

# **Response to referee comments on “Stratospheric ozone trends for 1985–2018: sensitivity to recent large variability” by W. T. Ball et al**

## **General comments relevant to both referees:**

We thank all referees (and the interactive comment of M. Diallo 16. March 2019) for their various important comments reproduced below, our replies are in blue text, while referee/reviewer comments are in black. Since almost all the points raised are specific points, rather than a generalized response, we respond to each one below point-by-point.

One change worth raising to the reviewers is that Fig. 4, showing annual mid-latitude and tropical changes in the QBO had a plotting error, and additionally, we changed the choice of altitude to 15 hPa as this better represents the message we are trying to make about the variability of ozone dependent on season and QBO phase. This also make the results consistent with Fig 2, with an upward ozone anomaly in the SH during a westerly phase QBO, the only extreme increase in that phase, that took place around the time of the only southern hemisphere sudden stratospheric warming in 2002.

## **References needed**

Two paper by Diallo et al. (2018) and (2019) recently published in ACP discusses the combined influence of QBO and ENSO on the UTLS ozone and water vapour distributions using a lagged multiple regression analysis. A reference to these paper should be included along with a discussion of the added scientific value of the results presented here.

We thank M. Diallo for making us aware of these recent papers. These papers have now been references twice in relevant discussions of ENSO and QBO impact on both variability and ozone in the lower stratosphere (and in general throughout).

The papers are accessible here:

1. Diallo, M., Riese, M., Birner, T., Konopka, P., Müller, R., Hegglin, M. I., Santee, M. L., Baldwin, M., Legras, B., and Ploeger, F.: Response of stratospheric water vapor and ozone to the unusual timing of El Niño and the QBO disruption in 2015–2016, *Atmos. Chem. Phys.*, 18, 13055–13073, <https://doi.org/10.5194/acp-18-13055-2018>, 2018.
2. Diallo, M., Konopka, P., Santee, M. L., Müller, R., Tao, M., Walker, K. A., Legras, B., Riese, M., Ern, M., and Ploeger, F.: Structural changes in the shallow and transition branch of the Brewer–Dobson circulation induced by El Niño, *Atmos. Chem. Phys.*, 19, 425–446, <https://doi.org/10.5194/acp-19-425-2019>, 2019.

## **Anonymous Referee #1**

***Received and published: 17 April 2019***

This manuscript is an update and extension of a similar analysis by Ball et al. (ACP, 2018), which reported evidence from satellite data that ozone in the lower stratosphere at latitudes less than 60 degrees has continued to decline since 1998 even though upper stratospheric ozone has started to recover in response to the Montreal Protocol bans of

many ozone-destroying substances. Since much more ozone is in the lower stratosphere than in the upper stratosphere, the lower stratospheric decline dominates. On the other hand, evidence for an increase in tropospheric ozone was reported that could potentially cancel out the lower stratospheric decline. In that 2018 paper, no conclusion about the cause of the continued lower stratospheric decline was drawn and possible explanations involving both dynamics and chemistry were only briefly discussed.

In this manuscript, the trend analysis is slightly updated by the addition of more data and a stronger conclusion is drawn that the cause of the continued decline in the lower stratosphere is dynamical in nature rather than chemical. Specifically, the manuscript agrees with a recent study by Chipperfield et al. (GRL, 2018) that a strong positive ozone anomaly in 2017 was driven by short-term dynamical transport of ozone. However, it is further shown here that this short-term increase was caused by the stratospheric quasi-biennial oscillation and that a long-term gradual decline in lower stratospheric ozone remains even when the QBO-related variability is taken into account. The manuscript also agrees with the Chipperfield et al. conclusion that short-lived chemical depletion cannot explain the long-term decline, which must therefore be dynamical in origin.

I have two main comments on the manuscript, at least one of which can be considered as major. If the authors can satisfactorily address these comments in their revision, publication can be recommended.

(1) The magnitude of the long-term trends in zonal mean extrapolar lower stratospheric ozone is not clearly stated in the abstract or in most parts of the text. This leaves readers in the dark about how important the declines are. According to Figure 2a, the overall quasi-global decrease over 1985-2018 (33 years) is roughly 3.5 DU, which is in the range of 1.0 to 1.5%, or roughly -0.5% per decade. For comparison, the increase in tropospheric column ozone in the same latitude range estimated by Ball et al. (2018; their Figure 4) is +1.68 +/- 0.11 DU per decade or roughly +0.5% per decade. Could the authors please add numbers like this to the abstract and conclusions sections? The present manuscript notes large uncertainties in the actual tropospheric ozone increase (lines 38-41). However, at least the magnitude of the estimated lower stratospheric ozone decreases and the strong possibility of compensating tropospheric ozone increases should be more explicitly stated. Then readers can judge for themselves the importance of the observed lower stratospheric ozone decline due to dynamical processes.

We are happy to include some of these numbers in the main text, but rather avoid making the abstract less fluid and more difficult to read; these numbers are primarily included in the discussion and conclusions. In addition, we include an extensive table of numbers (absolute levels in 1985 and 1998, and DU, % and %/decade changes over 1998-2018, 1985-1998, and 1985-2018).

In addition, to be clear, the values mentioned above by the reviewer should relate to the quasi-global *lower* stratosphere. As such, the 1985-2018 (34 years) change is 3.4 DU, with a background level in 1985 of 87.8 DU (for the lower stratospheric component) leads to a change of -3.8% over the period, or approximately -1.1% per decade; this can be split into -1.3% per decade before 1998 and 0.9% after.

Finally, we do not wish to speculate on the tropospheric changes in the context of contribution to offsetting since we do not know how tropospheric ozone was changing prior

to 2005 (based on Ball et al., 2018), and other work indicating global tropospheric changes do not go back to 1985 (Gaudel et al., 2018).

(2) In this manuscript, as well as in the published work of Ball et al. (2018), the seasonal and longitude dependences of the observed zonal mean ozone trends are not evaluated. By not evaluating these characteristics or at least reviewing previous work on these characteristics, the authors are missing some important clues for understanding the origin of the observed zonal mean ozone trends in the lower stratosphere. Beginning in the 1990's, a number of authors have found that there is both a seasonal and a longitude dependence of column ozone trends, most of which originates in the lower stratosphere, especially at middle latitudes in winter and spring. Some of these authors concluded that these dependences are a consequence of decadal variability of quasi-stationary ultra-long Rossby waves that propagate from the troposphere into the stratosphere (Hood and Zaff, JGR, 1995; Peters and Entzian, Meteorol. Z., 1996; Peters et al., Beitr. Phys. Atmos., 1996; Hood et al., JGR, 1997; Hood et al., JGR, 1999). Decadal climate variability was therefore implicated. This was an extremely controversial and unpopular conclusion at the time because the prevailing view was that the observed ozone trends in both the polar and extrapolar regions in the lower stratosphere were dominantly chemical in origin. Note that these quasi-stationary waves are not necessarily linear (even though most mathematical treatments make this assumption) so that the zonal mean ozone change is not necessarily zero. Moreover, there is an associated trend in the BDC, which also affects ozone amounts, because the BDC is driven by Rossby wave breaking and absorption in the stratosphere (e.g., Hood and Soukharev, JAS, 2005). The continued decline of lower stratospheric ozone reported in this manuscript may therefore imply that there is a long-term component of the variability of these ultra-long waves. Schneidereit and Peters (Atmosphere, v. 9, p. 468, 2018) have recently investigated the zonally asymmetric component of ozone trends over 1979-2016 and find negative trends over Europe in winter that are twice as large as the zonal mean trend. A strong negative trend in one area such as this could produce a net change in the zonal mean ozone if the waves are non-linear. These authors suggest that long-term changes in the Rossby wave train that propagates out of the tropics and is linked to Arctic warming may be the ultimate driver. If the authors are not able to investigate zonally asymmetric ozone trends or long-term changes in quasi-stationary waves and their breaking behavior in the stratosphere for the present manuscript, then they should at least include some discussion of this previous work with references to some of the above papers.

Significant decreases in total ozone at (northern) mid-latitudes as published by the International Ozone Panel Report in 1988 were believed to be caused by anthropogenic emission of ODSs, which ultimately led to the Montreal Protocol and its enforcements. Subsequently, several studies were published presenting results that stratospheric ozone changes were (partially) attributed to long-term climate variability or other drivers causing additional trends (e.g. shown by Schneidereit and Peters, Atmosphere, v. 9, p. 468, 2018) indicating that ozone changes at particular latitude-longitude sites have large (winter) total ozone trends. Appenzeller et al. (2000), and Weiss et al. (2000) have discussed how winter ozone negative trends can be strongly enhanced by the NAO/AO over central Europe (i.e. Arosa). The EU project CATNDIDOZ also looked at similar questions regarding spatial influences on zonally averaged and global trends.

However, in this study, we have focused on zonally symmetric trends, which is of course a simplification of the real world. While of course the reviewer is correct that, in reality, the variability has a longitudinal component, and investigating this is worth undertaking, we do not make that step for now. Indeed, considering the non-zonal component might be taken

into account more properly by using equivalent latitude (at least at higher latitudes rather than the lower that dominate this study) instead of geometric latitudes averaged over some bands, and would be a natural way to account for the geographical, longitudinal inhomogeneity of ozone. For now this remains outside of the scope of this manuscript.

Nevertheless, it is worth mentioning at least in a brief discussion the potential physical drivers, particularly for future work. As such, we have added the following into the manuscript. 'Part of the negative trends in northern hemispheric stratospheric ozone in the 1980s and 1990s at higher latitudes have been previously attributed to synoptic and planetary waves (Hood & Zaff, 1995; Hood et al., 1999) inducing large localised (e.g. over Europe) wintertime decreases in ozone that might in turn be driven by sea surface temperature and eddy flux changes on decadal or longer timescales, although most of these studies are limited to the end of the last century when ODSs remained an established primary driver of the decrease. Nevertheless, these dynamical changes do not in themselves determine a specific underlying driving force, although increasing anthropogenic greenhouse gases (GHGs) is an obvious, though unverified candidate (Hood & Soukharev, 2005; Peters & Entzian, 1999; Ball et al., 2018).

## **Anonymous Referee #2**

***Received and published: 22 April 2019***

### **GENERAL COMMENTS**

This paper presents a compelling update to the earlier work of Ball et al., and a much awaited response to the Chipperfield paper. While the science is excellent, unfortunately the writing is often too curt and the reader has to guess at what is being referred to (see many examples below). The writing, in many places, indicates a lack of clarity of thought, or perhaps just that the author is not writing what he means to say. I would strongly recommend that the author improves the quality of the writing. There are no major changes required for this paper to be ready for publication. Most of my suggested changes below are relatively minor. It is an excellent piece of analysis. Now it just needs to be communicated to its audience clearly.

We appreciate the author's positive comments. Additionally we thank the reviewer for the clear amount of time and effort that he/she has made to ensure that we do not miscommunicate our results, nor the success of the Montreal Protocol and its amendments.

### **SPECIFIC COMMENTS**

Line 1: I would suggest replacing 'The Montreal Protocol' with 'The Montreal Protocol, together with its amendments and adjustments'.

Replaced with 'The Montreal Protocol, and its subsequent amendments, ...'

Line 3: I would suggest that you reserve the word 'recovery' strictly to talk about the recovery of ozone from the effects of ODSs. When you are talking more generally about ozone increases, whether driven chemically or dynamically, talk rather about 'ozone increases' rather than 'ozone recovery'.

We agree that it is important to be clear on this issue. Following this, a similar suggestion by reviewer 3, and multiple specific suggestions regarding this point later in this review, we

have systematically gone through the manuscript to replace 'recovery' with 'increase'/'decrease' or to specify that the recovery has to do with 'ODSs' unless it is already clear.

This however raises an interesting point regarding terminology because, like the reviewer, we expect 'recovery from ODSs' to be already occurring everywhere due to the reduction of chlorine in the atmosphere, and yet we do not see signs of it in the lower stratosphere. If we assume that to be the case, then recovery is underway everywhere. This then suggests the need for new terminology in light of counteracting influence of GHGs. For example, perhaps 'restoration' to pre-ODS ozone-levels is a term that should be adopted to represent 'a return to levels prior to anthropogenic influences'. For now, we will leave this definition aside in the manuscript.

Line 12: I am struggling a bit with this 'still likely lower than in 1998 (probability ~80%)'. Surely it is either lower or it isn't. OK, maybe I will get to see later how this is nuanced by some statistical significance.

We do not use frequentist significance tests in this paper, but rather make straightforwardly interpretable Bayesian probability statements that reflect "the probability (or confidence) that X is true given the observed data, model and prior assumptions". In the case you highlight for example, we calculate the (Bayesian posterior) probability distribution for the overall change since 1998 given the observed data and model assumptions, and estimate from this the fraction of the PDF that is either negative or positive. In this case, the overall change since 1998 is likely negative with 80% probability, and 20% probability that it is positive (because those are the fractions of the probability distribution that are split by the zero line). Our use of Bayesian probabilities is intentional and makes for easily interpretable and robust statements. Frequentist significance statements are muddled by the fact that they are not "direct" inferential statements, so misinterpretation is easy (and commonplace), and too often lead to the poor practice of drawing binary inferential conclusions (X is proven/rejected) based on arbitrary thresholds (eg  $p < 0.05$ ).

Lines 16-17: With regard to the sentence 'These decreases do not reveal an inefficacy of the Montreal Protocol', an important point that can, and perhaps should, be made here is that tropical stratospheric ozone is almost certainly recovering (from the effects of ODS), and hence the Montreal Protocol is successful, while simultaneously still declining (due to other influences). The point to make is that recovery and declining ozone are not mutually exclusive.

We agree. However, since we do not diagnose either way the ODS-recovery and GHG-related-decline, nor attribute the ratio of these factors, we do not wish to speculate or assume that point; such quantification should be (and has been) performed using chemistry climate models. Since we make the point in the following sentence, that the Montreal Protocol has been beneficial, and that dynamics are probably in play in counteracting it, we do not think this needs adjusting.

Line 33: You need to be clear here what you mean by 'significant'. Do you mean statistically significantly different from zero at the 2 sigma level, or do you mean a more general 'significant' as in 'large'. It matters a lot in this specific context so I think that you should be clear.

Agreed, and this point was raised by another reviewer. We have added 'statistically' before 'significant'.

line 59: I think it would be plausible to say that climate change may be exacerbating some specific dynamical mechanism (or more than one) that affects ozone, but it feels incongruous to state that climate change, in and of itself, could be a mechanism for dynamically affecting ozone.

Agreed, and this point was raised by another reviewer. We have replaced 'climate change' with 'increasing anthropogenic greenhouse gases (GHGs)'.

Line 125: Delete 'point the reader to Laine et al. (2014) for details on this method and' since you have anyway cited the Laine et al., 2014 paper.

Done.

Line 135: This essentially assumes that the sensitivity of ozone to the regressors is time-dependent. What support is there for this assumption? Why might ozone have a certain sensitivity to EESC in 1985 and then a different sensitivity in 1995, or 2005? This ability for DLM to accommodate changes in the sensitivity of ozone to the regressors is presented as an advantage of DLM over MLR but I am not convinced that it is. In choosing DLM over MLR you are making some significant assumptions and I am not sure that you have support for those assumptions. Or is it just the amplitude and phase of the seasonal cycle that you allow to vary with time?

Our main reason for supporting DLM over MLR is its superiority over MLR in attaining a better estimate for trends in validation-test cases where the trend is known in advance (Ball et al., 2017) and because, after that validation, DLM provides more insight into the evolution of the trend than MLR does (when assuming linear or piecewise-linear trends). You are correct: in the DLM model used for this paper, only the amplitude and phase of the seasonal components are allowed to vary in time, not the other regressor coefficients. We note also that since, in ozone composites, the instruments change in time, so do the observing kernels and the exact region observed; this can introduce an artificial apparent change in amplitude of the seasonal cycle and motivates allowing for variation of seasonal (and regressor) parameters in time. We allow this because the seasonal part has the largest variance and if the seasonal cycle changes with time then removing a mean seasonal cycle may lead to a bias in the residuals over time; we see this also as an advantage. It may well be true for the other drivers, but this does not have a strong justification yet. It should be noted that the seasonal cycle, even when evolving, is required to be stationary over time to avoid leaking into the non-stationary trend component. Many of the issues related to MLR are also discussed in Petropavlovskikh et al., 2018 (SPARC Report).

Line 138-139: Well, unless EESC is selected as a descriptor of the long-term secular trend in which case it is a much more natural and appropriate descriptor than any pure statistical descriptor.

We agree that EESC is a key component of the long-term trend and it is hence natural to include it as a regressor, but even then it is an overly strong assumption to assert (in the model) that the only thing driving the background trend is EESC. We also note that the EESC will need a second component to allow for spatial variation in the inflection point, otherwise the same issue with respect to the piece-wise linear misrepresenting changes will occur. A GHG regressor would also make sense as an additional component of the background trend. However, here we simply aim to diagnose the long-term trend, whether ODS or GHG (or other) related, following the standard aim in this community. The DLM trend has the advantage that it does not tie the background trend to a particular prescription for the shape or a (possibly incomplete) set of components before the data are analyzed; it is agnostic about the shape or drivers of the trend. However, for the same reason, the DLM trend term does not elicit specific physical drivers of the background trend on its own. We



have added in bold/italic: 'Secondly, MLR that does not assume a driver for the long-term trends, e.g. for the influence of ODSs or GHGs typically assumes a fixed prescription for the shape'

Lines 143-144: I don't believe it is true that 'in practice MLR is often performed by first subtracting an estimated mean seasonal cycle'. I certainly don't. Most MLR-based analyses I have seen fit the annual cycle as a series of Fourier expansions along with all of the other regressors.

We agree that not all analyses subtract the seasonal cycle, e.g.: Kyrola et al., 2013 (ACP); Frith et al., 2017 (ACP); Petropavlovskikh et al., 2018 (SPARC). But many do and this was how, e.g.: Maycock et al., 2016, 2018 (ACP); Sofieva et al., 2017 (ACP); Steinbrecht et al., 2017 (ACP). We have added in bold/italic: '...and then making a post-hoc correction for autoregressive residuals, **although many do fit annual and semi-annual components.**'

Line 151: What is MCMC? I have not seen this acronym defined anywhere.

Agreed – we have added 'Monte Carlo Markov Chain' prior to the first mention of MCMC'.

Line 170: Can you give some indication of why the SAOD time series is not available beyond 2016?

The dataset had not been extended at the time of the manuscript. Thus, we have modified the sentence to read: '...for this analysis, the SAOD is **currently not available extended** beyond 2016, so we repeat the year 2016...'

Line 196: It wasn't clear to me what was meant by 'this group of spatial responses'.

Responses of what to what? Are you referring the latitude/pressure resolved trends plotted in Figure 1?

Yes. We have modified this sentence to read: 'any sensitivity of the end year to the state of these drivers should be encapsulated in ~~this group~~ **the set** of spatial responses **depending on the end year only (Fig. 1)**...'

Line 214: But the latitudinal extent of the changes you are seeing in observations is much wider than what is seen in the CCMs right?

Yes, though we cannot attribute that, with trend analysis only, to the BDC, which is why simply leave this as a statement, and only with respect to the MMM of CCMs (see WMO 2014 and 2018 reports), not to individual members of the MMM.

Line 240: But only in the lower stratosphere right?

An excellent point; we have modified the sentence: '...convincingly showed that the majority of post-1997 quasi-global **lower stratospheric** ozone variability...'

Line 253: I think that you need to read and cite Gray, L.J. and Pyle, J.A., A twodimensional model of the quasi-biennial oscillation of ozone, J. Atmos. Sci., 46, 203-220, 1989. Another paper that might be relevant is Bodeker, G.E.; Garny, H.; Smale, D.; Dameris, M. and Deckert, R., The 1985 Southern Hemisphere mid-latitude total column ozone anomaly, Atmos. Chem. Phys., 7, 5625–5637, 2007, especially if you are seeking clarification of the origin of the large mid-latitude changes in ozone that occur every few years.

We thank the reviewer for these suggestions, which we have integrated into this paragraph, including the addition of: '**Bodeker et al. (2007) previously identified large SH negative anomalies in 1985, 1997 and 2006 and related these to the QBO-Westerly phase**'. We note that it appears to us that the general relation between QBO and ozone is rather well known in the science community, although the connection of the event highlighted by Chipperfield

et al. (2018) and the additional ones of similar magnitude we have identified, to the QBO has not yet been made until here. We further believe this motivates the need to improve attribution methods to account for these interactions.

Line 311: It is not clear to me what you mean by 'governing each other'? Do you just mean 'governing each'?

You are correct, we do, so we have clarified this: '...the processes governing each other *timescale* are likely quite different.'

Line 325: This is worded in a very confusing way - please rewrite.

We have rewritten this as follows: 'This agrees with Chipperfield et al., 2018 who suggested the large ~~upswing~~ *rapid increase* of 2017 ~~that settled in 2018~~ affected trends, although this was mainly in the SH *and has subsequently showed little change over 2018.*'

Line 329: As \*what\* in years prior to 2010 are essentially unaffected by the addition of 2017 and 2018? Ozone in 2013 is unaffected by the addition of 2017 and 2018? You are referring to something being unaffected by the addition of 2017 and 2018 but I am not sure what that something is.

We agree this is confusing. The 'what' is '*the DLM-estimated change of ozone relative to 1998 in*'; this has been inserted at the start of the sentence.

Line 334: You need to make it clear that you are referring to the minimum in the DLM fit and not a minimum in the observed ozone.

We have clarified this: 'If that happens it is therefore possible that the non-linear trend estimates will likely decrease again, ~~with~~ *and* the emergent 2013 minimum *in the DLM non-linear trend estimate* seen in Fig. 2b *is* likely to shift to a later date or disappear.'

Line 336: Do you mean mid-latitude ozone excursions depend on the phasing of the QBO phase from westerly to easterly (or vice versa) on the phase of the annual cycle in ozone? If so, please consider wording as such. If not, please reword to be more clear.

We have added in bold/italic: 'Since mid-latitude ozone excursions depend on the QBO-seasonal interaction, *i.e. the QBO phase relative to the time of year, ...*'

Line 337: Can't you just have cross-terms in your regression model i.e. a QBO basis function modulated by a phase dependent seasonal cycle?

You could do that, but the phase of both QBO and seasonal terms will depend on location, and simply multiply terms does not work. As the phase of the two terms should not simply be estimated by a minimisation of, e.g., the residuals, but should have some underlying physical motivation connected to it, we do not do it here and leave it for future work.

Line 338: I don't think that is true. They can, they just need to include the appropriate regression model basis functions. Perhaps many MLR models currently in use do not, but that does mean that they fundamentally can't.

Absolutely. It is true given these predictors don't use it; but there is no standard way to deal with it yet, so it remains true until this has been resolved (see previous point above) and why we have stated the sentence with 'without predictors...'

Line 358: Uncertainties in what are consistently large?

We agree this is unclear, and have added in bold/italic: '...exhibits the largest sensitivity to the end year and uncertainties *in the change from 1998* are consistently...'



Line 359-360: The upper stratosphere is also sensitive to what in the tropics? Too often the explicit subject of a sentence or phrase is omitted in your writing which requires the reader to constantly be guessing at what you are referring to. This makes deciphering the narrative very tiring. There are many examples of this in my comments. The next one is on the same line (360):

We have clarified this as: 'The upper stratosphere is also sensitive *to the end year* in the tropics'.

Line 360: What has 'shifted from negative to positive'. And what, exactly is uncertain?

We have clarified this too: '~~... but~~ *and the end year* has shifted ~~the estimated ozone change~~ from negative to positive *with ncreasing end year*, although *the uncertainty* always ~~uncertain~~ *remains large*.'

Line 361: Uncertainties in what are smaller? And smaller compared to what?

We have clarified as: 'at mid-latitudes uncertainties *in the change since 1998* are smaller...'

Line 361: There has been a general shift in what towards more positive and significant increases? Please read the sentence that starts on line 359 and ends on line 362 i.e. "The upper stratosphere is also sensitive....SH and quasi-global estimates" and see whether, read as it stands, it would make sense to someone. Take that sentence to a colleague and ask them to tell you what it means. They may be horrified to read that the stratosphere has shifted from negative to positive. Maybe they always thought that the stratosphere was positive and may be alarmed that it has become negative. But at least they will know that whatever happened, its (whatever it is) is always uncertain. I would strongly suggest that you write in a way that prevents the reader from having to guess at things.

This is a fair criticism. We have amended the text to be clearer following the reviewer's suggestions.

Line 367: The statement that 'The quasi-global lower stratosphere continues to exhibit a monotonic decline' is not true. There are many things in the quasi-global lower stratosphere that are not continuing to exhibit a monotonic decline. A good example would be CO<sub>2</sub>. Please work to improve the precision of your writing.

Agreed – we have inserted 'ozone' into this sentence to clarify it is only ozone we are assessing.

Line 368: I was shocked to read that 'the whole stratosphere continues to remain lower than in 1998'! Is the sky really falling? <https://www.youtube.com/watch?v=NO04VXBIS0M>. Is there nothing we can do to lift the stratosphere?

We did not mean to shock the reviewer, nor future readers. As such, we have amended the sentence as: '~~...and~~ *ozone abundances integrated over the whole stratosphere continues to remain lower ...*'

Line 374: Regarding 'changes prior to the last five years are largely unaffected in the partial columns'. I would be horrified if it was possible that ozone prior to the last five years was affected by the addition of recent years. It would mean that someone, somewhere, has invented time travel. But perhaps that's not what you mean? It might be a good idea then to write \*exactly\* what you mean.

Again, the reviewer makes a fair point. We have modified the text here as follows. Figure 5 also confirms that the *gradients of the* non-linear curves are only affected by unmodelled variance in years close to the end points, *typically within the last five years of the partial*

*column timeseries considered here; the shape of the DLM curves changes prior to the final last five years of the DLM curves are largely unaffected in the partial columns.'*

Line 403: So are you really saying that the Montreal Protocol is working only in the upper stratosphere and not in the lower stratosphere. This will hugely concern policymakers. They will wonder why all the hard work they have done since 1987 in reducing emissions of CFCs, halons, HCFCs and other ODSs has only decreased their concentrations in the upper stratosphere. Could I put it to you that the Montreal Protocol has been effective in reducing ODS concentrations, and thereby concentrations of Cly and Bry throughout the atmosphere, and that, as a result, ozone throughout the atmosphere, including the lower stratosphere, is recovering from the effects of those ODSs. Is this recovery apparent in observations in the upper stratosphere? Apparently yes. I say apparently only in that (at least in this paper) a thorough attribution of the drivers of those ozone increases has not been done. Is this recovery apparent in observations in the lower stratosphere? No, clearly not? Why not? Well because other factors have been affecting ozone (not diagnosed in this paper) that are likely (we cannot be sure since a thorough attribution has not been done) overwhelming the increases brought about by reductions in concentrations of Cly and Bry. Wouldn't that be a more accurate picture to communicate to policy-makers?

In order to be precise, we have included the following at line 403:

*'An upper stratospheric increase is the expected result from long-term stratospheric chlorine reductions, a direct consequence of the Montreal Protocol and its amendments, though we do not explicitly attribute the cause of the increase to that here. Indeed, the Montreal Protocol and its amendments will have been effective in reducing ozone losses throughout the stratosphere through reductions in CFC emissions, HCFCs and other ODSs. The lack of a positive trend since 1998 in the lower stratosphere, as opposed to the one clear in the upper stratosphere, is likely the consequence of other factors such as dynamical changes (Wargan et al., 2018).'*

Line 412: What does it mean to be 'confident in the evolution of stratospheric ozone'? You're confident that ozone is evolving? I'm pretty confident it is too. It always has been. I don't need your paper for that. Maybe you mean that the aim of this work is to build confidence in our quantitative understanding of trends, and other long-term variability, in upper, middle and lower stratospheric ozone?

Agreed. We have replaced this sentence with: *'The aim of this work is to assess the current state of, and trends in, stratospheric ozone. Improved knowledge of such trends, and the relevant forcing mechanisms and associated variability, will help to better constrain CCM projections of ozone to the end of the 21<sup>st</sup> Century.'*

Line 414: I think that the best tools for studying long-term changes in ozone, and attributing the causes of those changes to known drivers, is the application of regression models to observations. Yes, models can be useful for attribution but they have little role, if any, in detecting the changes in the first place. I think that chemistry-climate models are the best tools for making projections of how ozone may change in the future.

We certainly agree that they do not have as strong a role in initial detections, especially in this case where the implication is models are not reproducing these trends. However, we still believe these remain an excellent tool. So we have inserted 'one of' in this sentence: 'Chemistry models resolving the stratosphere are *one of* the best tools for attribution and long-range studies of ozone'.

Lines 423-425: This sentence blurs the lines between observations and model output. Were the 2017 data from Chipperfield CTM output or observations? If it was model output I would

suggest replacing 'found lower stratospheric ozone had rapidly increased in 2017 back to 1998 levels' with 'found that model simulated lower stratospheric ozone increased in 2017 back to 1998 levels'.

As also mentioned by another reviewer, we have modified this sentence partly following your advice. 'A recent study (Chipperfield et al., 2018) used a CTM to reconstruct ~~extend~~ the ozone timeseries *beyond the observational record available at the time to 2017 and found that that model simulated a lower stratospheric ozone increase in 2017 back to 1998 levels*; this was attributed to dynamical variability.'

Line 429: Any idea why the CTM got it so wrong?

No, we do not know why.

Line 438: Does it enhance the positive trend in ozone, or does it enhance the recovery of ozone from ODSs - noting that those are two very different things?

Fair. Modified to reflect the former: 'We also find that the 2017--2018 addition enhances the *estimated* magnitude of the upper stratospheric ozone ~~recovery~~ *positive trend*'

Lines 439-440: How can the 'recovery' display a 'reduction'. That makes no sense to me. I can understand how ozone can reduce. I can even understand that ozone can reduce while simultaneously recovering from the effects of ODSs (I am not saying that that's what is happening here, but it is conceivable).

A reread is also partially confusing to us too. We have amended it, striking out 'recovery of the' to simply state that it still displays a reduction.

Line 447: By 'continues at all latitudes north of 30°S' do you mean continues to decrease at all latitudes north of 30°S?

Thank you for this clarification; 'to decrease' has been inserted.

Line 449: The seasonal-dependence of the QBO on what? Or do you mean the seasonal dependence of ozone on the QBO?

Yes; inserted 'ozone on'.

Line 455: This is the first mention of 'return dates'. What is meant by this? What is the 'return date' in a CCM?

Yes, this need clarifying: '...and therefore their return dates, i.e. a *return of ozone to the level it was in 1980 (WMO 2014, Dhomse et al., 2018, WMO 2018)*.'

Lines 456-457: What, exactly, do you mean by 'numerical inaccuracies'? Can you please add a sentence or two that elucidates this.

We meant 'numerical diffusion', which has replaced 'inaccuracies'.

Line 460-461: Wait a minute. I have seen no evidence anywhere that there has been a 'halt in the recovery in total column ozone' from the effects of ODSs. I have seen plenty of evidence that ozone in different regions of the atmosphere continues to decline (including this paper) but no attribution of this declines such that one could conclude that ozone is not recovering from the effects of ODSs. In fact it would devastate our understanding of atmospheric chemistry if it was found that decreasing concentrations of Cly and Bry had no impact whatsoever on the Cly and Bry cycles that destroy ozone. I see chemists leaping from buildings. So I strongly reject your conclusion that ozone, anywhere in the atmosphere, is not recovering from the effects of ODSs as you have presented no evidence at all to that effect. To make that call, you would need to do a robust attribution of changes in ozone to

date and demonstrate that the ozone changes attributed exclusively to changes in Cly and Bry have been negative. And that you have not done.

A fair point. We have clarified this as follows: 'The halt *in ODS-related ozone losses as a result of the Montreal Protocol and its amendments*, and *an* initial recovery in total column ozone is almost universally reproduced by CCMs...'

Line 464: Do you mean predictions or projections? I think that it is very dangerous to use models to make predictions.

We meant 'projections' and have swapped these words accordingly.

Line 468: Do you mean total column ozone or do you mean ozone in all parts of the atmosphere?

We meant total column, and have clarified this.

Line 470: I have no idea what you mean by 'super-recovery'. I fully understand how ozone can recover from the effects of ODSs. But I can't understand how it can 'superrecover'. I can understand how ozone could become higher than it was in the 1960s, but this has nothing to do with ODSs, and therefore nothing to do with 'recovery'. It results from CO<sub>2</sub>-induced cooling of the upper stratosphere. What then is 'superrecovery'?

Super-recovery is a term used by the WMO ozone assessment report (and other articles); from the WMO 2018 ozone assessment: '*For global mean total ozone columns, the return to 1980 values is faster and the possibility of super-recovery (i.e., the increase of ozone above historical levels) is higher for the RCPs with larger GHG increases.*' We have also modified the error of a return to '1960s' levels to '1980s'.' We have added a similar clause as the WMO to our manuscript: '*...continuing on to a 'super-recovery', i.e. that ozone will be higher by the end of the 21<sup>st</sup> Century than prior to 1980s levels...*'

Line 475: Ah, so the Montreal Protocol has effected a recovery in ozone from the effects of ODSs since the late 1990s? Your paper is communicating very mixed messages. Let me ask a very simple question: Is ozone in the lower stratosphere recovering from the effects of ODSs? If you answer yes, then what you have written elsewhere in the paper is wrong. If you answer no, then what you have written here is wrong because here you say that ozone declines would have been far worse without the Montreal Protocol which, to me, says that the Montreal Protocol has effected a recovery of ozone from the effects of ODSs. Please write clearly what you mean.

Agreed. Thanks to the time you have invested, and detailed comments, this manuscript should now be very clear and consistent by this point. We appreciate that.

#### GRAMMAR AND TYPOGRAPHICAL ERRORS

Line 2: Replace 'work suggests' with 'work has suggested'. Done.

Line 6: Replace 'wiped out' with 'offset'. 'wiped out' is too colloquial. Likewise on line 72. Done.

Line 10: Replace 'hemispheric' with 'hemisphere'. Done.

Line 18: Replace 'protocol's' with 'Protocol's'. Done.

Line 30: Replace 'its amendments' with 'its amendments and adjustments' just to be complete. Done.

Line 31: Replace 'coincided with' with 'led to'. They can't be coincident if they are separated by 11-13 years. Done.

Line 60: Replace 'negative trends' with 'negative trends in ozone'. Done.

Line 96: Replace 'This data was' with 'These data were'. Done.

Line 98: Replace 'in context' with 'in the context'. [Done](#).

Line 105: Replace 'the averaging the two products' with 'the averaging of the two products'. [Done](#).

Line 128: Be consistent in the way you spell timeseries. [Done](#).

Line 204: I would suggest replacing 'increase' with 'increase in SH mid-latitude lower stratospheric ozone' just to be completely clear (or whatever region Chipperfield reported the change over). [Done](#).

Line 210: Replace 'as more data is added' with 'as more data are added'. [Done](#).

Line 232: Either 'the identification criteria were' or 'the identification criterion was'. The word criteria is plural. [Done](#).

Line 277: Replace 'in context of these' with 'in the context of these'. [Done](#).

Line 250: Replace 'Equatorial variability related' with 'Equatorial variability in ozone related'. [Done](#).

Line 251: Replace 'decreases' with 'decreases in ozone'. [Done](#).

Line 251: Replace 'to that of the' with 'to that at'. [Done](#).

Line 311: Replace 'Whilst' with 'While', unless you really do want to be very British. [The lead author is very British; but we have made the amendment](#).

Line 317: I would suggest replacing 'DLM trends estimated' with 'DLM trends in lower stratospheric ozone estimated' just to be totally clear. [Done](#).

Line 351: Replace 'large resurgence in 2017' with 'large resurgence in ozone in 2017'. [Done](#).

Line 357: Replace 'that the middle-stratosphere exhibits' with 'that ozone trends in the middle-stratosphere exhibit'. [Done](#).

Line 365: Replace 'have made' with 'has made'. [Done](#).

Line 387: Replace 'tropical' with 'tropics'. [Done](#).

Line 393: Replace 'indicated' with 'indicate'. [Done](#).

Line 396: Replace 'exclude, 50–60°' with 'exclude 50–60°'. [Done](#).

Line 412-414: This sentence needs a lot of help. [A lot of help has been provided to this sentence; done](#).

Line 420: delete 'extremely'. [Done](#).

Line 454: Replace 'spread on' with 'spread in'. [Done](#).

Line 467: Replace 'is likely' with 'are likely'. [Done](#).

### **Anonymous Referee #3**

***Received and published: 26 April 2019***

Ball et al. provide an update on Ball et al., further examining trends in stratospheric ozone derived from satellite observations. Additionally, they discuss the recent results of Chipperfield et al. (2018), who modelled large increases in lower stratospheric ozone in a CTM, indicating that the observations used in this study give a smaller increase and suggest that this is consistent with interannual variability driven by the QBO. Further, they suggest that, if this is the case, then the lower stratospheric ozone increases will decrease in the near future. Despite these increases, longterm trends in lower stratospheric ozone trends remain negative. The analysis and discussion presented in the paper is of a high standard and explores an important and relevant topic within the scope of SCP, and as such merits publication following revision. I have several comments the authors should address before publication:

General Comments:

1. As a major point of consideration, the authors frequently use the term recovery, or lack of, when discussing ozone increases and decreases. However, recovery of stratospheric ozone is really reserved for increases of ozone resulting from reductions in stratospheric C<sub>ly</sub> expected due to the Montreal Protocol. Changes resulting from dynamical variability or stratospheric cooling resulting from CO<sub>2</sub> increases are not strictly recovery. As the manuscript does not explore all the drivers of ozone changes at different latitudes/altitudes, I feel the authors should refer only to ozone increases/decreases where they are not attributing ozone changes to change in ODS.

We accept this point. Repeating part of our response to reviewer 2's specific comment (regarding Line 3): we agree that it is important to be clear on this issue. Following this, a similar suggestion by reviewer 3, and multiple specific suggestions regarding this point later in this review, we have systematically gone through the manuscript to replace 'recovery' with 'increase'/'decrease' or to specify the recovery is to do with 'ODSs' unless it is already clear.

2. A second major comment is that the authors should state specifically what they are discussing in each section. Too often terms like decline/recovery/trend are used with no reference to ozone. This is particularly true in the sentence on L367-369, which states 'The quasi-global lower stratosphere continues to exhibit a monotonic decline that is still highly confident with 99% probability (Fig. 7 and Table 1), and the whole stratosphere continues to remain lower than in 1998. . .'. Further, more clarity should be provided so the reader knows when the authors are discussing ozone trends and ozone values.

The reviewer raises an important point, and we have made an effort to improve clarity throughout. Without writing an exhaustive list, the two examples were adjusted as such (bold/italic text):

- 'The quasi-global lower stratospheric **ozone** continues...'

- '...and **ozone integrated over** the whole stratosphere continues to remain lower than in 1998...'

3. There is no discussion in the manuscript on the chemical lifetime of ozone – the reason that dynamical variability plays such a key role in the lower stratosphere is that here the chemical lifetime is long. A brief description of this fact, with references, would add to the introduction.

We have added a brief discussion of this when discussing the CTM in the introduction:  
*"Indeed, chemistry and photochemistry play a dominant role over dynamical perturbations in the upper stratosphere as ozone lifetimes are short (~days), while ozone lifetimes of ~6-12 months in the lower stratosphere means that equator-to-mid-latitude transport of similar timescales plays an important (dominant) role there (London, 1980; Perliski & London, 1989; Brasseur & Solomon, 2005)."*

4. The authors frequently refer to 'climate change' as a potential driver of the examined dynamical variability. However, I feel they would be better served by using a phrase such as 'changes to stratospheric dynamics resulting from anthropogenic greenhouse gases,' which is closer to a mechanistic analysis. Climate change is itself a response to changing GHGs.

We agree that formally we are inaccurate using the 'climate change' as a forcing term; as such we have modified the following:



- 'Although the underlying driving force has not yet been determined, ~~climate change~~ **increasing anthropogenic greenhouse gases (GHGs)** is an obvious though unverified candidate.'
- '...rather than ~~forced by climate change~~ **a response to increasing GHGs...**'
- '...due to enhanced upwelling from the Brewer Dobson circulation (BDC) as a result of ~~climate change~~ **changes to stratospheric dynamics from increasing GHGs...**'
- '...it remains to be seen if this can be attributed to the ~~climate change~~ **anthropogenic GHG** induced upwelling of the BDC...'
- '...this is important given that the changing climate **due to anthropogenic GHG emissions** may impact inter-annual dynamical variability..."

5. I miss any discussion on why the CTM results of Chipperfield et al. and the observations presented in this study differ so greatly. Is there any consensus on why this is? Does it represent some failing of the chemistry in the CTM? Is the re-analysis dataset used not accurate? IS it within the uncertainties of the observations? Further discussion on this point would improve the manuscript.

The reviewer offers a series of excellent questions worthy of an answer. However, there is no answer now, and we believe this would require exploring the model, as well as developing improved uncertainty estimates. We do not have explicit uncertainties on the observations, though MLS usually shows high precision and is stable, so these are expected to be smaller than the difference over 2017 between the CTM and observations. The CTM shows periods of similar deviations with respect to the observations (see 2003, 2007-2008 in Fig 1c of Chipperfield et al., 2018 for the lower stratosphere, although in that plot there is not data from the observations to directly compare). It may be that the issues relate to the CTM parameters or to the reanalysis fields driving the dynamics; but this is not something we wish to speculate on, but should indeed be investigated. We have simply added to the end of the second paragraph in section 3.2 discussing the difference between observations and CTM: '**We do not know why the CTM and observations disagree in the magnitude of change for this period.**'

Specific comments:

L1: Add 'and its subsequent amendments' following Montreal Protocol  
Done.

L6: Replace 'wiped out'  
Replaced with 'offset'.

L18: Important here to say that dynamical variability is counteracting the effects of the Montreal Protocol on stratospheric ozone recovery, not on the regulation of halogenated ODS.

Agreed. Sentence now reads: 'Rather, they suggest other effects to be at work, mainly dynamical variability on long or short timescales, counteracting the **positive effects of the Montreal Protocol on stratospheric ozone recovery** ~~protocol's regulation of halogenated ozone depleting substances (hODS)~~'

L19: Swap '(30-60)' with 'variations'  
Done.

L33: Add 'statistically' before significant to indicate that you do not mean large increase

Done.

L33-43: The authors could cite here, or elsewhere in the manuscript, that these observations are supported by CCM studies (e.g. Meul et al., 2016; Keeble et al., 2017; Dhomse et al., 2018).

The suggested papers have varying degrees of conclusions regarding significant trends, the projection scenario and models used, and in what they specifically report, so we do not wish to overly complicate the introduction given the focus on observations only. However, we agree with the reviewer that this is worth pointing out, and so we have included the sentence: ***'The importance of considering tropospheric and stratospheric changes separately to understand changes in total column ozone has also been highlighted in recent studies using chemistry climate models (CCMs) (Meul et al., (2016), Keeble et al. 2017, Dhomse et al., 2018}.'***

L44-49: Stress that these composites are observations

Done: 'To assess trends in stratospheric ozone, composites ***of observations*** must be formed by merging multiple ozone ***observational*** timeseries into a long, multi-decadal record...'

L45: Use consistent spelling of timeseries throughout the manuscript

Done; replaced all instances of 'time-series' with 'timeseries'.

L60: Add 'ozone' between 'negative trends'

Done.

L93: Remove 'see'

Done.

L94: IS the Froidevaux et al. paper cited here now published? If not I would recommend removing it from the manuscript, instead saying that the v2.20 dataset used here is an update of Froidevaux et al. (2015).

Froidevaux et al, 2019 is now publish; we have updated the reference.

L125: delete 'point the reader to Laine et al. (2014) for details on this method and' – the paper is already cited in the sentence

Done.

L135-137: Why do the authors allow the seasonal component to vary but keep the regressors constant? Is it more likely that one should vary in time than the others? Some additional text explaining the rationale behind this decision is warranted.

We have added:

The reviewer raises a good point. Because the seasonal cycle has the largest variance by far, and because there is some evidence it modulates with time either due to different instrument sampling or real changes in the seasonal cycle, we allowed it to freely vary. However, we accept that this could also influence other regressors and in future these should be allowed to vary too, though we leave an adoption of that freedom to a future assessment of the impact on trend analysis from allowing such freedom. We have modified this part of the text to include the bold/italic in the following. "Here, we allow the amplitude and phase of the seasonal components to be dynamic, but keep the regressor amplitudes constant in time; ***we do this because the seasonal cycle in the observational composites can change over time either as a physical feedback of changing temperature and ozone, or***

*due to different observations exhibiting different seasonal amplitudes (not shown) that are a result of the observing instruments 'seeing' slightly different parts of the atmosphere or having different sampling. Due to the seasonal cycle having the largest variability of all modes we expect that, if left unaccounted for, the time varying seasonal modulation might have an influence other parts of the regression. In principle other regressor amplitudes could also have some time modulation for similar reasons. We leave an investigation of more flexible DLM models with dynamic regressor amplitudes to future work where a physically-motivated justification for such freedom can be investigated."*

L137-140: This is not true if EESC is used to represent the long-term trend.

We agree in part, however, we would argue it is more complicated and our assertion remains reasonable. The long term 'background' component, that which the DLM non-linear trend or a piece-wise linear trend is supposed to represent, is likely to represent the impact of GHG and ODSs (or EESC). Due to the need to account for a lag required in an EESC regressor, a second EESC curve is needed to represent a shift in the EESC; either way these are fixed shapes. The DLM does not require a fixed background trend prescription. Therefore, we would argue that our description is reasonable. Additionally, we would argue it is true that a piecewise linear trend does not represent the temporal evolution of EESCs, GHGs, and their combined effects, especially when the inflection date is fixed the same for all altitudes and latitudes.

L128-155: Are there any references in the literature to support the claims made in this section? There is a lot of literature on using MLR techniques when determining ozone trends, and any literature which assesses the DLM technique should also be cited.

It is true there is limited literature on this. Laine et al., 2014 and Ball et al., 2017 are the main ones for ozone, with the former comparing results DLM with MLR on real timeseries, and the latter considering idealized (simulated) test cases to compare performance. Ball et al., 2017 showed that, in the validation-test cases they considered (where the background trend is "known"), DLM out-performed MLR on trend reconstruction. A comprehensive inter-comparison of the two approaches is still needed, but the description as laid out here is formally correct, regarding the propagation of errors etc, so still provides a sound foundation for motivating DLM over MLR. We have cited these two papers again at an appropriate point in this section.

L151: What is MCMC – it does not appear to be defined in the manuscript.

MCMC means Monte Carlo Markov Chain, a type of algorithm for sampling probability distributions; this has been added to the manuscript.

L197-202: The authors should be clear here that they are discussing ozone trends

*This has been clarified.*

L204: Add 'in lower stratospheric ozone' between increase and reported

*Done.*

L225: Replace 'upswing' – also elsewhere in the manuscript.

*Done.*

L240-241: specifically in the lower stratosphere, where the lifetime of ozone is long – please clarify that in this sentence.

*Added the part sentence as the reviewer suggested here.*

L250: The authors could add here further discussion, with appropriate references, discussing other drivers of variability.

We have added the following:

‘...but the variability is less regular, unsurprisingly since the NH stratosphere is known to have additional variability , ***a consequence of greater sea-land contrast and more orography than in the SH. The NH thus exhibits stronger large scale wave activity and polar vortex and stratospheric variability (see Butchart et al. (2014) and Kidston et al. (2015) and references therein).***’

L273-287: This paragraph is very confusing and should be re-written to aid the readers understanding of the QBOs influence on the transport of ozone, and how this differs at different latitudes. This is a key point of the paper, and so spending some time clarifying this section will significantly improve the manuscript.

We have made amendments to this paragraph to improve understanding.

“We reiterate that the separation of positive and negative anomalies into those related to Easterly or Westerly QBO phases is clearest for the SH (Fig. 4a) and the corresponding, opposing, equatorial changes (Fig. 4b). The anti-correlated behaviour of anomalies between midlatitude and equatorial regions is consistent with previous studies investigating the relationship between the QBO and mid-latitude ozone variability (Zerefos et al., 1992; Randel et al., 1999; Strahan et al., 2015). We summarise the dynamical concept, in the context of these results, in the following (see Baldwin et al. (2001) and Choi et al. (2002) for detailed discussion). The QBO consists of downward propagating equatorial zonal winds; in the lower stratosphere this consists of a Westerly above an Easterly, or vice versa. For Westerly above Easterly (i.e. the 15 hPa QBO is Westerly as identified by blue lines in Fig. 4) leads to a shear that induces a anomalously downward motion of air, and adiabatic warming (Fig. 1 of Choi et al. (2002)) and also to an anomalous increase in ozone; for an Easterly above a westerly, this leads to anomalously rising air and adiabatic cooling together with an associated ozone decreases; an equator-to-mid-latitude circulation forms to conserve mass (Randel et al., 1999; Polvani et al., 2010, Tweedy et al., 2017). At sub-tropical and mid-latitudes, the return of this meridional circulation draws ozone-rich air from above down into ozone poor regions, anomalously enhancing ozone (yellow, Fig. 4a,c). When Easterlies lie over Westerlies (blue, Fig. 4), the opposite circulation is set up, and ozone anomalies reverse.”

Figure 4: Why does the red dashed line in the lower panels not have the same value where it intercepts the left and right hand side axes? The figure caption says the red dashed line is for January to October 2017, so the July value should be the same on both, as both are July 2017.

This is because each 12 month period is treated independently and normalised to September in each 12 month sequence. If there is a trend, or multi-year variability, or short-term fluctuations, it will not be the same level each year due to the normalisation in September. Of course, in reality they are connected and the same value in an absolute sense.

L311: Remove ‘other’

Instead replaced with ‘timescale’.

L325-334: The sensitivity of total column ozone trends to the choice of end year is also discussed by Weber et al. (2018) and Keeble et al. (2018), both of which could be cited here. Weber et al. has also now been published and the citation in the reference list and throughout the manuscript should be updated to reflect this.

We have included references, as well as Frith et al. (2014), to these sensitivity studies in the footnote that accompanied the point regarding the curve being ‘locked-in’ over certain timescales; we include Keeble et al., 2018, but note they did not demonstrate, though did discuss, the sensitivity of MLR to the data. However, we note that MLR sensitivity analyses are not directly comparable with the DLM technique in the sense that the whole period for which a linear trend is estimated in MLR is affected, whereas a limited timescale is affected in DLM due to a local change in the gradient of the non-linear curve close to the perturbation.

L373-284: The authors should clarify that here they are discussing ozone trends and not ozone values, which should not be affected by the addition of subsequent years.

Done; see additions to revised manuscript.

L387: change ‘tropical’ to ‘tropics’

Done.

L401-403: Care must be taken here not to equate the success of the Montreal Protocol with increases in ozone. The Montreal Protocol is working – look at decreases to anthropogenic Cl – but it is not possible to say, without exploring the drivers of the changes, that increasing ozone in the upper stratosphere is evidence of that success. See also major comment above for use of ozone recovery and ozone increases.

An important but subtle point raised by the reviewer, which we agree with. We have preceded this point with (bold/italic):

***‘An upper stratospheric increase is the expected result from long-term stratospheric chlorine reductions, a direct consequence of the Montreal Protocol and its amendments, though we do not explicitly attribute the cause of the increase to that here (for more on attribution see, for example WMO (2018). Indeed, the Montreal Protocol and its amendments will have been effective in reducing ozone losses throughout the atmosphere through reductions in CFC emissions, HCFCs and other ODSs. The lack of a positive trend since 1998 in the lower stratosphere, as opposed to the one clear in the upper stratosphere, is likely the consequence of other factors such as dynamical changes (Wargan et al., 2018). These results once again reinforce the result the conclusion that only the SH is affected by the 2017 ozone increase (lower stratosphere), that the Montreal Protocol is appears to be working (upper stratosphere), and that the decreases in the lower stratosphere at tropical and NH latitudes remain in place, but are not yet fully understood.’***

L412-414: Consider rewording this sentence for clarity – as it is currently written, it is very hard to follow what is meant.

Rewritten:

***‘The aim of this work is to assess the current state of, and trends in, stratospheric ozone. Improved knowledge of such trends, and the relevant forcing mechanisms and associated variability, will help to better constrain CCM projections of ozone to the end of the 21st Century.’***

L423-425: Clarify here that this is a result from a modelling study, not an extension of the observed record of ozone changes.

We have done this: ‘A recent study used a CTM to reconstruct ~~extend~~ the ozone timeseries **beyond the observational record available at the time to 2017** and found lower stratospheric ozone had rapidly increased in 2017 back to 1998 levels.’

L454: Replace ‘spread on’ with ‘spread in’

Done.

L464: Replace 'predictions' with 'projections'

Done.

L465-484: There is an important feedback here, as ozone changes also affect the dynamics. Several CCM studies have highlighted the impacts of ozone depletion on stratospheric circulation, and as ozone starts to recover due to reductions of ODS, stratospheric dynamics will respond.

A good suggestion to add. We have now included the following after mention of a change in large scale stratospheric circulation: *'Further, ozone is not a passive tracer, but dynamics responds to ozone changes (Li et al., 2018; Polvani et al., 2018; Abalos et al., 2019), as has been demonstrated most notably in the SH following ozone depletion and strengthening of the ozone hole (WMO, 2014; WMO, 2018); as ozone is expected to recover in the coming decades, the dynamics of the stratosphere are also expected to respond.'*



# **Stratospheric ozone trends for 1985–2018: sensitivity to recent large variability**

William T. Ball<sup>1,2</sup>, Justin Alsing<sup>3,4</sup>, Johannes Staehelin<sup>1</sup>, Sean M. Davis<sup>5</sup>,  
Lucien Froidevaux<sup>6</sup>, and Thomas Peter<sup>1</sup>

<sup>1</sup>Institute for Atmospheric and Climate Science, Swiss Federal Institute of Technology Zurich,  
Universitaetstrasse 16, CHN, CH-8092 Zurich, Switzerland

<sup>2</sup>Physikalisch-Meteorologisches Observatorium Davos World Radiation Centre, Dorfstrasse 33,  
7260 Davos Dorf, Switzerland

<sup>3</sup>Oskar Klein Centre for Cosmoparticle Physics, Stockholm University, Stockholm SE-106 91,  
Sweden

<sup>4</sup>Physics Department, Blackett Laboratory, Imperial College London, SW7 2AZ, UK

<sup>5</sup>NOAA Earth System Research Laboratory Chemical Sciences Division, Boulder, CO, USA

<sup>6</sup>Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA

*Correspondence to:* W. T. Ball (william.ball@env.ethz.ch)

**Abstract.** The Montreal Protocol , **and its subsequent amendments**, has successfully prevented catastrophic losses of stratospheric ozone, and signs of recovery are now evident. Nevertheless, recent ~~work suggests~~ **work has suggested** that ozone in the lower stratosphere (<24 km) continued to decline over 1998–2016, offsetting recovery at higher altitudes and preventing a statistically significant increase in quasi-global (60°S – 60°N) total column ozone. In 2017, a large lower stratospheric ozone resurgence over less than 12 months was estimated (using a chemistry-transport model; CTM) to have ~~wiped-out~~ **offset** the long-term decline in the quasi-global integrated lower stratospheric ozone column. Here, we extend the analysis of space-based ozone observations to December 2018 using the BASIC<sub>SG</sub> ozone composite. We find that the observed 2017 resurgence was only around half that modelled by the CTM, was of comparable magnitude to other strong inter-annual changes in the past, and restricted to southern ~~hemispheric~~ **hemisphere** mid-latitudes (SH; 60°S–30°S). In the SH mid-latitude lower stratosphere, the data suggest that by the end of 2018 ozone is still likely lower than in 1998 (probability ~80%). In contrast, tropical and northern hemisphere (NH) ozone continue to display ongoing decreases, exceeding 90% probability. Robust tropical (>95%, 30°S–30°N) decreases dominate the quasi-global integrated decrease (99% probability); the integrated tropical stratospheric column (1–100 hPa, 30°S–30°N) displays a significant overall **ozone** decrease, with 95% probability. These decreases do not reveal an inefficacy of the Montreal Protocol. Rather, they suggest other effects to be at work, mainly dynamical variability on long or short timescales, counteracting the **positive effects of the Montreal Protocol on stratospheric ozone recovery** ~~protocol's regulation of halogenated ozone-depleting substances (hODS)~~. We demonstrate that large inter-annual mid-latitude (30°–60°) variations, such as the 2017 resurgence, are driven by non-linear quasi-biennial oscillation (QBO) phase-dependent seasonal variability. However, this variability is not represented in current regression analyses. To understand if observed lower stratospheric **ozone** decreases are a transient or long-term phenomenon, progress needs to be made in accounting for this dynamically-driven variability.

## 1 Introduction

Ozone in the stratosphere acts as a protective shield against ultraviolet radiation that may harm the biosphere, and leads to cataracts, skin damage, and skin cancer in humans (Slaper et al., 1996; WMO, 2014, 2018). In the latter half of the 20<sup>th</sup> century, the emission of long-lived halogen-containing ozone depleting substance (hODSs) led to ~5% loss in quasi-global (60°S–60°N) integrated total column ozone (WMO, 2014), which represents the combined changes in tropospheric and stratospheric ozone contributions. The 1987 Montreal Protocol and its amendments **and adjustments** led to a reduction in hODSs that ~~coincided with~~ **resulted in** a halt in total column ozone losses around 1998–2000 (Harris et al., 2015; Chipperfield et al., 2017).

However, there is still no evidence of a **statistically** significant increase in total column ozone since 1998 (Chipperfield et al., 2017; Weber et al., 2018; Ball et al., 2018), despite a significant increase in upper stratospheric ozone (1–10 hPa) (Ball et al., 2017; Steinbrecht et al., 2017; Ball et al., 2018; Petropavlovskikh et al., 2019). Ball et al. (2018) and Ziemke et al. (2018) presented evidence, using OMI/MLS tropospheric column observations for 2005–2016, that tropospheric ozone had also increased significantly. However, large uncertainties remain in quasi-global tropospheric ozone trends, and the recent Tropospheric Ozone Assessment Report (TOAR) shows that different tropospheric ozone products give a wide range of trends, some even indicating negative changes (Gaudel et al., 2018). **The importance of considering tropospheric and stratospheric changes separately to understand changes in total column ozone has also been highlighted in recent studies using chemistry climate models (CCMs) (Meul et al., 2016; Keeble et al., 2017; Dhomse et al., 2018).** If tropospheric and upper stratospheric ozone have indeed both increased, then the observed flat trend in total column ozone implies that middle and lower stratospheric ozone should have decreased.

To assess trends in stratospheric ozone, composites **of observations** must be formed by merging multiple ozone **observational** timeseries into a long, multi-decadal record from which variability can be attributed, and long-term trends determined. Composites are subject to artefacts from merging different observing platforms. Multiple papers (Tummon et al., 2015; Harris et al., 2015; Steinbrecht et al., 2017; Ball et al., 2017, 2018) and a SPARC report (Petropavlovskikh et al., 2019) review, discuss, and attempt to account for the artefacts in the uncertainty budget.

Ball et al. (2018) integrated ozone over the whole stratosphere, i.e. the ozone layer, quasi-globally for pressure levels from 147–1 hPa (~13–48 km) at mid-latitudes (30°–60°), and 100–1 hPa (~16–48 km) between the sub-tropics (30°S–30°N), and found ozone to be lower in 2016 than in 1998 in multiple ozone composites. In their analysis, the lower stratosphere (147/100–32 hPa, ~13/17–24 km) was driving this decrease. The most significant decreases were in the tropics, but negative trends extended out into the mid-latitudes (Fig. 1d). Other studies have subsequently confirmed these negative trends (Zerefos et al., 2018; Wargan et al., 2018; Chipperfield et al., 2018). Evidence points towards dynamical variations driving changes (Chipperfield et al., 2018), perhaps in the form of enhanced isentropic mixing (Wargan et al., 2018). **Part of the negative trends in northern hemispheric stratospheric ozone in the 1980s and 1990s at higher latitudes have been previously attributed to synoptic and planetary waves (Hood and Zaff, 1995; Hood et al., 1999) inducing large localised (e.g. over Europe) wintertime decreases in ozone that might in turn be driven by sea surface temperature and eddy flux changes on decadal or longer timescales, although most of these studies are limited to the end of the last century when ODSs remained an established primary driver of the decrease. The El Niño Southern Oscillation (ENSO) and Quasi-Biennial Oscillation (QBO) are known to influence the dynamical variability in the lower stratosphere and may be a main player in driving inter-annual and decadal variability in this region (Diallo**

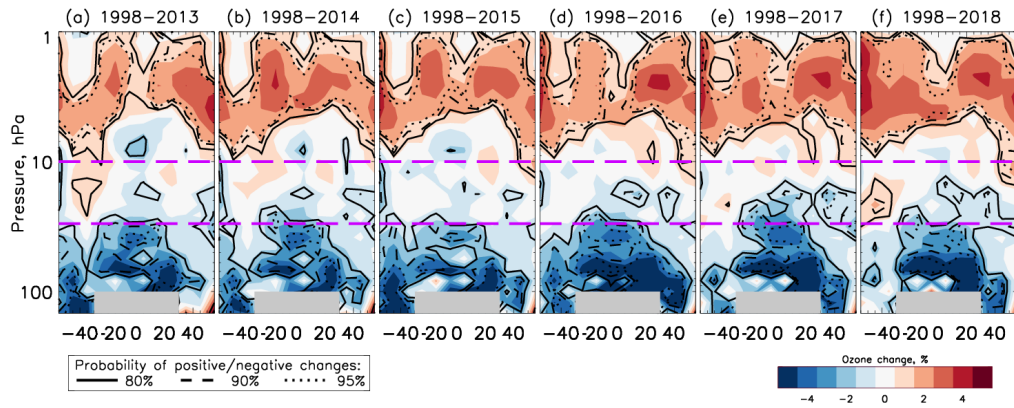


Figure 1: Zonally averaged ozone change between 1998 and end years (a) 2013 to (f) 2018. Red represents increases, blue decreases (%; right legend). Contours represent probability levels of positive or negative changes (left legend). Grey-shaded regions represent unavailable data. Pink dashed-lines delimit regions integrated to partial ozone columns in other figures.

et al., 2018, 2019). Nevertheless, these dynamical changes do not in themselves determine a specific underlying driving force, however the effect of ~~Although the underlying driving force has not yet been determined, climate change~~ **increasing anthropogenic greenhouse gases (GHGs)** is an obvious, though unverified candidate (Hood and Soukharev, 2005; Peters and Entzian, 1999) on specific mechanisms needs further study (Ball et al., 2018). On the other hand, Stone et al. (2018) showed that negative **ozone** trends could be simulated in the lower stratosphere over the same period in two of nine ensemble members of a coupled CCM as a result of natural variability interfering in the (linear) trend analysis **although none of the ensembles displayed the same widespread negative trends as detected in observations Ball et al. (2018)**. They suggested that an additional seven years of observations would lead to negative signals disappearing in favour of positive trends. The implication is then that the observed negative trend over the relatively short 19 year timeframe may be a temporary result from large natural variability (in the single realisation) of the real-world, rather than ~~forced by climate change~~ **a response to increasing GHGs**.

Chipperfield et al. (2018) used a chemistry transport model (CTM) to reconstruct ozone variability close to past real-world behaviour; transport in the CTM is driven by ERA-Interim (Dee et al., 2011) reanalysis fields. The results showed changes similar to those presented by Ball et al. (2018) up to December 2016. They extended their CTM analysis by an additional 12 months to find that the 1998–2016 **ozone** decline in the lower stratosphere (~2 DU; Ball et al. (2018)) was ~~wiped out~~ **offset** by a sudden increase of ozone in 2017, exceeding 8 DU quasi-globally. This was attributed almost entirely to dynamical changes and was primarily located in the southern hemisphere; **Froidevaux et al. (2019) have noted that ozone trends derived from Aura/MLS data over a shorter period (2005–2018) have a tendency towards slightly positive values in the SH, but not so elsewhere within**

the extra-polar regions. Chipperfield et al. (2018) they suggested that the lower stratospheric ozone decrease was a result of large natural variability that biased the trend analysis, and that the variability could be attributed to dynamics and not to chemical or photolytic changes, although the source of dynamical perturbations was not identified or the impact on trends quantified. **Thus, an assessment of this recent variability on trends, and an update to 2018 is needed and is a key aim of this study.**

Here, we update the observational analysis of Ball et al. (2018) to include data to the end of 2018 (Section 3.1). This allows us to assess the impact of the 2017 ozone increase in the lower stratosphere on the trend analysis, and to consider additional changes over 2018. We show that large ozone-increase events, with a duration and magnitude similar to that of 2017 (Chipperfield et al., 2018) have occurred regularly since 1985 at mid-latitudes (Section 3.2), and find the events are linked to a seasonally-dependent QBO effect (Section 3.3). We update partial column ozone trends from 2016 to 2018 in section 3.4. Finally, we consider the sensitivity of trends to the recent increase of stratospheric ozone (Sections 3.5-3.6) by considering six periods that start in 1985 and end between 2013 and 2018, in order to demonstrate where signals are robust to the end date, and where not. Such an analysis is essential to establish if the negative trends are a result of natural variability interfering in the trend analysis, and to take the first steps to account for what ~~may~~ **might** be driving the large, short-term variability.

## 2 Data and methods

### 2.1 Ozone Data

Although other ozone composites exist (Petropavlovskikh et al., 2019), we focus exclusively on data formed from merging ozone from SWOOSH (Davis et al., 2016) and GOZCARDS (Froidevaux et al. (2015); here we use the v2.20 update of Froidevaux et al. (2019)) using the so-called BASIC (BAYesian Integrated and Consolidated) approach (Ball et al., 2017) to account for artefacts in merged composites and improve trend estimates. ~~This data was~~ **These data were** referred to as ‘Merged-SWOOSH/GOZCARDS’ by Ball et al. (2018), but we refer here to it as BASIC<sub>SG</sub>. To briefly place the SWOOSH and GOZCARDS datasets in ~~the~~ context of BASIC<sub>SG</sub>, Figure S1 of Ball et al. (2018) presented 1998–2016 changes using SWOOSH or GOZCARDS alone; this figure reveals that these ozone composites show generally similar changes on large spatial scales, though there are clear differences on small scales, e.g. in the tropical upper stratosphere, and in the southern hemisphere lower stratosphere. Figure S2 of Ball et al. (2018) importantly demonstrates at 100 hPa in the tropical lower stratosphere that there are significant differences between SWOOSH and GOZCARDS in the late 1990s; this figure also shows that BASIC<sub>SG</sub> is able to account for the differences in a principled way that is not simply the averaging of the two products, which is particularly important for having confidence in an assessment of **ozone in** the lower stratosphere. We extend

BASIC<sub>SG</sub> from Ball et al. (2018) by two years to cover 1985–2018. This period is essentially an extension of the Aura Microwave Limb Sounder (Aura/MLS); both SWOOSH and GOZCARDS consider Aura/MLS exclusively after 2009.

We only consider BASIC<sub>SG</sub> here for the following reasons. First, as discussed in Ball et al. (2018), compared to the other composites it had the least apparent artefacts within the timeseries. The Stratosphere-troposphere Processes And their Role in Climate (SPARC) Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) report (Petropavlovskikh et al., 2019) indicates this method to be more robust to outliers than other composites. Second, BASIC<sub>SG</sub> is resolved in the lower stratosphere, which is not the case for all composites; for further discussion see Ball et al. (2018) and the SPARC LOTUS report (Petropavlovskikh et al., 2019). Additionally, SWOOSH and GOZCARDS are currently two of the most up-to-date composites available. Finally, we are interested here in the sensitivity of stratospheric ozone changes to different end years and, since Aura/MLS is arguably one of the best remote sensing platforms for ozone currently in operation (Petropavlovskikh et al., 2019), focusing only on BASIC<sub>SG</sub> provides an analysis, discussion, and interpretation that is free from the complications of considering multiple composites that have multivariate reasons for displaying different behaviour.

## 2.2 Regression analysis

As in Ball et al. (2018), we perform all timeseries analysis using dynamical linear modelling (DLM) (Laine et al., 2014) ; ~~we point the reader to Laine et al. (2014) for details on this method, and we provide a short overview here. We use~~ using the public DLM code DLMMC (available at <https://github.com/justinalsing/dlmmc>). **We provide a short overview of DLM here.**

Our DLM approach models the ozone timeseries as a (dynamical) linear combination of the following components: two seasonal components (with 6- and 12-month periods respectively), a set of regressor variables (i.e., proxy timeseries describing various known drivers), an auto-regressive (AR) process, and a smooth non-linear (non-parametric) background trend. DLM differs from traditional multiple linear regression (MLR) approaches in a number of key ways. Firstly, while MLR fits for a fixed (constant-in-time) linear combination of seasonal, regressor, and trend components, DLM can allow the amplitudes of the various components to vary dynamically in time, capturing richer phenomenology in the data. Here, we allow the amplitude and phase of the seasonal components to be dynamic, but keep the regressor amplitudes constant in time; **we do this because the seasonal cycle in the observational composites can change over time either as a physical feedback of changing temperature and ozone, or due to different observations exhibiting different seasonal amplitudes (not shown) that are a result of the observing instruments ‘seeing’ slightly different parts of the atmosphere or having different sampling. Due to the seasonal cycle having the largest variability of all modes we expect that, if left unaccounted for, the time varying seasonal modulation might have an influence on the regression. In principle other regressor**



165 **amplitudes could also have some time modulation for similar reasons; we leave an investigation of more flexible DLM models with dynamic regressor amplitudes to future work where a physically-motivated justification for such freedom can be investigated.** Secondly, MLR that **does not assume a driver for the long-term trends, e.g. for the influence of ODSs or GHGs)** typically assumes a fixed prescription for the shape of the background trend, e.g., a piecewise-linear  
170 or independent-linear trend with some fixed, pre-chosen inflection-date. These assumptions are both restrictive and give a poor representation of the smooth background trends we expect from nature (Laine et al., 2014; Ball et al., 2017). DLM addresses this by instead modelling the trend as a smooth, non-parametric, non-linear curve, where the ‘smoothness’ of the trend is controlled by a free parameter that is included in the fit (see supplementary materials Fig. S1). Thirdly, in practice MLR is often  
175 performed by first subtracting an estimated mean seasonal cycle, fitting the trend and regressor variables to the anomalies, and then making a post-hoc correction for auto-regressive residuals, **although many do fit annual and semi-annual components.** This procedure typically does not propagate the errors on the seasonal cycle and AR parameters in a rigorous way, leading to misrepresentation of uncertainties. DLM addresses this by inferring all components of the model simultaneously, and formally marginalizing over the uncertainties in all other parameters when reporting uncertainties on  
180 e.g., the trend. We use the same prior assumptions as described in Ball et al. (2018).

Probabilities of an overall increase (decline) in ozone between two dates (Figs. 1, 7, and Table 1) are computed as the fraction of Monte Carlo Markov Chain (MCMC) samples that show positive (negative) change. Credible intervals (Figs. 6, 8, 9) are computed as the central 95 and 99 percentiles  
185 of the MCMC samples. The use of ‘confidence’ or ‘significance’ is used in this paper interchangeably with ‘probability’ and refers specifically to Bayesian probabilities; it does not refer to the application of frequentist significance tests and/or confidence intervals.

We use the same regressors as Ball et al. (2018): solar (30 cm radio flux, F30) (Dudok de Wit et al., 2014)), volcanic (latitudinally resolved stratospheric aerosol optical depth, SAOD) (Thomason et al., 2017), ENSO (NCAR, 2013), and the Quasi-Biennial Oscillation, QBO, at 30 and 50 hPa <sup>1</sup>.  
190 In previous analyses, we considered the Arctic and Antarctic Oscillation, AO/AAO <sup>2</sup>, as proxies for Northern and Southern surface pressure variability only for partial column ozone analysis in their respective hemisphere; here we also consider them for the spatially-resolved analysis and in all cases use both AO and AAO simultaneously – they have little affect outside their respective regions, but  
195 we do not limit the possibility they may influence some variability in either hemisphere (Tachibana et al., 2018). We use a first order autoregressive (AR1) process (Tiao et al., 1990) to consider auto-correlation in the residuals. We remove a three year period following the Pinatubo eruption, i.e. June 1991 to May 1994, which is a year longer than the previous analysis, to avoid any effects of the eruptions that may have persisted. Another key point regarding the SAOD proxy is that, unlike the

<sup>1</sup>QBO indices: <http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/>

<sup>2</sup>AO/AAO indices: <http://www.cpc.ncep.noaa.gov/products/precip/CWlink/>

other proxies that have been fully updated to the end of 2018 for this analysis, the SAOD is **currently** not **available extended** beyond 2016, so we repeat the year 2016 for 2017 and 2018; if any deviations in the SAOD occurred during this period, our analysis will not account for this. Nevertheless, as can be seen in Fig. 1d here, in comparison to Fig. 1b of Ball et al. (2018), all of these adjustments to the procedure from Ball et al. (2018) have little impact on the estimated mean changes in ozone.

## 3 Results

### 3.1 Stratospheric ozone changes since 1998

Figure 1d shows the pressure-latitude, spatially-resolved 1998–2016 ozone change, reproducing Fig. 1b of Ball et al. (2018). Minor differences exist because the BASIC<sub>SG</sub> composite ~~has~~ **and DLM procedure have** been updated. Ozone in the lower stratosphere (delimited by the pink dashed line at 32 hPa, or 24 km) shows a marked and almost hemisphere-symmetric decrease, while upper stratospheric changes (>10 hPa, 32 km) are mainly positive; the middle stratosphere generally shows relatively flat **ozone** trends since 1998 with low probability of an overall change.

Figures 1e and f show the 1998–2017 and –2018 ozone changes, respectively. Four points of interest emerge from the comparison to 1998–2016: (i) while still negative, the magnitude of the lower stratospheric SH (60°S–30°S) **ozone** decrease has become smaller and less significant (~~~80% probability~~); (ii) tropical (30°S–30°N) and NH (30°N–50°N) changes remain negative and highly probable; (iii) the probability (and magnitude) of negative **ozone** trends over tropical and NH regions in the middle stratosphere (32–10 hPa) has increased; and (iv) the magnitude and probability of upper stratospheric **ozone increase** has strengthened. Importantly, Fig. 1 demonstrates the robustness of negative **ozone** trends in the lower, and positive trends in the upper, stratosphere irrespective of the final year of the analysis. Figures 1a–f present ozone changes from 1998 to end years 2013 through 2018, showing the sensitivity of ozone trends to six consecutive end years. These end years give insight into the sensitivity of the trends to large inter-annual variability. In particular, these six years encompass periods of both negative/Easterly and positive/Westerly phases of ENSO/QBO. These modes are major contributors to stratospheric variability (Zerefos et al., 1992; Tweedy et al., 2017; Tothir et al., 2018; Garfinkel et al., 2018; Diallo et al., 2018, 2019), and any sensitivity of the end year to the state of these drivers should be encapsulated in ~~this group~~ **the set** of spatial responses **depending on the end year only (Fig. 1)**, particularly if these modes were not well-captured by DLM predictors (Fig. 1). A lower stratosphere negative **ozone** trend is persistent for all end years. For 1998–2013, there is a highly probable negative trend in **ozone** in the SH lower stratosphere; the probability is retained until 2016, after which it reduces. The opposite is seen in the NH, where only a small region of probable ozone decrease exists for 1998–2013, and this strengthens with each panel until 2016, after which a highly probable decrease **of ozone** remains stable. There is no apparent switch from negative to positive **ozone** changes in these regions for any of the six end years.

The reduced probability of a SH decrease is related, as we will see in Section 3.2, to the rapid 2017 increase in **SH mid-latitude lower stratospheric ozone** reported by Chipperfield et al. (2018) using a CTM. However, Fig. 1 also confirms in observations that this is localised to south of 30°S and does not reveal coherent or consistent behaviour over time with the NH, suggesting that there may be large, hemispherically independent variability interfering with the trend estimates. Nevertheless, there are no signs as yet of an ozone increase underway in the quasi-global lower stratosphere.

Further, the decrease in ozone in the tropical lower stratosphere increases in magnitude and significance as more data is **are** added. The tropical lower stratospheric ozone is projected to decrease by the end of the century in all ~~chemistry-climate models (CCMs)~~ (Dhomse et al., 2018), due to enhanced upwelling from the Brewer Dobson circulation (BDC) as a result of ~~climate change~~ **changes to stratospheric dynamics from increasing GHGs** (Polvani et al., 2018). It is possible that this is a detection of the expected tropical lower stratosphere decline in ozone, earlier than expected (WMO, 2014). However, whilst the data show a significant decline, it remains to be seen if this can be attributed to the ~~climate change~~ **anthropogenic GHG** induced upwelling of the BDC.

### 3.2 On the rapid increase in ozone in 2017

Chipperfield et al. (2018) reported a rapid increase in the quasi-global lower stratospheric ozone in 2017, modelled using a CTM driven by ERA-Interim reanalysis to represent dynamical variability closer to that which occurred historically. The quasi-global, deseasonalised timeseries from BASIC<sub>SG</sub> is shown in Fig. 2a; 2017 is bounded by the vertical dashed lines and the large increase is highlighted in red from a minimum in November 2016 to a maximum reached 12 months later in October 2017.

The observed 2016–2017 increase in Fig. 2a was 5.5 DU, which is 63% of the 8.7 DU increase reported by Chipperfield et al. (2018). Split into three latitude bands, 60°–30°S, 30°S–30°N, and 30°–60°N (Figs. 2b–d), we find that the ~~upswing~~ **rapid increase** can be decomposed into a 12 DU increase in the SH, 3 DU in the tropics, and 6 DU in the NH. Weighting for latitude – 21, 58, and 21% respectively – the SH contribution accounts for nearly half of the quasi-global increase (2.5 DU, 1.9 DU, 1.3 DU). The overall increase is composed of two sub-periods, dominated by a NH increase until May 2017, and a SH increase over April–August 2017; the tropical region saw comparatively little change in the second period. **We do not know why the CTM and observations disagree in the magnitude of change for this period.**

Importantly, the rapid increase seen in 2017 is not unique. Four other quasi-global ‘events’ of this type are found over 1985–2018, shown in Fig. 2a; the identification ~~criteria~~ **criterion** was an increase of at least 90% of the 2017 increase occurring within a 13 month period. The decomposed timeseries (Fig. 2b–d) show that the large increases in the SH are *normal*, occurring regularly. They also occur in the NH, but not as regularly, and the tropical variability is much smaller than the mid-latitude variance. In addition to the large increases, there are also comparatively large negative swings in

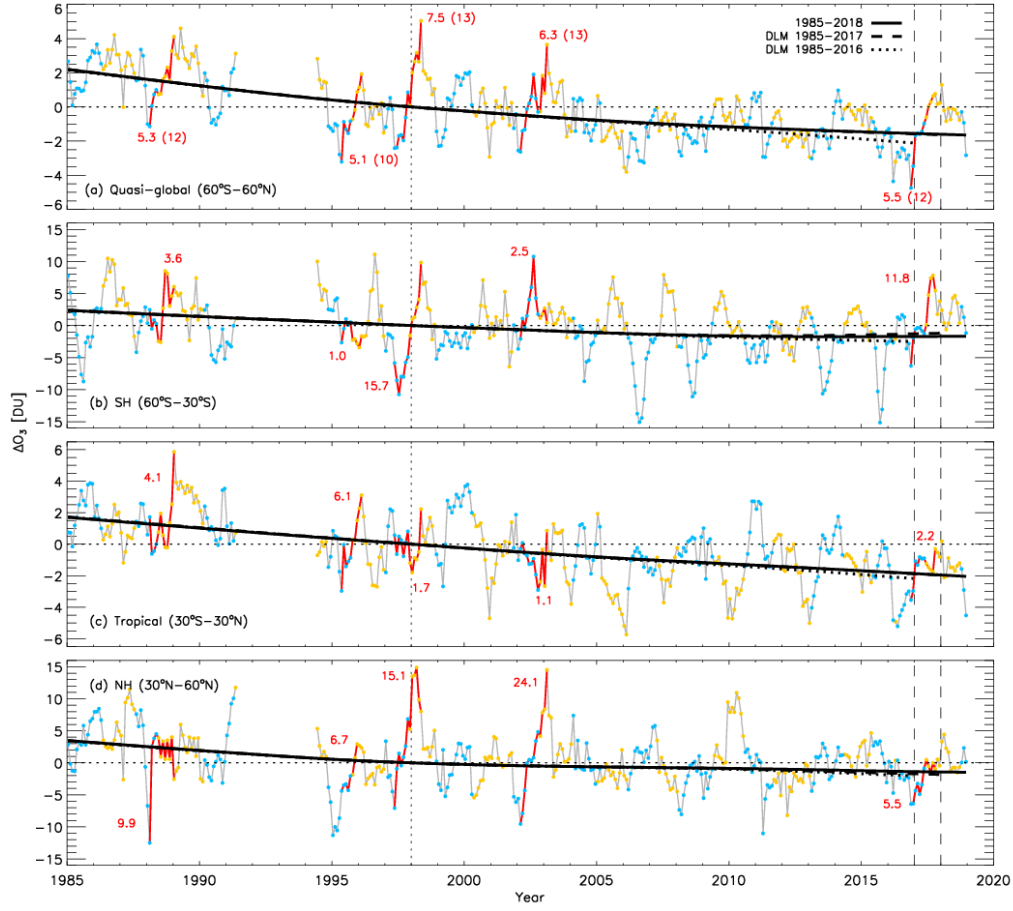


Figure 2: Lower stratospheric partial column ozone anomalies, (a) quasi-global ( $60^{\circ}\text{S}$ – $60^{\circ}\text{N}$ ), (b) southern hemisphere ( $60^{\circ}\text{S}$ – $30^{\circ}\text{S}$ , 147–30 hPa), (c) tropics ( $30^{\circ}\text{S}$ – $30^{\circ}\text{N}$ , 100–30 hPa), (d) northern hemisphere ( $30^{\circ}\text{N}$ – $60^{\circ}\text{N}$ , 147–30 hPa). The DLM non-linear trend is shown for 1985–2016 (dotted), 1985–2017 (dashed), and 1985–2018 (solid). Red lines represent contiguous periods identified in the quasi-global anomalies exceeding 90% of the magnitude of the November 2016 to October 2017 change within a 13 month period; the DU changes are written above or below each period; red periods in (b–d) are those identified in (a). Colour dots are plotted on each timeseries when the QBO at 30 hPa is either in an Easterly (yellow) or Westerly (blue) phase. The three-year period following the eruption of Mt. Pinatubo, June 1991 to May 1993, has been removed. Figures for the whole, upper and middle stratosphere are provided in the Supplementary Materials Figs. S2–4.

both SH and NH timeseries – one in the NH beginning in 2002 exceeds 24 DU. In the following section we argue that these large, rapid changes are driven by a non-linear seasonal-QBO effect.

### 3.3 Contribution of QBO to mid-latitude ozone variability

Chipperfield et al. (2018) convincingly showed that the majority of post-1997 quasi-global **lower stratospheric** ozone variability in Fig. 2a was dynamically controlled, **specifically in the lower stratosphere where the lifetime of ozone is long**. Given that the contributions from each sub-region (Fig. 2b–d) add up to the quasi-global change in 2017, it is reasonable to assume that dynamics controls much of the sub-decadal variability there too. The peaks (or troughs) in the SH are 2–3 years apart; the QBO has a similar periodicity and is known to have the largest inter-annual dynamical impact on ozone in the stratosphere (see Gray and Pyle (1989), Zerefos et al. (1992), and Toth et al. (2018), and references therein). Labelling each month in Fig. 2 with the 30 hPa QBO-Easterly or Westerly phase in yellow or blue dots, respectively, reveals that the large SH negative anomalies are almost always associated with a Westerly phase, while positive anomalies are associated with an Easterly phase; **Bodeker et al. (2007) previously identified large SH negative anomalies in 1985, 1997 and 2006 and related these to the QBO-Westerly phase. This also appears to be the case in the NH, but the variability is less regular, unsurprisingly since the NH stratosphere is known to have additional variability, a consequence of greater sea-land contrast and more orography than in the SH. The NH thus exhibits stronger large scale wave activity and polar vortex and stratospheric variability (see Butchart (2014) and Kidston et al. (2015) and references therein).** Equatorial variability in ozone related to the QBO phase at 30 hPa shows the opposite behaviour to that of the at mid-latitudes: decreases in ozone generally appear to occur with the Easterly phase and vice versa, and the return from maximum excursion (i.e. the sign of the gradient) appears to be more related to the change in phase.

Histograms of the ozone anomalies relative to the DLM trend line for each QBO phase at 30 hPa are shown in the upper row of Fig. 3. The shift in the histogram between QBO phases is clear in the SH; the NH displays little shift, again likely related to other drivers influencing NH ozone changes, though the extremes show a similar phase separation as in the SH. The difference between the QBO Easterly and Westerly histograms are shown in the bottom row, and make clear the correlation between QBO state and ozone anomalies.

To clarify this further, in Fig. 4 all 34 years in the 1985–2018 period are split into 13-month periods starting in January for the SH (upper row) and July for the NH (lower row), i.e. a few month prior to the onset of winter in the respective hemisphere; the latitudes plotted are refined to isolate clearer signals for (a) 50°–30°S, (c) 30°–50°N, and (b,d) 10°S–10°N. This refinement reduces the influence of polar variability on the 30°–50° band, and isolates the equatorial region to where the QBO variability is strongest; we note that the act of forming partial columns of ozone may reduce the integrated variability compared to counter-varying layers that would otherwise be resolved by pressure level. We find the use of the QBO phase at 70 15 hPa also better separates the events in this additional analysis. We find negative and positive **ozone** excursions in the lower stratosphere become clear in 13-month segments when they are bias-shifted to zero in March (a,b) and September (c,d)

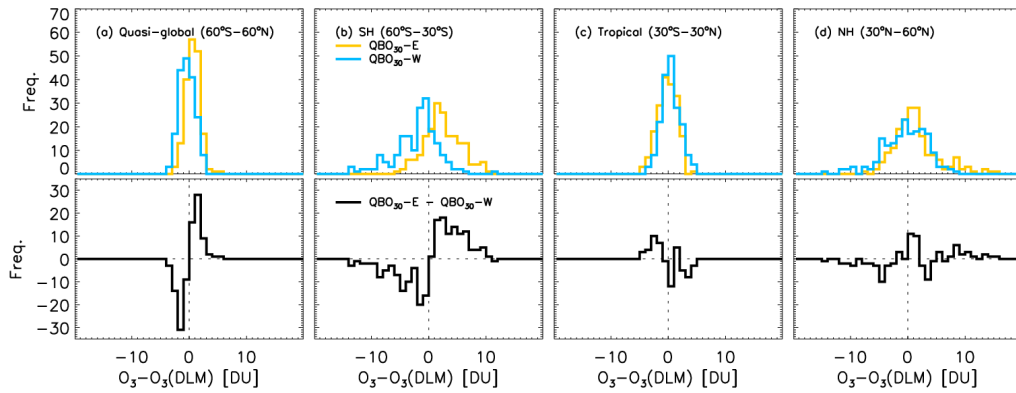


Figure 3: **(Upper row)** Histograms of ozone anomalies relative to the DLM non-linear trend line in Fig. 2 for months when the QBO at 30 hPa is either in an Easterly (yellow) or Westerly (blue) phase: (a) quasi-global (60°S–60°N), (b) southern hemisphere (60°–30°S, 147–30 hPa), (c) tropics (30°S–30°N, 100–30 hPa), (d) northern hemisphere (30°–60°N, 147–30 hPa). **(Lower row)** Difference between QBO Easterly and Westerly histograms from the upper row.

and then colour coded according to their QBO phase in April or October, respectively (vertical dotted line). The largest deviations are found to occur four **or five** months later (vertical dashed line), at the onset of hemispheric autumn (Holton and Tan, 1980; Dunkerton and Baldwin, 1991). **It is also interesting to note that the only large, positive QBO-Westerly anomaly that peaks four months later, in either hemisphere, occurs in the SH in 2002. This year is famous for having the only** observed sudden stratospheric warming in the southern hemisphere, and indicates that while the QBO-phase appears to dominate this distribution of anomalies, other processes can also sometimes dominate.

We reiterate that the separation of positive and negative anomalies into those related to Easterly or Westerly QBO phases is clearest for the SH (Fig. 4a) and the corresponding, opposing, equatorial changes (Fig. 4b). The anti-correlated behaviour of anomalies between mid-latitude and equatorial regions is consistent with previous studies investigating the relationship between the QBO and mid-latitude ozone variability (Zerefos et al., 1992; Randel et al., 1999; Strahan et al., 2015). We summarise the dynamical concept, in the context of these results, in the following (see Baldwin et al. (2001) and Choi et al. (2002) for detailed discussion). The QBO consists of downward propagating equatorial zonal winds; in the lower stratosphere this consists of a Westerly above an Easterly, or vice versa. For Westerly above Easterly (i.e. the 15 hPa QBO is Westerly as identified by blue lines in Fig. 4) leads to a shear that induces a anomalously downward motion of air, and adiabatic warming (Fig. 1 of Choi et al. (2002)) and also to an anomalous increase in ozone; for an Easterly above a westerly, this leads to anomalously rising air and adiabatic cooling together with an associated ozone decreases; an



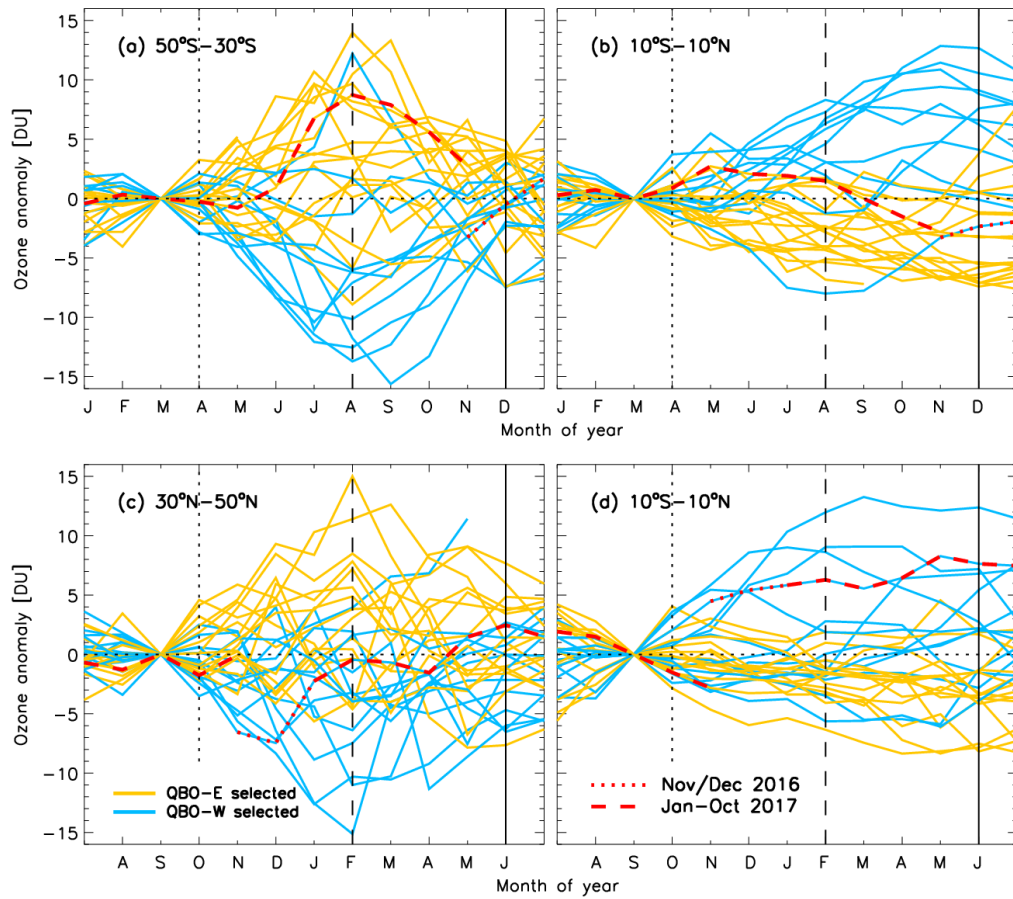


Figure 4: Lower stratospheric partial column ozone at (a)  $50^{\circ}$ – $30^{\circ}$ S, (b,d)  $10^{\circ}$ S– $10^{\circ}$ N, and (c)  $30^{\circ}$ – $50^{\circ}$ N. Each line represents a 13 month period starting in January (upper row) or July (lower row), all bias-shifted to zero in March (upper) or September (lower) and colour coded by the state of the QBO at 15 hPa in April (upper) or October (lower) so that QBO Easterly phases are yellow and Westerly blue. The period covering November to December 2016 is highlighted as a dotted-red line, while January to November 2017 is dashed-red.

equator-to-mid-latitude circulation forms to conserve mass (Randel et al., 1999; Polvani et al., 2010; Tweedy et al., 2017). At sub-tropical and mid-latitudes, the return of this meridional circulation draws ozone-rich air from above down into ozone poor regions, anomalously enhancing ozone (yellow, Fig. 4a,c). When Easterlies lie over Westerlies (blue, Fig. 4), the opposite circulation is set up, and ozone anomalies reverse.

The 2017 increase is highlighted in Fig. 4, with November 2016 to January 2017 shown as a dotted red line, and January to October 2017 as a red dashed line. Focusing on Fig. 4a in the SH, the increase onset during the Easterly phase is large, but as noted earlier larger excursions have occurred before and regularly (Fig. 2). A prolonged Westerly phase, following the breakdown of the expected

Table 1: Absolute change between 1998 and 2018 in Dobson Units, and probability (in brackets) of a positive or negative change in ozone (%) for integrated regions of the stratosphere. Blue text indicates ozone changes are negative, while red indicate positive changes; bold text indicates probabilities exceeding 90%.

Region	60°–30°S	50°–30°S	30°S–30°N	20°S–20°N	30°–50°N	30°–60°N	50°S–50°N	60°S–60°N
Whole	-0.5 (57)	-0.4 (56)	<b>-1.9 (95)</b>	<b>-2.5 (95)</b>	-3.2 (83)	-2.6 (77)	-1.1 (86)	-1.1 (86)
Upper	<b>+0.8 (100)</b>	<b>+0.8 (99)</b>	<b>+0.7 (96)</b>	+0.6 (85)	<b>+0.6 (92)</b>	<b>+0.6 (96)</b>	<b>+0.8 (100)</b>	<b>+0.8 (100)</b>
Middle	<b>+0.7 (78)</b>	<b>+0.8 (78)</b>	<b>-0.7 (94)</b>	<b>-0.9 (95)</b>	-0.5 (73)	-0.5 (72)	-0.4 (81)	-0.2 (73)
Lower	-1.6 (80)	-1.7 (82)	<b>-2.1 (99)</b>	<b>-2.1 (98)</b>	-1.9 (82)	-1.5 (76)	<b>-1.8 (99)</b>	<b>-1.7 (99)</b>

340 QBO pattern in 2016 (Osprey et al., 2016; Newman et al., 2016; Tweedy et al., 2017), may have contributed to a suppressed level of ozone in 2016 (note the single orange dot in 2016 in Fig. 2a signifying a brief Easterly QBO phase). The arrival of the Easterly phase proper in 2017 led to the ozone increase at mid-latitudes. The Westerly phase at 30 hPa began in late 2018 and ozone should, barring no further QBO breakdown, decrease again in 2019 in the SH mid-latitudes; the last three  
345 months of 2018 hint at such a decrease (Fig. 2).

Despite this variability, Fig. 1 indicates that the lower stratospheric negative trends **in ozone** could already be identified throughout the lower stratosphere before, and after, 2016. As such, the QBO breakdown event is likely not the primary cause of the negative **ozone** trends reported by Ball et al. (2018), but does appear to affect the robustness of the trend depending on the end year. We will  
350 investigate this end-year sensitivity in section 3.5.

### 3.4 Latitude-integrated lower stratospheric ozone trend estimates

While Chipperfield et al. (2018) applied ordinary least squares trend fits to timeseries using a single linear trend, this cannot be compared to multi-variate regression approaches, e.g. DLM and MLR. This is because the former simply asks what the trend in the data is, regardless of the forcing agents,  
355 while the latter attempts to separate known (usually quasi-periodic) drivers to distill out the trend that has (usually unknown) drivers of its own. The DLM non-linear trend estimates presented here are the first multivariate analysis applied to ozone timeseries that include the large ozone increase witnessed in 2017. It is important to be clear that long-term trends cannot be compared with single year changes; indeed, the processes governing each **other timescale** are likely quite different. Whilst  
360 **While** large short-term increases will likely bias the whole trend-line for that period under MLR analyses (with piecewise linear and independent linear trends – PWLT or ILT), DLM promises to be more robust in the sense that asymmetric fluctuations will only influence part of the smooth trend over a timescale fixed by the smoothness parameter  $\sigma_{trend}$  that controls how rapidly the trend is allowed to evolve (see Ball et al. (2017) and Fig. S1).

365 The DLM trends **in lower stratospheric ozone** estimated over 1985–2018 in Fig. 2 continue to be negative, monotonic trends up to 2018 in the quasi-global, tropical and NH regions, while the SH trend reaches a minimum in ~2013 before slowly rising. All integrated regions suggest the mean remains below the 1998 level (see Table 1; an extended supplementary materials Table S1 provides changes in DU, %, and %}decade for 1985–2018, 1985–1997, and 1998–2018), though  
 370 the probability of an overall decrease is 99% in the quasi-global, dominated by the tropics (99%), with probabilities of a decrease of 80% and 76% in the SH and NH respectively. Except in the SH, monotonic downward trends remain, with the addition of two years only affecting the gradient of the monotonic trends (compare dotted line for the year ending in 2016, with the dashed for 2017 and solid for 2018 in Fig. 2). This agrees with Chipperfield et al. (2018) who suggested the large  
 375 upswing **rapid increase** of 2017 ~~that settled in 2018~~ affected trends, although this was mainly in the SH **and has subsequently showed little change over 2018**. However, it has done little to reduce the overall probability of a decrease in the quasi-global timeseries (99%). Furthermore, the shape of the DLM curve is affected only near the end years, such that the period away from the end-date is relatively insensitive to a change in the end year and becomes ‘locked-in’<sup>3</sup>. As **the DLM-estimated**  
 380 **changes in ozone relative to 1998** in years prior to 2010 are essentially unaffected by the addition of 2017 and 2018, the data show robustly that lower stratospheric ozone did continue to decrease until at least 2010 in all regions. We speculate that the shift back to a QBO-Westerly phase will again decrease ozone at mid-latitudes in 2019 (which appears to have begun in October 2018, see Fig. 2). If that happens it is therefore possible that the non-linear trend estimates will likely decrease again,  
 385 **with and the emergent 2013 minimum in the DLM non-linear trend estimate** seen in Fig. 2b is likely to shift to a later date or disappear.

### 3.5 Sensitivity of DLM trends to the end year and non-linear seasonal-QBO effects

Since mid-latitude ozone excursions depend on the QBO-seasonal interaction, i.e. the QBO phase relative to the time of year, this is a non-linear mode of variability. Without predictors to represent  
 390 this non-linear behaviour, linear regression models (including both DLM and MLR) cannot capture these excursions and the variability can leak into, and bias, other predictors; most importantly, this may include the trend component of the regression model. In this section we examine the magnitude of this effect through a sensitivity analysis of the DLM trends to the end date of the data, both spatially and with partial columns.

395 Due to the magnitude of the mid-latitude, seasonally-dependent QBO ozone variability on short (two to three year) timescales, ILT or PWLT applied to the relatively short post-1997 timeseries will be sensitive to these large swings in ozone. For the smooth DLM trends on the other hand, we expect the last few years of the curve will be primarily affected, with the rest of the trend being stable. We

<sup>3</sup>In contrast, for MLR analyses, the entire trend-line is impacted by changes in the end year (Frith et al., 2014; Weber et al., 2018; Keeble et al., 2018); this is a good example of the inadequacy of using linear trends to describe these data.

demonstrate the affect on DLM in Fig. 5 where the partial column regions as presented in Ball et al. (2018) are shown for 10° bands, and quasi-globally (right column), over the whole stratospheric column (top), as well as upper, middle and lower. We show DLM curves estimated from six periods that start in 1985 and end in 2013–2018 as in Fig. 1; all curves are bias shifted to zero in 1998. This provides a visualisation of the sensitivity of the non-linear trends to the end year (and hence also the large resurgence **in ozone** in 2017). The uncertainties associated with a change between 1998 and the end year are presented in Fig. 6 with 95% (dark grey shading) and 99% credible intervals. The results specifically for the 1998–2018 change are combined and presented in Fig. 7 as probability distributions, in the same manner as in Fig. 2 of Ball et al. (2018), where blue and red colours represent negative and positive changes respectively, and numbers above each distribution are the probability of the change (fraction of the probability distributions) being negative.

From the panels of Fig. 5, it is clear that **ozone trends in** the middle-stratosphere exhibits the largest sensitivity to the end year and **the uncertainties in the change from 1998** are consistently large (Fig. 6); quasi-globally the change **since 1998** is negative for all end years, but does not exceed 95% probability. The upper stratosphere is also sensitive **to the end year** in the tropics (Fig. 5), ~~but~~ **and the end year** has shifted ~~the estimated ozone change~~ from negative to positive **with increasing end year**, although **the uncertainty** always ~~uncertain~~ **remains large** (Fig. 6); at mid-latitudes uncertainties **in the change since 1998** are smaller, but there has been a general shift towards more positive and significant increases, which is more-clearly reflected in the SH and quasi-global estimates. The evolution of the lower and whole stratospheric non-linear **ozone** trends mimic each other: south of 30°S, the end points of the negative changes have quickly increased in 2017 and 2018 **relative to 1998**, though remain negative in the lower stratosphere; at latitudes north of 30°S, the addition of 2017 and 2018 ~~have~~ **has** made little difference to the monotonic **ozone** decline; for 50–60°N, while flat, the additional years make little difference. The quasi-global lower stratosphere **ozone** continues to exhibit a monotonic decline that is still highly confident with 99% probability (Fig. 7 and Table 1), and **ozone abundances integrated over** the whole stratosphere **continues to** remain lower **in 2018** than in 1998, though this is now with a probability of 86%; these trends are dominated by the tropical contribution (58%, latitude weighted) to the quasi-global change, whereas **the** NH and SH contribute 21% each. Even so, the NH changes do not appear affected by the recent large seasonally-dependent QBO variability.

Figure 5 also confirms that the **gradients of the** non-linear curves are only affected by unmodelled variance in years close to the end points, **typically within the last five years of the partial column timeseries considered here; the shape of the DLM curves** ~~changes~~ prior to the **final** last five years **of the DLM curves** are largely unaffected ~~in the partial columns~~. Indeed, even with the large **ozone** increase in 2017 in the SH, we see that all trend-**curves** agree well prior to 2010; this is true in other panels, e.g. the middle stratosphere and tropical upper stratosphere. In the upper stratosphere the recovery onset remains robust, but in the SH lower stratosphere the large increase in 2017 results in

the non-linear trend curve having a local minimum emerge around 2013. As such we can infer that additional data are unlikely to affect the inferred **level change** of ozone in 2013, **relative to 1998**, or push the minima to earlier dates, because the affecting end year moves further away with more data. However, subsequent data might once again push ~~trends~~ **the changes since 1998** to lower levels, e.g.

440 if mid-latitudes do respond to a Westerly-phase QBO with ozone reducing sharply as it has done in the past (Fig. 2). We expand the idea of inferring the likely earliest minimum using the DLM with spatially-resolved data in the Supplementary Materials.

### 3.6 Update on ozone profiles

Briefly, in Fig. 8, we provide updated ozone change profiles for 1998–2018 using the standard latitudinal ranges for the SH (60°–35°S), ~~tropical~~ **tropics** (20°S–20°N), and NH (35°–60°N) (WMO, 2014, 2018; Steinbrecht et al., 2017; Petropavlovskikh et al., 2019). Fig. 8, also includes 1998–2016 and 1998–2017 profiles for comparison and shows that, for 1998–2018, confidence in an upper stratospheric **ozone** recovery **from ODSs** is clear for all latitude bands, including the tropics where it has previously remained below the 95% significance levels. The lower stratosphere shows negative

450 **ozone** changes at almost all levels, though these generally do not exceed a probability of 95%.

Ball et al. (2018), and Fig. 7, ~~indicated~~ **indicate** that the 50–60° zonal means in both hemispheres show little **ozone** change in the lower stratosphere in the last 21 years, while the tropical regions out to 30° show a strong decrease. By modifying the latitudinal extent of the profiles slightly, so that mid-latitudes cover 30–50° to exclude 50–60° and the tropics are widened to 30°S–30°N to include

455 the subtropics, the modified profiles are presented in Fig. 9. This provides some measure of the sensitivity to the latitudinal ranges chosen. Now we see the tropics show close to 95% confidence of an ozone decrease at all tropical lower stratospheric pressure levels, and there is increased confidence of an ozone reduction in the mid-latitude lower stratosphere. Further, the inclusion of higher latitude regions (20–30°) reinforces the tropical upper stratosphere **ozone** increase. **An upper stratospheric**

460 **increase is the expected result from long-term stratospheric chlorine reductions, a direct consequence of the Montreal Protocol and its amendments, though we do not explicitly attribute the cause of the increase to that here (for more on attribution see, for example, WMO (2018)).** **Indeed, the Montreal Protocol and its amendments will have been effective in reducing ozone losses throughout the atmosphere through reductions in CFC emissions, HCFCs and other**

465 **ODSs. The lack of a positive trend since 1998 in the lower stratosphere, as opposed to the one clear in the upper stratosphere, is likely the consequence of other factors such as dynamical changes (Wargan et al., 2018).** These results once again reinforce ~~the result~~ **the conclusion** that only the SH is affected by the 2017 **ozone** increase (lower stratosphere), that the Montreal Protocol **is appears** to be working (upper stratosphere), and that the decreases in the lower stratosphere at

470 tropical and NH latitudes remain in place, but are not yet fully understood.

## 4 Conclusions

Here, we have extended and analysed the BASIC<sub>SG</sub> stratospheric ozone composite from Ball et al. (2018) by two years to cover 1985–2018. BASIC<sub>SG</sub> merges two composites, SWOOSH and GOZ-CARDS. We perform a set of sensitivity tests, using dynamical linear modelling (DLM), on the post-1997 trend estimates to understand the impact of a recently reported, large increase in modelled ozone in the lower stratosphere in 2017 (Chipperfield et al., 2018), following almost two decades of persistently decreasing ozone.

~~The aim of this work is to build confidence in the current evolution of stratospheric ozone, which is essential to assess ozone trends from observations to assess and ensure model projections to the end of the 21<sup>st</sup> Century are accurate.~~ **The aim of this work is to assess the current state of, and trends in, stratospheric ozone. Improved knowledge of such trends, and the relevant forcing mechanisms and associated variability, will help to better constrain CCM projections of ozone to the end of the 21<sup>st</sup> Century.** Chemistry models resolving the stratosphere are **one of** the best tools for attribution and long-range studies of ozone, but different types exist: free-running CCMs generate their own model-dependent internal climate and variability; chemistry transport models (CTMs) use wind, temperature and surface pressure fields fully prescribed by reanalyses; and specified-dynamics CCMs (SD-CCMs) use reanalyses to nudge the internally-generated variability of the model closer to the historical variability in the real atmosphere while attempting to retain model dependent processes and internal consistency. CTMs and SD-CCMs can be ~~extremely~~ useful for attributing historical changes in ozone to evolving concentrations of CO<sub>2</sub> and ODSs (Solomon et al., 2016), or the Sun (Ball et al., 2016), by accounting for dynamical variability in observations.

A recent study (Chipperfield et al., 2018) used a CTM to reconstruct ~~extend~~ the ozone timeseries **beyond the observational record available at the time to 2017 and found that that model simulated a lower stratospheric ozone increase in 2017 back to 1998 levels;** this was attributed to dynamical variability. **Indeed, chemistry and photochemistry play a dominant role over dynamical perturbations in the upper stratosphere as ozone lifetimes are short (~days), while ozone lifetimes of ~6–12 months in the lower stratosphere means that equator-to-mid-latitude transport of similar timescales plays an important (dominant) role there (London, 1980; Perliski et al., 1989; Brasseur and Solomon, 2005).** CTMs can provide insight as to whether the changes might be driven by photochemistry, chemistry, or dynamics. However, because the dynamical fields are prescribed, the CTM cannot provide insight into the underlying dynamical driver of the long-term decreases or the 2017 increase. We show here that the 2017 increase simulated by the CTM (Chipperfield et al., 2018) was more than 60% larger than that observed, and that the 1998–2017 and –2018 (Fig. 1e and f) change remains negative (60°S–60°N), and significant in the tropics and some sub-regions of the NH (Fig. 1f). Neither free-running CCMs (WMO, 2014), nor SD-CCMs (Ball et al., 2018), have so far been demonstrated to accurately reproduce the long-term changes estimated from observations in ~~the lower stratosphere~~ **ic ozone** (Fig. 6).

The effect of the ozone increase in 2017 was small and the probability of an overall ozone decrease **in the lower stratosphere** remains at 99% for 1998–2018 (**-1.7 DU, or 2.0%**; see Table S1). We note that the lower stratospheric **ozone** trends are dominated by the tropical regions (30°S–30°N) where the decrease is robust to the end year over 2013–2018, with a probability of 99% (**-2.1 DU, -3.5%**) that it was lower in 2018 than in 1998. Nevertheless, mid-latitudes out to 50°N also indicate that the decrease persists (**-1.9 DU, -1.7%**). We also find that the 2017–2018 addition enhances the **estimated** magnitude of the upper stratospheric ozone ~~recovery~~ **positive trend**, but that the ~~recovery of the~~ quasi-global (60°S–60°N) ozone layer still displays a reduction since 1998, though the confidence in this has reduced from 95% in 2016 (Ball et al., 2018) to 86% in 2018 (**-1.1 DU, -0.4%**). Given the high probability of a decrease in tropical middle (94%) and lower (99%) stratospheric ozone, the whole tropical stratospheric **ozone** column indicates a highly probable decrease (95%) over 1998–2018 (**-1.9 DU, -0.8%**).

In general, uncertainties on changes since 1998 in partial columns have changed little over 2013–2018 (**Fig. 6**), a result likely due to the large fraction of unaccounted variance in the standard set of predictors used in regression analysis. Our analysis shows that ozone continued to decrease until a minimum in at least 2013 in the SH, and **has continued to decrease** at all latitudes north of 30°S. By comparing the phase of the QBO with large, 2–3 year inter-annual variability at mid-latitudes, the implication is that these large mid-latitude changes are related to the seasonal-dependence of **ozone** on the QBO, i.e. a non-linearity; if true, this could explain why regression models cannot capture this variability, since such non-linear behaviour is not included. The clarification of the origin of these large mid-latitude changes – occurring every few years – is a high priority.

CCMs are consistent in the sign of their projections, although lower stratospheric **ozone** variability can differ with observations and there is a large spread ~~on~~ **in** their sensitivity to hODSs (Douglass et al., 2012, 2014), and therefore their return dates, **i.e. a return of ozone to the level it was in 1980 (WMO, 2014; Dhomse et al., 2018; WMO, 2018)**. CCMs do a good job on many timescales, but due to historically different internal variability, and parametrized sub-grid scale processes and numerical ~~inaccuracies~~ **diffusion**, behaviour in some regions may not be well-reproduced (SPARC/WMO, 2010). It is clear from modelling studies that pre-Montreal Protocol ozone decreases can be attributed to ODS increases (WMO, 2014), and CCMs and CTMs generally reproduce the Antarctic ozone hole well (Solomon et al., 2016). The **halt in ODS-related ozone losses as a result of the Montreal Protocol and its amendments**, and **an** initial recovery **from ODSs** in total column ozone is almost universally reproduced by CCMs (SPARC/WMO, 2010), as is the upper stratospheric **ozone** recovery. But, **ozone** trends since 1998 in the lower stratosphere have not been demonstrated to be simulated in models in the mid-latitudes, most notably in the NH.

Future ~~predictions~~ **projections** tend to focus on how stratospheric ozone will evolve under a given global warming scenario; this is important given that the changing climate **due to anthropogenic GHG emissions** may impact inter-annual dynamical variability (Osprey et al., 2016; Newman et al.,

2016; Tweedy et al., 2017), and changes in the large-scale circulation in the stratosphere is **are** likely to modify future distributions of ozone (Chipperfield et al., 2017). **Further, ozone is not a passive tracer, but dynamics responds to ozone changes (Li et al., 2018; Polvani et al., 2018; Abalos et al., 2019), as has been demonstrated most notably in the SH following ozone depletion and strengthening of the ozone hole (WMO, 2014, 2018); as ozone is expected to recover in the coming decades, the dynamics of the stratosphere are also expected to respond.** The overall expectations are that total column ozone levels will return to ~~1960s~~ **1980s** levels globally by ~2050, in the Antarctic by 2100, and by ~2030 and ~2050 in Northern and Southern mid-latitudes, respectively, continuing on to a ‘super-recovery’, **i.e. that ozone will be higher by the end of the 21<sup>st</sup> Century than prior to 1980s levels** (Dhomse et al., 2018; WMO, 2014, 2018), **although this is predicated on future scenarios of hODSs decreases continuing as expected (Montzka et al., 2018).** However, it is neither clear whether the recent increase in SH lower stratospheric ozone will remain at higher levels or will reduce again in 2019 as the QBO shifts to a Westerly phase, nor why the NH continues to show a persistent decrease. Nonetheless, we note that the signal is small compared to the (i) large inter-annual variability, (ii) pre-2000 changes induced by ozone depleting substances, and (iii) ozone losses that would have occurred without the Montreal Protocol being enacted.

The ongoing negative trend of ozone in the lower stratospheric component of the total column also continues to pose a problem for global trends in tropospheric ozone. If tropospheric ozone has really increased over the last two decades, and stratospheric ozone was not decreasing or remained flat, then some component of the total column ozone must have been decreasing to balance the **ozone budget since it appears that total column ozone has remained steady in the past 5-10 years.** Alternatively, it is possible that the solution simply lies in very large observational uncertainties (Harris et al., 2015; Gaudel et al., 2018; Petropavlovskikh et al., 2019) and/or the inadequacies of linear regression techniques to attribute variability and identify trends. **In addition to potential future improvements in** merged observational records, this calls for a community push to improve detection and attribution techniques to solve an issue that is of great importance to the health of society, the biosphere, and the climate.

*Author contributions.* WTB designed the experiments; WTB and JA prepared and executed the BASIC algorithms; JA developed the DLM code and WTB and JA performed the DLM analysis; WTB conceived and performed the QBO analysis. SD and LF prepared and provided GOZCARDS and SWOOSH ozone datasets. WTB prepared the manuscript with contributions from all co-authors.

*Acknowledgements.* W.T.B. was funded by the SNSF project 200020\_182239 (POLE). ‘BASIC<sub>SG</sub>’ for 1985–2018 will be available for download from <https://data.mendeley.com/datasets/2mgx2xzzpk/3> following review of this manuscript. Work at the Jet Propulsion Laboratory was performed under contract with the National



580 Aeronautics and Space Administration. GOZCARDS ozone data contributions from Ryan Fuller (at JPL) are gratefully acknowledged.

## References

- Abalos, M., Polvani, L., Calvo, N., Kinnison, D., Ploeger, F., Randel, W., and Solomon, S.: New Insights on the Impact of Ozone-Depleting Substances on the Brewer-Dobson Circulation, *Journal of Geophysical Research (Atmospheres)*, 124, 2435–2451, doi:10.1029/2018JD029301, 2019.
- Baldwin, M. P., Gray, L. J., Dunkerton, T. J., Hamilton, K., Haynes, P. H., Randel, W. J., Holton, J. R., Alexander, M. J., Hirota, I., Horinouchi, T., Jones, D. B. A., Kinnersley, J. S., Marquardt, C., Sato, K., and Takahashi, M.: The quasi-biennial oscillation, *Reviews of Geophysics*, 39, 179–229, doi:10.1029/1999RG000073, 2001.
- Ball, W. T., Haigh, J. D., Rozanov, E. V., Kuchar, A., Sukhodolov, T., Tummon, F., Shapiro, A. V., and Schmutz, W.: High solar cycle spectral variations inconsistent with stratospheric ozone observations, *Nature Geoscience*, 9, 206–209, doi:10.1038/ngeo2640, 2016.
- Ball, W. T., Alsing, J., Mortlock, D. J., Rozanov, E. V., Tummon, F., and Haigh, J. D.: Reconciling differences in stratospheric ozone composites, *Atmospheric Chemistry & Physics*, 17, 12 269–12 302, doi:10.5194/acp-17-12269-2017, 2017.
- Ball, W. T., Alsing, J., Mortlock, D. J., Staehelin, J., Haigh, J. D., Peter, T., Tummon, F., Stuebi, R., Stenke, A., Anderson, J., Bourassa, A., Davis, S. M., Degenstein, D., Frith, S., Froidevaux, L., Roth, C., Sofieva, V., Wang, R., Wild, J., Yu, P., Ziemke, J. R., and Rozanov, E. V.: Evidence for a continuous decline in lower stratospheric ozone offsetting ozone layer recovery, *Atmospheric Chemistry & Physics*, 18, 1379–1394, doi:10.5194/acp-18-1379-2018, 2018.
- Bodeker, G. E., Garny, H., Smale, D., Dameris, M., and Deckert, R.: The 1985 Southern Hemisphere mid-latitude total column ozone anomaly, *Atmospheric Chemistry & Physics*, 7, 5625–5637, 2007.
- Brasseur, G. P. and Solomon, S.: *Aeronomy of the Middle Atmosphere: Chemistry and Physics of the Stratosphere and Mesosphere*, Dordrecht: Springer Netherlands, Editor: Mysak, L. A., 2005.
- Butchart, N.: The Brewer-Dobson circulation, *Reviews of Geophysics*, 52, 157–184, doi:10.1002/2013RG000448, 2014.
- Chipperfield, M. P., Bekki, S., Dhomse, S., Harris, N. R. P., Hassler, B., Hossaini, R., Steinbrecht, W., Thiéblemont, R., and Weber, M.: Detecting recovery of the stratospheric ozone layer, *Nature*, 549, 211–218, doi:10.1038/nature23681, 2017.
- Chipperfield, M. P., Dhomse, S., Hossaini, R., Feng, W., Santee, M. L., Weber, M., Burrows, J. P., Wild, J. D., Loyola, D., and Coldewey-Egbers, M.: On the Cause of Recent Variations in Lower Stratospheric Ozone, *Geophys. Res. Lett.*, 45, 5718–5726, doi:10.1029/2018GL078071, 2018.
- Choi, W., Lee, H., Grant, W. B., Park, J. H., Holton, J. R., Lee, K.-M., and Naujokat, B.: On the secondary meridional circulation associated with the quasi-biennial oscillation, *Tellus Series B Chemical and Physical Meteorology B*, 54, 395, doi:10.3402/tellusb.v54i4.16673, 2002.
- Davis, S. M., Rosenlof, K. H., Hassler, B., Hurst, D. F., Read, W. G., Vömel, H., Selkirk, H., Fujiwara, M., and Damadeo, R.: The Stratospheric Water and Ozone Satellite Homogenized (SWOOSH) database: a long-term database for climate studies, *Earth System Science Data*, 8, 461–490, doi:10.5194/essd-8-461-2016, 2016.
- Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-

J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Quarterly Journal of the Royal Meteorological Society*, 137, 553–597, doi:10.1002/qj.828, 2011.

625 Dhomse, S. S., Kinnison, D., Chipperfield, M. P., Salawitch, R. J., Cionni, I., Hegglin, M. I., Abraham, N. L., Akiyoshi, H., Archibald, A. T., Bednarz, E. M., Bekki, S., Braesicke, P., Butchart, N., Dameris, M., Deushi, M., Frith, S., Hardiman, S. C., Hassler, B., Horowitz, L. W., Hu, R.-M., Jöckel, P., Josse, B., Kirner, O., Kremser, S., Langematz, U., Lewis, J., Marchand, M., Lin, M., Mancini, E., Marécal, V., Michou, M., Morgenstern, O., O'Connor, F. M., Oman, L., Pitari, G., Plummer, D. A., Pyle, J. A., Revell, L. E., Rozanov, E., Schofield, R., Stenke, A., Stone, K., Sudo, K., Tilmes, S., Visionsi, D., Yamashita, Y., and Zeng, G.: Estimates of ozone return dates from Chemistry-Climate Model Initiative simulations, *Atmospheric Chemistry & Physics*, 18, 8409–8438, doi:10.5194/acp-18-8409-2018, 2018.

630 Diallo, M., Riese, M., Birner, T., Konopka, P., Müller, R., Hegglin, M. I., Santee, M. L., Baldwin, M., Legras, B., and Ploeger, F.: Response of stratospheric water vapor and ozone to the unusual timing of El Niño and the QBO disruption in 2015-2016, *Atmospheric Chemistry & Physics*, 18, 13 055–13 073, doi:10.5194/acp-18-13055-2018, 2018.

Diallo, M., Konopka, P., Santee, M. L., Müller, R., Tao, M., Walker, K. A., Legras, B., Riese, M., Ern, M., and Ploeger, F.: Structural changes in the shallow and transition branch of the Brewer–Dobson circulation induced by El Niño, *Atmospheric Chemistry & Physics*, 19, 425–446, doi:10.5194/acp-19-425-2019, 2019.

640 Douglass, A. R., Stolarski, R. S., Strahan, S. E., and Oman, L. D.: Understanding differences in upper stratospheric ozone response to changes in chlorine and temperature as computed using CCMVal-2 models, *Journal of Geophysical Research (Atmospheres)*, 117, D16306, doi:10.1029/2012JD017483, 2012.

Douglass, A. R., Strahan, S. E., Oman, L. D., and Stolarski, R. S.: Understanding differences in chemistry climate model projections of stratospheric ozone, *Journal of Geophysical Research (Atmospheres)*, 119, 4922–4939, doi:10.1002/2013JD021159, 2014.

645 Dudok de Wit, T., Bruinsma, S., and Shibasaki, K.: Synoptic radio observations as proxies for upper atmosphere modelling, *Journal of Space Weather and Space Climate*, 4, A06, doi:10.1051/swsc/2014003, 2014.

Dunkerton, T. J. and Baldwin, M. P.: Quasi-biennial Modulation of Planetary-Wave Fluxes in the Northern Hemisphere Winter., *Journal of Atmospheric Sciences*, 48, 1043–1061, doi:10.1175/1520-0469(1991)048<1043:QBMOPW>2.0.CO;2, 1991.

650 Frith, S. M., Kramarova, N. A., Stolarski, R. S., McPeters, R. D., Bhartia, P. K., and Labow, G. J.: Recent changes in total column ozone based on the SBUV Version 8.6 Merged Ozone Data Set, *Journal of Geophysical Research (Atmospheres)*, 119, 9735–9751, doi:10.1002/2014JD021889, 2014.

Froidevaux, L., Anderson, J., Wang, H.-J., Fuller, R. A., Schwartz, M. J., Santee, M. L., Livesey, N. J., Pumphrey, H. C., Bernath, P. F., Russell, III, J. M., and McCormick, M. P.: Global Ozone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS): methodology and sample results with a focus on HCl, H<sub>2</sub>O, and O<sub>3</sub>, *Atmospheric Chemistry & Physics*, 15, 10 471–10 507, doi:10.5194/acp-15-10471-2015, 2015.

660 Froidevaux, L., Kinnison, D. E., Wang, R., Anderson, J., and Fuller, R. A.: Evaluation of CESM1 (WACCM) free-running and specified-dynamics atmospheric composition simulations using global multispecies satellite data records, *Atmos. Chem. Phys.*, 19, 4783–4821, doi:10.5194/acp-19-4783-2019, 2019.

Garfinkel, C. I., Gordon, A., Oman, L. D., Li, F., Davis, S., and Pawson, S.: Nonlinear response of tropical lower-stratospheric temperature and water vapor to ENSO, *Atmospheric Chemistry & Physics*, 18, 4597–4615, doi:10.5194/acp-18-4597-2018, 2018.

665 Gaudel, A., Cooper, O. R., and etc, E.: Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, *Elem Sci Anth*, 6, 10, doi:10.1525/elementa.291, 2018.

Gray, L. J. and Pyle, J. A.: A two-dimensional model of the quasi-biennial oscillation of ozone, *Journal of Atmospheric Sciences*, 46, 203–220, doi:10.1175/1520-0469(1989)046<0203:ATDMOT>2.0.CO;2, 1989.

670 Harris, N. R. P., Hassler, B., Tummon, F., Bodeker, G. E., Hubert, D., Petropavlovskikh, I., Steinbrecht, W., Anderson, J., Bhartia, P. K., Boone, C. D., Bourassa, A., Davis, S. M., Degenstein, D., Delcloo, A., Frith, S. M., Froidevaux, L., Godin-Beekmann, S., Jones, N., Kurylo, M. J., Kyrölä, E., Laine, M., Leblanc, S. T., Lambert, J.-C., Liley, B., Mahieu, E., Maycock, A., de Mazière, M., Parrish, A., Querel, R., Rosenlof, K. H., Roth, C., Sioris, C., Staehelin, J., Stolarski, R. S., Stübi, R., Tamminen, J., Vigouroux, C., Walker, K. A.,  
675 Wang, H. J., Wild, J., and Zawodny, J. M.: Past changes in the vertical distribution of ozone - Part 3: Analysis and interpretation of trends, *Atmospheric Chemistry & Physics*, 15, 9965–9982, doi:10.5194/acp-15-9965-2015, 2015.

Holton, J. R. and Tan, H.-C.: The Influence of the Equatorial Quasi-Biennial Oscillation on the Global Circulation at 50 mb., *Journal of Atmospheric Sciences*, 37, 2200–2208, doi:10.1175/1520-0469(1980)037<2200:TIOTEQ>2.0.CO;2, 1980.  
680

Hood, L., Rossi, S., and Beulen, M.: Trends in lower stratospheric zonal winds, Rossby wave breaking behavior, and column ozone at northern midlatitudes, *J. Geophysical Res.*, 104, 24,321–24,339, doi:10.1029/1999JD900401, 1999.

Hood, L. L. and Soukharev, B. E.: Interannual Variations of Total Ozone at Northern Midlatitudes Correlated with Stratospheric EP Flux and Potential Vorticity., *Journal of Atmospheric Sciences*, 62, 3724–3740, doi:10.1175/JAS3559.1, 2005.  
685

Hood, L. L. and Zaff, D. A.: Lower stratospheric stationary waves and the longitude dependence of ozone trends in winter, *J. Geophysical Res.*, 100, 25,791–25,800, doi:10.1029/95JD01943, 1995.

Keeble, J., Bednarz, E. M., Banerjee, A., Abraham, N. L., Harris, N. R. P., Maycock, A. C., and Pyle, J. A.:  
690 Diagnosing the radiative and chemical contributions to future changes in tropical column ozone with the UM-UKCA chemistry-climate model, *Atmospheric Chemistry & Physics*, 17, 13 801–13 818, doi:10.5194/acp-17-13801-2017, 2017.

Keeble, J., Brown, H., Abraham, N. L., Harris, N. R. P., and Pyle, J. A.: On ozone trend detection: using coupled chemistry-climate simulations to investigate early signs of total column ozone recovery, *Atmospheric  
695 Chemistry & Physics*, 18, 7625–7637, doi:10.5194/acp-18-7625-2018, 2018.

Kidston, J., Scaife, A. A., Hardiman, S. C., Mitchell, D. M., Butchart, N., Baldwin, M. P., and Gray, L. J.: Stratospheric influence on tropospheric jet streams, storm tracks and surface weather, *Nature Geoscience*, 8, 433–440, doi:10.1038/ngeo2424, 2015.

Laine, M., Latva-Pukkila, N., and Kyrölä, E.: Analysing time-varying trends in stratospheric ozone time series using the state space approach, *Atmospheric Chemistry & Physics*, 14, 9707–9725, doi:10.5194/acp-14-9707-2014, 2014.  
700

- Li, F., Newman, P., Pawson, S., and Perlwitz, J.: Effects of Greenhouse Gas Increase and Stratospheric Ozone Depletion on Stratospheric Mean Age of Air in 1960-2010, *Journal of Geophysical Research (Atmospheres)*, 123, 2098–2110, doi:10.1002/2017JD027562, 2018.
- 705 London, J.: The Observed Distribution and Variations of Total Ozone, in: *Atmospheric Ozone and its Variation and Human Influences*, edited by Nicolet, M. and Aikin, A. C., p. 31, 1980.
- Meul, S., Dameris, M., Langematz, U., Abalichin, J., Kerschbaumer, A., Kubin, A., and Oberländer-Hayn, S.: Impact of rising greenhouse gas concentrations on future tropical ozone and UV exposure, *Geophys. Res. Lett.*, 43, 2919–2927, doi:10.1002/2016GL067997, 2016.
- 710 Montzka, S. A., Dutton, G. S., Yu, P., Ray, E., Portmann, R. W., Daniel, J. S., Kuijpers, L., Hall, B. D., Mondeel, D., Siso, C., Nance, J. D., Rigby, M., Manning, A. J., Hu, L., Moore, F., Miller, B. R., and Elkins, J. W.: An unexpected and persistent increase in global emissions of ozone-depleting CFC-11, *Nature*, 557, 413–417, doi:10.1038/s41586-018-0106-2, 2018.
- NCAR: The Climate Data Guide: Multivariate ENSO Index, Retrieved from <https://climatedataguide.ucar.edu/climate-data/multivariate-enso-index>, 2013.
- 715 Newman, P. A., Coy, L., Pawson, S., and Lait, L. R.: The anomalous change in the QBO in 2015-2016, *Geophys. Res. Lett.*, 43, 8791–8797, doi:10.1002/2016GL070373, 2016.
- Osprey, S. M., Butchart, N., Knight, J. R., Scaife, A. A., Hamilton, K., Anstey, J. A., Schenzinger, V., and Zhang, C.: An unexpected disruption of the atmospheric quasi-biennial oscillation, *Science*, 353, 1424–1427, doi:10.1126/science.aah4156, 2016.
- 720 Perliski, L. M., London, J., and Solomon, S.: On the interpretation of seasonal variations of stratospheric ozone, *Journal of Geophysical Research*, 37, 1527–1538, doi:10.1016/0032-0633(89)90143-8, 1989.
- Peters, D. and Entzian, G.: Longitude-Dependent Decadal Changes of Total Ozone in Boreal Winter Months during 1979-92., *Journal of Climate*, 12, 1038–1048, doi:10.1175/1520-0442(1999)012<1038:LDDCOT>2.0.CO;2, 1999.
- 725 Petropavlovskikh, I., Godin-Beekmann, S., Hubert, D., Damadeo, R., Hassler, B., and Sofieva, V.: SPARC/IO3C/GAW report on Long-term Ozone Trends and Uncertainties in the Stratosphere, SPARC/IO3C/GAW, SPARC Report No. 9, WCRP-17/2018, GAW Report No. 241, doi:10.17874/f899e57a20b, 2019.
- 730 Polvani, L. M., Sobel, A. H., and Waugh, D. W.: The Stratosphere: Dynamics, Transport, and Chemistry, Washington DC American Geophysical Union Geophysical Monograph Series, 190, doi:10.1029/GM190, 2010.
- Polvani, L. M., Abalos, M., Garcia, R., Kinnison, D., and Randel, W. J.: Significant Weakening of Brewer-Dobson Circulation Trends Over the 21st Century as a Consequence of the Montreal Protocol, *Geophys. Res. Lett.*, 45, 401–409, doi:10.1002/2017GL075345, 2018.
- 735 Randel, W. J., Wu, F., Swinbank, R., Nash, J., and O'Neill, A.: Global QBO Circulation Derived from UKMO Stratospheric Analyses., *Journal of Atmospheric Sciences*, 56, 457–474, doi:10.1175/1520-0469(1999)056<0457:GQCDFU>2.0.CO;2, 1999.
- Slaper, H., Velders, G. J. M., Daniel, J. S., de Groot, F. R., and van der Leun, J. C.: Estimates of ozone depletion and skin cancer incidence to examine the Vienna Convention achievements, *Nature*, 384, 256–258, doi:10.1038/384256a0, 1996.
- 740

Solomon, S., Ivy, D. J., Kinnison, D., Mills, M. J., Neely, R. R., and Schmidt, A.: Emergence of healing in the Antarctic ozone layer, *Science*, 353, 269–274, doi:10.1126/science.aae0061, 2016.

SPARC/WMO: SPARC Report on the Evaluation of Chemistry-Climate Models, SPARC, 2010.

Steinbrecht, W., Froidevaux, L., Fuller, R., Wang, R., Anderson, J., Roth, C., Bourassa, A., Degenstein, D.,

745 Damadeo, R., Zawodny, J., Frith, S., McPeters, R., Bhartia, P., Wild, J., Long, C., Davis, S., Rosenlof, K., Sofieva, V., Walker, K., Rahpoe, N., Rozanov, A., Weber, M., Laeng, A., von Clarmann, T., Stiller, G., Kramarova, N., Godin-Beekmann, S., Leblanc, T., Querel, R., Swart, D., Boyd, I., Hocke, K., Kämpfer, N., Maillard Barras, E., Moreira, L., Nedoluha, G., Vigouroux, C., Blumenstock, T., Schneider, M., Garcia, O., Jones, N., Mahieu, E., Smale, D., Kotkamp, M., Robinson, J., Petropavlovskikh, I., Harris, N., Hassler, B., Hubert, D., and Tummon, F.: An update on ozone profile trends for the period 2000 to 2016, *Atmos. Chem. Phys. Discuss.*, 2017, 1–24, doi:10.5194/acp-2017-391, <https://www.atmos-chem-phys-discuss.net/acp-2017-391/>, 2017.

Stone, K. A., Solomon, S., and Kinnison, D. E.: On the Identification of Ozone Recovery, *Geophys. Res. Lett.*, 45, 5158–5165, doi:10.1029/2018GL077955, 2018.

755 Strahan, S. E., Oman, L. D., Douglass, A. R., and Coy, L.: Modulation of Antarctic vortex composition by the quasi-biennial oscillation, *Geophys. Res. Lett.*, 42, 4216–4223, doi:10.1002/2015GL063759, 2015.

Tachibana, Y., Inoue, Y., Komatsu, K. K., Nakamura, T., Honda, M., Ogata, K., and Yamazaki, K.: Interhemispheric Synchronization Between the AO and the AAO, *Geophys. Res. Lett.*, 45, 13, doi:10.1029/2018GL081002, 2018.

760 Thomason, L., Ernest, N., Millan, L., Rieger, L., Bourassa, A., Vernier, J., Peter, T., Luo, B., and Arfeuille, F.: A global, space-based stratospheric aerosol climatology: 1979 to 2016, *Earth Syst. Sci. Data*, in preparation, doi:10.5067/GloSSAC-L3-V1.0, 2017.

Tiao, G. C., Xu, D., Pedrick, J. H., Zhu, X., and Reinsel, G. C.: Effects of autocorrelation and temporal sampling schemes on estimates of trend and spatial correlation, *Journal of Geophysical Research*, 95, 20 507–20 517, doi:10.1029/JD095iD12p20507, 1990.

765 Tohir, A. M., Portafaix, T., Sivakumar, V., Bencherif, H., Pazmiño, A., and Bègue, N.: Variability and trend in ozone over the southern tropics and subtropics, *Annales Geophysicae*, 36, 381–404, doi:10.5194/angeo-36-381-2018, 2018.

Tummon, F., Hassler, B., Harris, N. R. P., Staehelin, J., Steinbrecht, W., Anderson, J., Bodeker, G. E., Bourassa, A., Davis, S. M., Degenstein, D., Frith, S. M., Froidevaux, L., Kyrölä, E., Laine, M., Long, C., Penckwitt, A. A., Sioris, C. E., Rosenlof, K. H., Roth, C., Wang, H.-J., and Wild, J.: Intercomparison of vertically resolved merged satellite ozone data sets: interannual variability and long-term trends, *Atmospheric Chemistry & Physics*, 15, 3021–3043, doi:10.5194/acp-15-3021-2015, 2015.

770 Tweedy, O. V., Kramarova, N. A., Strahan, S. E., Newman, P. A., Coy, L., Randel, W. J., Park, M., Waugh, D. W., and Frith, S. M.: Response of trace gases to the disrupted 2015–2016 quasi-biennial oscillation, *Atmospheric Chemistry & Physics*, 17, 6813–6823, doi:10.5194/acp-17-6813-2017, 2017.

Wargan, K., Orbe, C., Pawson, S., Ziemke, J. R., Oman, L. D., Olsen, M. A., Coy, L., and Emma Knowland, K.: Recent Decline in Extratropical Lower Stratospheric Ozone Attributed to Circulation Changes, *Geophys. Res. Lett.*, 45, 5166–5176, doi:10.1029/2018GL077406, 2018.

- 780 Weber, M., Coldewey-Egbers, M., Fioletov, V. E., Frith, S. M., Wild, J. D., Burrows, J. P., Long, C. S., and  
Loyola, D.: Total ozone trends from 1979 to 2016 derived from five merged observational datasets - the  
emergence into ozone recovery, *Atmospheric Chemistry & Physics*, 18, 2097–2117, doi:10.5194/acp-18-  
2097-2018, 2018.
- WMO: Scientific Assessment of Ozone Depletion: 2014 Global Ozone Research and Monitoring Project Re-  
785 port, World Meteorological Organization, p. 416, geneva, Switzerland, 2014.
- WMO: Scientific Assessment of Ozone Depletion: 2018, Global Ozone Research and Monitoring  
Project–Report, World Meteorological Organization, p. 588, geneva, Switzerland, 2018.
- Zerefos, C., Kapsomenakis, J., Eleftheratos, K., Tourpali, K., Petropavlovskikh, I., Hubert, D., Godin-  
Beekmann, S., Steinbrecht, W., Frith, S., Sofieva, V., and Hassler, B.: Representativeness of single lidar  
790 stations for zonally averaged ozone profiles, their trends and attribution to proxies, *Atmospheric Chemistry  
& Physics*, 18, 6427–6440, doi:10.5194/acp-18-6427-2018, 2018.
- Zerefos, C. S., Bais, A. F., Ziomas, I. C., and Bojkov, R. D.: On the relative importance of quasi-biennial  
oscillation and El Nino/Southern Oscillation in the revised Dobson total ozone records, *J. Geophys. Res.*, 97,  
10, doi:10.1029/92JD00508, 1992.
- 795 Ziemke, J. R., Oman, L. D., Strode, S. A., Douglass, A. R., Olsen, M. A., McPeters, R. D., Bhartia, P. K., Froide-  
vaux, L., Labow, G. J., Witte, J. C., Thompson, A. M., Haffner, D. P., Kramarova, N. A., Frith, S. M., Huang,  
L.-K., Jaross, G. R., Seftor, C. J., Deland, M. T., and Taylor, S. L.: Trends in Global Tropospheric Ozone  
Inferred from a Composite Record of TOMS/OMI/MLS/OMPS Satellite Measurements and the MERRA-2  
GMI Simulation, *Atmospheric Chemistry and Physics Discussions*, 2018, 1–29, doi:10.5194/acp-2018-716,  
800 <https://www.atmos-chem-phys-discuss.net/acp-2018-716/>, 2018.

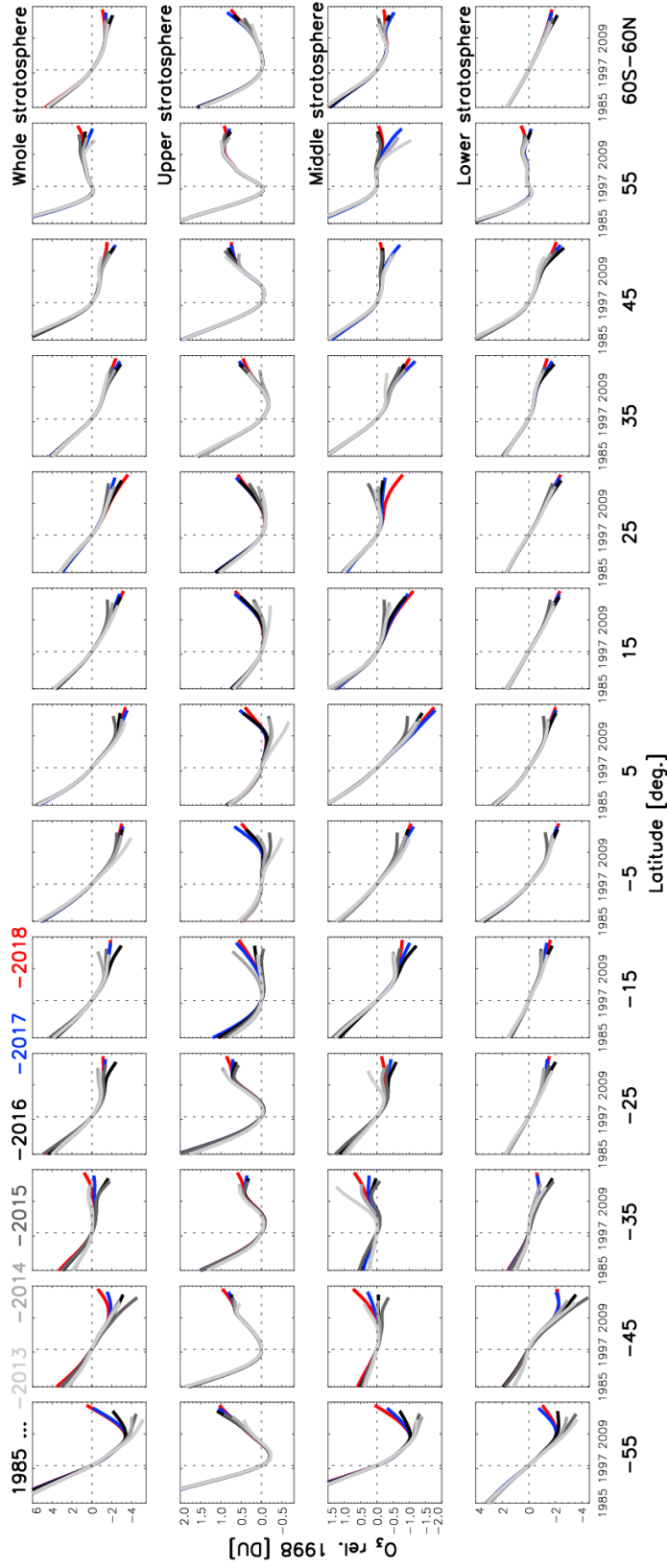


Figure 5: The partial column ozone non-linear trends estimated as a function of end year (2013 to 2018; dark to light colours), for each 10° latitude and quasi-global (left to right) and (top to bottom) the whole, upper, middle and lower stratosphere. Each sub-panel covers 1985–2018 and all curves are bias corrected to January 1998 (horizontal and vertical dotted lines). Uncertainties for each 1998–end-year change are given in Fig. 6.



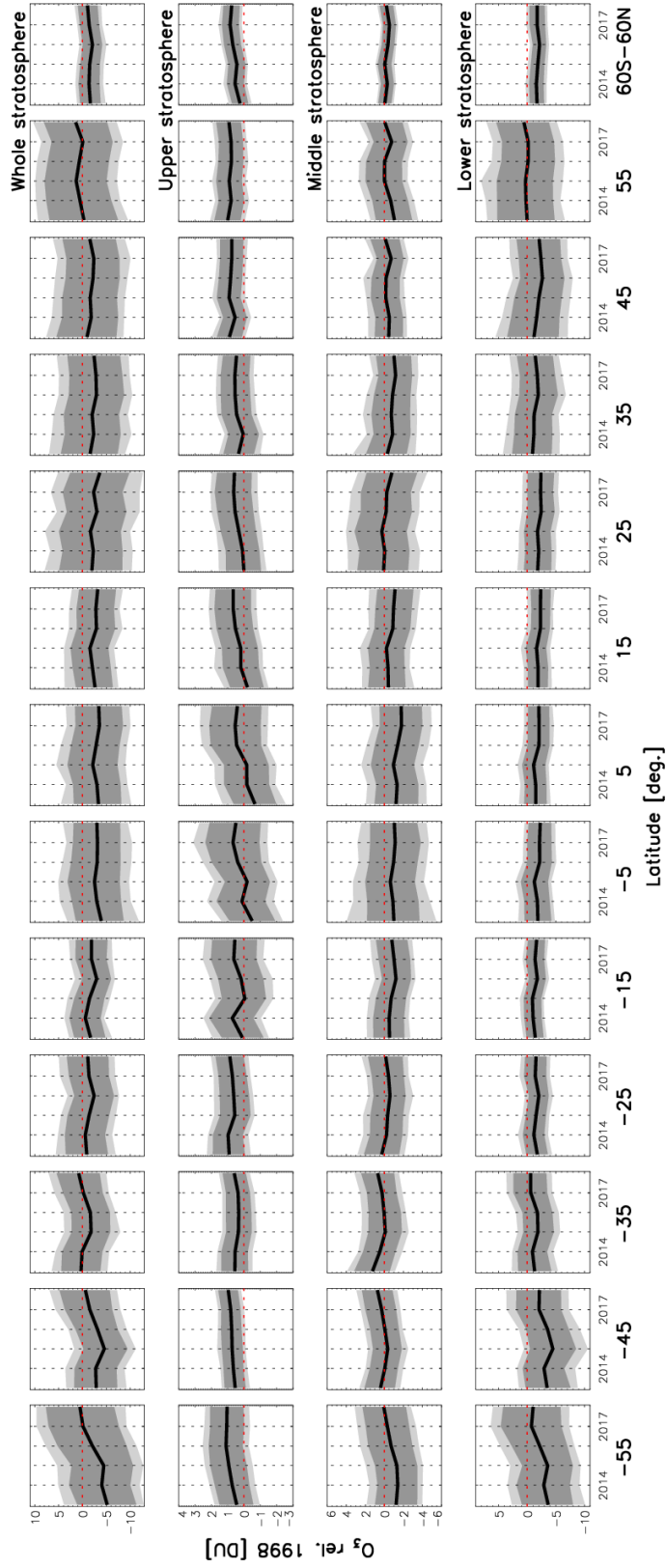


Figure 6: The partial column ozone changes between 1998 and the end-year from 2013 to 2018 (x-axis of each sub-panel) from the non-linear trends as in Fig. 5. Dark and light shading represent 95% and 99% credible intervals.

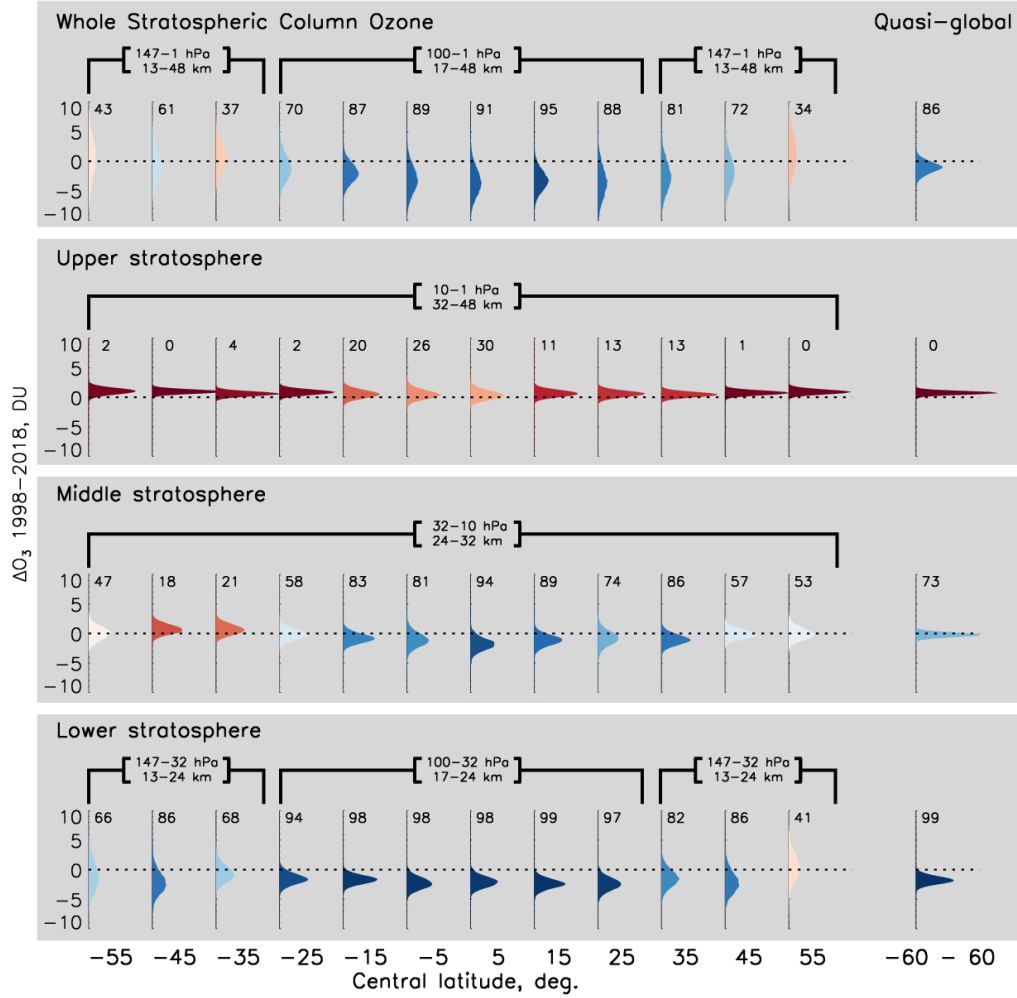


Figure 7: Posterior distributions (shaded) for the 1998–2018 partial column ozone changes. (Top) whole stratospheric column, (middle) upper and (bottom) lower stratosphere in 10° bands for all latitudes (left) and integrated from 60°S–60°N (‘Quasi-global’, right). The stratosphere extends deeper at mid-latitudes than equatorial (marked above each latitude). Numbers above each distribution represents the distribution-percentage that is negative; colours are graded relative to the percentage-distribution (positive, red-hues, with values <50; negative, blue).

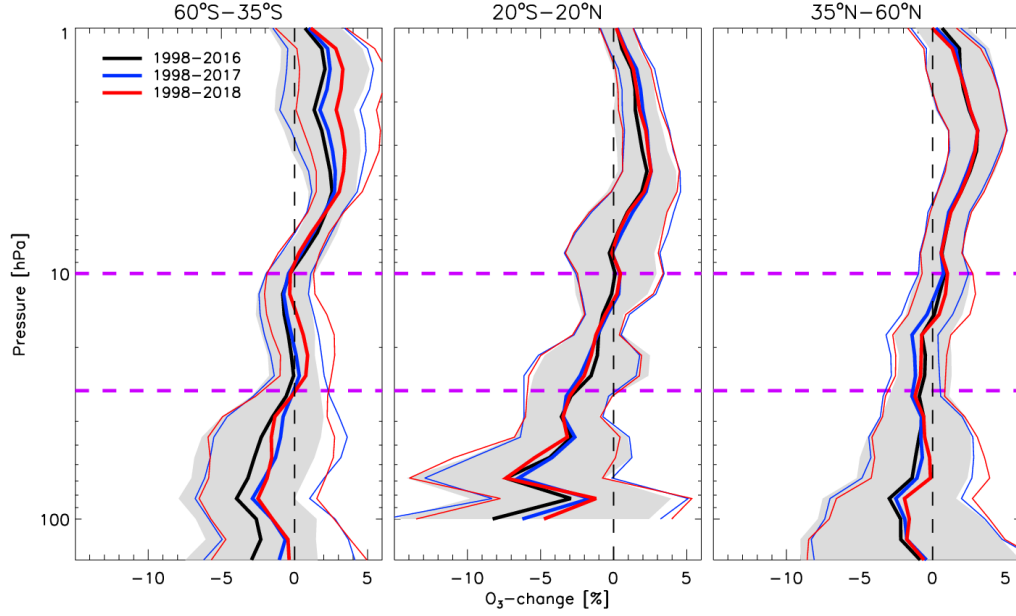


Figure 8: The ozone profiles for 1998 to an end-year of 2016 through 2018 (see legend) in the southern hemisphere (60°–35°S), the tropics (20°S–20°N), and the northern hemisphere (35°–60°N). Shading is for 2016 only. Uncertainties are 95% credible intervals. Pink lines indicate boundaries of partial columns in other figures.

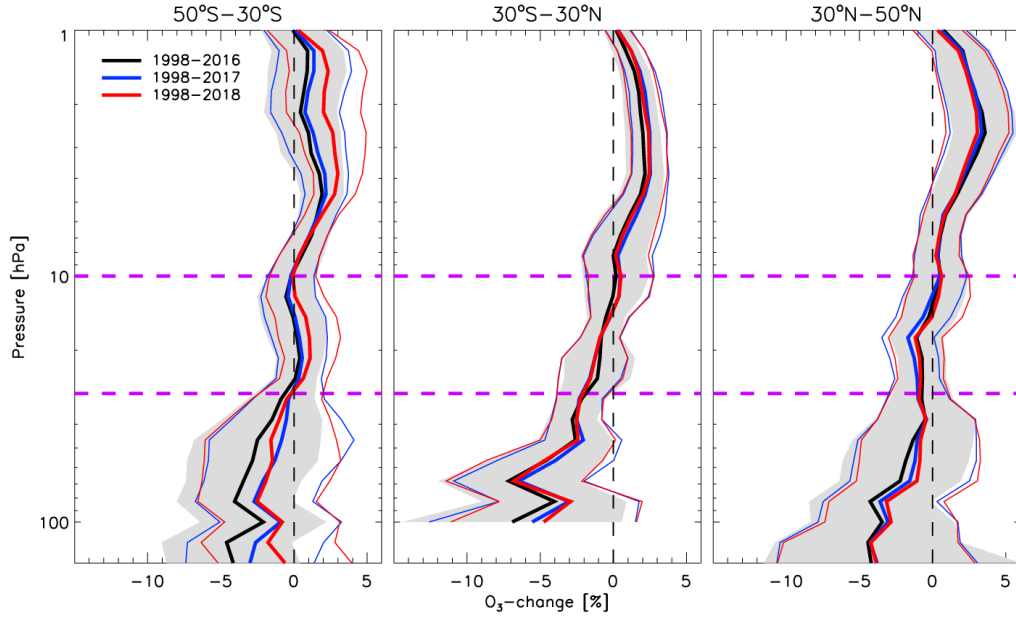


Figure 9: As for Fig. 8, but for the 50°S–30°S (SH), 30°S–30°N (tropics), 30°N–50°N (NH).

**Response to editor comments on second revision of “Stratospheric ozone trends for 1985–2018: sensitivity to recent large variability”  
by W. T. Ball et al**

**Response**

We thank the editor for his response and suggestions. We respond to specific points as follows (black are the editor’s comment, blue our response).

Thank you for addressing the reviewers' comments. I have one non-specific recommendation and one specific one. The non-specific one is that there are a lot of very long sentences, with colons or semi-colons in the middle. They could usefully be changed to full stops to make the text more accessible to the reader.

We were happy to make the paper clearer and have removed 32 instances of the semi-colon, as mentioned. We kept three cases where its use remained appropriate or clarity couldn’t easily be improved (near lines 207, 318 and 336).

The specific one is that you should have another look at the para on future projections on line 542 to see if the various points you make can be made more clearly / distinctly. The addition of text has made the logic less clear.

We have heavily modified the first half of this paragraph and believe it is clearer now.

I would also add a sentence around lines 57-60 re-raising the issue of longitudinal variations in trends as being worthy of future work - that can be shorter than what you have put in the intro.

We considered the editor’s suggestion, and while we agree it is worth pointing out this future avenue of research to open, we instead included it as follows. We modified the text to split the paragraph where the discussion moves from the negative ozone trends over to previous investigations of wave activity driving the changes longitudinally (break at line 62), and have inserted the suggested sentence at line 67.

# **Stratospheric ozone trends for 1985–2018: sensitivity to recent large variability**

William T. Ball<sup>1,2</sup>, Justin Alsing<sup>3,4</sup>, Johannes Staehelin<sup>1</sup>, Sean M. Davis<sup>5</sup>,  
Lucien Froidevaux<sup>6</sup>, and Thomas Peter<sup>1</sup>

<sup>1</sup>Institute for Atmospheric and Climate Science, Swiss Federal Institute of Technology Zurich,  
Universitaetstrasse 16, CHN, CH-8092 Zurich, Switzerland

<sup>2</sup>Physikalisch-Meteorologisches Observatorium Davos World Radiation Centre, Dorfstrasse 33,  
7260 Davos Dorf, Switzerland

<sup>3</sup>Oskar Klein Centre for Cosmoparticle Physics, Stockholm University, Stockholm SE-106 91,  
Sweden

<sup>4</sup>Physics Department, Blackett Laboratory, Imperial College London, SW7 2AZ, UK

<sup>5</sup>NOAA Earth System Research Laboratory Chemical Sciences Division, Boulder, CO, USA

<sup>6</sup>Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA

*Correspondence to:* W. T. Ball (william.ball@env.ethz.ch)

**Abstract.** The Montreal Protocol, and its subsequent amendments, has successfully prevented catastrophic losses of stratospheric ozone, and signs of recovery are now evident. Nevertheless, recent work has suggested that ozone in the lower stratosphere (<24 km) continued to decline over 1998–2016, offsetting recovery at higher altitudes and preventing a statistically significant increase in quasi-global (60°S – 60°N) total column ozone. In 2017, a large lower stratospheric ozone resurgence over less than 12 months was estimated (using a chemistry-transport model; CTM) to have offset the long-term decline in the quasi-global integrated lower stratospheric ozone column. Here, we extend the analysis of space-based ozone observations to December 2018 using the BASIC<sub>SG</sub> ozone composite. We find that the observed 2017 resurgence was only around half that modelled by the CTM, was of comparable magnitude to other strong inter-annual changes in the past, and restricted to southern hemisphere mid-latitudes (SH; 60°S–30°S). In the SH mid-latitude lower stratosphere, the data suggest that by the end of 2018 ozone is still likely lower than in 1998 (probability ~80%). In contrast, tropical and northern hemisphere (NH) ozone continue to display ongoing decreases, exceeding 90% probability. Robust tropical (>95%, 30°S–30°N) decreases dominate the quasi-global integrated decrease (99% probability); the integrated tropical stratospheric column (1–100 hPa, 30°S–30°N) displays a significant overall ozone decrease, with 95% probability. These decreases do not reveal an inefficacy of the Montreal Protocol. Rather, they suggest other effects to be at work, mainly dynamical variability on long or short timescales, counteracting the positive effects of the Montreal Protocol on stratospheric ozone recovery. We demonstrate that large inter-annual mid-latitude (30°–60°) variations, such as the 2017 resurgence, are driven by non-linear quasi-biennial oscillation (QBO) phase-dependent seasonal variability. However, this variability is not represented in current regression analyses. To understand if observed lower stratospheric ozone decreases are a transient or long-term phenomenon, progress needs to be made in accounting for this dynamically-driven variability.

## 1 Introduction

Ozone in the stratosphere acts as a protective shield against ultraviolet radiation that may harm the biosphere, and leads to cataracts, skin damage, and skin cancer in humans (Slaper et al., 1996; WMO, 2014, 2018). In the latter half of the 20<sup>th</sup> century, the emission of long-lived halogen-containing ozone depleting substances (hODSs) led to ~5% loss in quasi-global (60°S–60°N) integrated total column ozone (WMO, 2014), which represents the combined changes in tropospheric and stratospheric ozone contributions. The 1987 Montreal Protocol and its amendments and adjustments led to a reduction in hODSs that resulted in a halt in total column ozone losses around 1998–2000 (Harris et al., 2015; Chipperfield et al., 2017).

However, there is still no evidence of a statistically significant increase in total column ozone since 1998 (Chipperfield et al., 2017; Weber et al., 2018; Ball et al., 2018), despite a significant

increase in upper stratospheric ozone (1–10 hPa) (Ball et al., 2017; Steinbrecht et al., 2017; Ball et al., 2018; Petropavlovskikh et al., 2019). Ball et al. (2018) and Ziemke et al. (2018) presented evidence, using OMI/MLS tropospheric column observations for 2005–2016, that tropospheric ozone had also increased significantly. However, large uncertainties remain in quasi-global tropospheric ozone trends, and the recent Tropospheric Ozone Assessment Report (TOAR) shows that different tropospheric ozone products give a wide range of trends, some even indicating negative changes (Gaudel et al., 2018). The importance of considering tropospheric and stratospheric changes separately to understand changes in total column ozone has also been highlighted in recent studies using chemistry climate models (CCMs) (Meul et al., 2016; Keeble et al., 2017; Dhomse et al., 2018). If tropospheric and upper stratospheric ozone have indeed both increased, then the observed flat trend in total column ozone implies that middle and lower stratospheric ozone should have decreased.

To assess trends in stratospheric ozone, composites of observations must be formed by merging multiple ozone observational timeseries into a long, multi-decadal record from which variability can be attributed, and long-term trends determined. Composites are subject to artefacts from merging different observing platforms. Multiple papers (Tummon et al., 2015; Harris et al., 2015; Steinbrecht et al., 2017; Ball et al., 2017, 2018) and a SPARC report (Petropavlovskikh et al., 2019) review, discuss, and attempt to account for the artefacts in the uncertainty budget.

Ball et al. (2018) integrated ozone over the whole stratosphere, i.e. the ozone layer, quasi-globally for pressure levels from 147–1 hPa (~13–48 km) at mid-latitudes (30°–60°), and 100–1 hPa (~16–48 km) between the sub-tropics (30°S–30°N), and found ozone to be lower in 2016 than in 1998 in multiple ozone composites. In their analysis, the lower stratosphere (147/100–32 hPa, ~13/17–24 km) was driving this decrease. The most significant decreases were in the tropics, but negative trends extended out into the mid-latitudes (Fig. 1d). Other studies have subsequently confirmed these negative trends (Zerefos et al., 2018; Wargan et al., 2018; Chipperfield et al., 2018). Evidence points towards dynamical variations driving changes (Chipperfield et al., 2018), perhaps in the form of enhanced isentropic mixing (Wargan et al., 2018).

Part of the negative trends in northern hemispheric stratospheric ozone in the 1980s and 1990s at higher latitudes have been previously attributed to synoptic and planetary waves (Hood and Zaff, 1995; Hood et al., 1999) inducing large localised (e.g. over Europe) wintertime decreases in ozone. These changes in wave activity might be driven by sea surface temperature and eddy flux changes on decadal or longer timescales, although most of these studies are limited to the end of the last century when ODSs remained an established primary driver of the decrease. Since recent studies almost exclusively consider zonal mean ozone fields, this motivates re-investigation of longitudinal ozone changes (in future work). The El Niño Southern Oscillation (ENSO) and Quasi-Biennial Oscillation (QBO) are known to influence the dynamical variability in the lower stratosphere and may be a main player in driving inter-annual and decadal variability in this region (Diallo et al., 2018, 2019). Nevertheless, these dynamical changes do not in themselves determine a specific underlying driving force,

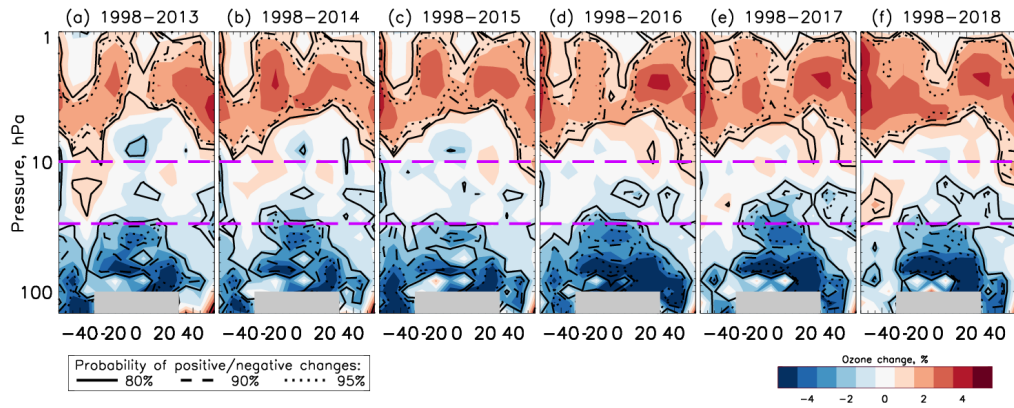


Figure 1: Zonally averaged ozone changes between 1998 and end years (a) 2013 to (f) 2018. Red represents increases, blue decreases (%; right legend). Contours represent probability levels of positive or negative changes (left legend). Grey-shaded regions represent unavailable data. Pink dashed-lines delimit regions integrated to partial ozone columns in other figures.

however the effect of increasing anthropogenic greenhouse gases (GHGs) (Hood and Soukharev, 2005; Peters and Entzian, 1999) on specific mechanisms needs further study (Ball et al., 2018). On the other hand, Stone et al. (2018) showed that negative ozone trends could be simulated in the lower stratosphere over the same period in two of nine ensemble members of a coupled CCM as a result of natural variability interfering in the (linear) trend analysis, although none of the ensembles displayed the same widespread negative trends as detected in observations (Ball et al., 2018). They suggested that an additional seven years of observations would lead to negative signals disappearing in favour of positive trends. The implication, then, is that the observed negative trend over the relatively short 19 year timeframe may be a temporary result from large natural variability (in the single realisation) of the real-world, rather than a response to increasing GHGs.

Chipperfield et al. (2018) used a chemistry transport model (CTM) to reconstruct ozone variability close to past real-world behaviour; transport in the CTM is driven by ERA-Interim (Dee et al., 2011) reanalysis fields. The results showed changes similar to those presented by Ball et al. (2018) up to December 2016. They extended their CTM analysis by an additional 12 months to find that the 1998–2016 ozone decline in the lower stratosphere ( $\sim 2$  DU; Ball et al. (2018)) was offset by a sudden increase of ozone in 2017, exceeding 8 DU quasi-globally. This was attributed almost entirely to dynamical changes and was primarily located in the southern hemisphere (SH). Froidevaux et al. (2019) have also noted that ozone trends derived from Aura/MLS data over a shorter period (2005–2018) have a tendency towards slightly positive values in the SH, but not so elsewhere within the extra-polar regions. Chipperfield et al. (2018) suggested that the lower stratospheric ozone decrease was a result of large natural variability that biased the trend analysis, and that the variability could be attributed to dynamics and not to chemical or photolytic changes, although the source of dynamical



perturbations was not identified or the impact on trends quantified. Thus, an assessment of this recent variability on trends, and an update to 2018 is needed and is a key aim of this study.

Here, we update the observational analysis of Ball et al. (2018) to include data to the end of 2018 (Section 3.1). This allows us to assess the impact of the 2017 ozone increase in the lower stratosphere on the trend analysis, and to consider additional changes over 2018. We show that large ozone-increase events, with a duration and magnitude similar to that of 2017 (Chipperfield et al., 2018) have occurred regularly since 1985 at mid-latitudes (Section 3.2), and find the events are linked to a seasonally-dependent QBO effect (Section 3.3). We update partial column ozone trends from 2016 to 2018 in section 3.4. Finally, we consider the sensitivity of trends to the recent increase of stratospheric ozone (Sections 3.5-3.6) by considering six periods that start in 1985 and end between 2013 and 2018, in order to demonstrate where signals are robust to the end date, and where not. Such an analysis is essential to establish if the negative trends are a result of natural variability interfering in the trend analysis, and to take the first steps to account for what might be driving the large, short-term variability.

## 2 Data and methods

### 2.1 Ozone Data

Although other ozone composites exist (Petrovskikh et al., 2019), we focus exclusively on data formed by merging ozone from SWOOSH (Davis et al., 2016) and GOZCARDS (Froidevaux et al. (2015); here we use the v2.20 update of Froidevaux et al. (2019)) using the so-called BASIC (BAYesian Integrated and Consolidated) approach (Ball et al., 2017) to account for artefacts in merged composites and improve trend estimates. These data were referred to as ‘Merged-SWOOSH/GOZCARDS’ by Ball et al. (2018), but we refer here to it as BASIC<sub>SG</sub>. To briefly place the SWOOSH and GOZCARDS datasets in the context of BASIC<sub>SG</sub>, Figure S1 of Ball et al. (2018) presented 1998–2016 changes using SWOOSH or GOZCARDS alone; this figure reveals that these ozone composites show generally similar changes on large spatial scales, though there are clear differences on small scales, e.g. in the tropical upper stratosphere, and in the SH lower stratosphere. Figure S2 of Ball et al. (2018) importantly demonstrates at 100 hPa in the tropical lower stratosphere that there are significant differences between SWOOSH and GOZCARDS in the late 1990s; this figure also shows that BASIC<sub>SG</sub> is able to account for the differences in a principled way that is not simply the averaging of the two products, which is particularly important for having confidence in an assessment of ozone in the lower stratosphere. We extend BASIC<sub>SG</sub> from Ball et al. (2018) by two years to cover 1985–2018. This period is essentially an extension of the Aura Microwave Limb Sounder (Aura/MLS); both SWOOSH and GOZCARDS consider Aura/MLS exclusively after 2009.

We only consider BASIC<sub>SG</sub> here for the following reasons. First, as discussed in Ball et al. (2018), compared to the other composites it had the least apparent artefacts within the timeseries. The

130 Stratosphere-troposphere Processes And their Role in Climate (SPARC) Long-term Ozone Trends  
and Uncertainties in the Stratosphere (LOTUS) report (Petropavlovskikh et al., 2019) indicates this  
method to be more robust to outliers than other composites. Second, BASIC<sub>SG</sub> is resolved in the  
lower stratosphere, which is not the case for all composites. For further discussion see Ball et al.  
(2018) and the SPARC LOTUS report (Petropavlovskikh et al., 2019). Additionally, SWOOSH  
135 and GOZCARDS are currently two of the most up-to-date composites available. Finally, we are  
interested here in the sensitivity of stratospheric ozone changes to different end years and, since  
Aura/MLS is arguably one of the best remote sensing platforms for ozone currently in operation  
(Petropavlovskikh et al., 2019), focusing only on BASIC<sub>SG</sub> provides an analysis, discussion, and  
interpretation that is free from the complications of considering multiple composites that have mul-  
140 tivariate reasons for displaying different behaviour.

## 2.2 Regression analysis

As in Ball et al. (2018), we perform all timeseries analysis using dynamical linear modelling (DLM)  
(Laine et al., 2014) using the public DLM code `dlmmc` (available at <https://github.com/justinalsing/dlmmc>). We provide a short overview of DLM here.

145 Our DLM approach models the ozone timeseries as a (dynamical) linear combination of the fol-  
lowing components. There are two seasonal components (with 6- and 12-month periods respec-  
tively), a set of regressor variables (i.e., proxy timeseries describing various known drivers), an  
auto-regressive (AR) process, and a smooth non-linear (non-parametric) background trend. DLM  
differs from traditional multiple linear regression (MLR) approaches in a number of key ways.

150 Firstly, while MLR fits for a fixed (constant-in-time) linear combination of seasonal, regressor, and  
trend components, DLM can allow the amplitudes of the various components to vary dynamically  
in time, capturing richer phenomenology in the data. Here, we allow the amplitude and phase of the  
seasonal components to be dynamic, but keep the regressor amplitudes constant in time. We do this  
because the seasonal cycle in the observational composites can change over time, either as a physical  
155 feedback of changing temperature and ozone or due to different observations exhibiting different sea-  
sonal amplitudes (not shown) that are a result of the observing instruments ‘seeing’ slightly different  
parts of the atmosphere or having different sampling. Due to the seasonal cycle having the largest  
variability of all modes we expect that, if left unaccounted for, the time varying seasonal modulation  
might have an influence on the regression. In principle other regressor amplitudes could also have  
160 some time modulation for similar reasons. However, we leave an investigation of more flexible DLM  
models with dynamic regressor amplitudes to future work where a physically-motivated justification  
for such freedom can be investigated.

Secondly, MLR that does not assume a driver for the long-term trends, e.g. for the influence of  
ODSs or GHGs, typically assumes a fixed prescription for the shape of the background trend, e.g.  
165 a piecewise-linear or independent-linear trend with some fixed, pre-chosen inflection-date. These

assumptions are both restrictive and give a poor representation of the smooth background trends we expect from nature (Laine et al., 2014; Ball et al., 2017). DLM addresses this by instead modelling the trend as a smooth, non-parametric, non-linear curve, where the ‘smoothness’ of the trend is controlled by a free parameter that is included in the fit (see supplementary materials Fig. S1).

170 Thirdly, in practice MLR is often performed by first subtracting an estimated mean seasonal cycle, fitting the trend and regressor variables to the anomalies, and then making a post-hoc correction for AR residuals, although many do fit annual and semi-annual components. This procedure typically does not propagate the errors on the seasonal cycle and AR parameters in a rigorous way, leading to misrepresentation of uncertainties. DLM addresses this by inferring all components of the model  
175 simultaneously, and formally marginalizing over the uncertainties in all other parameters when reporting uncertainties on, e.g., the trend. We use the same prior assumptions as described in Ball et al. (2018).

Probabilities of an overall increase (decrease) in ozone between two dates (Figs. 1, 7, and Table 1) are computed as the fraction of Monte Carlo Markov Chain (MCMC) samples that show positive  
180 (negative) change. Credible intervals (Figs. 6, 8, 9) are computed as the central 95 and 99 percentiles of the MCMC samples. The use of ‘confidence’ or ‘significance’ is used in this paper interchangeably with ‘probability’ and refers specifically to Bayesian probabilities; it does not refer to the application of frequentist significance tests and/or confidence intervals.

We use the same regressors as Ball et al. (2018): solar (30 cm radio flux, F30) (Dudok de Wit  
185 et al., 2014)), volcanic (latitudinally resolved stratospheric aerosol optical depth, SAOD) (Thomason et al., 2017), ENSO (NCAR, 2013), and the Quasi-Biennial Oscillation, QBO, at 30 and 50 hPa <sup>1</sup>. In previous analyses, we considered the Arctic and Antarctic Oscillation, AO/AAO <sup>2</sup>, as proxies for northern hemisphere (NH) and SH surface pressure variability only for partial column ozone analysis  
190 in their respective hemisphere; here we also consider them for the spatially-resolved analysis and in all cases use both AO and AAO simultaneously. The AO and AAO have little affect outside their respective regions, but we do not limit the possibility they may influence some variability in either hemisphere (Tachibana et al., 2018). We use a first order AR (AR1) process (Tiao et al., 1990) to consider auto-correlation in the residuals. We remove a three year period following the Pinatubo eruption, i.e. June 1991 to May 1994, which is a year longer than the previous analysis, to avoid  
195 any effects of the eruptions that may have persisted. Another key point regarding the SAOD proxy is that, unlike the other proxies that have been fully updated to the end of 2018 for this analysis, the SAOD is currently not extended beyond 2016, so we repeat the year 2016 for 2017 and 2018. If any deviations in the SAOD occurred during the 2017–2018 period our analysis will not account for this. Nevertheless, as can be seen in Fig. 1d here, in comparison to Fig. 1b of Ball et al. (2018), all of

<sup>1</sup>QBO indices: <http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/>

<sup>2</sup>AO/AAO indices: <http://www.cpc.ncep.noaa.gov/products/precip/CWlink/>

200 these adjustments to the procedure from Ball et al. (2018) have little impact on the estimated mean changes in ozone.

### 3 Results

#### 3.1 Stratospheric ozone changes since 1998

Figure 1d shows the pressure-latitude, spatially-resolved 1998–2016 ozone change, reproducing  
205 Fig. 1b of Ball et al. (2018). Minor differences exist because the BASIC<sub>SG</sub> composite and DLM procedure have been updated. Ozone in the lower stratosphere (delimited by the pink dashed line at 32 hPa, or 24 km) shows a marked and almost hemisphere-symmetric decrease, while upper stratospheric changes (>10 hPa, 32 km) are mainly positive. The middle stratosphere generally shows relatively flat ozone trends since 1998 with low probability of an overall change.

210 Figures 1e and f show the 1998–2017 and –2018 ozone changes, respectively. Four points of interest emerge from a comparison to 1998–2016: (i) while still negative, the magnitude of the lower stratospheric SH (60°S–30°S) ozone decrease has become smaller and less significant; (ii) tropical (30°S–30°N) and NH (30°N–50°N) changes remain negative and highly probable; (iii) the probability (and magnitude) of negative ozone trends over tropical and NH regions in the middle stratosphere  
215 (32–10 hPa) has increased; and (iv) the magnitude and probability of upper stratospheric ozone increase has strengthened. Importantly, Fig. 1 demonstrates the robustness of negative ozone trends in the lower, and positive trends in the upper, stratosphere irrespective of the final year of the analysis. Figures 1a–f present ozone changes from 1998 to end years 2013 through 2018, showing the sensitivity of ozone trends to six consecutive end years. These end years give insight into the sensitivity  
220 of the trends to large inter-annual variability. In particular, these six years encompass periods of both negative/Easterly and positive/Westerly phases of ENSO/QBO. These modes are major contributors to stratospheric variability (Zerefos et al., 1992; Tweedy et al., 2017; Toihr et al., 2018; Garfinkel et al., 2018; Diallo et al., 2018, 2019), and any sensitivity of the end year to the state of these drivers should be encapsulated in the set of spatial responses depending on the end year only (Fig. 1), particularly if these modes were not well-captured by DLM predictors. A lower stratosphere negative  
225 ozone trend is persistent for all end years. For 1998–2013, there is a highly probable negative trend in ozone in the SH lower stratosphere; the probability is retained until 2016, after which it reduces. The opposite is seen in the NH, where only a small region of probable ozone decrease exists for 1998–2013, and this strengthens with each panel until 2016, after which a highly probable decrease  
230 of ozone remains stable. There is no apparent switch from negative to positive ozone changes in these regions for any of the six end years.

The reduced probability of a SH decrease is related, as we will see in Section 3.2, to the rapid 2017 increase in SH mid-latitude lower stratospheric ozone reported by Chipperfield et al. (2018) using a CTM. However, Fig. 1 also confirms in observations that this is localised to south of 30°S and

235 does not reveal coherent or consistent behaviour over time with the NH, suggesting that there may be large, hemispherically independent variability interfering with the trend estimates. Nevertheless, there are no signs as yet of an ozone increase underway in the quasi-global lower stratosphere.

Further, the decrease in ozone in the tropical lower stratosphere increases in magnitude and significance as more data are added. The tropical lower stratospheric ozone is projected to decrease by the  
240 end of the century in all CCMs (Dhomse et al., 2018), due to enhanced upwelling from the Brewer Dobson circulation (BDC) as a result of changes to stratospheric dynamics from increasing GHGs (Polvani et al., 2018). It is possible that this is a detection in observations of the expected tropical lower stratosphere decline in ozone, earlier than expected (WMO, 2014). However, whilst the data show a significant decline, it remains to be seen if this can be attributed to the anthropogenic GHG  
245 induced upwelling of the BDC.

### 3.2 On the rapid increase of ozone in 2017

Chipperfield et al. (2018) reported a rapid increase in the quasi-global lower stratospheric ozone in 2017, modelled using a CTM driven by ERA-Interim reanalysis to represent dynamical variability closer to that which occurred historically. The quasi-global, deseasonalised timeseries from  
250 BASIC<sub>SG</sub> is shown in Fig. 2a. The year 2017 is bounded by the vertical dashed lines and the large increase is highlighted in red from a minimum in November 2016 to a maximum reached 11 months later in October 2017.

The observed 2016–2017 increase in Fig. 2a was 5.5 DU, which is 63% of the 8.7 DU increase reported by Chipperfield et al. (2018). Split into three latitude bands, 60°–30°S, 30°S–30°N, and  
255 30°–60°N (Figs. 2b–d), we find that the rapid increase can be decomposed into a 12 DU increase in the SH, 3 DU in the tropics, and 6 DU in the NH. Weighting for latitude – 21, 58, and 21% respectively – the SH contribution accounts for nearly half of the quasi-global increase (2.5 DU, 1.9 DU, 1.3 DU). The overall increase is composed of two sub-periods, dominated by a NH increase until May 2017, and a SH increase over April–August 2017. The tropical region saw comparatively  
260 little change in the second period. We do not know why the CTM and observations disagree in the magnitude of change for this period.

Importantly, the rapid increase seen in 2017 is not unique. Four other quasi-global ‘events’ of this type are found over 1985–2018, shown in Fig. 2a. The identification criterion for these events was an increase of at least 90% of the 2017 increase occurring within a 13 month period. The decomposed  
265 timeseries (Fig. 2b–d) show that the large increases in the SH are *normal*, occurring regularly. They also occur in the NH, but not as regularly, and the tropical variability is much smaller than the mid-latitude variance. In addition to the large increases, there are also comparatively large negative swings in both SH and NH timeseries – one in the NH beginning in 2002 exceeds 24 DU. In the following section we argue that these large, rapid changes are driven by a non-linear seasonal-QBO  
270 effect.

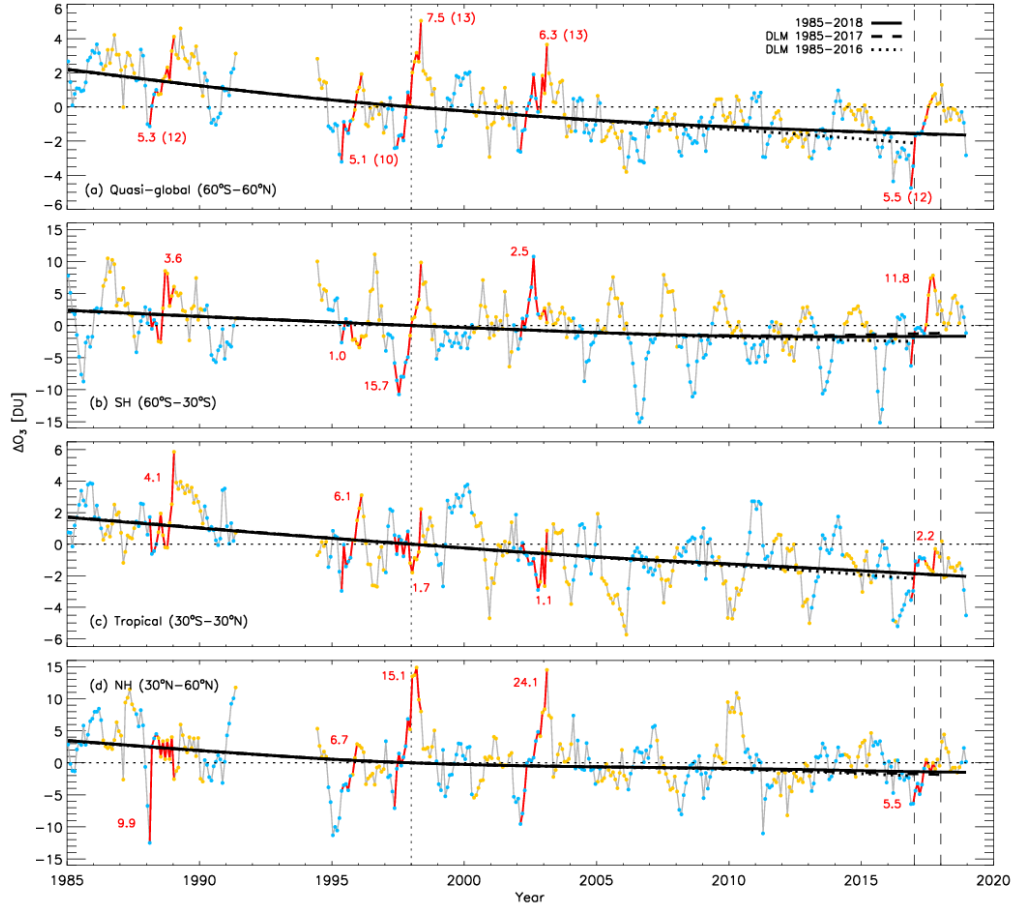


Figure 2: Lower stratospheric partial column ozone anomalies, (a) quasi-global ( $60^{\circ}\text{S}$ – $60^{\circ}\text{N}$ ), (b) southern hemisphere ( $60^{\circ}\text{S}$ – $30^{\circ}\text{S}$ , 147–30 hPa), (c) tropics ( $30^{\circ}\text{S}$ – $30^{\circ}\text{N}$ , 100–30 hPa), (d) northern hemisphere ( $30^{\circ}\text{N}$ – $60^{\circ}\text{N}$ , 147–30 hPa). The DLM non-linear trend is shown for 1985–2016 (dotted), 1985–2017 (dashed), and 1985–2018 (solid). Red lines represent contiguous periods identified in the quasi-global anomalies exceeding 90% of the magnitude of the November 2016 to October 2017 change within a 13 month period; the DU changes are written above or below each period; red periods in (b–d) are those identified in (a). Colour dots are plotted on each timeseries when the QBO at 30 hPa is either in an Easterly (yellow) or Westerly (blue) phase. The three-year period following the eruption of Mt. Pinatubo, June 1991 to May 1993, has been removed. Figures for the whole, upper and middle stratosphere are provided in the Supplementary Materials Figs. S2–4.

### 3.3 Contribution of QBO to mid-latitude ozone variability

Chipperfield et al. (2018) convincingly showed that the majority of post-1997 quasi-global lower stratospheric ozone variability in Fig. 2a was dynamically controlled, specifically in the lower stratosphere where the lifetime of ozone is long. Given that the contributions from each sub-region

(Fig. 2b–d) add up to the quasi-global change in 2017, it is reasonable to assume that dynamics controls much of the sub-decadal variability there too. The peaks (or troughs) in the SH are 2–3 years apart. The QBO has a similar periodicity and is known to have the largest inter-annual dynamical impact on ozone in the stratosphere (see Gray and Pyle (1989), Zerefos et al. (1992), and Toth et al. (2018), and references therein). Labelling each month in Fig. 2 with the 30 hPa QBO-Easterly or Westerly phase in yellow or blue dots, respectively, reveals that the large SH negative anomalies are almost always associated with a Westerly phase, while positive anomalies are associated with an Easterly phase; Bodeker et al. (2007) previously identified large SH negative anomalies in 1985, 1997 and 2006 and related these to the QBO-Westerly phase. This also appears to be the case in the NH, but the variability is less regular, unsurprisingly since the NH stratosphere is known to have additional variability as a consequence of greater sea-land contrast and more orography than in the SH. The NH thus exhibits stronger large scale wave activity and consequently polar vortex and stratospheric variability (see Butchart (2014) and Kidston et al. (2015) and references therein). Equatorial variability in ozone related to the QBO phase at 30 hPa shows the opposite behaviour to that at mid-latitudes: decreases in ozone generally appear to occur with the Easterly phase and vice versa, and the return from maximum excursion (i.e. the sign of the gradient) appears to be more related to the change in phase.

Histograms of the ozone anomalies relative to the DLM trend line for each QBO phase at 30 hPa are shown in the upper row of Fig. 3. The shift in the histogram between QBO phases is clear in the SH. The NH displays little shift, again likely related to other drivers influencing NH ozone changes, though the extremes show a similar phase separation as in the SH. The difference between the QBO Easterly and Westerly histograms are shown in the bottom row, and make clear the correlation between the QBO state and ozone anomalies.

To clarify this further, in Fig. 4 all 34 years in the 1985–2018 period are split into 13-month periods starting in January for the SH (upper row) and July for the NH (lower row), i.e. a few months prior to the onset of winter in the respective hemisphere. The latitudes plotted are refined to isolate clearer signals for (a) 50°–30°S, (c) 30°–50°N, and (b,d) 10°S–10°N. This refinement reduces the influence of polar variability on the 30°–50° band, and isolates the equatorial region to where the QBO variability is strongest. We note that the act of forming partial columns of ozone may reduce the integrated variability compared to counter-varying layers that would otherwise be resolved by pressure level. We find the use of the QBO phase at 15 hPa also better separates the events in this additional analysis. We find negative and positive ozone excursions in the lower stratosphere become clear in 13-month segments when they are bias-shifted to zero in March (a,b) and September (c,d) and then colour coded according to their QBO phase in April or October, respectively (vertical dotted line). The largest deviations are found to occur four or five months later (vertical dashed line), at the onset of hemispheric autumn (Holton and Tan, 1980; Dunkerton and Baldwin, 1991). It is also interesting to note that the only large, positive QBO-Westerly anomaly that peaks four

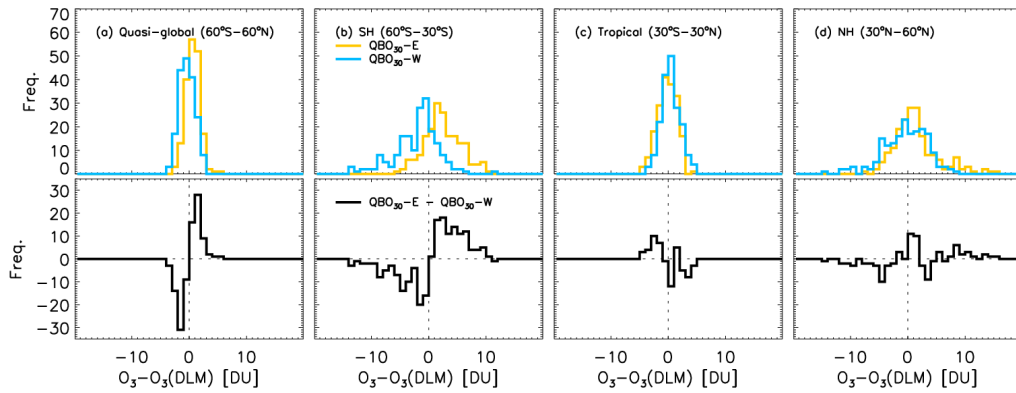


Figure 3: (**Upper row**) Histograms of ozone anomalies relative to the DLM non-linear trend line in Fig. 2 for months when the QBO at 30 hPa is either in an Easterly (yellow) or Westerly (blue) phase: (a) quasi-global (60°S–60°N), (b) southern hemisphere (60°–30°S, 147–30 hPa), (c) tropics (30°S–30°N, 100–30 hPa), (d) northern hemisphere (30°–60°N, 147–30 hPa). (**Lower row**) Difference between QBO Easterly and Westerly histograms from the upper row.

months later, in either hemisphere, occurs in the SH in 2002. This year is famous for having the only observed sudden stratospheric warming in the SH, and indicates that while the QBO-phase appears to dominate this distribution of anomalies, other processes can also sometimes dominate.

315 We reiterate that the separation of positive and negative anomalies into those related to Easterly or Westerly QBO phases is clearest for the SH (Fig. 4a) and the corresponding, opposing, equatorial changes (Fig. 4b). The anti-correlated behaviour of anomalies between mid-latitude and equatorial regions is consistent with previous studies investigating the relationship between the QBO and mid-latitude ozone variability (e.g. Zerefos et al. (1992); Randel et al. (1999); Strahan et al. (2015)). We  
 320 summarise the dynamical concept, in the context of these results, in the following (see Baldwin et al. (2001) and Choi et al. (2002) for detailed discussion). The QBO consists of downward propagating equatorial zonal winds; in the lower stratosphere this consists of a Westerly above an Easterly, or vice versa. A Westerly above Easterly (i.e. the 15 hPa QBO is Westerly as identified by blue lines in Fig. 4) leads to a shear that induces an anomalous downward motion of air, and adiabatic warming  
 325 (Fig. 1 of Choi et al. (2002)) and also leads to an anomalous increase in ozone. For an Easterly above a westerly, this leads to anomalously rising air and adiabatic cooling together with an associated ozone decrease. An equator-to-mid-latitude circulation forms to conserve mass (Randel et al., 1999; Polvani et al., 2010; Tweedy et al., 2017). At sub-tropical and mid-latitudes, the return of this meridional circulation draws ozone-rich air from above down into ozone poor regions, anomalously  
 330 enhancing ozone there (yellow, Fig. 4a,c). When Easterlies lie over Westerlies (blue, Fig. 4), the opposite circulation is set up, and ozone anomalies reverse.



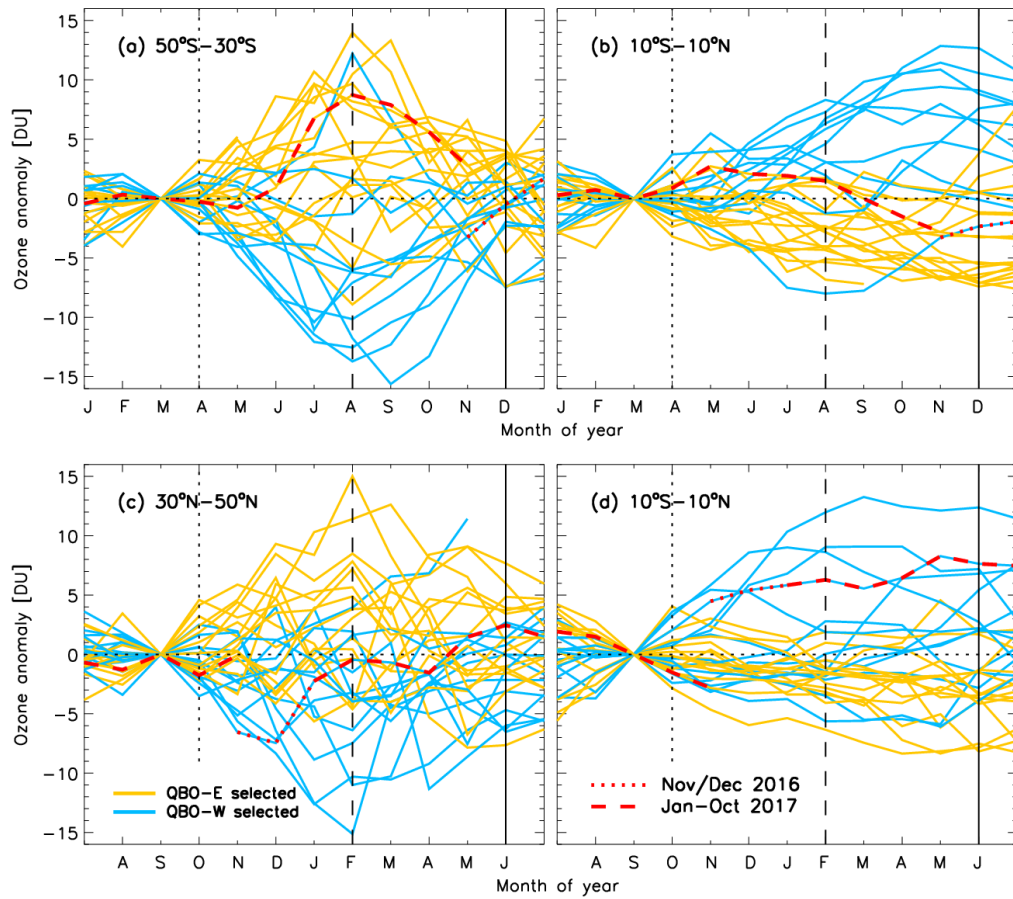


Figure 4: Lower stratospheric partial column ozone at (a) 50°–30°S, (b,d) 10°S–10°N, and (c) 30°–50°N. Each line represents a 13 month period starting in January (upper row) or July (lower row), all bias-shifted to zero in March (upper) or September (lower) and colour coded by the state of the QBO at 15 hPa in April (upper) or October (lower) so that QBO Easterly phases are yellow and Westerly blue. The period covering November to December 2016 is highlighted as a dotted-red line, while January to November 2017 is dashed-red.

The 2017 increase is highlighted in Fig. 4, with November 2016 to January 2017 shown as a dotted red line, and January to October 2017 as a red dashed line. Focusing on Fig. 4a in the SH, the increase onset during the Easterly phase is large, but as noted earlier larger excursions have occurred  
 335 before and regularly (Fig. 2). A prolonged Westerly phase, following the breakdown of the expected QBO pattern in 2016 (Osprey et al., 2016; Newman et al., 2016; Tweedy et al., 2017), may have contributed to a suppressed level of ozone in 2016 (note the single orange dot in 2016 in Fig. 2a signifying a brief Easterly QBO phase). The arrival of the Easterly phase proper in 2017 led to the ozone increase at mid-latitudes. The Westerly phase at 30 hPa began in late 2018 and ozone should,

Table 1: Absolute change between 1998 and 2018 in Dobson Units, and probability (in brackets) of a positive or negative change in ozone (%) for integrated regions of the stratosphere. Blue text indicates ozone changes are negative, while red indicate positive changes; bold text indicates probabilities exceeding 90%.

Region	60°–30°S	50°–30°S	30°S–30°N	20°S–20°N	30°–50°N	30°–60°N	50°S–50°N	60°S–60°N
Whole	-0.5 (57)	-0.4 (56)	<b>-1.9 (95)</b>	<b>-2.5 (95)</b>	-3.2 (83)	-2.6 (77)	-1.1 (86)	-1.1 (86)
Upper	<b>+0.8 (100)</b>	<b>+0.8 (99)</b>	<b>+0.7 (96)</b>	+0.6 (85)	<b>+0.6 (92)</b>	<b>+0.6 (96)</b>	<b>+0.8 (100)</b>	<b>+0.8 (100)</b>
Middle	+0.7 (78)	+0.8 (78)	<b>-0.7 (94)</b>	<b>-0.9 (95)</b>	-0.5 (73)	-0.5 (72)	-0.4 (81)	-0.2 (73)
Lower	-1.6 (80)	-1.7 (82)	<b>-2.1 (99)</b>	<b>-2.1 (98)</b>	-1.9 (82)	-1.5 (76)	<b>-1.8 (99)</b>	<b>-1.7 (99)</b>

340 barring no further QBO breakdown, decrease again in 2019 in the SH mid-latitudes; the last three months of 2018 hint at such a decrease (Fig. 2).

Despite this variability, Fig. 1 indicates that the lower stratospheric negative trends in ozone could already be identified throughout the lower stratosphere before, and after, 2016. As such, the QBO breakdown event is likely not the primary cause of the negative ozone trends reported by Ball et al.  
345 (2018), but does appear to affect the robustness of the trend depending on the end year. We will investigate this end-year sensitivity in section 3.5.

### 3.4 Latitude-integrated lower stratospheric ozone trend estimates

While Chipperfield et al. (2018) applied ordinary least squares trend fits to timeseries using a single linear trend, this cannot be compared to multi-variate regression approaches, e.g. DLM and MLR.  
350 This is because the former simply asks what the trend in the data is, regardless of the forcing agents, while the latter attempts to separate known (usually quasi-periodic) drivers to distill out the trend that has (usually unknown) drivers of its own. The DLM non-linear trend estimates presented here are the first multivariate analysis applied to ozone timeseries that include the large ozone increase witnessed in 2017. It is important to be clear that long-term trends cannot be compared with single  
355 year changes and, indeed, the processes governing each timescale are likely quite different. While large short-term increases will likely bias the whole trend-line for that period under MLR analyses (with piecewise linear and independent linear trends – PWLT or ILT), DLM promises to be more robust in the sense that asymmetric fluctuations will only influence part of the smooth trend over a timescale fixed by the smoothness parameter  $\sigma_{trend}$  that controls how rapidly the trend is allowed to  
360 evolve (see Ball et al. (2017) and Fig. S1).

The DLM trends in lower stratospheric ozone estimated over 1985–2018 in Fig. 2 continue to be negative, monotonic trends up to 2018 in the quasi-global, tropical and NH regions, while the SH trend reaches a minimum in ~2013 before slowly rising. All integrated regions suggest that mean

ozone remains below the 1998 level (see Table 1<sup>3</sup>), though the probability of an overall decrease is 99% in quasi-global ozone, dominated by the tropics (99%), with probabilities of a decrease of 80% and 76% in the SH and NH respectively. Except in the SH, monotonic downward trends remain, with the addition of two years only affecting the gradient of the monotonic trends (compare the dotted line for the year ending in 2016, with the dashed for 2017 and solid for 2018 in Fig. 2). This agrees with Chipperfield et al. (2018) who suggested the large rapid increase of 2017 affected trends, although this was mainly in the SH and has subsequently showed little change over 2018. However, it has done little to reduce the overall probability of a decrease in the quasi-global timeseries (99%). Furthermore, the shape of the DLM curve is affected only near the end years, such that the period away from the end-date is relatively insensitive to a change in the end year and becomes ‘locked-in’<sup>4</sup>. This is a good example of the inadequacy of using linear trends to describe these data. As the DLM-estimated changes in ozone relative to 1998 in years prior to 2010 are essentially unaffected by the addition of 2017 and 2018, the data show robustly that lower stratospheric ozone did continue to decrease until at least 2010 in all regions. We speculate that the shift back to a QBO-Westerly phase will again decrease ozone at mid-latitudes in 2019 (which appears to have begun in October 2018, see Fig. 2). If that happens, it is therefore possible that the non-linear trend estimates will likely decrease again, and the emergent 2013 minimum in the DLM non-linear trend estimate seen in Fig. 2b is likely to shift to a later date or disappear.

### 3.5 Sensitivity of DLM trends to the end year and non-linear seasonal-QBO effects

Since mid-latitude ozone excursions depend on the QBO-seasonal interaction, i.e. the QBO phase relative to the time of year, this is a non-linear mode of variability. Without predictors to represent this non-linear behaviour, linear regression models (including both DLM and MLR) cannot capture these excursions and the variability can leak into, and bias, other predictors. Most importantly, this bias may include the trend component of the regression model. In this section we examine the magnitude of this effect through a sensitivity analysis of the DLM trends to the end date of the data, both spatially and with partial columns.

Due to the magnitude of the mid-latitude, seasonally-dependent QBO ozone variability on short (two to three year) timescales, ILT or PWLT applied to the relatively short post-1997 timeseries will be sensitive to these large swings in ozone. For the smooth DLM trends on the other hand, we expect the last few years of the curve will be primarily affected, with the rest of the trend being stable. We demonstrate the impact on DLM in Fig. 5 where the partial column regions as presented in Ball et al. (2018) are shown for 10° bands, and quasi-globally (right column), over the whole stratospheric column (top), as well as upper, middle and lower. We show DLM curves estimated from six periods

<sup>3</sup>an extended supplementary materials Table S1 provides changes in DU, %, and % per decade for 1985–2018, 1985–1997, and 1998–2018.

<sup>4</sup>In contrast, for MLR analyses, the entire trend-line is impacted by changes in the end year (Frith et al., 2014; Weber et al., 2018; Keeble et al., 2018)

that start in 1985 and end in 2013–2018, as in Fig. 1. All curves are bias shifted to zero in 1998. This provides a visualisation of the sensitivity of the non-linear trends to the end year (and hence also the large resurgence in ozone in 2017). The uncertainties associated with a change between 1998 and the end year are presented in Fig. 6 with 95% (dark grey shading) and 99% credible intervals. The results specifically for the 1998–2018 change are combined and presented in Fig. 7 as probability distributions, in the same manner as in Fig. 2 of Ball et al. (2018), where blue and red colours represent negative and positive changes respectively, and numbers above each distribution are the probability of the change (fraction of the probability distributions) being negative.

From the panels of Fig. 5, it is clear that ozone trends in the middle-stratosphere exhibit the largest sensitivity to the end year, and the uncertainties in the change from 1998 are consistently large (Fig. 6). Quasi-globally the middle stratosphere change since 1998 is negative for all end years, but does not exceed 95% probability. The upper stratosphere is also sensitive to the end year in the tropics (Fig. 5), and the end year shifts the estimated ozone change from negative to positive with increasing end year, although the uncertainty always remains large (Fig. 6). At mid-latitudes uncertainties in the change of upper stratospheric ozone since 1998 are smaller, but there has been a general shift towards more positive and significant increases, which is more-clearly reflected in the SH and quasi-global estimates.

The evolution of the lower and whole stratospheric non-linear ozone trends mimic each other. South of 30°S, the end points of the negative changes have quickly increased in 2017 and 2018 relative to 1998, though remain negative in the lower stratosphere. At latitudes north of 30°S, the addition of 2017 and 2018 has made little difference to the monotonic ozone decline and for 50–60°N, where changes are flat, the additional years make little difference. The quasi-global lower stratospheric ozone continues to exhibit a monotonic decline that is still highly confident with 99% probability (Fig. 7 and Table 1), and ozone abundances integrated over the whole stratosphere continues to remain lower in 2018 than in 1998, though this is now with a probability of 86%; these trends are dominated by the tropical contribution (58%, latitude weighted) to the quasi-global change, whereas the NH and SH contribute 21% each. Even so, the NH changes do not appear affected by the recent large seasonally-dependent QBO variability.

Figure 5 also confirms that the gradients of the non-linear curves are only affected by unmodelled variance in years close to the end points, typically within the last five years of the partial column timeseries considered here. The shape of the DLM curves prior to the final five years of the DLM curves are largely unaffected. Indeed, even with the large ozone increase in 2017 in the SH, we see that all trend-curves agree well prior to 2010. This is also true in other panels, e.g. the middle stratosphere and tropical upper stratosphere. In the upper stratosphere the recovery onset remains robust, but in the SH lower stratosphere the large increase in 2017 results in the non-linear trend curve having a local minimum emerge around 2013. As such we can infer that additional data are unlikely to affect the inferred change of ozone in 2013, relative to 1998, or push the minima to earlier

dates, because the affecting end year moves further away with more data. However, subsequent data  
435 might once again push the changes since 1998 to lower levels, e.g. if mid-latitudes do respond to  
a Westerly-phase QBO with ozone reducing sharply as it has done in the past (Fig. 2). We expand  
the idea of inferring the likely earliest minimum using the DLM with spatially-resolved data in the  
Supplementary Materials.

### 3.6 Update on ozone profiles

440 Briefly, in Fig. 8, we provide updated ozone change profiles for 1998–2018 using the standard latitu-  
dinal ranges for the SH (60°–35°S), tropics (20°S–20°N), and NH (35°–60°N) (WMO, 2014, 2018;  
Steinbrecht et al., 2017; Petropavlovskikh et al., 2019). Fig. 8, also includes 1998–2016 and 1998–  
2017 profiles for comparison and shows that, for 1998–2018, confidence in an upper stratospheric  
ozone recovery from ODSs is clear for all latitude bands, including the tropics where it has pre-  
445 viously remained below the 95% significance levels. The lower stratosphere shows negative ozone  
changes at almost all levels, though these generally do not exceed a probability of 95%.

Ball et al. (2018), and Fig. 7, indicate that the 50–60° zonal means in both hemispheres show  
little ozone change in the lower stratosphere in the last 21 years, while the tropical regions out  
to 30° show a strong decrease. By modifying the latitudinal extent of the profiles slightly, so that  
450 mid-latitudes cover 30–50° to exclude 50–60° and the tropics are widened to 30°S–30°N to include  
the subtropics, the modified profiles are presented in Fig. 9. This provides some measure of the  
sensitivity to the latitudinal ranges chosen. Now we see the tropics show close to 95% confidence of  
an ozone decrease at all tropical lower stratospheric pressure levels, and there is increased confidence  
of an ozone reduction in the mid-latitude lower stratosphere. Further, the inclusion of higher latitude  
455 regions (20–30°) reinforces the tropical upper stratosphere ozone increase.

An upper stratospheric increase is the expected result from long-term stratospheric chlorine re-  
ductions, a direct consequence of the Montreal Protocol and its amendments, though we do not  
explicitly attribute the cause of the increase to that here (for more on attribution see, for example,  
WMO (2018)). Indeed, the Montreal Protocol and its amendments will have been effective in re-  
460 ducing ozone losses throughout the atmosphere through reductions in CFC emissions, HCFCs and  
other ODSs. The lack of a positive trend since 1998 in the lower stratosphere, as opposed to the one  
clear in the upper stratosphere, is likely the consequence of other factors such as dynamical changes  
(Wargan et al., 2018). These results once again reinforce the conclusion that only the SH is affected  
by the 2017 ozone increase (lower stratosphere), that the Montreal Protocol appears to be working  
465 (upper stratosphere), and that the decreases in the lower stratosphere at tropical and NH latitudes  
remain in place, but are not yet fully understood.

## 4 Conclusions

Here, we have extended and analysed the BASIC<sub>SG</sub> stratospheric ozone composite from Ball et al. (2018) by two years to cover 1985–2018. BASIC<sub>SG</sub> merges two composites, SWOOSH and GOZ-CARDS. We perform a set of sensitivity tests, using dynamical linear modelling (DLM), on the post-1997 trend estimates to understand the impact of a recently reported, large increase in modelled ozone in the lower stratosphere in 2017 (Chipperfield et al., 2018), following almost two decades of persistently decreasing ozone.

The aim of this work is to assess the current state of, and trends in, stratospheric ozone. Improved knowledge of such trends, and the relevant forcing mechanisms and associated variability, will help to better constrain CCM projections of ozone to the end of the 21<sup>st</sup> Century. Chemistry models resolving the stratosphere are one of the best tools for attribution and long-range studies of ozone, but different types exist. Free-running CCMs generate their own model-dependent internal climate and variability. Chemistry transport models (CTMs) use wind, temperature and surface pressure fields fully prescribed by reanalyses. And, specified-dynamics CCMs (SD-CCMs) use reanalyses to nudge the internally-generated variability of the model closer to the historical variability in the real atmosphere while attempting to retain model dependent processes and internal consistency. CTMs and SD-CCMs can be useful for attributing historical changes in ozone to evolving concentrations of CO<sub>2</sub> and ODSs (Solomon et al., 2016), or the Sun (Ball et al., 2016), by accounting for dynamical variability in observations.

A recent study (Chipperfield et al., 2018) used a CTM to reconstruct the ozone timeseries beyond the observational record available at the time to 2017 and found that that model simulated a lower stratospheric ozone increase in 2017 back to 1998 levels. This increase was attributed to dynamical variability. Indeed, chemistry and photochemistry play a dominant role over dynamical perturbations in the upper stratosphere as ozone lifetimes are short (~days), while ozone lifetimes of ~6–12 months in the lower stratosphere means that equator-to-mid-latitude transport of similar timescales plays an important (dominant) role there (London, 1980; Perliski et al., 1989; Brasseur and Solomon, 2005). CTMs can provide insight as to whether the changes might be driven by photochemistry, chemistry, or dynamics. However, because the dynamical fields are prescribed, the CTM cannot provide insight into the underlying dynamical driver of the long-term decreases or the 2017 increase. We show here that the 2017 increase simulated by the CTM (Chipperfield et al., 2018) was more than 60% larger than that observed, and that the 1998–2017 and –2018 (Fig. 1e and f) change remains negative (60°S–60°N), and significant in the tropics and some sub-regions of the NH (Fig. 1f). Neither free-running CCMs (WMO, 2014), nor SD-CCMs (Ball et al., 2018), have so far been demonstrated to accurately reproduce the long-term changes estimated from observations in lower stratospheric ozone (Fig. 6).

The effect of the ozone increase in 2017 was small and the probability of an overall ozone decrease in the lower stratosphere remains at 99% for 1998–2018 (–1.7 DU, or 2.0%; see Table S1). We note that the lower stratospheric ozone trends are dominated by the tropical regions (30°S–30°N) where

the decrease is robust to the end year over 2013–2018, with a probability of 99% (-2.1 DU, -3.5%)  
 505 that it was lower in 2018 than in 1998. Nevertheless, mid-latitudes out to 50°N also indicate that the  
 decrease persists (-1.9 DU, -1.7%). We also find that the 2017–2018 addition enhances the estimated  
 magnitude of the upper stratospheric ozone positive trend, but that the quasi-global (60°S–60°N)  
 ozone layer still displays a reduction since 1998, though the confidence in this has reduced from 95%  
 in 2016 (Ball et al., 2018) to 86% in 2018 (-1.1 DU, -0.4%). Given the high probability of a decrease  
 510 in tropical middle (94%) and lower (99%) stratospheric ozone, the whole tropical stratospheric ozone  
 column indicates a highly probable decrease (95%) over 1998–2018 (-1.9 DU, -0.8%).

In general, uncertainties on changes since 1998 in partial columns have changed little over 2013–  
 2018 (Fig. 6), a result likely due to the large fraction of unaccounted variance in the standard set of  
 predictors used in regression analysis. Our analysis shows that ozone continued to decrease until a  
 515 minimum in at least 2013 in the SH, and has continued to decrease at all latitudes north of 30°S. By  
 comparing the phase of the QBO with large, 2–3 year inter-annual variability at mid-latitudes, the  
 implication is that these large mid-latitude changes are related to the seasonal-dependence of ozone  
 on the QBO, i.e. a non-linearity. If true, this could explain why regression models cannot capture this  
 variability, since such non-linear behaviour is not included. The clarification of the origin of these  
 520 large mid-latitude changes – occurring every few years – is a high priority.

CCMs are consistent in the sign of their projections, although lower stratospheric ozone variabil-  
 ity can differ with observations and there is a large spread in their sensitivity to hODSs (Douglass  
 et al., 2012, 2014), and therefore their return dates, i.e. a return of ozone to the level it was in 1980  
 (WMO, 2014; Dhomse et al., 2018; WMO, 2018). CCMs do a good job on many timescales, but  
 525 due to historically different internal variability, and parametrized sub-grid scale processes and nu-  
 merical diffusion, behaviour in some regions may not be well-reproduced (SPARC/WMO, 2010).  
 It is clear from modelling studies that pre-Montreal Protocol ozone decreases can be attributed to  
 ODS increases (WMO, 2014), and SD-CCMs and CTMs generally reproduce the Antarctic ozone  
 hole well (Solomon et al., 2016). The halt in ODS-related ozone losses as a result of the Montreal  
 530 Protocol and its amendments, and an initial recovery from ODSs in total column ozone is almost  
 universally reproduced by CCMs (SPARC/WMO, 2010), as is the upper stratospheric ozone recov-  
 ery. But, negative ozone trends since 1998 in the lower stratosphere have not been demonstrated to  
 be simulated in models in the mid-latitudes, most notably in the NH.

Future projections tend to focus on how stratospheric ozone will evolve under a given global  
 535 warming scenario. This is important given that anthropogenic GHG emissions that are changing the  
 climate may impact inter-annual dynamical variability in the stratosphere (Osprey et al., 2016; New-  
 man et al., 2016; Tweedy et al., 2017). Changes are also expected in the large-scale circulation of the  
 stratosphere, and these are likely to modify future distributions of ozone (Chipperfield et al., 2017).  
 Further, ozone is not a passive tracer, and the large scale long-term changes in ozone are expected  
 540 to feedback on the aforementioned dynamics (Li et al., 2018; Polvani et al., 2018; Abalos et al.,

2019). Such a feedback has been demonstrated, most notably, in the SH following ozone depletion and the growth of the ozone hole (WMO, 2014, 2018). Now, as ozone is expected to recover in the coming decades, the dynamics of the stratosphere are also expected to respond. The overall future expectations are that total column ozone levels will return to 1980s levels globally by ~2050, in the Antarctic by 2100, and by ~2030 and ~2050 in Northern and Southern mid-latitudes, respectively. The mid-latitudes are expected to continue on to a ‘super-recovery’, i.e. that ozone will be higher by the end of the 21<sup>st</sup> Century than prior to 1980s levels (Dhomse et al., 2018; WMO, 2014, 2018), although this is predicated on future scenarios of hODSs decreases continuing as expected (Montzka et al., 2018). However, it is neither clear whether the recent increase in SH lower stratospheric ozone will remain at higher levels or will reduce again in 2019 as the QBO shifts to a Westerly phase, nor why the NH continues to show a persistent decrease. Nonetheless, we note that the signal is small compared to the (i) large inter-annual variability, (ii) pre-2000 changes induced by ozone depleting substances, and (iii) ozone losses that would have occurred without the Montreal Protocol being enacted.

The ongoing negative trend of ozone in the lower stratospheric component of the total column also continues to pose a problem for global trends in tropospheric ozone. If tropospheric ozone has really increased over the last two decades, and stratospheric ozone was not decreasing or remained flat, then some component of the total column ozone must have been decreasing to balance the ozone budget since it appears that total column ozone has remained steady in the past 5-10 years. Alternatively, it is possible that the solution simply lies in very large observational uncertainties (Harris et al., 2015; Gaudel et al., 2018; Petropavlovskikh et al., 2019) and/or the inadequacies of linear regression techniques to attribute variability and identify trends. In addition to potential future improvements in merged observational records, this calls for a community push to improve detection and attribution techniques to solve an issue that is of great importance to the health of society, the biosphere, and the climate.

*Author contributions.* WTB designed the experiments. WTB and JA prepared and executed the BASIC algorithms. JA developed the DLM code and WTB and JA performed the DLM analysis. WTB conceived and performed the QBO analysis. SD and LF prepared and provided GOZCARDS and SWOOSH ozone datasets. WTB prepared the manuscript with contributions from all co-authors.

*Acknowledgements.* W.T.B. was funded by the SNSF project 200020\_182239 (POLE). ‘BASIC<sub>SG</sub>’ for 1985–2018 will be available for download from <https://data.mendeley.com/datasets/2mgx2xzzpk/3> following review of this manuscript. Work at the Jet Propulsion Laboratory was performed under contract with the National Aeronautics and Space Administration. GOZCARDS ozone data contributions from Ryan Fuller (at JPL) are gratefully acknowledged.



## 575 References

- Abalos, M., Polvani, L., Calvo, N., Kinnison, D., Ploeger, F., Randel, W., and Solomon, S.: New Insights on the Impact of Ozone-Depleting Substances on the Brewer-Dobson Circulation, *Journal of Geophysical Research (Atmospheres)*, 124, 2435–2451, doi:10.1029/2018JD029301, 2019.
- Baldwin, M. P., Gray, L. J., Dunkerton, T. J., Hamilton, K., Haynes, P. H., Randel, W. J., Holton, J. R., Alexander, M. J., Hirota, I., Horinouchi, T., Jones, D. B. A., Kinnersley, J. S., Marquardt, C., Sato, K., and Takahashi, M.: The quasi-biennial oscillation, *Reviews of Geophysics*, 39, 179–229, doi:10.1029/1999RG000073, 2001.
- 580 Ball, W. T., Haigh, J. D., Rozanov, E. V., Kuchar, A., Sukhodolov, T., Tummon, F., Shapiro, A. V., and Schmutz, W.: High solar cycle spectral variations inconsistent with stratospheric ozone observations, *Nature Geoscience*, 9, 206–209, doi:10.1038/ngeo2640, 2016.
- 585 Ball, W. T., Alsing, J., Mortlock, D. J., Rozanov, E. V., Tummon, F., and Haigh, J. D.: Reconciling differences in stratospheric ozone composites, *Atmospheric Chemistry & Physics*, 17, 12 269–12 302, doi:10.5194/acp-17-12269-2017, 2017.
- Ball, W. T., Alsing, J., Mortlock, D. J., Staehelin, J., Haigh, J. D., Peter, T., Tummon, F., Stuebi, R., Stenke, A., Anderson, J., Bourassa, A., Davis, S. M., Degenstein, D., Frith, S., Froidevaux, L., Roth, C., Sofieva, V., Wang, R., Wild, J., Yu, P., Ziemke, J. R., and Rozanov, E. V.: Evidence for a continuous decline in lower stratospheric ozone offsetting ozone layer recovery, *Atmospheric Chemistry & Physics*, 18, 1379–1394, doi:10.5194/acp-18-1379-2018, 2018.
- 590 Bodeker, G. E., Garny, H., Smale, D., Dameris, M., and Deckert, R.: The 1985 Southern Hemisphere mid-latitude total column ozone anomaly, *Atmospheric Chemistry & Physics*, 7, 5625–5637, 2007.
- 595 Brasseur, G. P. and Solomon, S.: *Aeronomy of the Middle Atmosphere: Chemistry and Physics of the Stratosphere and Mesosphere*, Dordrecht: Springer Netherlands, Editor: Mysak, L. A., 2005.
- Butchart, N.: The Brewer-Dobson circulation, *Reviews of Geophysics*, 52, 157–184, doi:10.1002/2013RG000448, 2014.
- Chipperfield, M. P., Bekki, S., Dhomse, S., Harris, N. R. P., Hassler, B., Hossaini, R., Steinbrecht, W., Thiéblemont, R., and Weber, M.: Detecting recovery of the stratospheric ozone layer, *Nature*, 549, 211–218, doi:10.1038/nature23681, 2017.
- 600 Chipperfield, M. P., Dhomse, S., Hossaini, R., Feng, W., Santee, M. L., Weber, M., Burrows, J. P., Wild, J. D., Loyola, D., and Coldewey-Egbers, M.: On the Cause of Recent Variations in Lower Stratospheric Ozone, *Geophys. Res. Lett.*, 45, 5718–5726, doi:10.1029/2018GL078071, 2018.
- 605 Choi, W., Lee, H., Grant, W. B., Park, J. H., Holton, J. R., Lee, K.-M., and Naujokat, B.: On the secondary meridional circulation associated with the quasi-biennial oscillation, *Tellus Series B Chemical and Physical Meteorology B*, 54, 395, doi:10.3402/tellusb.v54i4.16673, 2002.
- Davis, S. M., Rosenlof, K. H., Hassler, B., Hurst, D. F., Read, W. G., Vömel, H., Selkirk, H., Fujiwara, M., and Damadeo, R.: The Stratospheric Water and Ozone Satellite Homogenized (SWOOSH) database: a long-term database for climate studies, *Earth System Science Data*, 8, 461–490, doi:10.5194/essd-8-461-2016, 2016.
- 610 Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-

- 615 J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Quarterly Journal of the Royal Meteorological Society*, 137, 553–597, doi:10.1002/qj.828, 2011.
- Dhomse, S. S., Kinnison, D., Chipperfield, M. P., Salawitch, R. J., Cionni, I., Hegglin, M. I., Abraham, N. L., Akiyoshi, H., Archibald, A. T., Bednarz, E. M., Bekki, S., Braesicke, P., Butchart, N., Dameris, M., Deushi, M., Frith, S., Hardiman, S. C., Hassler, B., Horowitz, L. W., Hu, R.-M., Jöckel, P., Josse, B., Kirner, O., 620 Kremser, S., Langematz, U., Lewis, J., Marchand, M., Lin, M., Mancini, E., Marécal, V., Michou, M., Morgenstern, O., O'Connor, F. M., Oman, L., Pitari, G., Plummer, D. A., Pyle, J. A., Revell, L. E., Rozanov, E., Schofield, R., Stenke, A., Stone, K., Sudo, K., Tilmes, S., Visionsi, D., Yamashita, Y., and Zeng, G.: Estimates of ozone return dates from Chemistry-Climate Model Initiative simulations, *Atmospheric Chemistry & Physics*, 18, 8409–8438, doi:10.5194/acp-18-8409-2018, 2018.
- 625 Diallo, M., Riese, M., Birner, T., Konopka, P., Müller, R., Hegglin, M. I., Santee, M. L., Baldwin, M., Legras, B., and Ploeger, F.: Response of stratospheric water vapor and ozone to the unusual timing of El Niño and the QBO disruption in 2015-2016, *Atmospheric Chemistry & Physics*, 18, 13 055–13 073, doi:10.5194/acp-18-13055-2018, 2018.
- 630 Diallo, M., Konopka, P., Santee, M. L., Müller, R., Tao, M., Walker, K. A., Legras, B., Riese, M., Ern, M., and Ploeger, F.: Structural changes in the shallow and transition branch of the Brewer–Dobson circulation induced by El Niño, *Atmospheric Chemistry & Physics*, 19, 425–446, doi:10.5194/acp-19-425-2019, 2019.
- Douglass, A. R., Stolarski, R. S., Strahan, S. E., and Oman, L. D.: Understanding differences in upper stratospheric ozone response to changes in chlorine and temperature as computed using CCMVal-2 models, *Journal of Geophysical Research (Atmospheres)*, 117, D16306, doi:10.1029/2012JD017483, 2012.
- 635 Douglass, A. R., Strahan, S. E., Oman, L. D., and Stolarski, R. S.: Understanding differences in chemistry climate model projections of stratospheric ozone, *Journal of Geophysical Research (Atmospheres)*, 119, 4922–4939, doi:10.1002/2013JD021159, 2014.
- Dudok de Wit, T., Bruinsma, S., and Shibasaki, K.: Synoptic radio observations as proxies for upper atmosphere modelling, *Journal of Space Weather and Space Climate*, 4, A06, doi:10.1051/swsc/2014003, 2014.
- 640 Dunkerton, T. J. and Baldwin, M. P.: Quasi-biennial Modulation of Planetary-Wave Fluxes in the Northern Hemisphere Winter., *Journal of Atmospheric Sciences*, 48, 1043–1061, doi:10.1175/1520-0469(1991)048<1043:QBMOPW>2.0.CO;2, 1991.
- Frith, S. M., Kramarova, N. A., Stolarski, R. S., McPeters, R. D., Bhartia, P. K., and Labow, G. J.: Recent 645 changes in total column ozone based on the SBUV Version 8.6 Merged Ozone Data Set, *Journal of Geophysical Research (Atmospheres)*, 119, 9735–9751, doi:10.1002/2014JD021889, 2014.
- Froidevaux, L., Anderson, J., Wang, H.-J., Fuller, R. A., Schwartz, M. J., Santee, M. L., Livesey, N. J., Pumphrey, H. C., Bernath, P. F., Russell, III, J. M., and McCormick, M. P.: Global Ozone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS): methodology and sample results with 650 a focus on HCl, H<sub>2</sub>O, and O<sub>3</sub>, *Atmospheric Chemistry & Physics*, 15, 10 471–10 507, doi:10.5194/acp-15-10471-2015, 2015.
- Froidevaux, L., Kinnison, D. E., Wang, R., Anderson, J., and Fuller, R. A.: Evaluation of CESM1 (WACCM) free-running and specified-dynamics atmospheric composition simulations using global multispecies satellite data records, *Atmos. Chem. Phys.*, 19, 4783–4821, doi:10.5194/acp-19-4783-2019, 2019.

- 655 Garfinkel, C. I., Gordon, A., Oman, L. D., Li, F., Davis, S., and Pawson, S.: Nonlinear response of tropical lower-stratospheric temperature and water vapor to ENSO, *Atmospheric Chemistry & Physics*, 18, 4597–4615, doi:10.5194/acp-18-4597-2018, 2018.
- Gaudel, A., Cooper, O. R., and etc, E.: Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, *Elem*
- 660 *Sci Anth*, 6, 10, doi:10.1525/elementa.291, 2018.
- Gray, L. J. and Pyle, J. A.: A two-dimensional model of the quasi-biennial oscillation of ozone, *Journal of Atmospheric Sciences*, 46, 203–220, doi:10.1175/1520-0469(1989)046<0203:ATDMOT>2.0.CO;2, 1989.
- Harris, N. R. P., Hassler, B., Tummon, F., Bodeker, G. E., Hubert, D., Petropavlovskikh, I., Steinbrecht, W., Anderson, J., Bhartia, P. K., Boone, C. D., Bourassa, A., Davis, S. M., Degenstein, D., Delcloo, A., Frith, S. M., Froidevaux, L., Godin-Beekmann, S., Jones, N., Kurylo, M. J., Kyrölä, E., Laine, M., Leblanc, S. T., Lambert, J.-C., Liley, B., Mahieu, E., Maycock, A., de Mazière, M., Parrish, A., Querel, R., Rosenlof, K. H., Roth, C., Sioris, C., Staehelin, J., Stolarski, R. S., Stübi, R., Tamminen, J., Vigouroux, C., Walker, K. A., Wang, H. J., Wild, J., and Zawodny, J. M.: Past changes in the vertical distribution of ozone - Part 3: Analysis and interpretation of trends, *Atmospheric Chemistry & Physics*, 15, 9965–9982, doi:10.5194/acp-15-9965-2015, 2015.
- 670 Holton, J. R. and Tan, H.-C.: The Influence of the Equatorial Quasi-Biennial Oscillation on the Global Circulation at 50 mb., *Journal of Atmospheric Sciences*, 37, 2200–2208, doi:10.1175/1520-0469(1980)037<2200:TIOTEQ>2.0.CO;2, 1980.
- Hood, L., Rossi, S., and Beulen, M.: Trends in lower stratospheric zonal winds, Rossby wave breaking behavior, and column ozone at northern midlatitudes, *J. Geophysical Res.*, 104, 24,321–24,339, doi:10.1029/1999JD900401, 1999.
- 675 Hood, L. L. and Soukharev, B. E.: Interannual Variations of Total Ozone at Northern Midlatitudes Correlated with Stratospheric EP Flux and Potential Vorticity., *Journal of Atmospheric Sciences*, 62, 3724–3740, doi:10.1175/JAS3559.1, 2005.
- 680 Hood, L. L. and Zaff, D. A.: Lower stratospheric stationary waves and the longitude dependence of ozone trends in winter, *J. Geophysical Res.*, 100, 25,791–25,800, doi:10.1029/95JD01943, 1995.
- Keeble, J., Bednarz, E. M., Banerjee, A., Abraham, N. L., Harris, N. R. P., Maycock, A. C., and Pyle, J. A.: Diagnosing the radiative and chemical contributions to future changes in tropical column ozone with the UM-UKCA chemistry-climate model, *Atmospheric Chemistry & Physics*, 17, 13 801–13 818, doi:10.5194/acp-17-13801-2017, 2017.
- 685 Keeble, J., Brown, H., Abraham, N. L., Harris, N. R. P., and Pyle, J. A.: On ozone trend detection: using coupled chemistry-climate simulations to investigate early signs of total column ozone recovery, *Atmospheric Chemistry & Physics*, 18, 7625–7637, doi:10.5194/acp-18-7625-2018, 2018.
- Kidston, J., Scaife, A. A., Hardiman, S. C., Mitchell, D. M., Butchart, N., Baldwin, M. P., and Gray, L. J.: Stratospheric influence on tropospheric jet streams, storm tracks and surface weather, *Nature Geoscience*, 8, 433–440, doi:10.1038/ngeo2424, 2015.
- 690 Laine, M., Latva-Pukkila, N., and Kyrölä, E.: Analysing time-varying trends in stratospheric ozone time series using the state space approach, *Atmospheric Chemistry & Physics*, 14, 9707–9725, doi:10.5194/acp-14-9707-2014, 2014.

- 695 Li, F., Newman, P., Pawson, S., and Perlwitz, J.: Effects of Greenhouse Gas Increase and Stratospheric Ozone Depletion on Stratospheric Mean Age of Air in 1960-2010, *Journal of Geophysical Research (Atmospheres)*, 123, 2098–2110, doi:10.1002/2017JD027562, 2018.
- London, J.: The Observed Distribution and Variations of Total Ozone, in: *Atmospheric Ozone and its Variation and Human Influences*, edited by Nicolet, M. and Aikin, A. C., p. 31, 1980.
- 700 Meul, S., Dameris, M., Langematz, U., Abalichin, J., Kerschbaumer, A., Kubin, A., and Oberländer-Hayn, S.: Impact of rising greenhouse gas concentrations on future tropical ozone and UV exposure, *Geophys. Res. Lett.*, 43, 2919–2927, doi:10.1002/2016GL067997, 2016.
- Montzka, S. A., Dutton, G. S., Yu, P., Ray, E., Portmann, R. W., Daniel, J. S., Kuijpers, L., Hall, B. D., Mondeel, D., Siso, C., Nance, J. D., Rigby, M., Manning, A. J., Hu, L., Moore, F., Miller, B. R., and Elkins, J. W.: An  
705 unexpected and persistent increase in global emissions of ozone-depleting CFC-11, *Nature*, 557, 413–417, doi:10.1038/s41586-018-0106-2, 2018.
- NCAR: The Climate Data Guide: Multivariate ENSO Index, Retrieved from <https://climatedataguide.ucar.edu/climate-data/multivariate-enso-index>, 2013.
- Newman, P. A., Coy, L., Pawson, S., and Lait, L. R.: The anomalous change in the QBO in 2015-2016, *Geophys. Res. Lett.*, 43, 8791–8797, doi:10.1002/2016GL070373, 2016.
- 710 Osprey, S. M., Butchart, N., Knight, J. R., Scaife, A. A., Hamilton, K., Anstey, J. A., Schenzinger, V., and Zhang, C.: An unexpected disruption of the atmospheric quasi-biennial oscillation, *Science*, 353, 1424–1427, doi:10.1126/science.aah4156, 2016.
- Perliski, L. M., London, J., and Solomon, S.: On the interpretation of seasonal variations of stratospheric ozone,  
715 *Journal of Geophysical Research*, 37, 1527–1538, doi:10.1016/0032-0633(89)90143-8, 1989.
- Peters, D. and Entzian, G.: Longitude-Dependent Decadal Changes of Total Ozone in Boreal Winter Months during 1979-92., *Journal of Climate*, 12, 1038–1048, doi:10.1175/1520-0442(1999)012<1038:LDDCOT>2.0.CO;2, 1999.
- Petropavlovskikh, I., Godin-Beekmann, S., Hubert, D., Damadeo, R., Hassler, B., and Sofieva,  
720 V.: SPARC/IO3C/GAW report on Long-term Ozone Trends and Uncertainties in the Stratosphere, SPARC/IO3C/GAW, SPARC Report No. 9, WCRP-17/2018, GAW Report No. 241, doi:10.17874/f899e57a20b, 2019.
- Polvani, L. M., Sobel, A. H., and Waugh, D. W.: The Stratosphere: Dynamics, Transport, and Chemistry, Washington DC American Geophysical Union Geophysical Monograph Series, 190, doi:10.1029/GM190, 2010.
- 725 Polvani, L. M., Abalos, M., Garcia, R., Kinnison, D., and Randel, W. J.: Significant Weakening of Brewer-Dobson Circulation Trends Over the 21st Century as a Consequence of the Montreal Protocol, *Geophys. Res. Lett.*, 45, 401–409, doi:10.1002/2017GL075345, 2018.
- Randel, W. J., Wu, F., Swinbank, R., Nash, J., and O'Neill, A.: Global QBO Circulation Derived from UKMO Stratospheric Analyses., *Journal of Atmospheric Sciences*, 56, 457–474, doi:10.1175/1520-0469(1999)056<0457:GQCDFU>2.0.CO;2, 1999.
- 730 Slaper, H., Velders, G. J. M., Daniel, J. S., de Groot, F. R., and van der Leun, J. C.: Estimates of ozone depletion and skin cancer incidence to examine the Vienna Convention achievements, *Nature*, 384, 256–258, doi:10.1038/384256a0, 1996.

- Solomon, S., Ivy, D. J., Kinnison, D., Mills, M. J., Neely, R. R., and Schmidt, A.: Emergence of healing in the Antarctic ozone layer, *Science*, 353, 269–274, doi:10.1126/science.aae0061, 2016.
- SPARC/WMO: SPARC Report on the Evaluation of Chemistry-Climate Models, SPARC, 2010.
- Steinbrecht, W., Froidevaux, L., Fuller, R., Wang, R., Anderson, J., Roth, C., Bourassa, A., Degenstein, D., Damadeo, R., Zawodny, J., Frith, S., McPeters, R., Bhartia, P., Wild, J., Long, C., Davis, S., Rosenlof, K., Sofieva, V., Walker, K., Rahpoe, N., Rozanov, A., Weber, M., Laeng, A., von Clarmann, T., Stiller, G., Kramarova, N., Godin-Beekmann, S., Leblanc, T., Querel, R., Swart, D., Boyd, I., Hocke, K., Kämpfer, N., Maillard Barras, E., Moreira, L., Nedoluha, G., Vigouroux, C., Blumenstock, T., Schneider, M., Garcia, O., Jones, N., Mahieu, E., Smale, D., Kotkamp, M., Robinson, J., Petropavlovskikh, I., Harris, N., Hassler, B., Hubert, D., and Tummon, F.: An update on ozone profile trends for the period 2000 to 2016, *Atmos. Chem. Phys. Discuss.*, 2017, 1–24, doi:10.5194/acp-2017-391, <https://www.atmos-chem-phys-discuss.net/acp-2017-391/>, 2017.
- Stone, K. A., Solomon, S., and Kinnison, D. E.: On the Identification of Ozone Recovery, *Geophys. Res. Lett.*, 45, 5158–5165, doi:10.1029/2018GL077955, 2018.
- Strahan, S. E., Oman, L. D., Douglass, A. R., and Coy, L.: Modulation of Antarctic vortex composition by the quasi-biennial oscillation, *Geophys. Res. Lett.*, 42, 4216–4223, doi:10.1002/2015GL063759, 2015.
- Tachibana, Y., Inoue, Y., Komatsu, K. K., Nakamura, T., Honda, M., Ogata, K., and Yamazaki, K.: Interhemispheric Synchronization Between the AO and the AAO, *Geophys. Res. Lett.*, 45, 13, doi:10.1029/2018GL081002, 2018.
- Thomason, L., Ernest, N., Millan, L., Rieger, L., Bourassa, A., Vernier, J., Peter, T., Luo, B., and Arfeuille, F.: A global, space-based stratospheric aerosol climatology: 1979 to 2016, *Earth Syst. Sci. Data*, in preparation, doi:10.5067/GloSSAC-L3-V1.0, 2017.
- Tiao, G. C., Xu, D., Pedrick, J. H., Zhu, X., and Reinsel, G. C.: Effects of autocorrelation and temporal sampling schemes on estimates of trend and spatial correlation, *Journal of Geophysical Research*, 95, 20 507–20 517, doi:10.1029/JD095iD12p20507, 1990.
- Tohir, A. M., Portafaix, T., Sivakumar, V., Bencherif, H., Pazmiño, A., and Bègue, N.: Variability and trend in ozone over the southern tropics and subtropics, *Annales Geophysicae*, 36, 381–404, doi:10.5194/angeo-36-381-2018, 2018.
- Tummon, F., Hassler, B., Harris, N. R. P., Staehelin, J., Steinbrecht, W., Anderson, J., Bodeker, G. E., Bourassa, A., Davis, S. M., Degenstein, D., Frith, S. M., Froidevaux, L., Kyrölä, E., Laine, M., Long, C., Penckwitt, A. A., Sioris, C. E., Rosenlof, K. H., Roth, C., Wang, H.-J., and Wild, J.: Intercomparison of vertically resolved merged satellite ozone data sets: interannual variability and long-term trends, *Atmospheric Chemistry & Physics*, 15, 3021–3043, doi:10.5194/acp-15-3021-2015, 2015.
- Tweedy, O. V., Kramarova, N. A., Strahan, S. E., Newman, P. A., Coy, L., Randel, W. J., Park, M., Waugh, D. W., and Frith, S. M.: Response of trace gases to the disrupted 2015–2016 quasi-biennial oscillation, *Atmospheric Chemistry & Physics*, 17, 6813–6823, doi:10.5194/acp-17-6813-2017, 2017.
- Wargan, K., Orbe, C., Pawson, S., Ziemke, J. R., Oman, L. D., Olsen, M. A., Coy, L., and Emma Knowland, K.: Recent Decline in Extratropical Lower Stratospheric Ozone Attributed to Circulation Changes, *Geophys. Res. Lett.*, 45, 5166–5176, doi:10.1029/2018GL077406, 2018.

- Weber, M., Coldewey-Egbers, M., Fioletov, V. E., Frith, S. M., Wild, J. D., Burrows, J. P., Long, C. S., and Loyola, D.: Total ozone trends from 1979 to 2016 derived from five merged observational datasets - the emergence into ozone recovery, *Atmospheric Chemistry & Physics*, 18, 2097–2117, doi:10.5194/acp-18-2097-2018, 2018.
- WMO: Scientific Assessment of Ozone Depletion: 2014 Global Ozone Research and Monitoring Project Report, World Meteorological Organization, p. 416, geneva, Switzerland, 2014.
- WMO: Scientific Assessment of Ozone Depletion: 2018, Global Ozone Research and Monitoring Project–Report, World Meteorological Organization, p. 588, geneva, Switzerland, 2018.
- Zerefos, C., Kapsomenakis, J., Eleftheratos, K., Tourpali, K., Petropavlovskikh, I., Hubert, D., Godin-Beekmann, S., Steinbrecht, W., Frith, S., Sofieva, V., and Hassler, B.: Representativeness of single lidar stations for zonally averaged ozone profiles, their trends and attribution to proxies, *Atmospheric Chemistry & Physics*, 18, 6427–6440, doi:10.5194/acp-18-6427-2018, 2018.
- Zerefos, C. S., Bais, A. F., Ziomas, I. C., and Bojkov, R. D.: On the relative importance of quasi-biennial oscillation and El Nino/Southern Oscillation in the revised Dobson total ozone records, *J. Geophys. Res.*, 97, 10, doi:10.1029/92JD00508, 1992.
- Ziemke, J. R., Oman, L. D., Strode, S. A., Douglass, A. R., Olsen, M. A., McPeters, R. D., Bhartia, P. K., Froidevaux, L., Labow, G. J., Witte, J. C., Thompson, A. M., Haffner, D. P., Kramarova, N. A., Frith, S. M., Huang, L.-K., Jaross, G. R., Seftor, C. J., Deland, M. T., and Taylor, S. L.: Trends in Global Tropospheric Ozone Inferred from a Composite Record of TOMS/OMI/MLS/OMPS Satellite Measurements and the MERRA-2 GMI Simulation, *Atmospheric Chemistry and Physics Discussions*, 2018, 1–29, doi:10.5194/acp-2018-716, <https://www.atmos-chem-phys-discuss.net/acp-2018-716/>, 2018.

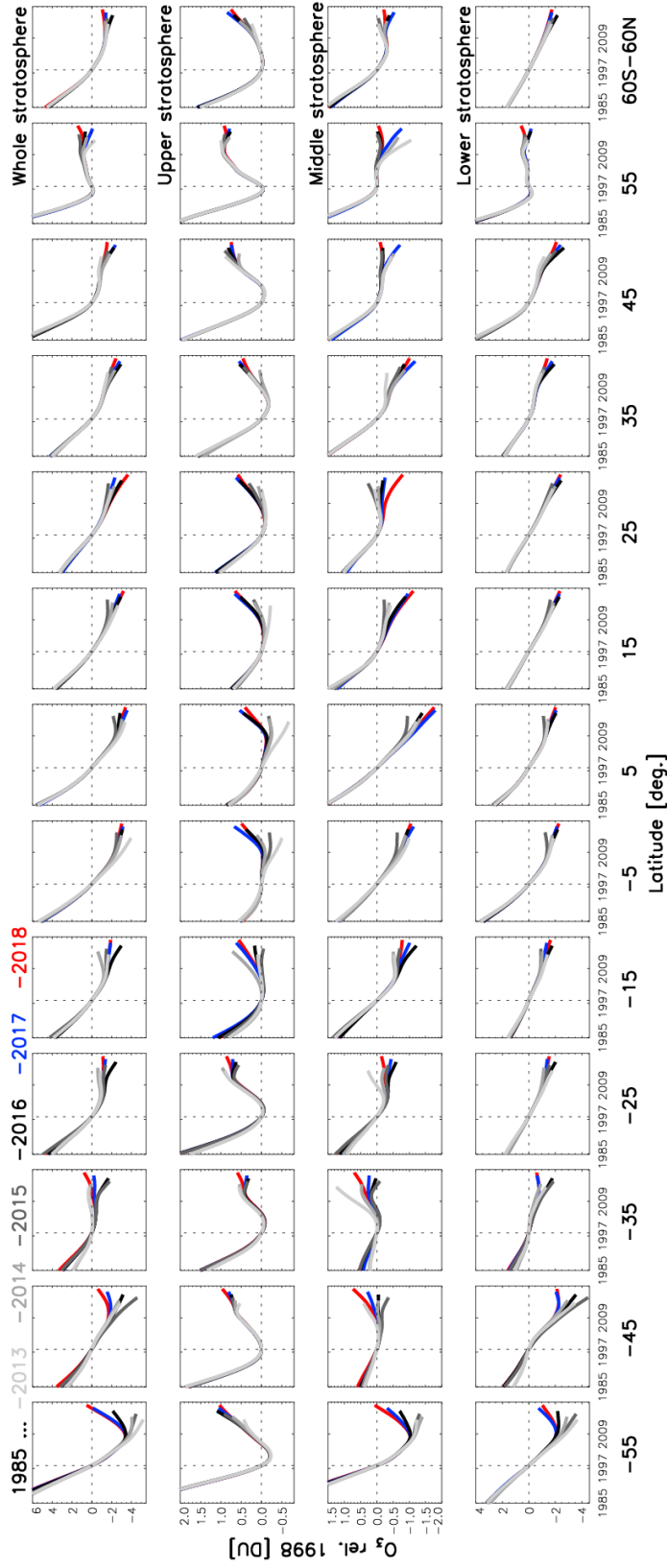


Figure 5: The partial column ozone non-linear trends estimated as a function of end year (2013 to 2018; dark to light colours), for each 10° latitude and quasi-global (left to right) and (top to bottom) the whole, upper, middle and lower stratosphere. Each sub-panel covers 1985–2018 and all curves are bias corrected to January 1998 (horizontal and vertical dotted lines). Uncertainties for each 1998–end-year change are given in Fig. 6.

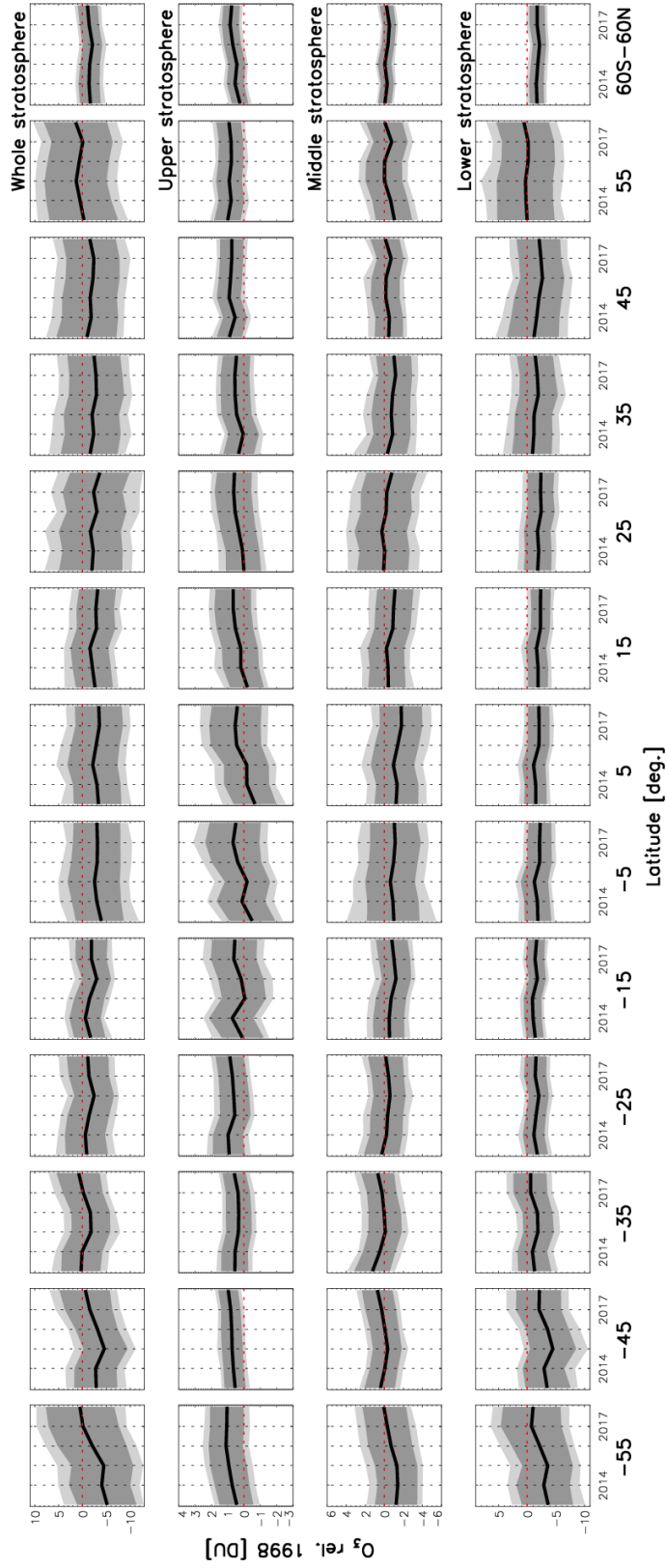


Figure 6: The partial column ozone changes between 1998 and the end-year from 2013 to 2018 (x-axis of each sub-panel) from the non-linear trends as in Fig. 5. Dark and light shading represent 95% and 99% credible intervals.



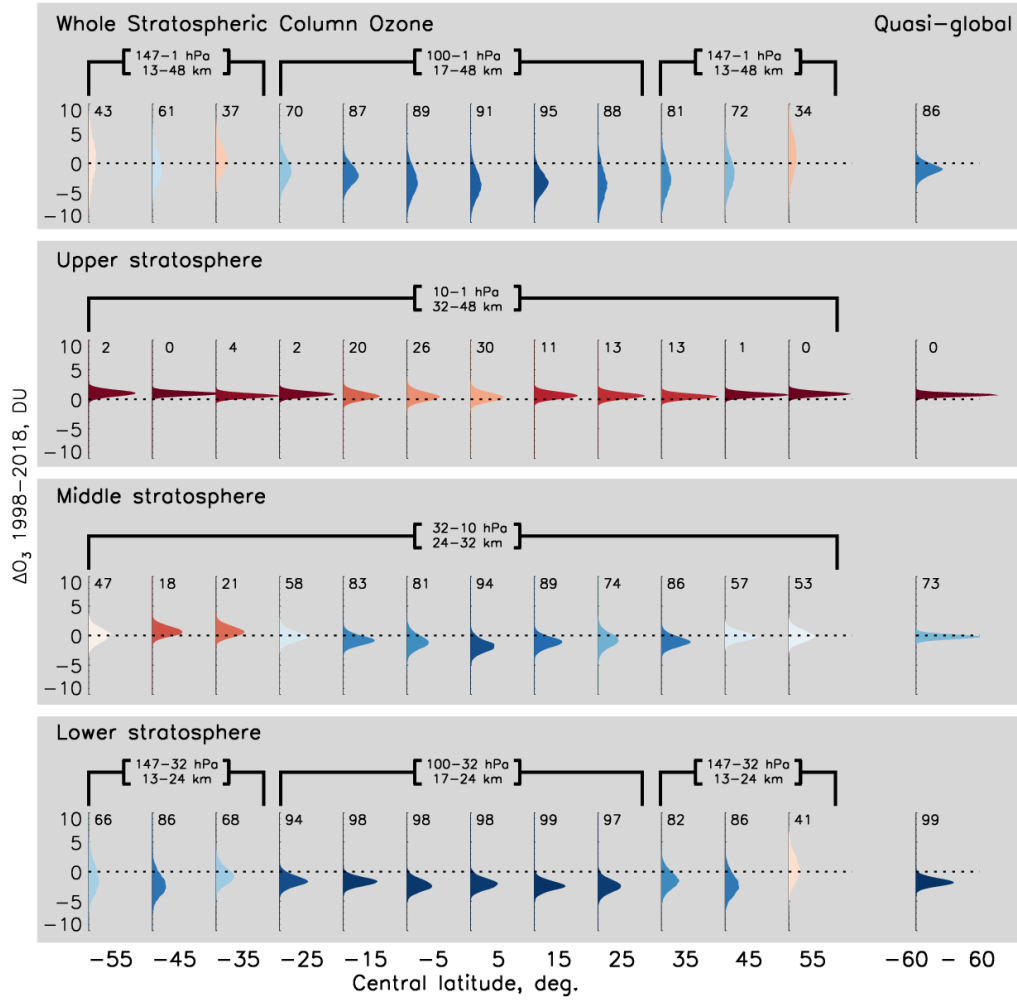


Figure 7: Posterior distributions (shaded) for the 1998–2018 partial column ozone changes. (Top) whole stratospheric column, (middle) upper and (bottom) lower stratosphere in 10° bands for all latitudes (left) and integrated from 60°S–60°N (‘Quasi-global’, right). The stratosphere extends deeper at mid-latitudes than equatorial (marked above each latitude). Numbers above each distribution represents the distribution-percentage that is negative; colours are graded relative to the percentage-distribution (positive, red-hues, with values <50; negative, blue).

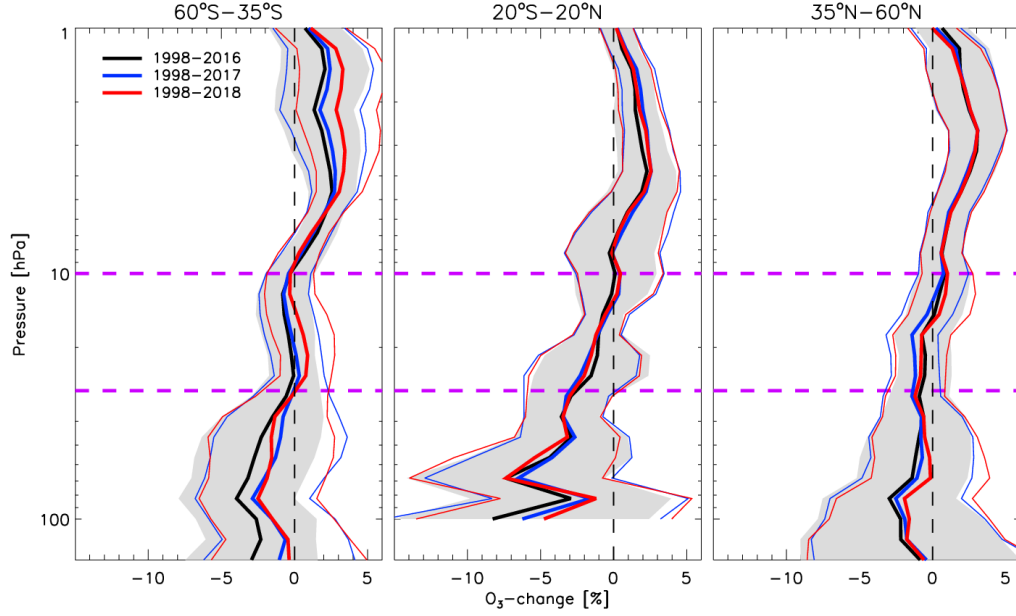


Figure 8: The ozone profiles for 1998 to an end-year of 2016 through 2018 (see legend) in the southern hemisphere (60°–35°S), the tropics (20°S–20°N), and the northern hemisphere (35°–60°N). Shading is for 2016 only. Uncertainties are 95% credible intervals. Pink lines indicate boundaries of partial columns in other figures.

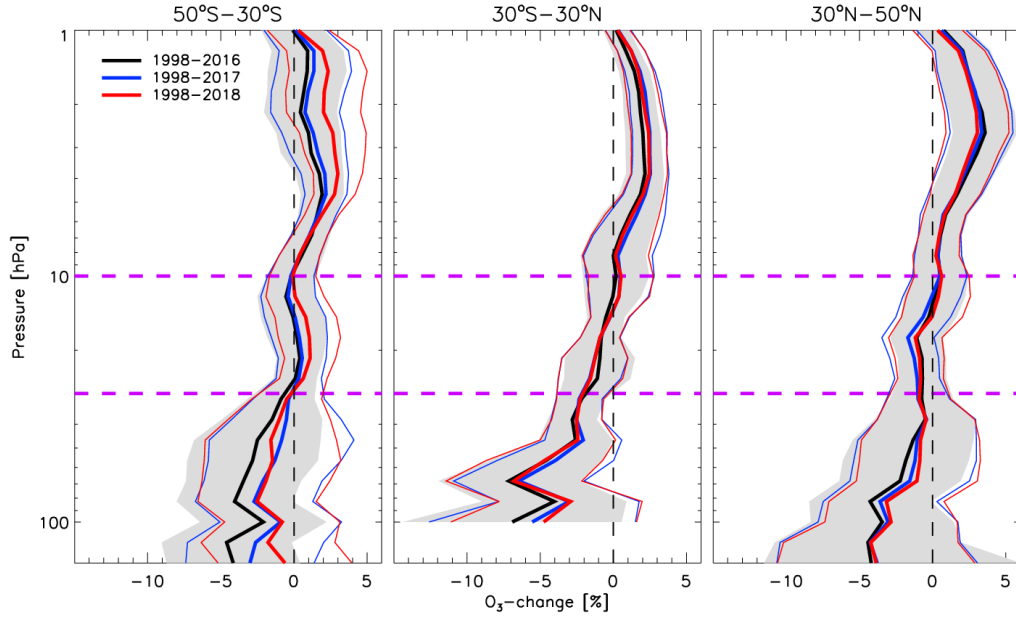


Figure 9: As for Fig. 8, but for the 50°S–30°S (SH), 30°S–30°N (tropics), 30°N–50°N (NH).