Response to Editor

Dear Editor Burrows, thank you for clearing our paper. We made a small revision to the paper and abstract in response to this last issue

> *Line 13: Reviewer 1 noted that "will be removed 20% faster than current projections" in the abstract is unclear, and I agree. In particular, the abstract text does not clarify which "current projections" are being referred to here. Please revise.*

In response to your request and that of the reviewer, we added a simple calculation and a figure in the Supplement that we should have done earlier. A section was added to the text in the last section:

"For example, if we extrapolate the observed growth in N2O burden over the last two decades (+2.9 %/decade), we reach 419 ppb by 2100, a value between RCP6.0 and RCP8.5. If we use a constant lifetime of 120 y to derive annual emissions and then re-project future N2O using a declining lifetime (-2.1 %/decade), this value drops to 391 ppb (see Fig. S2). Over the 21st Century, the N2O increase drops 27% from 103 ppb to 75 ppb, corresponding to a drop in effective radiative forcing of 0.09 W m-2, equivalent to about 6.6 ppm CO2 (Forster et al., 2021). Such differences are substantial when trying to tune our mitigation strategies to achieve climate change goals."

And the sentence in the abstract was edited:

" If the observed trends in lifetime and implied emissions continue, then the change in N_2O over the 21st Century will be 27% less than those projected with a fixed lifetime, and the impact on global warming and ozone depletion will be proportionately lessened."

Response to RC1

This is an incisive, thorough, and constructive review, thank you.

We have made adjustments or added material in response to almost all your comments. The quoted changes listed below were our original fixes. After incorporating changes in response to the other ACP reviewer and an independent reviewer, a final read-through has resulted in a number of editorial fixes and so the quotes below may be slightly different (but not in content). The additional review asked for the N2O budget tables (which come directly from these calculations) and so a short paragraph and supplemental table was added at the end.

> **RC 1**
>
> *This paper uses 16 years of MLS observations to infer a decreasing atmospheric lifetime of N2O. This is based on calculation of the N2O photochemical loss rates using a radiation scheme constrained by MLS temperature and ozone. The overall N2O loss rate derived from MLS data is increasing at a relatively faster rate than the total atmospheric N2O burden, hence the inferred decreasing lifetime.*
>
> *Within the details of the calculations, the calculated N2O loss frequencies are decreasing. In itself that would lead to a longer lifetime and relatively larger atmospheric N2O in the middle and lower atmosphere (due to vertical mixing).*
>
> *This is an interesting paper and suitable for ACP. The short paper is based on a simple, novel idea and is generally well written. However, I do find that some of the text is confusing and (maybe in the attempt to be concise) lacks some precision. I think the most interesting part of the paper is the use of N2O (a photochemically active species) to infer past circulation changes as an alternative to species like SF6. I think that some of the discussion tends to oversell the implications and ignore some other past work.*

Thanks for the excellent summary. We address and correct most of the issues you raise below; it has significantly improved the paper. Minor response here: The MLS dataset from Aug 2004 through Dec 2021 is 17+ years. The decrease in the loss frequency is small, and thus the much greater trend in the loss of N2O is due to higher N2O abundances. This is clearly identified with more rapid upward transport (mostly advection, not mixing) in the tropics rather than reduced mixing between tropics and extra-tropics. A paper submitted in parallel by Strahan to GRL shows that this increase in N2O is clearly modeled with their CTM using the MERRA-2 met fields. Since our paper was delayed, we can now add a short discussion and reference to that paper *(Strahan, S. E., Coy, L., Douglass, A. R., & Damon, M. R. (2022). Faster tropical upper stratospheric upwelling drives changes in ozone chemistry. GRL, 49, e2022GL101075. https://doi.org/10.1029/2022GL101075).*

> *My comments are below.*
>
> ***General Points***
>
> *1) MLS data. There are known issues with the MLS N2O data. These are mentioned in the results section (Line 156) but I think it would be good to add something upfront in the MLS data*

description section (Section 2). What do the uncertainties in the MLS data (esp N2O) imply for uncertainties in the derived lifetimes?

The issue of uncertainty in the MLS-derived lifetime was evaluated in Section 4 of the original paper that calculated the lifetime *(Prather, Hsu, DeLuca, Jackman, Oman, Douglass, Fleming, Strahan, Steenrod, Søvde, Isaksen, Froidevaux, and Funke (2015) Measuring and modeling the lifetime of nitrous oxide including its variability, J. Geophys. Res. Atmos., 120, 5693–5705. doi: 10.1002/2015JD023267.)*. We add a sentence here where the lifetime calculation is described (old line 105):

"Uncertainty in the absolute N2O lifetime is estimated at ±8 % (1-sigma), based primarily on the absolute calibration of MLS N2O and the chemical kinetics uncertainty, see Section 4 of P2015."

The bigger issue here regarding trends is the possible drift in calibration, but that is ruled out for these calculations by the excellent cross-calibration with ACE-FTS (Livesey et al., 2021). That paper is discussed, noting the specific figure in the paper.

2) The photolysis of N2O is temperature dependent due to T-dependent cross sections. Is this a factor in the decreasing J rate? I had to read the paper several times to try and get clear in my mind the implications of a decreasing J rate. This would lead to larger middle atmosphere N2O (as observed) but these increases would also mix down to the troposphere to give a similar overall rate of growth. Is that correct? If so I think this implication could be made clearer.

A very good point. You drove me to re-calculate the J-N2O values in the key-loss region 1-20 hPa. The ratio of J@300K to J@200K is consistently 1.50 through this altitude range. This translates into +0.4 %/K (ln(1.5)/100). Using Maycock (2018 GRL) temperature trends for this region over this period, -0.5 K/decade, results in -0.2 %/decade for J-N2O. We calculate -1.0 %/decade and thus the dominant effect is the increase in overhead O3 column as stated. We add the following discussion:

"Another source of declining J-N2O could be the reduction in its photolysis cross sections because of colder temperatures. We calculate that the relative change in J in this altitude region is +0.4 %/K, and combining with the observed temperature trend of about -0.5 K/decade (Maycock et al., 2018), we estimate the trend in J from temperature dependent cross sections is -0.2 %/decade and thus is a small contribution to the total. This trend in J of about -1 %/decade reduces the N2O loss and makes disagreement between the burden and loss trends even greater."

*3) The idea of an increased circulation removing CFCs more rapidly has been around for a while. I believe the original paper was: Butchart, N., Scaife, A. Removal of chlorofluorocarbons by increased mass exchange between the stratosphere and troposphere in a changing climate. Nature **410**, 799–802 (2001). https://doi.org/10.1038/35071047. This should be cited.*

Thank you, I missed the Butchart and Scaife paper, and this was an oversight. The only reason I can think of is that they talk about CFCs but their model has no chemistry in it. It would have been more in my view if they had run CFCs even with simplified chemistry in their climate model. For example, Hamilton and Fan *(2000, Effects of the stratospheric quasi-biennial oscillation* *on long-lived greenhouse gases in the troposphere, J. Geophys. Res.-Atmos., 105, 20581–20587)* is a core reference for me.

Butchart & Scaife are correct, were early to recognize this impact, and should have been included. What is spooky here is the prescient nature of their results: 3 %/dec increase in the 68 hPa tropical air mass flux is very close to the 2 %/dec found here for the decrease in N2O lifetime (CFC-12 should be the same). Our fix in the middle of second paragraph is to insert:

"Two decades ago this mechanism was proposed by Butchart and Scaife (2001) for the CFCs using dynamical diagnostics of the BDC from a climate model. Now we have observed and quantified this effect for a major greenhouse gas."

> *4) There is a message through the paper that this increasing BDC is beneficial for the environment. However, a main consequence of the increased BDC is smaller tropical column ozone and increased surface UV in a region which has a naturally small ozone column, high population and which has not yet experienced significant depletion. This impact needs to be mentioned. Also, even if N2O emissions are removed a bit faster in the future it will be a small effect and those new emissions will still have a negative impact on climate and ozone.*

We did not declare the enhanced BDC is beneficial for the environment but rather that it does produce a negative climate-N2O feedback. That is correct. If the N2O lifetime drops by 20%, then it does clear out the old N2O more quickly, and it also reduces the GWP for new emissions by the same amount. It has a fairly large impact because it affects the natural emissions. The issue is not that it makes N2O emissions 'safe' but rather reduces their climate and ozone impacts.

We already noted that enhanced BDC is expected to increase tropospheric ozone through increased stratosphere-troposphere exchange of ozone (a bad effect). That has been assessed recently. The issue of how tropical surface UV will respond to the enhanced BDC accompanied by climate change is 'known' as you say, but it is a more complex problem involving ozone changes throughout the atmosphere. It is beyond the scope of this work, we do not have the necessary data sets to calculate it.

> ***Other Specific Points***
>
> *Line 9. Abstract. I think that this should give the numbers for the decreased lifetime to show the magnitude of the effect.*

Done.

> *Lines 9-11. 'Because N2O abundances in the … shorter'. I don't think the logic of this sentence necessarily follows. What is sure is that the inferred lifetime is decreasing because the calculated atmospheric loss rate is increasing more rapidly than the estimated total atmospheric burden. If there was no circulation change but there was a decrease in the loss frequency (J), then that could also lead to relatively more N2O in parts of the middle atmosphere, couldn't it?*

We stand by this sentence as being correct and expressed as clearly as possible. As noted below, the lifetime is not 'inferred' but calculated based on the product of J and N2O. We do not understand the last part of your comment: if only J decreased, leaving more N2O, then the product loss is not going to increase faster than J decreased.

> *Line 13. 'will be removed 20% faster than current projections'. Where is this information from? I tried to follow this point highlighted in the abstract to the main text. The 20% figure appears to be from line 193, which seems to be a rough estimate of how much the lifetimes might decrease. If so, this seems very speculative to highlight in the abstract. It depends on the extrapolation of a short trend over a much longer timescale and models show that any circulation changes are very dependent on the GHG scenario used. Also, what are the 'current projections'? As stated in the paper, CCMs do predict this speeding up of the circulation so those projections include this effect. Do you mean simple estimates that employ a fixed present-day lifetime in budget calculations? This needs to be made clearer. This and the point below seem to be overselling the implications of this work.*

> *Line 15. 'negative feedback'. Is this really a negative feedback? It depends on what is behind the 'climate-driven' change in the BDC. How much contribution do N2O and CFCs make? I.e. to what extent is N2O affecting its own lifetime by this mechanism? If the circulation effect is mainly CO2 driven then does this affect the residence time of CO2? I.e. is there a negative feedback for CO2?*

Quantifying the feedback would require calculating the global warming per added kg of N2O and then scaling that to the BDC change. We do not want to try to quantify this. N2O is driving part of the global warming that drives the BDC change. That is enough to be able to make the statement.

> *Lines 25-26. 'Observational metrics for an enhanced BDC…but these observations run counter…'. This sentence is confusing. The reality is (I think) that models predict an enhanced BDC but the observations show an unchanged or decreasing BDC (with large uncertainty). The way that the sentence is constructed confuses this. The metrics are for a 'changing' BDC (either way). Also, please be specific with the sign of the model change.*

Correct. That is much better wording and we have revised using your suggestion.

> *Line 33. Need to insert some words. It is not the increased N2O which is shortening its lifetime, but the fact that that it is increasing in abundance leads to the inference that its lifetime is shortening. E.g. "… leads to the inference of a shorter lifetime…".*

There is some confusion here: the shorter lifetime is calculated based on the burden divided by the loss – it is not inferred. We have tried to make this clearer with a minor revision: *"…N2O increases through the middle tropical stratosphere, **relatively greater than the rate of tropospheric increases,** lead to a shorter lifetime…"*

> *Line 36. Consequences of an enhanced BDC. I think that a consequence that is often the focus is the decrease in tropical column ozone. This decrease would have important implications for tropical surface UV flux… I think it should be included in this paragraph.*

Tropical surface UV may be more important but this topic is beyond this paper. See discussion above.

Line 61. Missing word? "square root of the product of the values…"

Yes, corrected as suggested.

Line 64. The meaning of the word "lacking" is vague. It could mean missing or it could mean poor (lacking in quality). I assume that the meaning here is missing but please rewrite to be clear.

We changed it to "not reported". That should be clear.

Lines 62-64. Despite all the words I don't think that that latitudinal resolution of the gridded data is clear, nor what is done at latitudes beyond 86 degrees. I assume that there is no data so the profile from 86 degrees is extended to the pole. The start of this paragraph (line 50 onwards) should give the resolution of the starting gridded monthly MLS data.

We have introduced the MLS gridding at the beginning and have made clear how the weighted loss is calculated in the next paragraph: "*Each latitude profile is weighted by the area of the Earth ±2 ° latitude on either side except for latitudes 1 (90 °S-82 °S) and 43 (82 °N-90 °N).*" Thus the polar areas are included and need not be singled out.

Line 70. Say if the tropospheric values are constant in time.

We have revised this sentence: *"Below 100 hPa, we add 6 pressure layers for the purpose of the chemical model integration with typical tropospheric values (320 ppt); however, these layers have negligible impact on N_2O loss because very little of the critical ultraviolet radiation reaches below 100 hPa."*

Lines 72-73. 'atomic oxygen radical'. I would suggest referring to O(1D) as 'electronically excited atomic oxygen'.

Yes, that is better. Corrected.

Line 100. Conversion factor 4.78 Tg/ppbv. If I understand correctly this figure relates the global mean surface N2O vmr to the total N2O in the atmosphere. There must be the assumption of the stratospheric profile shape in the region where N2O is lost photochemically. Any circulation changes (or loss rate changes) would change this profile shape and could affect this conversion. How big is that effect? Some comment on this should be added.

The reference given (Prather et al., 2012) does a thorough evaluation of the possible uncertainties in calculating lifetimes, including the 'fill factor' that you note above. It is not based on assumptions but on the observed stratospheric and tropospheric distributions. The changes discussed here do not significantly change the fill factor.

Line 126 'The cause of the lifetime trends can be … an increase in the abundance of N2O'. Here the language is imprecise. The cause of the lifetime trend is either J rate changes or circulation *changes. The increase in N2O is a consequence of that which leads to the diagnosis of a decreased lifetime. Please rewrite for clarity/accuracy.*

We are not sure that the reviewer is not reading too much into the word "cause" here. Even the J rate change is "caused" by something else. We have revised to avoid a misunderstanding: *"The <u>immediate</u> cause of the lifetime trends <u>(i.e., enhanced N2O loss)</u> can be a change in the photochemical loss frequency or the abundance of N2O."*

Line 134. What about the effect of stratospheric cooling on T-dependent N2O cross sections? This should be mentioned and quantified (if only to show that it is not a large factor).

Yes, as noted in your main points above, we examined this and found it small. Text now reads: *"Another source of declining J-N2O could be the reduction in its photolysis cross sections because of colder temperatures. We calculate that the relative change in J in this altitude region is +0.4 %/K, and combining with the observed temperature trend of about -0.5 K/decade (Maycock et al., 2018), we estimate the trend in J from temperature dependent cross sections is -0.2 %/decade and thus a small contribution to the total. This trend in J of about -1 %/decade reduces the N2O loss and makes disagreement between the burden and loss trends even greater."*

Line 177. Satellite observations of what? This paper is using satellite observations of N2O to get the opposite result. The reader won't necessarily know the nuances here.

Yes, corrected to: *"satellite observations of AoA tracers…"*

Line 199. 'straightforward diagnostic'. Models have the option of including simple AoA tracers which are a much more direct diagnostic of a changing circulation. The lifetimes are clearly more complicated as they include changes in overhead ozone, temperature etc. The SPARC lifetime report did include estimates of lifetime changes in 2100. Those results are in Chipperfield et al. (2014), which should be mentioned. The modelled changes in lifetimes by 2100 were complicated to unpick (and having had a quick look at the paper are not 20%).

I agree that knowing what the lifetime changes are would be useful (as a lifetime) but the diagnostic should not be oversold as a way to understand models.

*Chipperfield, M.P., Q. Liang, S.E. Strahan, O. Morgenstern, S.S. Dhomse, N.L. Abraham, A.T. Archibald, S. Bekki, P. Braesicke, G. Di Genova, E.L. Fleming, S.C. Hardiman, D. Iachetti, C.H. Jackman, D.E. Kinnison, M. Marchand, G. Pitari, J.A. Pyle, E. Rozanov, A. Stenke and F. Tummon, Multi-model estimates of atmospheric lifetimes of long-lived Ozone-Depleting Substances: Present and future, J. Geophys. Res., **119**, 2555-2573, doi:10.1002/2013JD021097, 2014.*

This is also an important reference that we missed. A new section is added*: "Chipperfield et al. (2014) analyzed the change in CFC and N2O atmospheric lifetimes with climate change (year 2100 versus 2000). For the five chemistry-climate models that calculated the N2O lifetime, results were ambiguous: 2 increased, 1 decreased, and 2 were unchanged, all with absolute changes less than 0.5 %/decade. These results are not necessarily inconsistent with the projections here because the modeled scenarios included other chemical changes in the* *stratosphere and adopted a middling climate scenario (RCP 4.5). Chipperfield et al. also found that AoA metrics were a poor predictor of N2O lifetime (see their Fig. 8-9). "*

> *Line 201. 'wrinkle' is very colloquial and probably confusing to the non-native speakers. I would suggest keeping the language clear and simple.*

OK, have changed to 'oddity' which translates more cleanly.

> *Line 204. Typo? proportionally?*

Correct, and corrected.

Response to RC 2

Thank you very much for a concise and most useful review. It has helped us re-focus on the key points and hopefully made it easier for the reader.

We have made adjustments or added material in response to almost all your comments. The quoted changes listed below were our original fixes. After incorporating changes in response to the other ACP reviewer and an independent reviewer, a final editorial read-through has resulted in a number of editorial fixes and so the quotes below may be slightly different (but not in content). The added review asked for the N2O budget tables (which come directly from these calculations) and so a short paragraph and supplemental table was add at the end.

> RC2
>
> This is a concise, generally well-presented analysis of the changing N2O stratospheric sink and lifetime. I appreciated the brevity but also think some of the sections could be expanded a bit to improve clarity. The analysis has interesting and important implications, including for climate change-driven changes in the BDC and for the clearance of ozone-depleting substances from the stratosphere. I recommend publication with some minor revisions.

Thank you, and thanks for the most helpful suggestions.

> Abstract
>
> The second sentence repeats part of the first sentence and is also a bit awkward, since "it" has no clear singular antecedent. Perhaps rewrite as,
>
> "This [DECREASE] is occurring because the N2O abundances in the middle tropical stratosphere, where N2O is photochemically destroyed, ARE increasING at a faster rate than the bulk N2O in the lower atmosphere."

Thank you, great sentence. It is adopted with minor edits noted above.

> The result that the N2O lifetime is decreasing, even despite the reduced photolysis coefficient in the upper stratosphere due to ozone recovery, seems like an important concept to include in the abstract, since it lends further support to the importance of the increased BDC in driving the decline in N2O lifetime.

OK, we added a clause to the sentence following the one above: *"Because the chemical loss frequency of N2O in the critical region is decreasing,"*

> The projection to 2100 is quantitative (20% increase) but somewhat speculative, while only a qualitative statement is made about the 2005-2021 trend, even though the calculations over that period are solidly grounded in data. It might be better to include a quantitative estimate of the 2005-2021 trend.

We have added the rate of decrease (-2.1 ±1.2 %/decade) to the first sentence. Good point.

> Line 15, I would suggest removing "but relatively minor" since it seems to belittle the findings of this paper and also is not really developed in the body of the manuscript. In general, I think a stronger concluding sentence, which sums up the important implications of this work, would serve the Abstract better.

OK, removed the qualifier, but could not come up with a stronger concluding sentence.

Other comments

Line 26, "but these observations run counter to the climate model projections (Karpechko et al., 2018; Abalos et al., 2021; Garney et al., 2022)" Can the authors spell out more clearly what the observations are showing for those less familiar with this literature, e.g., are the SF6 observations suggesting no change in the BDC, or are they showing a slower BDC?

Yes, that was obscure. We changed it based on RC1 to *"While models predict an enhanced BDC, the SF6 observations indicate an unchanged or decreasing BDC, but with large uncertainty (...refs"*

Line 59, space => spaced

Done.

Line 63, should the second 86 be 84? Otherwise, it doesn't make sense that the dataset extends from 84N to 84S.

This section on data set latitudes was confusing and has been rewritten in this paragraph and the next.

Lines 87-94, This argument for the minimal impact of the solar cycle impact might belong in the Results/Discussion rather than the Methods, e.g., grouped together with discussion of other uncertainties like calibration drifts.

Good idea, we moved it to the end of Section 3.

Line 101, please clarify which source files have changed.

P2015 used the GOZCARDS data, 5 degree latitude bins. (noted)

Line 170, Please include a summary statement to wrap up this paragraph. As currently written, the impact of possible calibration drifts is not clear. Since the conclusions of this paper depend in large part on the relative trends in N2O in the middle/upper stratosphere vs. lower in the atmosphere, it seems important to leave the reader with a clear statement.

Good idea. We added: *"Thus, any calibration drift that impacts the lifetime (i.e., occurring in the critical region 3 to 30 hPa and 30°S to 30°N) is negligible compared to the increasing trend in N_2O loss (+5.0 %/decade), or it is slightly negative, which if corrected would further increase the trend."*

Line 204, proportion => proportionally

Done.

Paragraph starting at line 201, This is an interesting side note. Would this change be detected in a decrease in the NOy/N2O tracer correlation slope in the lower stratosphere? e.g., as discussed in Nevison et al. (1999), GBC, 13, 737-742.

Very interesting. We added this thought and reference to the section. Further, we added a brief summary of the overlapping work on the MLS trends in N2O by Strahan that was co submitted and just appeared in GRL. We have added a short paragraph highlighting that study and its overlap with ours.

Strahan, S. E., Coy, L., Douglass, A. R., & Damon, M. R. (2022). Faster tropical upper stratospheric upwelling drives changes in ozone chemistry. Geophysical Research Letters, 49, e2022GL101075. https://doi.org/10.1029/2022GL101075