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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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 the reviewer for their time in reviewing the manuscript and for their comments. (I will identify reviewers comments in the reply below asC, my reply as R)

C: This article needs to be greatly shortened and re-organized for clarity before publication.

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

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reviewers suggestions we will integrate section 5.1 and 3.3 to avoid a mid-paper summary. 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.

C: Further research is needed to convince the scientific community that the simulated ozone is credible. The problem is that the authors claimed to use Synoz but tweaked Synoz inappropriately by prescribing the concentration in the source region. It is likely that the simulated stratospheric ozone is way off and the STE fluxes are not realistic. The authors should either use Linoz (McLinden et al., 2000) to redo the simulation or validate the simulated ozone (e.g. zonal mean latitude-height distributions) and overhead column ozone distributions by comparing to the observations. There are plenty of satellite data (TOMS, MLS, ...) with global coverage out there that the authors can use to validate the basic profiles of the simulated ozone.

R: We respectfully disagree that we tweaked Synoz inappropriately. However, we do agree with the overall consensus of the reviewers that our methodology needs a better explanation that we include in the paper. 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.

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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. Because 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

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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.

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 reviewer suggests that "It is likely that the simulated stratospheric ozone is way off and the STE fluxes are not realistic", but does not support these claims. In fact the model shows very little bias against the measurements (Table 2), except at the surface where the simulated ozone overestimates the measurements. This is a common model bias (e.g., see Pozzoli et al., 2011) and errors in the stratospheric ozone flux are unlikely to be the cause. Also note the correlations between the simulated and observed ozone tend to be high. In light of the comments from Reviewer #2 and those of Jennifer Logan we will present additional model-measurement comparisons in the paper's supplement where we show the simulated seasonal cycle versus measurements and the simulated vertical ozone profile against measurements. We believe that the model simulation is reasonable and credible. The new figures to be included in the paper and give below are: (i) the 3-month smoothed monthly mean ozone timeseries over the key measurement regions at 500 hPa including Canada, the Eastern US, Northern Europe and Central Europe); (ii) the average annual cycle of ozone in the model versus measurements over the key measurement regions at 500 hPa; (iii) the vertical profile of ozone versus the measurements in the key measurement regions. We did not examine Japan as we do not utilize the tropospheric measurements there.

The results presented in these figures suggest that the simulations produce a satisfactory simulation of tropospheric ozone.The results shown here are very similar to those given by Hess and Lamarque (2007) using the MOZART-2 chemical transport model. This is notable as the simulation reported in Hess and Lamarque (2007) used Synoz as specified in McLinden et al. (2000). The amplitude of the simulated seasonal cycle at 500 hPa is less than observed (Figures 1-4). The average simulated ozone maximum is generally one month too late (two months over the Eastern US). Simulated ozone is generally significantly overestimated at the surface (except over Northern Europe) with small overestimates throughout most of the rest of the troposphere. This bias in ozone is common to many global model simulations (Pazzoli et al., 2011; Ellingsen et al., 2008).

We agree that a comparison with satellite data would be fruitful, but there is no equivalent derived product at 500hpa or at 150hpa or at the surface with detailed profiles over the record examined. Satellite data give limited vertical information on tropospheric ozone profiles and have limited ability to capture STE events (Tang et al., 2011). In addition, satellites that do give vertical information do not provide data over the length of record we were able to analyze. We could possibly compare with tropospheric ozone column, but cannot compare with total column, as we do not have a realistic stratospheric ozone column. Retrievals of unbiased tropospheric ozone column measurements are also difficult due to biases with respect to clouds, due to the sun synchronous observations. In summary we believe that a comparison with satellite data would be valuable, but it would necessarily be part of another study.

C: This article mainly focuses on the trend in time series. Why isn't there any discussion on the simulated downward trend since 2003 for tropospheric ozone in Figure 1. I find it perplexing that the annual NOx emission is held fixed throughout the simulation, yet there is a clear downward trend in tropospheric ozone.

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R: We show a downward trend in tropospheric ozone concentrations north of 30 N since 2003. There may be a number of explanations for this trend besides emission trends including: (1) changes in ozone production efficiency, (2) changes in the lightning distribution, (3) changes in the net ozone flux into or out of the region north of 30 N. Unfortunately we did not output the necessary variables to complete a full analyses of this trend. However, we should add, changes in tropospheric ozone are not the focus of this paper. We have focused on changes in ozone trends and changes in stratospheric ozone. Changes in ozone trends can be observed by available measurements and changes in the stratospheric component can be inferred to some extent by correlations with 150 hPa ozone. We know of no observational analogue to changes in the tropospheric component of ozone. While it would be interesting to pursue the cause of these changes in more detail we believe it is outside the scope of this paper (which as the reviewer suggests is long enough). Nevertheless we will comment on this downward trend and its possible causes in more detail in the revised paper.

C: If the upward stratospheric ozone trend seen in the simulation is indeed due to the strengthening of Brewer-Dobson circulation, isn't it relatively straightforward to verify it by plotting the time series of the downward mass fluxes of the BDC (defined by the residual circulation) from the meteorological data?

R: While this might be a valuable exercise we do not believe it is straightforward. The change in ozone we obtained is an integrated effect of changes in the BDC and possibly changes in stratospheric mixing. The change is likely due the changes in the BDC and mixing processes as stratospheric air is brought downwards by the BDC. Changes in the BDC are not linked in a straightforward manner to changes in the ozone flux. Is it the integrated change in BDC over the N.H. that is important for ozone flux, or the change over a range of latitudes? And at what time of year is it important to examine this change? This is an interesting topic, but we believe it is outside the scope of the present paper.

C: Going back to point (1), I find this article very difficult to read through because it is

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very wordy and lack of focus. I believe that the authors can and should do a lot better job on presentation. In this regard, the authors can benefit from the other reviewer's comments.

R: Please see comments to point (1) above.

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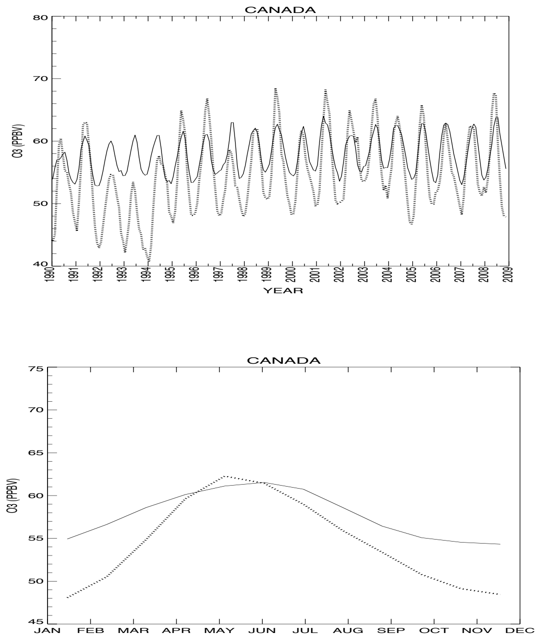

Figure 1. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the six Canadian ozonesonde stations. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 1.

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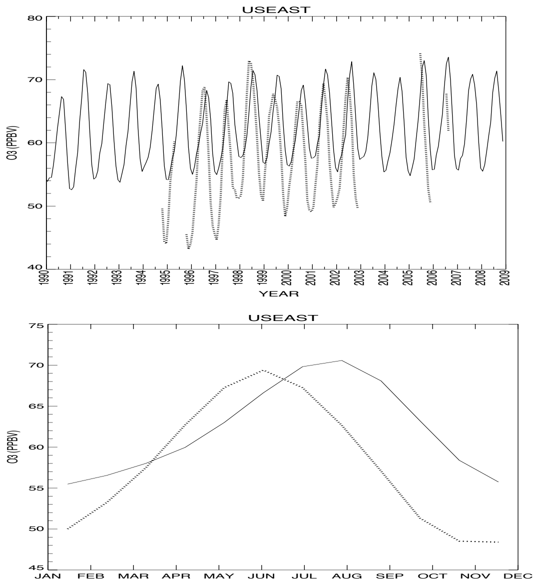

Figure 2. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the MOZAIC cluster and ozonesonde soundings for the Eastern US. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 2.

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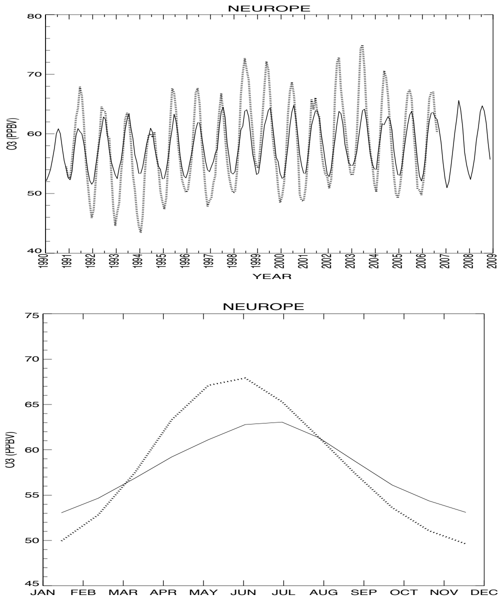

Figure 3. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the three ozonesonde soundings over Northern Europe. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 3.

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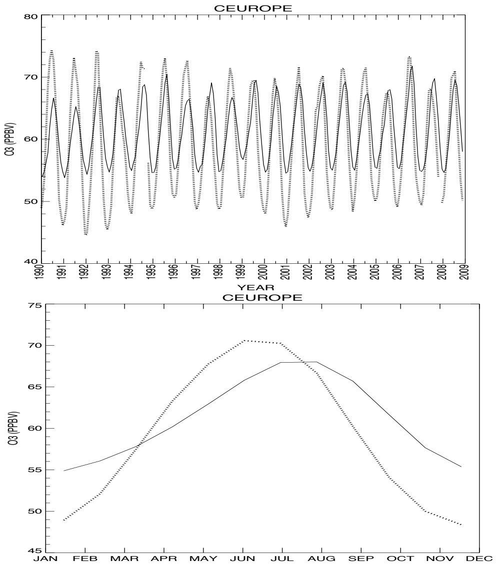

Figure 4. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the five ozonesonde soundings and MOZAIC cluster over Central Europe. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 4.

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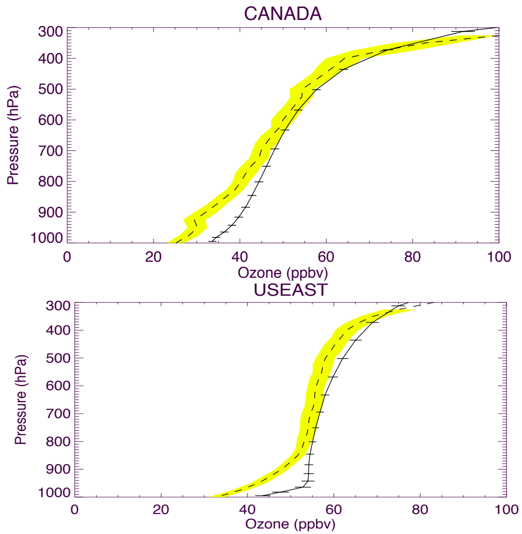

Figure 5. a) Average (1990 and 2009) simulated (dashed) and measured (dashed) vertical profiles of ozone (ppbv) over a) the six Canadian ozonesonde stations and b) the Eastern US ozonesonde station and MOZAIC cluster. Measured standard deviation (width of yellow shading) and simulated standard deviation (horizontal lines) is given as the standard deviation (±σ) of the average 12-month smoothed ozone data.

Fig. 5.

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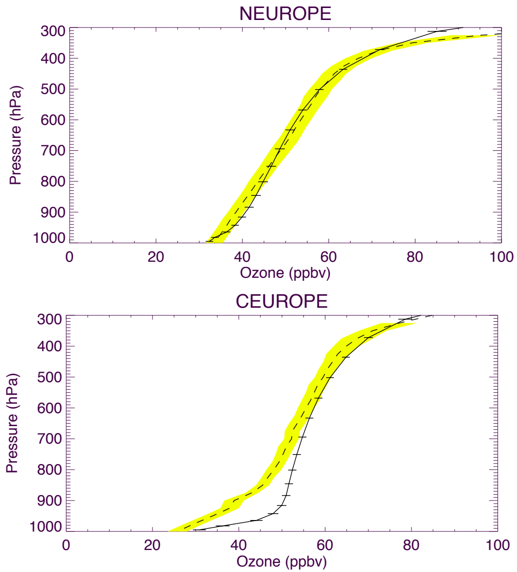

Figure 6. a) Average (1990 and 2009) simulated (dashed) and measured (dashed) vertical profiles of ozone (ppbv) over a) the three Northern European ozonesonde stations and b) the over the five ozonesonde soundings and MOZAIC over Central Europe. Measured standard deviation (width of yellow shading) and simulated standard deviation (horizontal lines) is given as the standard deviation (±σ) of the average 12-month smoothed ozone data.

Fig. 6.

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