Dear Michel,

below you find our detailed response to both reviewers as published in the online discussion. A PDF of the manuscript with track changes is attached after the replies.

Best wishes,

Mark

Replies to Reviewers #1 and #2

Weber et al., Global total ozone recovery trends attributed to ODS changes derived from five merged ozone datasets, doi:10.5194/acp-2021-1058

Reviewer #1

Reviewer comments are provided here with our replies written in italics

1. Short resume

Weber et al. present a comprehensive analysis of trends in total ozone, focusing primarily on the period since the turnaround in ozone-depleting substances. This is an update and extension of earlier work published in 2018. In contrast to latter publication, the authors now claim the detection of increases (0.4%/decade) in near-global (60S--60N) total ozone since 1996, with high confidence (>3-4\$\sigma\$). Positive trends over broad mid-latitude region in both hemispheres (35N--60N and 35S--60S), about 0.5--0.7%/decade, are significant as well although close to the 2sigma detection threshold.

The dynamical process terms (Arctic and Antarctic Oscillation, Brewer-Dobson circulation) in the regression model play a central role in this detection, especially at northern mid-latitudes. The authors deliberately chose not to detrend the dynamical terms prior to regression, in order to account for any long-term changes in AO, AAO and BDC. In doing so, they find that trends become less negative before 1996 and more positive since 1996 across large regions of the low- and mid-latitudes. This more complete attribution results in a higher significance of the trends, especially in the northern hemisphere where the 2sigma detection threshold was passed. Hence, the authors conclude that dynamical changes appear to counterbalance the recovery of ozone in the mid-latitude NH.

The authors furthermore explain the positive recovery trend of total ozone as a result of changes in ozone-depleting substances. Indeed, the ratio of the rate of increase and decrease in ODS concentrations is consistent with the rate of depletion and recovery of total ozone across all 5° latitude bands between 60S and 60N.

2. Recommendation

This paper provides an important update to previous assessments of long-term changes in total ozone. It is very well written and accessible to a large scientific audience. The methodology is sound and the presented results support the claims made by the authors. I highly recommend publication of this work in ACP if my remarks below have been addressed.

3. Major comments

Ordered in order of appearance in the text.

3.1 Extension of GOME-type backwards in time (Sect. 2.7)

I understand the importance of covering a sufficiently long period, but is this backwards extension for GOME-type data records still needed now that more than two solar cycles have been completed since 1995? Doesn't this break the independence between SBUV and GOME-type estimates? By how much does the negative trend in the SBUV period influence the recovery trend estimates during the GOME-type period? Have you tested the sensitivity of the resulting trend to the choice of NASA COH or NASA MOD, and without the extension?

 \rightarrow We try to stay consistent with the W18 paper, where we also applied for these extensions. The main idea of extending to the full-time period is to have as close as possible the same impact from all proxy terms, not only the solar term, during the full-time period. Since we use independent linear trends before and after the ODS peak, the impact of early trends on the late period trends is minimised.

Avoiding data gaps is important but preserving data quality/stability is perhaps even more important under high aerosol backgrounds. Could you elaborate why gaps are more important or, if that is not the case, comment on the stability of both SBUV records after Pinatubo?

 \rightarrow Calculated annual means were accepted as valid if at least 80% of the monthly means were contributing (10 months minimum) and 80% of the 5° zonal means were available in the broader zonal bands. If these conditions are not met we consider the annual mean as not representative for the given year and should be excluded from the MLR. The consistency of the SBUV data records with other total ozone data has been documented, e.g. Chou et al, 2014.

3.2 No reference to how trend errors are estimated (Sect. 3)

Many trend estimates (Fig. 3) are close to the 2sigma threshold. The computation of MLR coefficient uncertainties, therefore, deserves some attention, this is missing right now. Please explain how MLR parameter errors are computed or refer to relevant publications. \rightarrow All uncertainties are given as 2sigma and sigma is calculated from the least-squares fit. This is a standard approach and is described in many statistics textbooks.

Somewhat related to this, was there any consideration of including reported measurement errors in the regression? \rightarrow Measurement errors were not accounted for. Not for all merged datasets, an uncertainty estimate is provided from merging the data.

3.3 Annual time series

p.7, l.180: Could you motivate the choice for analysis of annual mean time series instead of monthly mean data? Is there an impact on the trend estimates and their significance? Please refer to relevant publications. \rightarrow The main reason for using annual means is that this does not require corrections for auto-correlation (mentioned in the text). Adding auto-correlation terms in the regression will not alter the trends but increases uncertainties in the fit coefficients (and trends). The short-term variability is not the focus of our MLR except that we try to minimise the residual of the regressed timeseries.

3.4 Robustness of attribution to dynamical processes (Sect. 5)

Previous work by the authors (Weber et al, 2018) also considered terms for dynamical processes in the MLR. At the time, however, no significant positive trends were detected (Fig. 9). \rightarrow see our general comment in the beginning of the reply to reviewer #2

It would be enlightening to discuss whether the four additional years of data have truly helped to attribute ozone changes more robustly to dynamical changes. Or, whether it is plausible that the current attribution is subject to geophysical variability (and measurement uncertainty). \rightarrow see our general comment in the beginning of the reply to reviewer #2

4. Minor comments

p.1, l.12-13: Near-global trend values disagree with quoted values in Section 4. Please revise. \rightarrow Numbers in the abstract have been adjusted to the values shown in Fig. 1.

p.3, l.82: "Annual mean timeseries of all five merged datasets are in very good agreement". Somewhat subjective, please add a number. \rightarrow add: "... to within a few DU"

p.5, l.132: The evolution in satellite quality has been described adequately. This is missing in the WOUDC section. Surely, there must have been progress in the calibration of these instruments or the coherence of the network since the work by Fioletov in 2008. If so, could you update this section accordingly? \rightarrow The ground-based network calibration procedures have been established a long time ago and there are no major changes in the network operation. The same is true for the WOUDC data set that is regularly generated by the WOUDC. We added a reference to a recent paper where the differences between satellite and ground-based data are discussed on a global scale. We added a reference to Garane et al.

p.6, l.142-143: "[...] can be estimated with a precision comparable with satellite-based data sets (~1%)." A reference would be appropriate. \rightarrow Comparison of satellite and ground-based data sets is discussed in the following paper: Chiou, E. W., Bhartia, P. K., McPeters, R. D., Loyola, D. G., Coldewey-Egbers, M.,

Fioletov, V. E., Van Roozendael, M., Spurr, R., Lerot, C., and Frith, S. M.: Comparison of profile total ozone from SBUV (v8.6) with GOME-type and ground-based total ozone for a 16-year period (1996 to 2011), Atmos. Meas. Tech., 7, 1681–1692, https://doi.org/10.5194/amt-7-1681-2014, 2014. It was referenced in line 140 and we added this reference to line 143 (at the end of the last sentence)

p.6, l.150: Remove "from the past into the future" as the statement "between 1960 and 2100" is more than sufficient. \rightarrow *done*

p.6, l.154-156: I am sorry, I did not get the point of "The multi-dataset mean was then added back to each dataset, such that all bias corrected timeseries are provided in units of the total column amounts (W18). However, the trend results derived from them are identical to those derived using anomaly timeseries." Could this be clarified a bit better for the non-expert? \rightarrow This procedure means that the bias-corrected time series differ from anomaly timeseries by a constant offset (multi-instrument mean). The bias correction has no influence on the calculated trends but makes the data more legible in the plots.

p.6, l.154: "to the mean". The 1998-2008 mean at the global or local level? \rightarrow all data are annual mean zonal means and for each zonal band considered an average for the period 1998-2008 was calculated for each dataset and a mean over all datasets (multi-instrument mean) calculated

p.6, l.165: See comment below, the second term in Eq. 1 should be b_1 (t-t_0) \rightarrow this is not correct, since t_0-t is positive (t_0>t), b_1 will be negative if ozone declines.

p.6, l.166: "coefficients b_1 and [...]" This is inconsistent with the notation in Eq 1. Sign of first trend term (t0-t) implies that positive b_1 values represent a decline in ozone. Please change this. The factors $X_1(t)$ and $X_2(t)$ define the decline/recovery periods. \rightarrow see previous statement

p.6, Eq.2 and 3: Figure 1 suggests that the "recovery" period starts in 1996, so the turnaround is defined as $t_0=1996$. If this is correct, then the notation in Eq. 2 and 3 should be changed to $X_1(t)=1$ for $t < t_0$ and $X_2(t)=1$ if $t \leq t_0$ (and vice versa for $X_i=0$). The trend model is not continuous at t_0 , hence $\leq<$ or \leq and $t = a difference. <math>\rightarrow$ This was indeed not consistent and has been corrected at several places. The early period is t < t0, late period t >= t0. So the first period includes $t_0=1996$ and the late period starts with 1996. As mentioned in the text the shift of t_0 back and forward did not change the trend estimates.

p.7, l.185-187: Is there any particular reason why you haven't used GloSSAC v2 (Kovilakam et al., 2020)? \rightarrow We actually tested the Glossac dataset, but we found only negligible differences in the trend estimates. This is likely due to the fact that the El Chichon and Mt Pinatubo eruptions dominate the stratospheric aerosol optical depth proxy timeseries. This effect is even enhanced since we use two proxies to separate both major volcanic events.

p.8, Table 2: EHF is missing from this list. Where can it be downloaded? \rightarrow As mentioned in the text, the eddy heat flux was calculated by us from the ERA5 reanalysis data and was not taken from an external source. A description of how to derive the eddy heat flux from reanalysis data is given in W18.

p.8, Eq.4: "BDCn" and "BDCs" should be explained in the text. \rightarrow added, "The BDCn and BDCs are 100 hPa eddy fluxes in the northern (n) and southern hemisphere (s)."

p.8, I.208: "the linear trend terms best approximate EESC related trends". Can a match between ozone trend and EESC expectations really validate the choice of terms in the MLR? There is a risk of a circle reasoning here. If the improved agreement with EESC expectations is motivating the choice of terms in the MLR model then you can't use this same agreement again to conclude a causal relation between trend and EESC. \rightarrow We only assume that all trends not related to ODS changes are contained in the proxy terms. The linear trend before and after the ODS peak is independent, but It turns out that the trend ratio before and after the ODS peak is consistent with the rate changes of EESC to within the uncertainties from the regression. However, we know that there are feedbacks between ODS (ozone) and climate (dynamics). Therefore, the linear trends will only approximate the ODS related contribution to ozone changes.

p.8, l.215-216: This phrase is not entirely clear on whether or not you use the detrended proxy. This choice is so central to this paper that it must be very clearly stated. \rightarrow We added, "For these reasons, we do not detrend the proxy timeseries in this study".

p.9, Fig. 1: chi^2 is the sum of "the squared differences median timeseries minus MLR" \rightarrow We changed to ".. sum square of differences between median and MLR timeseries' divided by ..."

p.9, l.219: "MLR prediction after fitting" would be clearer than "MLR result from applying". \rightarrow better: "MLR timeseries derived from"

p.9, I.220: To me, "after 1996" suggests 1996 is not included. What about replacing "after 1996" by "since 1996" throughout the manuscript? \rightarrow see earlier comment. It should be "after 1995" or "since 1996", similarly "before 1996" and "until 1995". We changed accordingly.

p.9, l.224: "recovery from reductions in ODS" would be more clear on the effect of ODS on ozone. \rightarrow *done*

p.11, l.260: Replace "from applying" by "when applying"? \rightarrow *leave it as is.*

p.11, l.260: It is somewhat unexpected to regress a "super" merged timeseries rather than average the trends from individual records. What is the rationale? Also, the sample size is just N=3, for 1979-1995, so won't the "super"merge-then-regress method lead to more uncertainty in the MLR parameters than the regress-then-average approach? \rightarrow In Table 3 we present the trends of the median timeseries' as well as the median and mean of the individual trends. The numbers are nearly the same.

p.12, Table 3 (caption): The periods in the caption are inconsistent with information in Figs 1 and 2. The first trend period stops in 1995, the second starts in 1996. Hence, it should be 1979-1995 and 1996-2000. \rightarrow *done*

p.12, Table 3: For each latitude belt, the occurrences of "mean/median trend >1996" should be >=1996, in order to be in line with Fig. 1 and 2. \rightarrow changed to t>1995

p.12, Table 3: The error notation was confusing for me, I haven't seen this specific notation very often. For instance, what does -1.9(13)" mean? Is it -1.9+-0.13 or -1.9+-1.3 or ...? I find an explicit notation such as "+0.4+-0.2" much more effective. I recommend using this throughout this table and also the manuscript. \rightarrow It is a common way to put uncertainties in the brackets, but I agree that this is not so widely used in the atmospheric science community. In order to keep the table compact, we will remain with our notation. p.12, l.265: "One notable change from W18 is that the tropical trends during the ODS rising phase are now more negative (down to -1%/decade) while before they were mainly close to zero. This may be caused by the additional proxy terms used in this study". The pre-1996 data have been available for a very long time now. Has this effect never been looked into before? If so, please refer to relevant work. - \rightarrow see our general comment in the beginning of the reply to reviewer #2

p.12, l.270: Please replace the "maybe" (conditional) by an "is" (certainty). Trend uncertainty scales with $n^{-3/2}$ (e.g., Weatherhead et al., 2000) so the eight more years in the recovery period already lead to ~45% smaller trend error. This seems not too far from the observed factor 2 reduction of the error in Table 3 and Fig. 3. \rightarrow *done*

p.13, l.274: "The expected tropical recovery [...]". Estimated mid-lat NH recovery trends are too small compared to EESC prediction as well. \rightarrow added "In the NH extratropics the expected ODS related recovery is slightly higher than the observed trends, but also agree within the uncertainties of the observed trends."

p.15, I.320: "NH total ozone has been steadily declining..." conflicts with the first phrase of this paragraph "stable ozone levels in NH since 2000". Please clarify the text. \rightarrow The stable levels refer to annual means at NH middle latitudes as shown in Figure 2 (added "middle latitude"), while the decline in Figure 6 is shown for March only and also includes polar latitudes.

p.15, l.324: "with larger springtime polar ozone losses"? \rightarrow done

p.15, l.325: Remove "recent" from "A recent downward trend". Perhaps you meant that this was recently reported? Ball et al report a continuous decline since the 1980s, not a recent decline. \rightarrow *done*

p.18, l.332: Quoted recovery trend value (11%/decade) conflicts with that in Figure 7 (12%/decade). Please correct. \rightarrow *done*

p.19, Table 4: Same comment on error notation as in Table 3 (p.12). \rightarrow see earlier comment.

p.19, I.367: The Gaudel paper is about differences between tropospheric ozone data records. So probably not the best reference when the message is about consistency between tropo/strato/total ozone. \rightarrow We removed this sentence, as we did not mention tropospheric ozone at all in the paper. When using annual mean zonal mean averages contribution of tropospheric ozone is likely to be small, but may become more important when looking at regional trends.

5. Technical corrections

 \rightarrow all done

p.1, l.10: Remove "on" from "[...] is indeed on slowly [...]".

p.1, I.12: Remove "in absolute numbers".

p.1, l.15: Add "-" to "chemistry-climate models".

p.2, I.30: Typo "stratosphere".

p.2, l.38: Remove "agreement" from "Montreal Protocol agreement".

p.3, l.75: Replace "in large part" by e.g. "largely".

p.3, I.79: Replace "Observations Zénithales" by "Observation Zénithale".

p.4, l.87: Replace "are processed using the same V8.7 retrieval algorithm" by e.g. "are retrieved using the same V8.7 algorithm".

p.4, l.108: Type "[...] shift to an equivalent [...]".

p.5, l.130-132: Double occurrence of ground-based. First one could be removed, e.g. "The WOUDC zonal mean ...".

p.7, l.175: Add "." after "W18)".

p.7, l.189: Replace "there are not sufficient number of months" by e.g. "there are not enough months" or "there is not a sufficient number of months".

p.7, I.194: Replace MLR "equation" by MLR "model"?

p.8, l.212: Remove "the possibility", as it is a bit redundant.

p.8, l.212: Replace "MLR results" by "MLR fit residuals" perhaps? This is a bit clearer as the MLR parameter estimates are MLR results as well.

p.9, l.218: "five bias-corrected" instead of "bias-corrected five".

p.11, l.242-243: Maybe you forgot to remove the newline between paragraphs?

p.11, l.251: Add a "+" sign to the quoted values at start of this line.

p.11, l.256: Remove ' after "timeseries".

p.11, l.261: Add "/decade " after +0.5%"

p.12, Table 3 (caption): Remove "and" from caption "[...] in bold have an absolute [...]"

p.12, Table 3 (caption): Add "prediction" at the end of "and mod_ithe MLR".

p.12, Table 3: Add \$+\$ to trend value \$\geq\$1996 for median time series near-global.

p.12, Table 3: The quoted r^2 value for WOUDC in 20S-20N band is single digit (0.7), should be double (0.70).

p.13, l.276: Remove "on" from "elucidate further on".

p.13, I.285: Type "Fig. 4a" should be "Fig. 4".

p.15, Fig.5 (caption): There is a missing word in "Negative values an anti-correlation [...]".

p.15, l.311: Add "s" to "chemical effect"?.

p.15, l.316: Add full stop at end of phrase.

p.17, Fig.7 (caption): Capitalise "See".

p.18, l.331: "Earlier signs of ozone recovery have been", should be plural.

p.18, l.331: Add "," in between "Now with".

p.18, I.332-333: "During September, the Antarctic ozone hole usually grows and [...]".

p.18, l.340: Remove "as shown in Fig. 7". A bit redundant, you already referred to the figure in the previous phrase.

p.18, I.344: Replace "globally" by "global"?

p.18, l.352: Add "," in between "tropics recovery".

p.18, l.354: Add "," in between "Arctic large".

p.19, l.363: "chemistry-climate models".

Reviewer #2

Reviewer comments are provided here with our replies written in italics.

The manuscript "Global total ozone recovery trends derived from five merged ozone datasets" by M. Weber provides an update to a study published by the first author in 2018, with four more years of data added to the five analyzed datasets (four satellite datasets and one dataset comprised of ground-based measurements). A multiple linear regression is applied to annual mean data from the period 1979 to 2020 to determine total column ozone (TCO) trends in different broad latitudinal bands for the period in which concentrations of ozone-depleting substances (ODSs) increased in the atmosphere, and for the period after the peak concentrations had been reached. The multiple linear regression includes next to the typical proxies also several dynamical variables (e.g. a proxy for the Brewer-Dobson circulation (BDC) or the Antarctic/Arctic Oscillation (AAO/AO)) which is one of the main differences to other trend analyses based on TCO data. The authors find with this method significant positive trends (related to the reduction in ODSs in the atmosphere) for the period 1997-2020 for the near-global mean (60S-60N), as well as for the Northern hemisphere mid-latitudes for which the trend is near zero if the dynamical proxies are not included in the regression.

The manuscript is very well written and well structured, mostly the data and methods are explained in enough detail to allow the reader to understand what is going on (in a few cases I found the description slightly too short and I have mentioned them in the details below), and the topic lays clearly within the scope of the ACP journal. There are a few minor things that I commented about below that are easy to fix, but there are two main points that I think need careful adjustment of the manuscript or some additional thought.

\rightarrow We address these points (see specific replies below).

I recommend the publication of the manuscript after revisions.

Two main points:

Attempting an attribution with a multiple linear regression that includes non-orthogonal proxies is tricky. Especially if several proxies include a trend. The hope then is, that the regression is able to separate the trend contribution from the different proxies based on the additional variability the proxies provide. However, it is possible that trends are not assigned correctly to the different proxies which would falsify the signal of the trend that if of interest, in this case here, the trend caused by ODSs and not by changes in dynamical variables. The authors argue that with the addition of the dynamical proxies the variability of the time series' are matched better by the regression results. There are two points that make me somewhat doubtful of this statement: (1) the pre-1996 trends change clearly with the introduction of the dynamical proxies (Figures 3 and 4) although the main trend signal should be coming from ODS-related changes in this period; (2) the signal from the SH Brewer Dobson circulation proxy in the NH polar regions that cannot really be explained. I think the manuscript needs more discussion of these points to strengthen the claim that the addition of the dynamical proxies can indeed robustly isolate the ODS-related trends. For the first point I raised I would suggest to check the older literature about regression results for the pre-1996 period where dynamical proxies have been used. I have added two references in the comments below that might be worth checking out. And there might even be more that could be checked and where the results could be compared to the pre-1996 ODS-related

trends calculated here. For the second point I raised I think it would be helpful to do some sensitivity test to check the robustness of the trend results and the contribution of the individual proxies: (I) not using the trend proxy but JUST the dynamical proxies, how do their contributions change if at all; (II) use some of the dynamical proxies only in the regions where they occur, e.g. AAO only in the SH, AO only in the NH, etc.; how does the contribution of these proxies change (if at all), and how does the ODS-related trend change? I think these sensitivity test will go a long way to show the robustness of the results presented here in this manuscript.

 \rightarrow We added two new tables to summarise the results from new sensitivity tests we carried out. New Table 4 shows different MLR settings applied to the median total ozone timeseries in broad zonal bands (as defined in Table 3). Here the results from the standard and full MLR are listed. In addition, we applied an iterative MLR approach where statistically insignificant terms (2sigma criterion) are successively excluded before the final MLR run. In order to document the changes from the MLR fits to the period up to and including 2016 as in W18, the results of the different MLR settings applied to the current data for the shorter period is provided in Table S1 (Supplement). Note that the results in Table S1 may differ from W18 as the merged datasets have been updated and data before 2017 may have changed as well.

The following can be concluded from these additional sensitivity tests:

"The inclusion of the dynamical proxies generally improved the MLR fit (r2 and chi values). Except for the NH zonal band (35N-60N) the various MLR settings yield nearly the same post ODS-peak trends for all broad zonal bands (new Table 4). There are, however, larger changes in the trends before the middle 1990s. In the extratropics the early-period trends are lower (-4.0%/decade vs. -1.9%/decade in the NH and -3.1%/decade vs. 1.9%/decade in the SH) in the standard retrieval. This means that atmospheric dynamics and transport changes contributed to lower early-period extratropical total ozone trends in the standard regression (due to the lack of these dynamical terms in the MLR). The opposite is the case in the tropics where the early-period trends in the standard MLR are slightly higher than in the full MLR. This opposite behavior is consistent with ozone transport patterns due to the Brewer-Dobson circulation.

The only significant changes in the post ODS-peak trends are seen in the NH extratropics. In the standard MLR this trend is zero, while the full and iterative MLR show trends of a half per cent per decade. The sum of the ODS-related trend (full MLR) and atmospheric dynamics contribution (difference in the trends between full and standard MLR) cancel to result in a zero trend in the standard MLR. The negative dynamical trend contribution in the NH is further discussed later in the paper. The correlation between regression and observations are substantially lower in the standard retrieval (r2=0.74 vs. 0.88) which means that the standard MLR seems not to capture all variability and changes in total ozone.

The results shown in Table 4 are compared with the results from the MLR applied to the period limited up to 2016 (same period as in W18) as shown in Table S1 (Supplement). Results from the shorter time period are nearly identical to those shown in Table 3. There is one notable change. The uncertainties of the NH trends from the full MLR up to 2020 are reduced such that these trends have become barely significant (2sigma). The Post-ODS-peak trend of the standard MLR is slightly positive up to 2016 but statistically insignificant and within the uncertainties not different from the current results."

I think it is really important to clarify throughout the manuscript (including the title!) what kind of trends the authors talk about. Mostly, the trends that are discussed are the trends that are attributed to the reduction of ODSs in the atmosphere WITHOUT any contribution of dynamics to the trend. In many places this is not totally clear since the trends are only called "recovery trends". However, for me this is the main point of the manuscript and the difference to other studies. It would therefore be extremely important and very helpful if the authors could be more specific in how they name the trends throughout the manuscript (e.g. instead of referring in the abstract in line 11 to "The near global trend of the median of all datasets..." it would be better to be more specific and refer to "The near global ODSrelated trends ...", and specifying this in the title like "Global total ozone recovery trends attributed to ODS changes derived from five merged ozone datasets")

 \rightarrow We agree. The title has been changed accordingly and we made appropriate changes in the text in order to refer to ODS-related rather than recovery trends.

Minor comments:

Line 10: "... is indeed on slowly recovering..." – remove the "on". \rightarrow done

Line 16: data from which phase of CCMI? Please specify. \rightarrow add "(Phase 1 CCMI REF-C2 scenario)"

Line 71: It is not clear in this section what the spatial coverage of the described datasets is. I assume 90S-90N since also polar regions are analyzed. Please add this information to the dataset descriptions. \rightarrow added at the end of the paragraph (*I. 82*): "All datasets cover the entire earth except for months and latitudes under polar night conditions (winter months)."

Line 72: "ground-based" instead of "ground" \rightarrow done

Line 78: "ground based Brewers, ..." - remove the "ground based" since it is already mentioned at the beginning of the sentence. \rightarrow *done*

Line 80: Add also here the information from which phase of the CCMI project simulations was analyzed. \rightarrow changed to "Phase 1 CCMI Initiative" (add "Phase 1")

Line 129: It is not clear how and by whom the ground-based dataset was updated. The references for the dataset are relatively old, therefore it would be good to add a few words on how the dataset was updated to the year 2020. \rightarrow The data set is a data product provided by the WOUDC and updated regularly. It is available from https://woudc.org/archive/Projects-Campaigns/ZonalMeans/. We added this information after the text in line 131: "The data set is a data product provided by the WOUDC and updated regularly"

Line 135: The word "belt" is used here, although it is only explained in the following sentence what exactly is meant by it. This should be switched to make it clearer for the reader what is meant by "belt". \rightarrow We replaced the corresponding sentences (lines 134-137) with "Then, for each station and for each month the deviations from the climatology were calculated, and a zonal mean value for a particular month was estimated as a mean of these deviations. The calculations were done for 5°-wide latitudinal zones. In order to take into account various densities of the network across regions, the deviations of the stations were first averaged over 5° by 30° cells, and then the zonal mean was calculated by averaging these first set of averages over the 5°-wide latitudinal zone." Line 154: the data were bias-corrected. It would be nice to give here a range of biases that needed to be adjusted. I understand that the biases can be different for the broad latitude bands and datasets, but some kind of number/range would be nice here. \rightarrow *The various biases between datasets are irrelevant and do not change the derived trends.*

Line 169: "applies" should be "apply" \rightarrow done

Line 175: "." is missing after the parenthesis. \rightarrow done

Line 176: The year 1996 is the time for maximum EESC concentrations for which region of the globe? Tropics? Everything besides the polar regions? \rightarrow "... and some years later (t0=2000) in the polar regions" is replaced by "except for the polar regions (>60°) where t0=2000" and removed the next sentence.

Line 177: It would be good to give the exact latitude ranges here which define the polar regions. \rightarrow see the previous comment.

Line 190: The end of the sentence is slightly misleading. I would add "for these years" before "were calculated" to clarify that only for the years with too many missing data no annual means were calculated. \rightarrow change second sub phrase after "and" to: "and for these years annual mean data were treated as missing data,"

Line 226: What about the pre-1996 trends? Did they stay very similar to W18 as well? \rightarrow see reply to the general comment above.

Line 248: "agree" instead of "agrees" \rightarrow done

Line 255-257: It might be nice to add here a table with the trends reported from W18 and calculated here. It would provide a nice overview of things that changed and things that stayed roughly the same (just for the multi-observational median, not each individual dataset) \rightarrow see reply to the general comment above and New Table 4 and S1.

Line 269: "ground-based" instead of "ground"? \rightarrow done

Line 285: Are there any studies that report on trends pre-1996 based on regression methods that use also dynamical proxies? There is one looking at ozone soundings at Payerne (Weiss et al., JGR, Vol. 106, D19, 22685-22694, 2001), and one looking at individual TCO station measurements (Maeder et al., 2007, https://doi.org/10.1029/2006JD007694) but there might be even more analyzing total column ozone data with dynamical proxies. As mentioned above, I think it would be helpful to provide an estimate how well the ODS-related trends compare with earlier findings for the pre-1996 period since they did change quite a bit with the introduction of the dynamical proxies. \rightarrow see reply to the general comment above. The older studies mainly used a piecewise linear trend (PLT) model and thus are difficult to compare. In W18 we discuss the various trend models and our decision to use preferably the ILT method in W18 (and this study).

Line 305/306: Couldn't this signal be a spurious regression result where the attribution did not work properly between the trend proxy and the dynamical proxies also including a trend? I think some sensitivity test (as mentioned above) would be helpful here to test the robustness of this signal. \rightarrow MARK (see general comments) \rightarrow see reply to the general comment above. It appears that the post-ODS trends are in most cases unchanged regardless of the number of extra terms used in the MLR. The linear trend term is the only low-frequency term in the MLR equations, while the dynamical proxies have some high-frequency contributions. This makes the trend estimates rather robust and less sensitive to the various other terms used in the MLR.

Line 316: "." missing after the parenthesis. \rightarrow *done*

Line 331: "have" instead of "has" \rightarrow done

Line 366-368. This sentence seems somehow out of place here. I think it needs a little more explanation and detail. \rightarrow We omit this sentence, as we did not discuss the possible impact of tropospheric ozone on column trends. The impact is possibly rather small when using annual and zonal means.

Global total ozone recovery trends <u>attributed to ODS changes</u> derived from five merged ozone datasets

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Abstract. We report on updated trends using different merged zonal mean total ozone datasets from satellite and ground-based
observations for the period from 1979 to 2020. This work is an update from the trends reported in Weber et al. (2018) using the same datasets up to 2016. Merged datasets used in this study include NASA MOD v8.7 and NOAA Cohesive Data (COH) v8.6.

- 4 both based on data from the series of Solar Backscatter UltraViolet (SBUV), SBUV-2, and Ozone Mapping and Profiler Suite (OMPS) satellite instruments (1978–present) as well as the Global Ozone Monitoring Experiment (GOME)-type Total Ozone
- 6 (GTO-ECV) and GOME-SCIAMACHY-GOME-2 (GSG) merged datasets (both 1995–present), mainly comprising satellite data from GOME, SCIAMACHY, OMI, GOME-2A, -2B, and TROPOMI. The fifth dataset consists of the annual mean zonal
- 8 mean data from ground-based measurements collected at the World Ozone and UV Radiation Data Center (WOUDC). Trends were determined by applying a multiple linear regression (MLR) to annual mean zonal mean data. The addition of
- 10 four more years consolidated the fact that total ozone is indeed on slowly recovering in both hemispheres as a result of phasing out ozone depleting substances (ODS) as mandated by the Montreal Protocol. The near global ODS-related ozone trend of the
- 12 median of all datasets after $\frac{1996 \text{ was } 0.51995 \text{ was } 0.4\pm0.2}{1995 \text{ was } 0.4\pm0.2}$ (2 σ) %/decade, which is in absolute numbers roughly a third of the decreasing rate of $\frac{1.41.5\pm0.6}{1.41.5\pm0.6}$ %/decade from 1978 until $\frac{1996.1995}{1995}$. The ratio of decline and increase is nearly identical to that
- 14 of the EESC (equivalent effective stratospheric chlorine or stratospheric halogen) change rates before and after 1996 which confirms-1995 confirming the success of the Montreal Protocol. The observed trends-total ozone timeseries are also in very
- 16 good agreement with the median of 17 chemistry climate models from <u>CCMI (Chemistry Climate Model Initiative CCMI-1</u> (Chemistry-Climate Model Initiative Phase 1) with current ODS and GHG (greenhouse gas) scenarios (REF-C2 scenario).
- 18 The positive ODS related trends in the NH after <u>1996-1995</u> are only obtained with a sufficient number of terms in the MLR accounting properly for dynamical ozone changes (Brewer-Dobson circulation, AO, AAO). A standard MLR (limited to solar,
- 20 QBO, volcanic, and ENSO) leads to zero trends showing that the small positive ODS related trends have been balanced by negative trend contributions from atmospheric dynamics resulting in nearly constant total ozone levels since 2000.

22 1 Introduction

The stratospheric ozone layer protects the biosphere from harmful UV radiation. How much UV reaches the surface depends,

- 24 among other factors like clouds, on the overhead total ozone column. The discovery of the Antarctic ozone hole (Chubachi, 1984; Farman et al., 1985; Solomon et al., 1986) raised the awareness of the need to protect the ozone layer that culminated in
- 26 the 1985 Vienna Convention and a commitment to take actions. One of the actions was the signing of the Montreal Protocol in 1987 that started the phaseout of ozone depleting substances (ODS), which are sufficiently long-lived to reach the stratosphere
- 28 and release active halogens that destroy ozone (e.g. Solomon, 1999). As a consequence of the Montreal Protocol and its later amendments stratospheric halogens started to decline in the middle 1990s (e.g. Anderson et al., 2000; Solomon et al., 2006).
- 30 A corresponding ozone increase has been detected from satellite and ground-based observations, particularly in the upper stratogenere (Braesicke et al., 2018, and references therein).
- 32 Changes in total ozone column are representative of lower stratospheric ozone changes as the majority of ozone resides in the lower stratosphere ("ozone layer"). Lower stratospheric ozone is sufficiently long-lived to be influenced by transport
- 34 and circulation changes. The rapid increase in northern hemisphere total ozone in the late 1990s (Harris et al., 2008) revealed the important role of ozone transport via the Brewer-Dobson (BD) circulation. These circulation changes also cause large
- 36 variability on inter- and intra-annual time scales in lower stratospheric ozone and the total column (e.g. Fusco and Salby, 1999; Randel et al., 2002; Dhomse et al., 2006; Harris et al., 2008; Weber et al., 2011) and make detection of ozone recovery
- 38 challenging. Apart from the observed variability, zonal mean total ozone levels in both hemispheres remained stable since about the year 2000 (e.g. Weber et al., 2018). The success of the Montreal Protocol agreement is nevertheless undisputed as
- 40 the earlier decline in total ozone was successfully stopped (M\u00e4der et al., 2010; Braesicke et al., 2018). Global and continuous ozone observations from satellites through 2020 now span a total time period of forty-two years,
- 42 of which 25 years cover the period after the stratospheric halogen peak (around 19961995). The added years should help in improving the statistical significance of ozone recovery after the middle 1990s (Weatherhead et al., 2000). This paper reports
- 44 on updated zonal mean total ozone trends from Weber et al. (2018) (abbreviated to W18 in the following) by adding four more years of data (2017-2020) to five merged total ozone datasets. In our earlier study ozone recovery trends in the extratropics were
- 46 on the order of ± 0.5 %/decade. The derived trends depend on the proper treatment of dynamical processes in the multi-linear regression. Changes in circulation and ozone transport, in part due to increasing greenhouse gas levels (GHG), have variability
- 48 on decadal and longer time scales and can therefore mask ODS related recovery trends. Longer data records are helpful to further disentangle the various processes responsible for long-term changes in ozone. In this work we focus specifically on
- 50 trend estimates directly related to ODS changes in order to evaluate the direct impact from the Montreal Protocol. The main results from our earlier paper (W18) were latitude dependent annual mean total ozone trends from the middle 1990s
- 52 to 2016, which were reported to be on average +0.5 %/decade in the extratropics and only significant in the SH (W18). Since W18 was published there were three recent studies on global and regional ozone column trends (Bozhkova et al., 2019; Krzyścin and Barane
- 54 (Bozhkova et al., 2019; Krzyścin and Baranowski, 2019; Coldewey-Egbers et al., 2022). Krzyścin and Baranowski (2019) derived total ozone column trends from a multivariate linear regression (MLR) applied to the Multi-Sensor Reanalysis-2 (MSR-2)

- 56 total ozone dataset up to 2017 (van der A et al., 2015). In their MLR they split the entire period from 1978 to 2017 into three periods with separate trends (either independent or piecewise linear). The choice of two inflection points were chosen from fits
- 58 having minimum fit root mean square (rms) errors. As stratospheric halogens are declining steadily since the middle 1990s the interpretation of the segmented trends is difficult. Trends of the first period (before middle 1990s) are in agreement with W18
- 60 and this study.

Bozhkova et al. (2019) applied a regression to TOMS and OMI total ozone at northern hemispheric mid-latitudes using the

- 62 approach by Bloomer et al. (2010), first applied to surface ozone and temperature data at selected stations in the US. Without using any proxy data the regression estimates trends of the seasonality expressed as Fourier series. Attribution of physical and
- 64 chemical processes to the long-term changes are therefore not possible as also stated by the authors. Latitude and longitude dependent total ozone trends are reported by **?** Coldewey-Egbers et al. (2022) derived from the ESA/DLR GTO-ECV dataset,
- 66 which is one of the five observational datasets used in this study. They report significant positive linear trends after 1995 over large regions in the extratropical southern hemisphere, while in the tropics and NH they are mostly insignificant. Consequently,
- 68 they only reported significant zonal mean positive trends in the SH.

In Section 2 the updates in the five merged datasets are briefly discussed. In Section 3 the multiple linear regression (MLR)

- as used in our trend analysis is described and discussed. Section 4 presents the total ozone trend results in broad zonal bands: near-global, southern and northern hemispheric extratropics, and tropics. In Section 5 latitude dependent annual mean total
- 72 ozone trends are presented and discussed. Polar ozone trends for the months where polar ozone losses are largest (e.g. during ozone hole season) are presented in Section 6. In Section 7 a summary and final remarks are given.

74 2 Total ozone datasets

Five merged total ozone datasets are used in this study of which one dataset is based upon ground-based observations.

- 76 All others are based on satellite observations. Two different merged datasets are derived from the series of SBUV and SBUV-2 satellite instruments (SBUV MOD V8.7 from NASA and SBUV COH V8.6 from NOAA) operating continuously since the
- 78 late 1970s. The other two merged datasets are based in large part_largely upon the series of European satellite spectrometers GOME, SCIAMACHY, GOME-2A, and GOME-2B with different retrieval and merging algorithms applied (University of
- Bremen GSG and ESA/DLR GTO-ECV datasets). These datasets start in 1995.
 The ground-based ground-based dataset is the monthly mean zonal mean data from the network of ground-based Brewers,
- 82 Dobsons, SAOZ (Système d'Analyse par Observations Zénithales), and filter instruments collected at the World Ozone and UV Data Center (WOUDC) (Fioletov et al., 2002). In addition a brief description of the model data from the CCMI Phase 1
- 84 initiative is given. The sources of observational data are listed in Table 1 and brief descriptions of the datasets are given in the following. Annual mean timeseries of all five merged datasets are in very good agreement with each other (see to within a few)
- 86 DU (see also Fig. 2.58 in Weber et al. (2021)). All datasets cover the entire earth except for months and latitudes under polar night conditions.

Table 1. Source of merged total ozone datasets.

Dataset	Start year	Source
NASA SBUV MOD V8.7	1970	http://acdb-ext.gsfc.nasa.gov/Data_services/merged/
NOAA SBUV COH V8.6	1978	ftp://ftp.cpc.ncep.noaa.gov/SBUV_CDR/
GSG	1995	http://www.iup.uni-bremen.de/gome/wfdoas
GTO	1995	http://atmos.eoc.dlr.de/gome/gto-ecv.html
WOUDC	1964	http://woudc.org/archive/Projects-Campaigns/ZonalMeans/

88 2.1 NASA SBUV MOD V8.7

The NASA Merged Ozone Data (MOD) time series is constructed using data from the Nimbus 4 BUV, Nimbus 7 SBUV,

90 and six NOAA SBUV-2 instruments numbered 11, 14, and 16-19, and the Ozone Mapping and Profiler Suite Nadir Profiler (OMPS-NP) instrument aboard the Suomi-NPP satellite (Frith et al., 2014, 2022). The instruments are of similar design, and

92 measurements from each are processed using the same V8.7 retrieval algorithm. To maintain consistency over the entire time series the individual instrument records are analyzed with respect to each other and absolute calibration adjustments are applied

94 as needed based on comparison of radiance measurements during periods of instrument overlap (DeLand et al., 2012). Version 8.7 uses the same core algorithm as Version 8.6 (Bhartia et al., 2013) but includes new inter-instrument calibration

- 96 adjustments for instrument records since 2000 (NOAA-16 SBUV/2 though through OMPS NP) based on a new approach to radiance intercomparisons across overlapping instruments (Kramarova et al., 2022). Version 8.7 also incorporates an updated
- 98 a-priori with improved tropospheric representation based on GMI model output, and diurnal adjustments to ensure the a-priori profile correctly reflects the local solar time of each measurement (Ziemke et al., 2021). A post-retrieval diurnal correction is
- 100 applied to adjust each instrument record to an equivalent measurement time of 1:30pm (Frith et al., 2020). Remaining offsets between instruments exist (mostly below 5% for layers, below 1% for total ozone), but their cause is not understood. We
- 102 therefore do not make adjustments to the data. Rather we set limitations on the data included in the merged product based on data quality analysis by the instrument team and on comparisons with independent measurements (DeLand et al., 2012;
- 104 Kramarova et al., 2013, 2022). For merging, data are averaged during periods with multiple operational instruments. The Version 8.7 MOD data contains monthly zonal mean ozone profiles in mixing ratio on pressure levels and in Dobson units on
- 106 layers. The total ozone is then provided as the sum of the layer data.

2.2 NOAA SBUV COH V8.6

- 108 The NOAA COH (cohesive) dataset is a simple extension in time of the dataset appearing in W18. The data includes v8.6 SBUV on Nimbus 7, v8.6 SBUV/2 from NOAA 9, 11, 16 to 19, and v2r3-v2r2 OMPS Nadir Profiler (NP) on Suomi-NPP as
- 110 available from NESDIS STAR. The merging approach differs from NASA MOD in two important ways. NASA MOD averages data from all relevant satellites in any time period for which the data meets certain quality criteria. NOAA COH uses data from

- 112 a single 'best' satellite in any time period. Which satellite is used depends on known data quality issues, on minimizing the solar zenith angle of the measurement, and on maximizing global coverage. NOAA COH does not shift to a an equivalent
- 114 measurement time (1:30pm), but performs an adjustment between data from differing satellites. For post 2000 data, where drift of the measurement time is minimized, the data are all adjusted to NOAA 18. For data 1999 and prior, the inter-satellite
- 116 overlap is often short, the satellite drift often significant, we choose only to adjust NOAA 9 to the two branches NOAA 11 prior and after the NOAA 9 time period. The total ozone is calculated from the sum of the adjusted profile layer data. By vertical
- 118 integration many of the layer adjustments to a large extent cancel such that the final total ozone product is altered by less than 1%, and in most cases by less than 0.5%, from the original satellite datasets.

120 2.3 University of Bremen GSG

The merged GOME, SCIAMACHY, GOME-2A and -2B (GSG) total ozone timeseries (Kiesewetter et al., 2010; Weber et al., 2011, 2018) consists of total ozone data that were retrieved using the University of Bremen Weighting Function DOAS (WF-DOAS) algorithm (Coldewey-Egbers et al., 2005; Weber et al., 2005; Orfanoz-Cheuquelaf et al., 2021). The merging of the

- 124 data has been described in W18. The most recent modification was to replace GOME-2A data after January 2015 with data from GOME-2B (2012-present) which has a better global coverage after changes in the GOME-2A scanning pattern. Latitude
- 126 dependent bias corrections for GOME-2B were applied from the overlapping period 2014-2020 with GOME-2A.

2.4 DLR/ESA GTO-ECV

- 128 The latest version of the GOME-type Total Ozone Essential Climate Variable (GTO-ECV) data record (Coldewey-Egbers et al., 2015; ?; Gauge et al., 2015, 2022; Garane et al., 2018) has been generated as part of the European Space Agency's Climate
- 130 Change Initiative+ ozone (ESA_CCI+ ozone) project. Total columns from six sensors (GOME, SCIAMACHY, OMI, GOME-2A, GOME-2B, and TROPOMI), retrieved with the GOME Direct Fitting (GODFIT) version 4 algorithm (Lerot et al., 2014;
- Garane et al., 2018), were combined into a coherent record that covers the period 1995-2020. OMI was used as a reference instrument and the other sensors were adjusted by means of latitude and time dependent correction factors determined from
 overlap periods.

2.5 WOUDC data

- 136 The WOUDC ground-based zonal mean data set (Fioletov et al., 2002) was formed from ground-based measurements by Dobson, Brewer, SAOZ instruments, and filter ozonometers available from the WOUDC. The overall performance of the ground-
- 138 based network was discussed by Fioletov et al. (2008) and the present state of the network is described by Garane et al. (2019) This data set is provided by the WOUDC and updated regularly.
- First, ground-based measurements were compared with an ozone "climatology" (monthly means for each point of the globe) estimated from satellite data for 1978–1989. Then, for each station and for each month the deviations from the climatology

- 142 were calculated, and the belt's a zonal mean value for a particular month was estimated as a mean of these deviations. The calculations were done for 5° latitudinal belts.
- [°]-wide zonal bands. In order to take into account various densities of the network across regions, the deviations of the stations were first averaged over 5° by 30° cells, and then the <u>belt-zonal</u> mean was calculated by averaging these first set of
- 146 averages over the belts^{5°} -wide zonal band. Then the zonal averages were smoothed by approximating them using Legendre polynomials.
- The WOUDC data set was compared with merged satellite time series and demonstrated a good agreement (Chiou et al., 2014). Estimates based on relatively sparse ground-based measurements, particularly in the tropics and southern hemisphere,
- 150 may not always reproduce monthly zonal mean fluctuations well. However, seasonal (and longer) averages can be estimated with a precision comparable with satellite-based data sets ($\sim 1\%$) (Chiou et al., 2014).

152 2.6 Chemistry climate Chemistry-climate model data

In this study output from the chemistry–climate models (CCMs) and chemistry-transport models (CTMs) participating in phase 1 of CCMI (Chemistry-Climate Model Initiative) are used (Eyring et al., 2013). An overview of the models, together with details particular to each model and an overview of the available simulations, is given in Morgenstern et al. (2017) along

- with a detailed description of the full forcings used in the reference simulations (Eyring et al., 2013; Hegglin et al., 2016).Here we have used median total column ozone from 17 models taking part in the REF-C2 experiment, an internally consistent
- seamless simulation from the past into the future between 1960 and 2100.

2.7 Data preparation

- 160 From the zonal mean monthly mean data in 5° latitude steps (all datasets) annual means were calculated. Wider zonal bands (like $35^{\circ}N-60^{\circ}N$) were averaged from the 5° data using area weights (see W18). All annual mean zonal mean timeseries were
- 162 bias corrected by subtracting the difference to the mean of all datasets during the 1998-2008 period. The multi-dataset mean was then added back to each dataset, such that all bias corrected timeseries are provided in units of the total column amounts
- 164 (W18). However, the trend results derived from them are identical to those derived using anomaly timeseries. Like in our earlier study, the GSG and GTO-ECV timeseries were extended from 1995 back to 1979 using the bias corrected
- 166 NOAA data. This way one ensures that all terms other than the trend terms are determined from the full time (1979-2020) period. The NOAA data was here preferred over the NASA data, as the former has shorter data gaps after the major volcanic
- 168 eruption from Mt Pinatubo in 1991 and subsequent years.

3 Multiple linear regression

170 The standard MLR model is identical to the one used in W18 and includes two independent linear trend terms (before and after the ODS related turnaround year $t_0 = \frac{19961995}{1995}$), two aerosol terms (Mt. Pinatubo 1992 and El Chichón 1983), solar cycle 172 term, two QBO terms (50 and 10 hPa), and ENSO (El Niño-Southern Oscillation):

$$y(t) = [a_1 + b_1 \cdot (t_0 - t)] X_1(t) + [a_2 + b_2 \cdot (t - t_0)] X_2(t) + \alpha_{sun} \cdot S(t) + \alpha_{qbo50} \cdot Q_{50}(t) + \alpha_{qbo10} \cdot Q_{10}(t) + \alpha_{ENSO} \cdot E(t) + \alpha_{ElChichón} \cdot A_1(t) + \alpha_{Pinatubo} \cdot A_2(t) + P(t) + \epsilon(t).$$
(1)

- 174 y(t) is the annual mean zonal mean total ozone timeseries and t the year of observations. The coefficients b_1 and b_2 are the linear trends before and after t_0 . In order to make both trends independent of each other (or disjoint), two y-intercepts (a_1 and a_2)
- are added. The multiplication of the independent variable t with X_i(t) in the first four terms of Eq. 1 describes mathematically that the first two terms only applies apply to the period before and the third and fourth terms to the period after the turnaround
 year. X₁(t) and X₂(t) are given by
 - $X_1(t) = \begin{cases} 1 & \text{if } t \le t_0 \\ 0 & \text{if } t > t_0 \end{cases}$

180 and

$$X_{2}(t) = \begin{cases} 0 & \text{if } t \le t_{0} \\ 1 & \text{if } t > t_{0} \end{cases},$$
(3)

(2)

- 182 respectively. The independent trends before and after t_0 are favored over the use of piecewise linear trends or the use of EESC as a proxy timeseries (see detailed discussions in W18). The maximum of the effective equivalent stratospheric chlo-
- rine (EESC) was reached at about the year $t_0 = 1996$ (Newman et al., 2007) and some years later ($t_0 \sim 2000$) in the polar regions (Newman et al., 2006, 2007). Therefore t_0 was set to 1996 globally, $t_0 = 1995$ (Newman et al., 2007) except for the
- polar regions, where $t_0=2000$ was selected region (> 60°) where $t_0=2000$ (Newman et al., 2006, 2007). The contributions from the QBO, 11-year solar cycle, and stratospheric aerosols are standard in total ozone MLR analyses (e.g. Staehelin et al.,
- 188 2001; Reinsel et al., 2005). $\epsilon(t)$ is the residual from fitting the coefficients to match the regression model (right side) to the observations. By using annual mean total ozone, auto-correlation is very low here (below 0.1 in absolute value for a shift by
- 190 one year) so that no further additional auto-regression term as commonly used for monthly mean ozone timeseries is needed (e.g. Dhomse et al., 2006; Vyushin et al., 2007).
- 192 The stratospheric aerosols are dominated by the major volcanic eruptions from El Chichón (1982) and Mt. Pinatubo (1991). Enhanced aerosols in the lower stratosphere lasting for a few years impact both ozone chemistry and transport (Schnadt Poberaj
- 194 et al., 2011; Dhomse et al., 2015). The stratospheric aerosol optical depth (SAOD) at 550 nm from Sato et al. (1993) is used as the explanatory variable before 1990 (includes the El Chichón event), while newer data from the WACCM model (Mills et al.,
- 196 2016) is used for the period after 1990 (includes Mt. Pinatubo major volcanic eruption and the series of more minor volcanic eruptions from the last decade). Missing years after 2015 were filled with background values from the late 1990s.
- As mentioned in W18 there are not is not a sufficient number of months and/or 5° latitude bands available in the SBUV data records for some years and thus no for these years annual means were calculated treated as missing data. Annual means were

Variable	Proxy	Source			
S(t)	Bremen composite Mg II index	http://www.iup.uni-bremen.de/UVSAT/Datasets/mgii			
	(Snow et al., 2014)				
$QBO_{50}(t), QBO_{10}(t)$	Singapore wind speed at 50 and $10 \mathrm{hPa}$	http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/qbo.dat			
	(update from Naujokat, 1986)				
E(t)	MEI (ENSO) Index	https://www.esrl.noaa.gov/psd/enso/mei/			
	(Wolter and Timlin, 2011)				
AO(t), AAO(t)	Antarctic Oscillation (AAO), Arctic	http://www.cpc.ncep.noaa.gov/products/precip/CWlink/daily_			
	Oscillation (AO)	ao_index/teleconnections.shtml			
$A_1(t)$	stratospheric aerosol depth at 550nm	https://data.giss.nasa.gov/modelforce/strataer/tau.line_2012.			
	(t < 1990)	12.txt			
	(update from Sato et al., 1993)				
$A_2(t)$	stratospheric aerosol depth at 550nm	http://dx.doi.org/10.5065/D6S180JM			
	from WACCM model ($t \ge 1990$)				
	(Mills et al., 2016)				

Table 2. Sources of explanatory variables / proxy timeseries used in the MLR.

200 only used in the regression if at least 80% of the 5° bands of the data were contained in the broad zonal bands and 80% of months available in that year. If annual means of the years 1982 and 1983 are missing, the "El Chichon" term is not used in the

202 MLR, similarly if missing all years from 1991 to 1994, the "Pinatubo" term is excluded in the MLR.

The MLR equation, Eq. 1, without the P(t) term has been commonly applied for determining trends from ozone profile data

- 204 (e.g. Bourassa et al., 2014; ?; Harris et al., 2015; Tummon et al., 2015; Sofieva et al., 2017; Steinbrecht et al., 2017)(e.g. Bourassa et al., 2 . The extra term P(t) in Eq. 1 accounts for additional factors of dynamical variability that have been used in different combina-
- tions and definitions (e.g. accumulated, time-lagged) in the past. It includes contributions from the Arctic (AO) and Antarctic Oscilation (AAO), and the Brewer-Dobson circulation (BDC) (e.g. Reinsel et al., 2005; Mäder et al., 2007; Chehade et al.,
- 208 2014; Weber et al., 2018). The BDC terms are usually described by the eddy heat flux at 100 hPa that is considered a main driver of the BDC (Fusco and Salby, 1999; Randel et al., 2002; Weber et al., 2011). The term P(t) is given as follows:

210
$$P(t) = \alpha_{AO} \cdot AO(t) + \alpha_{AAO} \cdot AAO(t) + \alpha_{BDCn} \cdot BDCn(t) + \alpha_{BDCs} \cdot BDCs(t).$$
(4)

In W18 the AAO term was not included. Table 2 summarises the sources of the proxy data used here. The <u>BDCn and BDCs</u> are 100 hPa eddy fluxes in the northern (n) and southern hemisphere (s). The calculation of the BDC proxy from the monthly

mean eddy heat fluxes is described in detail in W18. In this study the eddy heat flux data come were derived from the ERA-5

214 reanalysis (Hersbach et al., 2020).

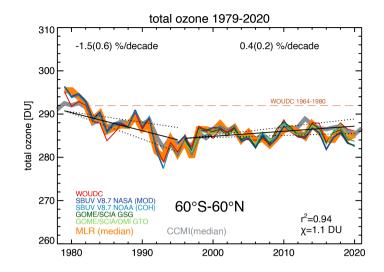


Figure 1. Near global (60°S-60°N) total ozone timeseries of five bias corrected merged datasets. The thick orange line is the result from applying the full MLR (Eqs. 1 and 4) to the median timeseries. The square of the correlation between observations and MLR is given by r^2 . χ^2 is the sum square of the <u>differences between</u> median timeseries minus observational and MLR timeseries divided by the degrees of freedom (difference between the number of years, *n*, and number of parameters used in the MLR, *m*). The solid lines indicate the linear trends before and after the ODS peak, respectively. The dotted lines indicate the 2σ uncertainty of the MLR trend estimates. Trend numbers are indicated for the pre- and post-ODS peak period in the top part of the plot. Numbers in parentheses are the 2σ trend uncertainty. The orange dashed line shows the mean ozone level from 1964 until 1980 from the WOUDC data. The thick grey line is the median of 17 chemistry-climate models from the CCMI initiative.

One may argue that the addition of P(t) will lead to some overfitting by the MLR. We justify this addition as it enables us to obtain MLR fits matching the extreme events like very high annual mean ozone in the NH in 2010 and the very large warming events above Antarctica in 2002 and 2019 with unusually high ozone. The better the dynamical variations are represented in the MLR, the more likely we can separate out dynamical trend contributions and the linear trend terms best approximate EESC

- related trends. In our previous study only selected terms from P(t) were used dependent on their significance in specific zonal bands. Retaining all terms in all MLRs leads to smoother behavior in the latitude dependent ozone response.
- The various proxy time series, in particular the atmospheric dynamics related ones, are partially correlated. One way to improve upon this is the possibility to orthogonalize them. Doing so will not change the MLR fit results, but some contributions from the original proxy terms will be redistributed among the proxies that were orthogonalized. It is also common to detrend
- the proxy time series. In that case all linear changes of the various processes or proxies will be added up in the linear trend term which makes attribution impossible. The For these reasons we do not detrend nor orthogonalise the proxy timeseries in
- 226 <u>this study. Our</u> goal here is that linear changes of all the processes as expressed by the various proxy terms shall be excluded from the linear trend terms such that the linear trends can be attributed as close as possible to ODS changes.

228 4 Total ozone trends in broad zonal bands

Figure 1 shows the near-global mean timeseries (60° S- 60° N) of the bias-corrected five five bias-corrected merged datasets. The

- thick orange line is the MLR result timeseries from applying the full regression model (Eqs. 1 and 4) to the median of the five timeseries. 94% of the variability in total ozone is well captured by the full MLR. A positive trend of $+0.4\pm0.2(2\sigma)$ %/decade
- after $\frac{1996}{1995}$ is derived. This trend is about one third of the absolute trend during the phase of increasing ODS before $\frac{1996}{1995}$ which is -1.5 ± 0.6 %/decade. The ratio of trends before and after $\frac{1996}{1995}$ is very close to the ratio of rate changes in
- 234 the effective equivalent stratospheric chlorine (EESC) before and after the middle 1990s (Dhomse et al., 2006; Newman et al., 2007). Therefore, the observed linear trend of roughly half a percent per decade up to 2020 can be interpreted as the recovery
- 236 from changes attributed to reductions in ODS following the Montreal Protocol. This ODS related recovery appears statistically robust (to within 2σ), even though the ozone levels have stayed more or less constant apart from the year-to-year variability
- since the year 2000. The magnitude of the post ODS-peak trend remained unchanged from W18. The trend results vary only slightly if the turnaround year (19961995) of the ODS change is shifted by one year back and forward. Even if the MLR fit of
- 240 the post ODS-peak period is limited to years after 2000, the recovery ODS-related trend remains robust at +0.5(0.3) %/decade. The current near-global ozone level (2017-2020) is about 2.3% below the average from the 1964-1980 time period, the latter
- 242 derived from the WOUDC data (see Fig. 1). Recovery of total ozone to the 1980 level is generally not expected before about the middle of this century (Braesicke et al., 2018). The near-global total ozone timeseries from the median of the seventeen CCMI
- 244 chemistry-climate models is in very good agreement with the observations from which we conclude that the chemical and dynamical changes in total ozone under current ODS and greenhouse gas (GHG) scenarios are well understood and consistent
- 246 with observations.

Figure 2 shows the ozone time series in the northern (NH) and southern hemisphere (SH) as well as in the tropics. Again, the current ozone levels are well below the 1964-1980 mean, specifically -3.6% and -4.7% in the NH and SH (35° - 60° latitudes),

respectively. The lower value in the SH is due to the influence from the spring Antarctic ozone hole, which exhibits the largest

- 250 local ozone depletion and leads to mixing of ozone depleted air into the middle latitudes (Atkinson et al., 1989; Millard et al., 2002). Recovery-ODS-related trends are +0.5(0.5) and +0.7(0.6) %/decade in the NH and SH, respectively. Within the trend
- uncertainty, the 1-to-3 ratio in the linear trends before and after the ODS peak in <u>1996-1995</u> are close to the ratio of the rate change in the EESC in both hemispheres.
- In the tropics the linear trend after 1996 1995 is close to zero and insignificant (Fig. 2 and ?)(Fig. 2 and Coldewey-Egbers et al., 2022). Table 3 summarises the MLR results in the broad zonal bands from the individual datasets and the median timeseries as well
- as the mean and median of the individual trends.

In most cases the results from the individual datasets are highly consistent in particular for the near-global time series. All

- datasets indicate significant near-global recovery ODS-related trends of around half a percent per decade. The trend derived from the NASA data is a bit lower at +0.2 %/decade. The median and mean trends of all datasets agrees here with
- the trends of the median timeseries as shown in Figs. 1 and 2. For the narrower zonal bands not all datasets show significant trends after 1996. 1995. The NASA and GSG datasets show low recovery lower trends in the NH (+0.3 and +0.1 %/decade,

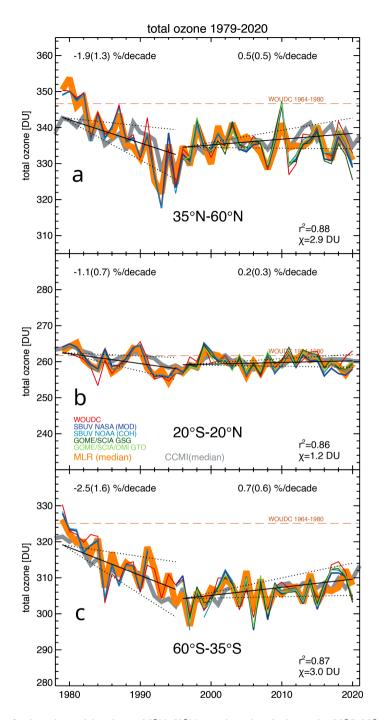


Figure 2. Same as Fig. 1, but for broad zonal bands, a) $35^{\circ}N-60^{\circ}N$ (northern hemisphere), b) $20^{\circ}S-20^{\circ}N$ (tropics), and c) $35^{\circ}S-60^{\circ}S$ (southern hemisphere).

- respectively), while all others are between ± 0.5 and ± 0.7 %/decade and significant. In the SH all recovery trends agree to within one tenth %/decade (+0.7 %/decade), except for the NOAA dataset showing a somewhat higher trend of ± 1.00 %/decade.
- 264 In the tropies the recovery trends are close to zero with the exception of the GSG and GTO datasets that have very small and barely significant positive recovery trends of $+0.3\pm0.3$ %/decade. The variations in the trend results from the different datasets
- 266 is most likely due to some residual drifts in the datasets that are not accounted for in the data merging. With the use of the full MLR with all terms and with four years added in the timeseries', the ozone trends in the various zonal bands before and after
- 268 1996 remain quite similar to the results reported in W18, but uncertainties are slightly reduced.

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- 272 terms and with four years added in the timeseries, the ozone trends in the various zonal bands after 1995 remain quite similar to the results reported in W18, but uncertainties are slightly reduced.
- Table 4 shows different MLR settings applied to the median total ozone timeseries in broad zonal bands (as defined in Table 3). Here the results from the standard and full MLR are listed. In addition, we applied an iterative MLR approach where
- 276 statistically insignificant terms (2σ criterion) from Eq. 4 and the El Niño term are successively excluded before the final MLR run. The inclusion of the dynamical proxies generally improved the MLR fit (r2 and chi values). Except for the NH zonal band
- 278 (35N-60N) the various MLR settings yield nearly the same post ODS-peak trends for all broad zonal bands (Table 4). There are, however, larger changes in the trends before the middle 1990s. In the extratropics the early-period trends are lower in the
- 280 standard retrieval.(-4.0 versus -1.9 %/decade in the NH and -3.1 versus -1.9 %/decade in the SH). This means that atmospheric dynamics and transport changes contributed to lower early-period extratropical total ozone trends in the standard regression
- 282 (due to the lack of these dynamical terms in the MLR). The opposite is the case in the tropics where the early-period trends in the standard MLR are slightly higher than in the full MLR. This opposite behavior is consistent with ozone transport patterns
- 284 due to the Brewer-Dobson circulation.

It appears that the post-ODS trends are in most cases unchanged regardless of the number of extra terms used in the MLR.

286 The linear trend term is the only low frequency term in the MLR equations, while the dynamical proxies have some high frequency contributions. This makes the trend estimates rather robust and less sensitive to the various other terms used in the

288 MLR. The only significant changes in the post ODS-peak trends are seen in the NH extratropics. In the standard MLR this trend is zero, while the full and iterative MLR show trends of a half percent per decade. The sum of the ODS-related trend (full

- 290 MLR) and atmospheric dynamics contribution (difference in the trends between full and standard MLR) cancel to result in a zero trend in the standard MLR. The negative dynamical trend contribution in the NH is further discussed later in the paper. The
- 292 correlation between regression and observations are substantially lower in the standard retrieval ($r^2 = 0.74$ versus 0.88), which indicates that the standard MLR seems not to capture all variability and changes of total ozone. The results shown in Table 4
- are compared with the results from the MLR applied to the period through 2016 (same period as in W18) as shown in Table S1 (Supplement). Results from the shorter time period are nearly identical to those shown in Table 3. There is one notable
- 296 change. The uncertainties of the NH trends from the full MLR up to 2020 are reduced such that these trends have become

barely significant (2sigma). The Post-ODS-peak trend of the standard MLR is slightly positive up to 2016 but statistically

- 298 insignificant and within the uncertainties not different from the current results.
 In order to document the changes from the MLR fits (Table 4) to results from the period up to and including 2016 (as in W18).
- 300 the different MLR settings were applied to the current data for the shorter period as summarised in Table S1 (Supplement). Note that the results in Table S1 may differ from W18 as the merged datasets have been updated and data before 2017 may
- 302 have changed as well. Results from the shorter time period are nearly identical to those shown in Table 4. There is one notable change. The uncertainties of the NH trends from the full MLR up to 2020 are reduced such that these trends have become barely
- significant (2σ) . The post ODS-peak trend of the standard MLR is slightly positive up to 2016 but statistically insignificant and within the uncertainties not different from the current results.

306 5 Latitude dependent total ozone trends

Latitude dependent trends in steps of 5° are shown from 60°S to 60°N for all five merged datasets (thin lines) in Fig. 3. The 308 two thick blue and red lines are the results before and after 1996-1995 from applying the full MLR to the median timeseries including 2σ uncertainties shown as error bars. In the extratropics the recovery ODS-related trends are on the order of +0.5%

- 310 $\frac{\%}{\text{decade}}$ with 2σ uncertainties of about the same magnitude. In the SH the recovery trends continuously increase to nearly +1.3 %/decade in the 55°S -60°S band while in the NH the recovery trends remain unchanged up to the highest latitudes shown.
- 312 In the tropics recovery trends are close to zero. One notable change from W18 is that the tropical trends during the ODS rising phase are now more negative (down to -1 %/decade) while before they were mainly close to zero. This may be caused by the
- 314 additional proxy terms used in this study.

After $\frac{1996}{1995}$ all trends of all datasets are in good agreement to within ± 0.3 %/decade. There are some notable differences

- in the northern subtropical and northern tropical trends for the WOUDC data (up to +1 %/decade) compared to the other datasets, which is most likely caused by larger uncertainties due to the sparsity of ground-ground-based data at these latitudes.
- The trend uncertainties are generally larger for the early period before 1996, which in part may be is caused by the different lengths of the periods before 1996 (17 years) and after 1996 1995 (25 years).
- The dashed pink line shows the expected recovery <u>ODS-related</u> trends when applying the 1-to-3 ratio (corresponding to the rate change of the EESC) to the trends before 1996. It agrees quite well in the extratropics with the independent linear
- 322 trend estimates and therefore give us confidence that ozone is responding to the long-term ODS decline. The expected tropical recovery ODS-related trends are slightly positive while the MLR regressions suggest rather near zero trends, but they still agree
- 324 within their uncertainties. In the NH extratropics the expected ODS related trend is slightly higher than the observed trends, but also agree within the uncertainties of the observed trends.
- In order to elucidate further on the interpretation of the independent linear trends after 1996 as recovery trends 1995, we repeated the analyses using the standard MLR which excludes several terms responsible for changes in atmospheric dynamics
- and transport (Eq. 1 with P(t) = 0). The latitude dependent trends from the standard MLR are shown in Fig. 4. While the recovery trends observed trends for both MLRs are nearly unchanged in the SH, the NH recovery trends are reduced to zero in

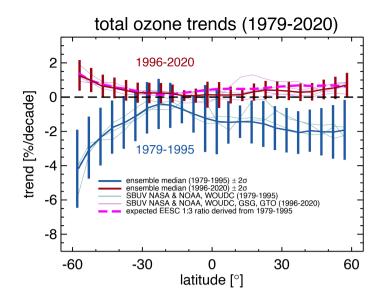


Figure 3. Latitude dependent ozone trends in steps of 5° from applying the full MLR (Eqs. 1 and 4) to the median timeseries of the five merged total ozone datasets. Trends and 2σ standard deviations are shown in blue for the time period before 1996-and in red after 1996. 1995. The thin lines show the trends of the individual total ozone datasets. The pink dashed lined are the post-ODS peak trends as expected from the 1-to-3 ratio (corresponding to changes in the stratospheric halogen) applied to the median timeseries' trends before 1996.

- the NH extratropics. On the other hand the tropical trends before 1996 are closer to zero. The expected recovery-ODS-related trends (from the 1-to-3 EESC ratio) have become larger with increases to +1.5 %/decade at the higher latitudes now in both
- 332 hemispheres. The most obvious result is that the independent linear trends after <u>1996_1995</u> in the NH being close to zero now clearly deviate from the expected 1-to-3 ratio. It appears that the additional atmospheric dynamics terms in the regression
- 334 balance the positive recovery trends from the full MLR which explains why total ozone in the NH appears more or less stable during the last two decades (panel a of Fig. 2).
- The declining trends in the NH before 1996 (Fig. 4a) are stronger in the standard MLR and are comparable to the SH (about -4 %/decade near 60° latitude). On the other hand ODS related trends are expected to be somewhat stronger in the SH as the
- 338 influence from polar ozone losses on mid-latitude ozone is thought to be larger in the SH, since Arctic ozone losses are more sporadic and generally smaller. In that regard the trends from the full MLR seem to support this notion.
- 340 The comparisons of trends from the standard and full MLR reveal that the NH <u>ODS-related</u> ozone recovery is balanced by long-term changes in atmospheric dynamics (circulation and transport changes) or in other words the near zero linear post
- 342 ODS-peak trends are caused by the combination of ODS-related recovery and dynamical changes. These two signals are more clearly separated in the full MLR. Before discussing this further, we will take a look at the contributing factors or terms in the
- 344 MLR. Figure 5 shows the maximum response of the various terms in Eqs. 1 and 4 as a function of latitude (from the fit to the median timeseries).

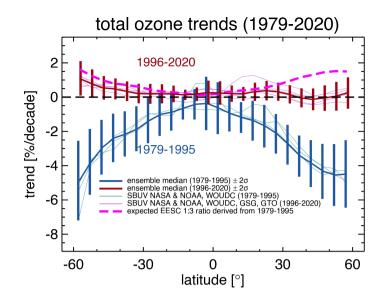


Figure 4. Same as Fig. 3, but from applying the standard MLR (Eq. 1 and P(t) = 0)).

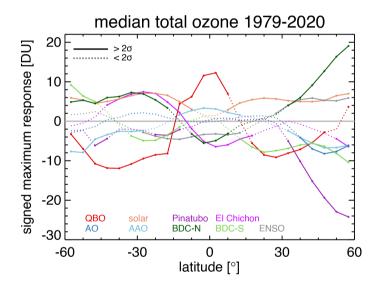


Figure 5. Signed maximum timeseries contribution from various terms in the full MLR equation applied to the median of the five merged total ozone datasets. Solid line indicates values of fit coefficients that are larger than their 2σ uncertainty. The sign of the BDCs proxy time series is reversed, so that in both hemispheres positive BDC term values correspond to enhanced Brewer-Dobson circulation. Negative values an anti-correlation of the ozone response to the proxy. For instance, positive solar contributions mean high solar activity leads to more ozone.

Well-known factors like solar activity and QBO show the expected behaviour, i.e. more ozone during solar maximum at all latitudes (see e.g. W18) and the opposite sign in the QBO response between inner tropics and extratropics (Bowman, 1989; Baldwin et al., 2001). The solar response is of similar magnitude at all latitudes, which means that the solar effect in

the lower stratosphere is mostly indirect via changes in temperature and associated atmospheric circulation changes (e.g.?)

350 (e.g. Dhomse et al., 2022).

In the NH the BDC and and AO mostly contribute to ozone variability. Interestingly, there is an influence from the BDC from

- 352 one hemisphere to the other in both directions. BDC-N results in opposite responses in the tropics and NH extratropics. This is expected from the planetary waves driving the BDC leading to ascent in the tropics (lower ozone) and descent in the polar
- region (higher ozone) (e.g. Randel et al., 2002; Weber et al., 2011). The correlation of ozone anomalies in the NH winter/spring to SH total ozone was reported by Fioletov and Shepherd (2003) and is believed to explain the positive response in SH total
- 356 ozone. Somewhat surprising is the impact of the SH BDC on NH ozone with a negative ozone response, for which we have no explanation.
- 358 The major volcanic eruption of Mt. Pinatubo in 1991 had a stronger impact on the NH reducing ozone for several years after the event, while ozone advection apparently balanced the surface acid particle (aerosol) related ozone losses in the SH
- 360 (Schnadt Poberaj et al., 2011; Aquila et al., 2013; Dhomse et al., 2015). The second large major volcanic eruption from spring 1982 lead to aerosol related ozone loss in the tropics and NH, while surprisingly a positive ozone response in the SH is seen
- 362 possibly related to some atmospheric circulation changes compensating chemical effect effects from the El Chichon eruption. In contrast to Mt. Pinatubo, which spread sulfuric acid particles into both hemispheres, enhanced aerosols from El Chichon
- 364 were confined to lower latitudes in the NH (McCormick and Swissler, 1983) consistent with the region of negative ozone response shown in Fig. 5.
- 366 The main reason for stable ozone levels observed in the NH at NH mid-latitudes since 2000 were identified to stem from the balancing of the positive observed recovery ODS-related trend by negative trends due to circulation changes and ozone
- transport (see Figs. 2 and 3). The change in the BDC-n proxy and AO over the last 55 years is shown in Figure 6 along with March total ozone northward of 40°N. The variability in the extratropical annual mean is usually dominated by the variability
- 370 in winter/spring, where BDC maximizes in the seasonal cycle. Apart from the strong drop in ozone in the 1990s related to the major volcanic eruption and associated circulation changes, NH total ozone has been steadily declining over the last 55 years
- 372 (about 25 DU). This decline is coherent with an overall positive shift of the AO index. A weakening of the BDC is also seen but appears less clear than for the AO.
- A positive shift in the AO and a weakening of the BDC results in a strengthening of the polar vortex, which is associated with larger polar ozone losses (Lawrence et al., 2020). Hu et al. (2018) linked a recent strengthening of the stratospheric Arctic
- 376 vortex in part to a warming of sea surface temperatures in the central northern Pacific. A recent downward trend in extratropical lower stratospheric ozone has been reported by Ball et al. (2018) that could be consistent with the total ozone observations.
- 378 Other studies with many different ozone profile datasets did not show significant trends in the lower stratosphere due to very large variability and lower accuracy of the satellite data in this altitude region (Sofieva et al., 2017; Steinbrecht et al., 2017;
- 380 Arosio et al., 2019).

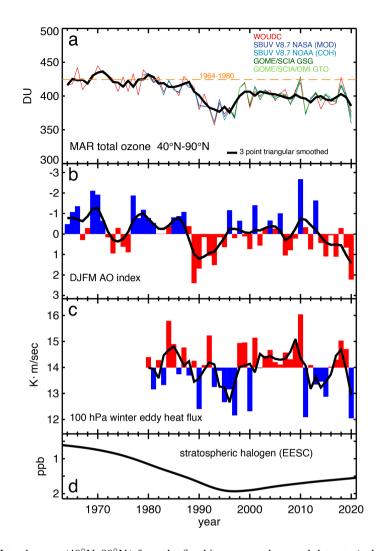


Figure 6. Panel a: March NH total ozone $(40^{\circ}N - 90^{\circ}N)$ from the five bias-corrected merged datasets (colored) and the smoothed median timeseries (thick black line). Panel b: DJFM Arctic oscillation (AO) index. Black lines shows the three-point triangular smoothed timeseries. Note the inverted y-scaleseries. Panel c: 100 hPa winter eddy heat flux September to March average (BDCn proxy) with black line showing the three-point triangular smoothed timeseries. Panel d: Inverted stratospheric halogen timeseries in ppb representive for middle latitudes (Newman et al., 2007).

6 Trends in polar spring

- Earlier signs of ozone recovery has been were observed in September above Antarctica (Solomon et al., 2016, W18). Now, with four more years of data this recovery of about 11+12 %/decade remains robust (see panel b of Fig. 7). During September,
 the Antarctic ozone hole size usually increases and reaches its maximum in late September and early October. In a typical
 - Antarctic winter, ozone is completely destroyed in the lower stratosphere, which may explain why no recovery is yet observed

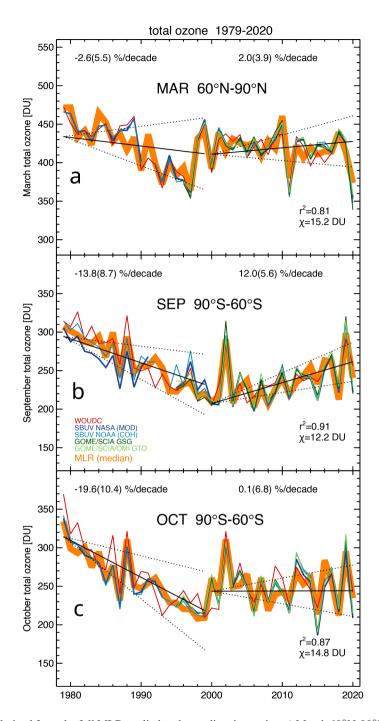


Figure 7. Polar ozone trends derived from the full MLR applied to the median timeseries. a) March 60° N- 90° N, b) September and c) October 60° S- 90° S. see Fig. 1 for more details.

- 386 in October over the polar cap (panel c in Fig. 7). Despite the lack of recovery in the late ozone hole season, several Several diagnostics clearly indicate a healing of Antarctic ozone as a consequence of the Montreal Protocol. Stone et al. (2021) show
- that the onset of the Antarctic ozone hole has been shifted to later dates despite the larger than average ozone holes observed in recent years (e.g. 2015 and 2020).
- Panel a of Fig. 7 shows the March ozone timeseries above the Arctic with ozone recovery ODS-related ozone trends not statistically different from zero. The trend results in the polar regions as shown in Fig. 7 basically confirm the results from W18
- and Langematz et al. (2018). Table 5 summarises the polar trends for the individual datasets and the mean timeseries. Within the trend uncertainties all datasets are in very good agreement.

394 7 Summary and conclusions

We derived globally total ozone recovery-trends from five merged total ozone datasets using a multiple linear regression with independent linear trend (ILT) terms before and after the turnaround in stratospheric halogens in the middle 1990s (~2000 in the polar regions). When properly accounting for dynamical changes via atmospheric circulation and transport, these retrieved

- 398 trends may be interpreted as recovery trends directly related to changes in the stratospheric halogens (and ODS) as a response to the Montreal Protocol and Amendments phasing out ozone depleting substances.
- For the near-global average we see small recovery <u>ODS-related</u> trends of about ±0.5 %/decade with main contributions from the extratropics in both hemispheres. The ratio of ozone trends after and before the turnaround year is in very good agreement
 with the trend ratios in stratospheric halogens or ODS.

In the tropicsrecovery, trends are not statistically different from zero. In line with earlier observations (Solomon et al.,

2016, W18), polar ozone recovery has been only identified in September above Antarctica, which is connected to the observed delay in the onset of the Antarctic ozone hole (Stone et al., 2021). In the Arctic, large interannual variability still prevents the
 detection of early signs of recovery.

Although we showed that <u>ODS-related</u> ozone recovery is evident at NH middle latitudes, the total ozone levels in the 408 NH extratropics have been more or less stable since about 2000. Our regression results show that the recovery positive ODS-related trend here is balanced by changes in ozone transport. A long-term positive drift in the AO index over the last

- 410 55 years is indicative of a strengthening of the Arctic vortex (Hu et al., 2018; Lawrence et al., 2020; von der Gathen et al., 2021) and reduced winter/spring transport of ozone into middle and high latitudes. This result may be consistent with the
- 412 observed decline in lower stratospheric ozone in the extratropics as reported by Ball et al. (2018) and Wargan et al. (2018), Wargan et al. (2018) and Ball et al. (2020) mainly attribute this decline to enhanced horizontal mixing with the tropical region,
- 414 where lowermost stratospheric ozone decreases (Thompson et al., 2021). Other studies and datasets, however, do so far not confirm the long-term decline in the lower stratosphere Arosio et al. (2019); Steinbrecht et al. (2017); Sofieva et al. (2017)
- 416 (Arosio et al., 2019; Steinbrecht et al., 2017; Sofieva et al., 2017), which may be in part due to the larger uncertainties of satellite observations in this altitude region. From chemistry climate-models chemistry-climate models it is expected that the BDC
- 418 and ozone will increase as a result of greenhouse gases and with a strengthening of the BDC due to increasing GHG, tropical

ozone declines and extratropical ozone increases in the lower stratosphere. Most models so far cannot explain the observed

420 extratropical decline in lower stratospheric ozone (Dietmüller et al., 2021).

Another point which will be important is to show consistency between total ozone trends and both stratospheric and

- 422 tropospheric ozone trends. The ozone satellite datasets still show significant differences and opposite signs in trends (Gaudel et al., 2018)
- 424 *Data availability.* The sources of the various datasets and proxy time series (explanatory variables) used in this study are summarised in Tables 1 and 2.
- 426 *Competing interests.* No competing interests are present.

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Table 3. <u>1979-1996-1979-1995</u> and <u>1997-2020-1996-2020</u> annual mean total ozone trends in various broad zonal bands. Uncertainties are given as 2σ and; trends in bold are have an absolute magnitude equal or larger than 2σ . r^2 is the square Pearson correlation between timeseries of observations and MLR and χ the residual defined as $\chi^2 = \sum_i (\text{obs}_i - \text{mod}_i)^2 / (n - m)$, where obs_i are the observations and mod_i the MLR model, n, the number of data (years) in the timeseries, and m, the number of parameters fitted. All results are obtained using the full MLR.

zonal bands	MLR/ (2017-2020) minus (1964 -1980)		median	NASA	NOAA	GSG	GTO	WOUDC
60° S- 60° N	full	trend ≥1996 ≥1995	+0.4(2)	+0.2(2)	+0.5(3)	+0.4(3)	+0.5(3)	+0.6(3)
		[%/dec.]						
near global	-2.3%	trend <1996 <u>≤1995</u>	-1.5(6)	-1.2(7)	-1.5(7)	—		-1.1(7)
		[%/dec.]						
		r^2	0.94	0.94	0.93	0.92	0.93	0.89
		χ [DU]	1.1	1.1	1.2	1.3	1.2	1.3
					trend >1995			.4(3)
				median	trend >199:	5 [%/dec.]	+0	.4(3)
35°N-60°N	full	trend ≥1996 <u>≥1995</u>	+0.5(5)	+0.3(5)	+0.7(5)	+0.1(6)	+0.6(6)	+0.6(6)
		[%/dec.]						
NH	-3.6%	trend <1996 <1995	-1.9(13)	-1.5(12)	-1.9(12)	_	_	-1.9(15)
		[%/dec.]						
		r^2	0.88	0.90	0.89	0.88	0.87	0.85
		χ [DU]	2.9	2.7	2.7	3.0	3.0	3.3
				mean trend >1995 [%/dec.]			+0.5(6)	
				median trend >1995 [%/dec.]			+0.6(6)	
20° S- 20° S	full	trend ≥1996 - <u>≥1995</u>	+0.2(3)	-0.2(3)	+0.1(3)	+0.3(3)	+0.3(3)	+0.4(5)
		[%/dec.]						
tropics	-1.1%	trend <1996 <1995	-1.1(7)	-1.2(7)	-1.0(7)			-0.6(12)
		[%/dec.]						
		r^2	0.86	0.89	0.87	0.85	0.82	0.7- 0.70
		χ [DU]	1.2	1.1	1.1	1.2	1.3	1.9
				mean	trend >1995	5 [%/dec.]	+0	.2(3)
				median	trend >1995	5 [%/dec.]	+0	.3(3)
35°S-60°S	full	trend ≥1996 ->1995	+0.7(6)	+0.6(6)	+1.0(7)	+0.7(7)	+0.8(6)	+0.8(7)
		[%/dec.]						
SH	-4.7%	trend <1996-<1995	-2.5(16)	-2.5(16)	-2.4(17)			-2.6(19)
		[%/dec.]						
		r^2	0.87	0.88	0.89	0.87	0.88	0.82
		χ [DU]	3.0	3.0	3.1	3.1	3.0	3.6
				mean	trend >1995	5 [%/dec.]	+0	.8(7)
				median	trend >1995	5 [%/dec.]	+0	.8(7)

bold numbers: statistical significance at 2σ

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Table 4. Different MLR settings applied to the broad zonal mean median ozone timeseries. For explanations of terms see Table 3. Standard MLR is based upon Eq. 1 with P(t) = 0. Iterative MLR means that only terms of P(t) and El-Nino term are fitted when they are statistically significant (2σ) , while full MLR includes all terms in Eq. 1 and P(t).

zonal bands	MLR	parameters added	$r^2_{\sim\sim}$	$\overset{\chi}{[{{}{}{}{}{}{\overset$	$\underbrace{\text{trend } (t \leq 1995)}_{[\%/\text{decade}]}$	$\underbrace{\text{trend } (t \ge 1996)}_{[\%/\text{decade}]}$
<u>60° S-60° N</u>	full iterative standard	all BDCs, BDCn, AAO ~	$\underbrace{\begin{array}{c} 0.94\\ 0.93\\ 0.89\end{array}}_{0.89}$	$\begin{array}{c} \underbrace{1.1} \\ 1.1 \\ 1.1 \\ 1.3 \\ \end{array}$	-1.5(6) -1.5(6) -2.2(6)	+0.4(2) +0.4(2) +0.3(3)
<u>35N°S-60°N</u>	full iterative standard	all all ~	$\underbrace{\begin{array}{c} 0.88\\ 0.88\\ 0.74\end{array}}_{0.74}$	$\begin{array}{c} 2.9\\ \widetilde{2.9}\\ \widetilde{4.0}\\ \widetilde{4.0}\end{array}$	-1.9(13) -1.9(13) -4.0(15)	+0.5(5) +0.5(5) 0.0(7)
<u>20S°S-20°N</u>	full iterative standard	all BDCs, ENSO	$\begin{array}{c} \underbrace{0.86} \\ 0.84\\ \hline 0.78 \end{array}$	$\begin{array}{c} \underbrace{1.2}\\ 1.2\\ \underbrace{1.2}\\ \underbrace{1.4}\\ \end{array}$	-1.1(7) -0.7(6) -0.8(7)	+0.2(3) +0.3(3) +0.2(3)
<u>35S°S-60°S</u>	full iterative standard	all AAO ∼	$\begin{array}{c} \underbrace{0.87} \\ 0.83\\ \hline 0.86\\ \hline \end{array}$	$\begin{array}{c} 3.0\\ 3.3\\ \widetilde{3.1}\\ \widetilde{3.1}\end{array}$	-2.5(16) -3.1(13) -3.1(14)	+0.7(6) +0.8(6) +0.8(7)

bold numbers: statistical significance at 2σ

Table 5. Polar total ozone trends in March (NH), September (SH), and October (SH) before and after 2000. Uncertainties are provided for 2σ and trends in bold indicate statistical significance. r^2 is the square Pearson correlation and χ the residual (see caption of Table 3). The results were obtained from the full MLR.

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zonal bands	MLR		median	NASA	NOAA	GSG	GTO	WOUDC
60°N-90°N March	full	trend \geq 2000 [%/dec.] trend <2000 [%/dec.] r^2 χ [DU]	+2.0(39) -2.6(55) 0.81 15.2	+3.0(35) -0.3(51) 0.85 13.2	$ \begin{array}{r} +3.4(37) \\ 0.0(54) \\ 0.85 \\ 14.0 \\ \end{array} $	+3.1(40) 	+3.6(37) 	+1.3(44) -2.3(61) 0.76 17.0
60S°S-90°S September	full	trend \geq 2000 [%/dec.] trend <2000 [%/dec.] r^2 χ [DU]	+12.0(56) -13.8(87) 0,91 12.2	+11.0(65) -8.9(100) 0.85 14.2	+10.1(68) -11.6(105) 0.87 14.6	+12.2(57) 	+11.2(57) 	+10.9(62) -19.1(107) 0.89 13.3
60° S-90° S October	full	trend \geq 2000 [%/dec.] trend <2000 [%/dec.] r^2 χ [DU]	+0.1(68) - 19.6(104) 0.87 14.8	-1.9(68) - 19.4(104) 0.87 14.8	+0.1(65) - 21.0(100) 0.88 14.2	+0.9(71) 	+0.5(72) 	+4.1(91) - 18.9(138) 0.81 19.7

bold numbers: statistical significance at 2σ

Supplement

Table S1. Different MLR settings applied to the broad zonal mean median ozone timeseries but limited to the period 1979-2016 (last four years removed compared to Table 4). For explanations of terms see Table 3. Standard MLR is based upon Eq. 1 with P(t) = 0. Iterative MLR means that only terms of P(t) and El-Nino term are fitted when they are statistically significant (2σ) , while full MLR includes all terms in Eq. 1 and P(t).

zonal bands	MLR	parameters added	$r^2_{\sim\sim}$	$\begin{array}{c} \chi \\ [DU] \end{array}$	$\frac{\text{trend } (t \leq 1995)}{[\%/\text{decade}]}$	$\underbrace{\text{trend } (t \ge 1996)}_{[\%/\text{decade}]}$
60°S-60°N	full iterative standard	all BDCs, BDCn, AAO	$\begin{array}{c} 0.95\\ \hline 0.95\\ \hline 0.79\\ \hline \end{array}$	$\begin{array}{c} \underbrace{1.0} \\ \underbrace{1.0} \\ \underbrace{1.2} \\ \underbrace{1.2} \end{array}$	-1.6(6) -1.6(6) -2.2(6)	+0.4(3) +0.4(2) +0.3(3)
35N°S-60°N	full iterative standard	all all ~	$\begin{array}{c} 0.88\\ \hline 0.88\\ \hline 0.76\\ \hline \end{array}$	$\begin{array}{c} 3.0\\ \overline{3.0}\\ \overline{4.0}\\ \overline{4.0}\end{array}$	$\begin{array}{c} -2.0(14) \\ -2.0(14) \\ -4.0(15) \end{array}$	$\begin{array}{c} +0.5(7) \\ +0.5(7) \\ +0.3(9) \end{array}$
205° S-20° N	full iterative standard	all BDCs, ENSO	$\begin{array}{c} 0.87\\ \hline 0.85\\ \hline 0.79\end{array}$	$\begin{array}{c} 1.2\\ 1.2\\ 1.2\\ 1.4\\ \end{array}$	-1.1(8) -0.6(6) -0.8(7)	+0.2(4) +0.2(4) +0.2(4)
<u>35S°S-60°S</u>	full iterative standard	all AAO ∼	$\begin{array}{c} 0.92\\ \widetilde{0.90}\\ \widetilde{0.79}\\ \end{array}$	$\begin{array}{c} 2.6\\ 2.6\\ \phantom{00000000000000000000000000000000000$	-2.7(14) -3.0(12) -3.2(16	+0.6(6) +0.6(6) +0.6(7)

bold numbers: statistical significance at 2σ

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