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Comment on acp-2021-635

Anonymous Referee #2

Referee comment on "From the middle stratosphere to the surface, using nitrous oxide to constrain the stratosphere–troposphere exchange of ozone" by Daniel J. Ruiz and Michael J. Prather, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-635-RC2, 2021

This paper suggests a set of metrics based on observations to test stratosphere-troposphere exchange (STE) of ozone and other tracers in models, which is a relevant topic given the importance of STE for the tropospheric burden of these species and implications for climate radiative forcing. While there are some promising results in this paper, presentation of these should be improved before I can recommend this study for publication. See detailed comments below.

Major comments

Methodology is not explained in a way that readers can easily follow and should be introduced in a section separate from the results. At least for the model and observational details used. You reference surface observations for the first time in L278 only. Details of how to generate the metrics may remain in the main body of the manuscript. Particularly, the paragraph starting at L109 introduces a new and rather complex methodology that is not substantiated. I suggest adding some figures to illustrate how the gradients of these tracers and their scaled versions across the tropopause in the end look like. This is important because I cannot easily assess if some of the surprising results you find are due to this tracer scaling (see below comments). The e90 tropopause is not that widely applied and should be explained in further detail. How would your results change if using a different tropopause? This information again is important if you want your metrics be used by the wider modelling community who may not have the e90 tropopause implemented in their models. When calculating global mean STE do you apply a latitude-weighting? If so, please add to the methodology. This would allow the reader to better judge the realism of the approach.

The results should also be better placed into context of the already published literature. Discussion of whether your results confirm previous estimates would strengthen your results and provide support for your methodology and the suggested metrics table. See detailed comments below.

Minor comments

Abstract: The abstract should be improved (I guess mostly a language issue) and state clearer and self-containing results that don't require reading the whole manuscript to understand. For example, '*The STE flux of O3, however, is predominantly northern hemispheric, but observational constraints show that this is only caused by the Antarctic ozone hole.*' I know what you mean but it's not written with a clear logic, the Antarctic ozone hole doesn't affect the NH STE flux directly, just its relative magnitude. Also you say '*we show that metrics founded on observations can better constrain the STE O3 ….*' better than what?

L35 To call stratospheric ozone influx is *driving* climate change and surface air pollution is overstated, since certainly fossil fuel emissions are the main cause for these. Statement should be weakened. Some references added to provide justification.

L36-37 Add citations to Lelieveld and Dentener (2000) and Lamarque and Hess (1999). Also, what is a regular seasonal cycle? I would argue that regularity is not maintained at a longer timescales where ozone depletion has affected the N2O-O3 relationship.

L45-46 Statement needs to be backed up with references of how N2O-O3 relationships were used for model-observation metrics.

L92-93 This sounds exaggerated to say '*this method is extremely robust*' without providing the basis for the statement. Also, I would like to see a more critical discussion and some added caveats of the method used. After all, a CTM is likely not a sufficiently sophisticated tool to be used for the investigation of the seasonal cycles at the surface, given that N2O has strong sources from soils that show large geographical and seasonal variations (see Butterbach-Bahl et al 2013).

L129-130 Please provide references supporting the statement, or was this meant to be describe your methodology? Again, the methodology should be better separated from your results.

L133-135 Please put your results into context with previous literature. The ozone estimate seems at the lower end of the range indicated by earlier observations studies (Murphy and Fahey 1994; Gettelman et al 1997) and modelling studies (Young et al., 2013; Stevenson et al 2006; Hegglin and Shepherd 2009; Kawase et al 2011)

L137 this explanation is not clear to me, that is why the budget of cF11 is about twice as large as that of cN2O. If it is destroyed faster, shouldn't it be smaller not larger? Or since it is a budget that balances loss and sources, it may be illogical to make this comparison to begin with since it is determined by the realism of the tracers in your model?

L140-144 Please put your results into context with previous literature. Sprenger and Wernli 2003 and Skerlak et al 2014 have illustrated that the subtropics are the main places of where stratosphere-to-troposphere transport happens and need to be referred to as well.

L146 There is no causal link between the small tropical fluxes to the interhemispheric asymmetry of the NH and SH, which this sentence seems to imply. Improve language.

L189-191 Why would O3 photochemical destruction only reach down to 16 km?

L235-137 Please improve language.

L239-240 This is not correct. There is ample evidence from aircraft observations that polar vortex air mass processing and mixing into the LMS after polar vortex break-up is observed in trace gas distributions also in the NH.

L241-242 This is another limitation of the methodology applied that should be mentioned already in the methods section.

L252-254 Don't you build in this tight correlation through your scaling of the tracers? Note your methodology in how you scale the two tracers was not 100% clear to me so I hope you could improve on the description in the methods section.

L258-259 It would be good to specify here that this is only true for these long-lived trace gases.

L341-350 I really struggle with the logic here. Soil emissions do have a seasonality and thus I cannot see how your model results want to imply that there is no seasonality. Again, I am suspicious that the scaling you apply to your tracers is responsible for this result rather than this to reflect what happens in the real atmosphere.

L352-361 These results seem to be in contradiction with Lamarque and Hess 1999 and also a recent study by Williams et al. 2019, who used a stratospheric ozone tracer to investigate STE impact on surface ozone seasonality. Please compare your results to these studies and provide a discussion of where the differences may come from.

L373-376 Do you not contradict yourself here with your earlier result that interannual variability driven mostly by the QBO leads to surprisingly similar amplitudes in the NH and SH?

L400-435 Hegglin and Shepherd (2007) have used ACE-FTS O3/N2O correlations in an extensive comparison to a CCM, so should be cited here. This study reveals how sampling issues can affect the interpretation of tracer-tracer correlations using ozone. In particular, the ACE-FTS instrument exhibits a strong sampling bias with unequal sampling of seasons and hemispheres. Undersampling the full correlation space (since your monthly ACE-FTS data will not have sampled all latitudes in your considered latitude range evenly) is likely to impact your results. The differences (or even apparent agreement!) in the slopes between observations and your model may thus be at least partially explained by this sampling bias. A discussion of the limitation of your approach should thus be added.

L441-445 This is a well-known feature and existing references should be added and discussed to highlight this (Lamarque and Hess 1999; Williams et al. 2019; Hegglin and Shepherd 2009)

L468-475 This evaluation does not provide much depth since it doesn't clearly link total column ozone to STE (and to provide metrics for STE was the main goal of your paper I thought). Your statement '*is clearly somehow connected*' expresses this weakness. I would like to see how it is connected otherwise I suggest to remove this section from the paper. You may introduce this differently, that is start with this evaluation since a model must represent the right ozone distribution to get STE ozone right?

L493-494 This is not a result of your study and should be attributed to literature that have discussed this (Meul et al. 2018; Hegglin and Shepherd 2009; Zeng et al. 2010)

L507 This table should include earlier references who suggest the same metrics as tests for model STE.

Additional references:

Butterbach-Bahl, K., Baggs, E.M., Dannenmann, M., Kiese, R. and Zechmeister-Boltenstern, S., 2013. Nitrous oxide emissions from soils: how well do we understand the processes and their controls?. Philosophical Transactions of the Royal Society B: Biological Sciences, 368(1621), p.20130122.

Hegglin, M. I. and Shepherd, T. G., 2009, Large climate-induced changes in ultraviolet index and stratosphere-to-troposphere ozone flux, Nat. Geosci., 2, 687–691, https://doi.org/10.1038/NGEO604.

Hegglin, M.I. and Shepherd, T.G., 2007. O3â□□N2O correlations from the Atmospheric Chemistry Experiment: Revisiting a diagnostic of transport and chemistry in the stratosphere. Journal of Geophysical Research: Atmospheres, 112(D19).

Kawase, H., Nagashima, T., Sudo, K. and Nozawa, T., 2011. Future changes in tropospheric ozone under Representative Concentration Pathways (RCPs). Geophysical Research Letters, 38(5).

Lelieveld, J. and Dentener, F. J., 2000, What controls tropospheric ozone?, J. Geophys. Res.-Atmos., 105, https://doi.org/10.1029/1999JD901011.

Meul, S., Langematz, U., Kröger, P., Oberländer-Hayn, S. and Jöckel, P., 2018. Future changes in the stratosphere-to-troposphere ozone mass flux and the contribution from climate change and ozone recovery. Atmospheric Chemistry and Physics, 18(10), pp.7721-7738.

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Williams, R.S., Hegglin, M.I., Kerridge, B.J., Jöckel, P., Latter, B.G. and Plummer, D.A., 2019. Characterising the seasonal and geographical variability in tropospheric ozone, stratospheric influence and recent changes. Atmospheric Chemistry and Physics, 19(6), pp.3589-3620.

Zeng, G., Morgenstern, O., Braesicke, P. and Pyle, J.A., 2010. Impact of stratospheric ozone recovery on tropospheric ozone and its budget. Geophysical Research Letters, 37(9).