Atmospheric Chemistry and Physics Discussions

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Comment on acp-2022-650

Anonymous Referee #1

Referee comment on "Observed changes in stratospheric circulation: decreasing lifetime of N_2O, 2005–2021" by Michael J. Prather et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-650-RC1, 2022

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.

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?

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.

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.

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.

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.

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?

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?

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.

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…”.

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.

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

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.

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.

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

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

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.

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.

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).

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.

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.

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

Line 204. Typo? proportionally?