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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Stratospheric impact on tropospheric ozone variability and trends: 1990–2009" *by* P. G. Hess and R. Zbinden

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The authors wish to thank anonymous reviewer #3 for their in-depth review and their constructive comments. (I will identify reviewers comments in the reply below as C, my reply as R)

C: This important paper shows strong evidence that the stratosphere is a major source of large-scale trends and variability in northern hemisphere tropospheric ozone. The paper is important not least because this is a controversial topic, with divergent views across the research community. Perhaps I am being a little unfair, but these views are often due to vested interests in measurements from particular sites, or measurement by particular techniques, or indeed model results from a particular model (as here). In this paper, ozone measurements from a wide range of platforms (sondes,

aircraft - MOZAIC, and surface sites) across mid- to high- northern latitudes are used in a consistent manner. Model simulations are from a single model: the Community Atmosphere Model with chemistry, CAM-chem. In general, I find that the relatively good agreement between the model and the observations provides quite compelling evidence that the stratosphere does exert a strong influence on tropospheric ozone. Where caveats exist, the authors have generally pointed to them. However, I would like the authors to address in a little more detail two important aspects.

The first is how the model simulates stratospheric ozone (using Synoz) and stratosphere-troposphere exchange (STE). The authors acknowledge that Synoz is relatively simple parameterization, and do check for the influence of any spin-up issues in Section 3. But given that the source of stratospheric ozone is crucial to the whole paper, some more details of the mechanism, including the interannual variability in STE and its location, and how reasonable this is, should be provided.

R: The stratospheric tracer synthetic tracer (Synoz) is described in McLinden et al. (2000). Synoz is a passive ozone-like tracer released into the equatorial stratospheric ozone production region (in our simulations defined between 10 and 70 hPa and 30 S – 30 N) at a rate equivalent to the cross-tropopause flux of ozone (specified in our simulations as 500 Tg/year). In our simulations below 500 hPa Synoz is relaxed to 25 ppbv with a timescale of 2 days. Ozone is set equal to Synoz above the tropopause ensuring the stratosphere to troposphere flux of ozone is equal to that of Synoz. At steady-state the cross-tropopause flux of Synoz is equal to its specified production rate (i.e. 500 Tg/year). Synoz has been used for many years in tropospheric chemical models with a high degree of success. For example Synoz was used in the GEOS-chem model until linearized ozone chemistry (LINOZ, see McLinden et al. 2000) was introduced in a beta version 24 February, 2010.

One of the advantages using Synoz as specified in McLinden et al (2000) is that the cross-tropopause flux of ozone is not sensitive to details of the stratospheric simulations. It is also not sensitive to interannual changes in stratospheric circulation. Be-

cause the production rate of Synoz production is fixed, as the stratospheric mean meridional circulation increases (decreases) an airmass spends less (more) time in the region where Synoz production is specified. This implies that as the circulation increases (decreases) the concentration of Synoz transported out of the equatorial source region (approximately equal to the amount of time that an airmass within the equatorial source region) decreases (increases). Thus, the flux of Synoz transported out of the equatorial source region (roughly proportional to the strength of the mean meridional circulation times the concentration of Synoz) is rather insensitive to changes in circulation strength.

In reality, as the stratospheric mean meridional circulation increases, ozone transport out of the ozone production region should also increase. Specifying the concentration of a Synoz like tracer (we will denote this tracer Synoz*) within the equatorial production region (instead of its production) implies that its flux out of the equatorial source region will be sensitive to the strength of the circulation; an increase (decrease) in the stratospheric meridional mass flux will increase (decrease) the transport of Synoz*. This is what we have done in this paper. The parameterization used in this paper was implemented as follows. (1) First we equilibrate the concentration of Synoz (McLinden et al„ 2000) by running the model on the order of 10 years. (2) The equilibrated concentration of Synoz in the defined equatorial region (30 S – 30 N between and 10 and 70 hPa) is saved during a test-year producing an annual record of Synoz* concentrations within the defined equatorial region within that year. (3) We re-run the test year, but instead of specifying the production rate of Synoz we specify concentrations of Synoz* within the equatorial production region (obtained from step 2). We check that the two methods of specifying Synoz produce the same result during the given test year, and indeed they do. In other words the simulated concentrations of Synoz are very similar to those of Synoz*. (4) We use the specified concentrations of Synoz* within the equatorial region during all subsequent years. As stated above this allows us to parameterize the impact of the interannual variability of the stratospheric circulation on ozone while still retaining the Synoz methodology. The stratospheric-tropospheric flux of ozone during the test

year should be very close to 500 Tg/year. The stratospheric-tropospheric flux of ozone during subsequent years using Synoz* should respond to changes in the strength of the simulated stratospheric Brewer-Dobson circulation in comparison to the test year.

C: The second aspect is the role of seasonality. All of the analysis is based on 12-month running averages, so any seasonality is removed. This may well be a sensible approach, but it is well known that STE has quite strong seasonality, as does tropospheric chemistry, and that measured ozone trends differ between seasons. Some comment on what could be gleaned from further examination of seasonality would be appreciated.

R: We agree that an analysis that takes seasonality into account would be very valuable. However, since we got such a strong signal without taking seasonality into account we did not do an extensive analysis of the seasonality of the signal. We intend to repeat the simulations in a model that resolves stratospheric dynamics and chemistry and will look in more detail at the seasonality of the trends in the new simulation. In the new supplement we investigate the seasonality of the ozone signal in the model and in the data as suggested by Jennifer Logan (see our reply to Jennifer).

C: My final comment is that although the paper is well written, it is overly long (it took me many sittings to get through it). There is undoubtedly a great deal of important information and analysis, but I think it could be written more concisely, and this would make it more accessible to the wider audience that it deserves. I strongly support its publication in ACP, given due attention to these comments and the specific ones listed below.

R: Following the suggestions of the reviewers the authors will extensively rewrite and condense the paper. We will present the data first followed by the modeling details (at the suggestion of Jennifer Logan). We will also include a number of additional tables to summarize the results and thus make the text easier to read. Following the 1st reviewers suggestions we will integrate section 5.1 and 3.3 to avoid a mid-paper sum-

mary. We also will simply the abstract to make it easier to read. In addition following the recommendation of reviewer 1 we will move the section discussing the equilibration of the model to the appendix and remove most of the paragraph in which we discuss components of the Mace Head ozone trend.

P22720

C: L19-24: Values are quoted for measured trends (necessarily) averaged over specific sites, but then for simulated trends over whole regions. It is then noted that the simulated trends differ when sampled at the measurement sites. I think it would be more sensible to quote the directly comparable values, and then additionally mention that the simulated values for the whole region differ from the simulated values sampled at the measurement sites.

R: We agree with this comment and follow this recommendation in the revised version.

C: P22721 (and throughout): It is Mace Head, not Macehead!

R: Thank you.

C: P22722âĂĺ Should trends be variability?âĂĺ

P22723âĂĺ

C: 1% per year.

R: We put the trends in terms percentage change per year because they often seem to be expressed that way in the literature.

C: L10: the net ozone response to climate change. Although the Stevenson et al (2006) study included 26 models, only a subset (10) simulated the impact of future climate change.

R: Thank you. This will be noted in the revised version.

C: L13: from -> between

R: Thank you. Fixed.

P22724

C: L10: clarify what is meant by 'net ozone flux' – presumably the net stratosphere to troposphere ozone flux.

R: Thank you. That is correct.

P22726

C: L21: analyses (and also next page, l4)

R: Thank you.

P22727

C: L10: I think it would be clearer to say 'higher underlying ozone' than 'more ozone'.

R: Thank you.

C: L12-18: Is 'regionally robust' equivalent to 'sufficiently determined'. I suggest for clarity, just use one term throughout.

R: Thank you. We did imply the same meaning in the text.

P22728

C: L2: similar to

R: Thank you.

C: L10-11: It is not that useful to know that CAM-chem has been compared to measurements – it would be more useful to know if it compared well (ideally some sort of quantitative measure).

R: Agreed. We believe the model provides good simulations of tropospheric ozone, and is certainly on a par with other global models. But this is all rather qualitative. The

interested reader can also look up the comparisons in the various papers. It is difficult to sum up the results from these various papers using a few quantitative numbers. Additional comparisons with measurements are given in the new supplement (see Figures attached to the review by Jennifer Logan).

C: L14-16: Reword this sentence. L17: has -> haveâĂĺ

R: Thank you.

P22729

C: L4-19: What is the interannual variability in STE simulated using Synoz? Does Synoz simulate STE in the correct locations? Given the crucial nature of simulating STE for the main conclusion of the paper, more information about exactly how Synoz is implemented is required here.

R: See the description of the implementation of Synoz above. As detailed in the expanded supplement to our revised paper (see figures accompanying our response to Jennifer Logan), and within the paper itself (Table 2) the methodology adopted here produces reasonable simulations of tropospheric ozone and its variability. In addition the STE (Stratospheric Tropospheric Exchange) of ozone using this parameterization is only a measure of changes in the stratospheric circulation and not of changes in stratospheric chemistry. This allows us to document the importance of circulation changes. The Synoz methodology does a good job capturing the location and variability of ozone due to the NAO – much of the ozone variability due to the NAO is due to STE and is captured in a model simulation using Synoz (Hess and Lamarque, 1997). We did not produce precise estimates of the interannual variability of STE – only that of the ozone in the troposphere transported through STE. In planned future simulations using a model with a resolved stratosphere and troposphere we will more precisely determine variability in the STE itself and its location.

C: L25: north of 30; delete concentration.

R: Thank you.

C: P22730âĂĺ L9: north of 30.

R: Thank you. âĂĺ C: P22732 L7: observational

R: Thank you.

C: P22733âĂĺ L22: delete the.

R: Thank you.

P22736

C: L1-12: It is interesting that Central Europe seems more variable than other regions – could this reflect a greater anthropogenic influence?

R: It is not clear what is going on in Central Europe. A recent analysis of European measurements submitted by Jennifer Logan to JGR should help better explicate trends and variability over Central Europe.

P22737

C: L23: overall.

R: Thank you.

P22738

C: L1: the overall

R: Thank you.

C: L29: I'm not sure 'unfortunately' is the correct word here – it is just the way the Japanese measurements are.

R: Thank you.

P22739

C: I note there is some discussion of the simulations in Section 4, which is about observations. I recommend just keep it to observations, and save this discussion for later.

R: We agree. As discussed above we will substantially rewrite the paper.

P22741

C: L4: Whilst strictly the 'interannual variability of emissions is constant', it is probably more usefully said to be 'zero'.

R: Thanks.

C: L10: Minor style suggestion – here you use 'R=0.64' etc. whereas earlier you write 'correlation of 0.64'. Keep it consistent.

R: Thanks.

P22743

C: L23-24: minima

R: Thanks.

C: P22761: Figure 1 Clarify that '12-month running mean . . . plotted as the annual average of monthly devi- ations' is what you refer to in the main text as 'AAMD'.

R: Thanks.

C: P22764: Figure 4

It may be worth mentioning that the mean curves in Figure 4 are the same as the individual curves in Figure 3b. I wondered if it might make sense to reverse the order of these two figures.

R: We agree it would make sense to reverse these figures and to explicitly point out their relation.

P22770: Figure 10

C: The lower panel should not be entitled 'Anthropogenic ozone' – tropospheric is better – i.e. it must include ozone produced from natural sources in the troposphere.

R: Thanks.

REFERENCES

Hess, P. G., and J.-F. Lamarque (2007), Ozone source attribution and its modulation by the Arctic oscillation during the spring months, J. Geophys. Res., 112, D11303, doi:10.1029/2006JD007557.

McLinden, C. A., Olsen, S. C., Hannegan, B., Wild, O., Prather, M. J., and Sundet, J.: Stratospheric ozone in 3-D models: A simple chemistry and the cross-tropopause flux, J. Geophys. Res., 105, 14653–14665, 2000.

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