockey-stick	two steps

^{*a*} called HARMOZ. ^{*b*} drift-corrected OSIRIS data. ^{*c*} OSIRIS and GOMOS data had significant drifts (Tegtmeier et al., 2013; Harris et al., 2015; Hubert et al., 2016). ^{*d*} ground-based trends as independent verification, but not included in average trend. ^{*e*} ground-based trends (weighted) included in average trend.

Proxy	description	URL
trend	linear increase over entire time period.	
change of trend	"hockey stick": 0 before 01/1997, linear	
	increase after.	
QBO	10 and 30 hPa equatorial zonal wind	http://www.geo.fu-berlin.de/en/met/ag/strat/produkte/
	from Singapore radio-sondes, as com-	qbo/index.html
	piled by FU Berlin. To account	
	for annual variation and phase	
	of the QBO influence, QBO(10),	
	$QBO(10) \cdot \sin(j), \qquad QBO(10) \cdot \cos(j),$	
	QBO(30), QBO(30) $\cdot \sin(j)$, and	
	$\underline{QBO(30)} \cdot \cos(j)$ are fitted, with	
	$j = 2\pi \cdot (\text{month mod } 12)/12.$	
Solar Cycle	solar radio flux at 10.7 cm, observed at	ftp://ftp.geolab.nrcan.gc.ca/data/solar_flux
	Penticton, Canada.	
El-Nino	Multivariate ENSO index from Wolter	https://www.esrl.noaa.gov/psd/enso/mei/
	and Timlin (2011).	
Aerosol	stratospheric aerosol optical depth fol-	https://data.giss.nasa.gov/modelforce/strataer/
	lowing Sato et al. (1993).	

Table 4. Proxy time series used for the multiple linear regression in Eq. 2 in this study.

	this study_	Harris et al. (2015)	<u>WMO (2014)</u>
standard weighted mean (SWM)	not used	used	used
assumed drift uncertainty (2σ)	not used	4 or 6% decade	<u>1 or 2% decade</u>
number of data sets in SWM	$\bar{\sim}$	$\overset{6^a}{\sim}$	$\overset{9^{b}}{\sim}$
joint distribution (J)	used	used	not used
number of data sets in J	7^a_{\sim}	6^{a}	~

Table 5. Approaches taken to obtain the average trend and its uncertainty estimate in the present study, in Harris et al. (2015), and inWMO (2014). Boldface indicates the approach used for the results plotted in Fig. 5.

 $^a\,$ only satellite based data sets included in average. $^b\,$ ground-based data sets were also included. Ozone sondes not used at levels higher than 10 hPa / 31-km.

Table 6. Average 2000 to 2016 ozone profile trends, obtained from individual trends for the GOZCARDS, SWOOSH, SAGE + OSIRIS, SAGE + CCI + OMPS(2D), SAGE + MIPAS + OMPS(v2), SBUV-NASA, and SBUV-NOAA satellite data sets. Given are mean trend TR and standard deviation 1σ of the individual trends, in percent per decade. Bold numbers indicate average trends TR larger than $2\sigma/\sqrt{3} \approx 1.15\sigma$, i.e. statistically significant with 95% confidence, assuming that the 7 data sets give 3 independent realizations of individual trends TR(i). The SBUV-NASA and SBUV-NOAA data sets are used only at levels above 40 hPa (23 km). Therefore, $2\sigma/\sqrt{2} \approx 1.41\sigma$ is applied as threshold for bold face at the 50 and 70 hPa levels.

level	60°S 1	to 35°S	20° S t	to 20°N	35°N	to 60°N	60°S t	to 60°N
(hPa)	TR	1σ	TR	1σ	TR	1σ	TR	1σ
1	1.0	0.9	1.0	0.7	1.3	1.2	1.1	0.7
2	1.9	1.1	1.6	0.6	2.5	1.1	1.8	0.6
5	1.9	1.1	1.5	1.4	1.6	1.1	1.6	1.2
10	0.7	0.9	0.7	0.7	0.6	0.7	0.8	0.7
20	0.4	0.5	-0.2	0.9	-0.1	0.8	0.0	0.7
50	0.2	0.6	-0.3	1.3	0.5	0.7	0.0	0.8
70	-0.2	1.4	-0.7	2.8	-0.8	1.3	-0.6	1.9

All values are % per decade.