

## ***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 take this opportunity to Jennifer Logan for her in-depth review of this paper and her many constructive comments. (I will identify reviewers comments in the reply below as C, my reply as R)

C: The data analysis is based on correlations of ozone at 150 hPa, in the lowermost stratosphere (LMS), and at 500 hPa. Previous studies have used such an approach, but did the correlations differently. Tarasick et al. (2005) used annual averages and found correlations of  $r=0.66$  for mean Canadian time series for both Arctic stations and for those at 53–59 N; Ordonez et al. (2007) found correlations for high altitude sites in Europe and sonde data at 150 hPa with  $r=0.77$  for 12 month running means; more importantly they showed that the correlations were largest in winter and spring ( $r=0.58$ -

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0.78) and were absent or not significant in summer and autumn. Terao et al. (2008) used 3 month running means of anomalies and found significant correlations for some Canadian and European sonde sites, and showed that the correlations were driven by the behavior in winter-spring.

R: Strong correlations between the stratospheric ozone and tropospheric ozone have been noted previously as summarized above by Jennifer Logan. One insight of this paper is to show that in many cases and on an interannual timescale these correlations are manifested on the large scale over many regions.

C: Given the strong seasonality in the last two studies cited by the authors, I do not understand why the present analysis relies solely on 12 month running means of monthly anomalies. It would be more interesting to examine the seasonality of the correlations, and see if it varies from region to region, given that some are more remote from anthropogenic emissions than others. We know that there is seasonality in STE and in its influence on tropospheric ozone, and this would be expected to manifest itself in the correlations. Ozone at 150 hPa is highly variable in winter and spring (the seasonal maximum), with much less variability in summer and autumn when ozone is much lower. Thus the interannual variability (IAV) in the 12 month running average anomalies is dominated by IAV in the seasonal maximum in spring. This is less of an issue at 500 hPa, when ozone is highest in summer, and the variability is not so dominated by one season. I expect that the correlations in this paper are driven mainly by stratospheric ozone in spring. I recommend that the authors examine the seasonality of the correlation by region.

R: We agree that an analysis that takes seasonality into account would be very valuable. However, we feel the study is long enough and prefer to leave seasonality to future work. Since the annual-averaged signal is strong we prefer to publish these results as presented. We intend to repeat the simulations in a model that resolves stratospheric dynamics and chemistry and will look in more detail at the question of seasonality (amongst other things).

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C: The overall time series at 150 hPa is formed from a mean for Canada (mean latitude 67 N), N. Europe (72 N), and central Europe (50 N), so it is for an overall mean latitude of 63 N. Given these relatively high latitudes, it is hardly a surprise that the three regions are highly correlated with each other. In the troposphere, a mean record is formed for Canada, N. Europe, and the eastern US (mean latitude ~40 N, but only ~12 years of data), so the mean latitude is 60 N or 70 N, depending if there is a robust data set for the eastern U.S. Clearly, the analysis of the overall record is for fairly high latitudes (~60 N). There is a huge difference in the behavior of ozone at 30 N and 60 N for both 150 hPa and for 500 hPa, when the monsoon season affects ozone at lower latitudes.

R: We are not sure that it is hardly a surprise that the three northern regions correlate with each other. In fact it is rather interesting that they do. It is true that the high latitude stations dominate the robust measurement signal we see. Nor do we disagree that the monsoon impacts 30 N, at least locally – e.g., over Japan. However, the modeled signal suggests, in fact, that the signal at the high latitude stations reflects the overall hemispheric averaged signal north of 30 N. In addition, the MOZAIC measurements (see supplement) suggest a high degree of correlation even at more southern latitudes (e.g., the Eastern US, Japan and Europe). Nevertheless, we agree with Jennifer that much of the signal we see is high latitude and will clearly state this in the revised manuscript.

C: The authors find that the correlation of their mean records at 150 and 500 hPa are highly significant, with  $r^2 = 0.7$ , and lower for the individual regions ( $r^2=0.42-0.56$ ) which can only be Canada and N. Europe (p22738, l8-12).

R: We correlated the mean records at 150 hPa (Canada, Northern European and Central European region) with: (i) the mean of the 500 hPa records (Canada, Northern European and Eastern US) and (ii) the Canadian, Northern European and Eastern US records separately. We will clarify this and put many of the relevant correlations into some tables to help clarify the text.

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C: (I hope this is what is meant, as on p. 22739 l. 6, it is said that the correlation is 0.4-0.5, and correlation means  $r$ , while explained variance is  $r^2$ ; clarify).

R: The correlations on 22739 are in reference to the correlations between the measurement regions at 500 hPa (e.g., between the 500 hPa Canadian region and the 500 hPa Eastern US region). Again this needs to be clarified and will likely be incorporated into a table.

C: A key question is the spatial extent to which this high correlation applies. As noted above, the mean latitude of the composite time series at 500 hPa is 60 N, with the eastern US data (40 N) showing similarities to the higher latitude time series in 1995-2002, but not thereafter. If this strong correlation for the mean time series applies for 50-90 N, it covers the northern 25% of the hemisphere, but it cannot be assumed to apply to 30-50 N on the basis of the results shown here.

R: This is correct. We state this uncertainty in the text (page 22738, line 16-19): “The measurements only sample the stratosphere and troposphere over a few distinct regions. To what extent are these regions and their correlation indicative of the simulated hemispheric-wide correlation (i.e., 30–90 N) between tropospheric ozone and its stratospheric component? Without additional data it is impossible to say definitively.”

We also note this in the conclusions “Outside these three regions we could not determine from the measurements alone whether the stratospheric ozone signal explains a significant fraction of the 500 hPa tropospheric ozone variability north of 30 N” .

Here the model simulation is of help. We state (22730, lines 23-19) “Ozone [where we are referring to simulated ozone] averaged regionally over the individual sites within a region (see Table 1) also gives a good representation of the overall variations in ozone north of 30 N at 500 hPa with the exception of Japan. The 500 hPa simulated ozone variability averaged north of 30 N explains only 32 % of the regional variability over the Japanese measurement sites; otherwise the overall averaged 500 hPa [simulated] ozone variations explain more than 77 % of the variability for the Canadian, Northern

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European and the US regions.”

In the revised text we will more clearly show the relationship between model results and the measurements and clearly state that the measurement sites only account for a relatively small fractional area of the hemisphere.

#### Model

C: The CTM in this study, CAM-chem, is driven by NCEP meteorology. It has a complete treatment of the troposphere in terms of emissions and chemistry, and keeps emissions the same each year. The model does not simulate stratospheric chemistry. Rather than using the SYNOZ approach, which assumes a constant ozone flux from the stratosphere each year, the authors say they specify the concentration of the “Synoz tracer” in the tropical stratosphere (details not given). It is far from clear how this works, or exactly how it was done.

R: The authors will include a more detailed description of how Synoz was used in these model simulations. This description is summarized here: 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

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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 air mass 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 air mass 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

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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: Since the key model results depend crucially on this aspect of the model (STE), the authors must evaluate their stratospheric tracer by comparison with observations. They must show time series of Synoz at 150 hPa, and also evaluate its vertical profile in the lower stratosphere over the sonde sites they use. The model study presented here has little validity if they do not find the model results for the LMS to be reasonable. These comparisons should be done on monthly mean ozone time series or anomalies (3 month smoothing is fine), not on 12 month smoothed anomaly time series. Their approach cannot be expected to capture the post-Pinatubo behavior, but the model should capture dynamical variability in the LMS in 1990-1991 and after 1995. If it doesn't, it corrupts the model results in the troposphere.

R: In the supplement of the revised paper and included below we show: (i) 3-month smoothed monthly mean ozone timeseries over the key measurement regions at 500 hPa (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.

These results show suggest that the simulations produce a satisfactory simulation of tropospheric ozone. These results 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

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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)

On the other hand we do not show the time series of Synoz at 150 hPa. The stratospheric tracer cannot be evaluated with comparisons with observations because there are no observations of Synoz. There is a long tradition of using Synoz in tropospheric models (including by GEOS-chem until a few years ago). Synoz is designed to capture the correct stratospheric-tropospheric exchange, but not necessarily the magnitude of the ozone in the lower stratosphere. While other more sophisticated approaches may be preferable, we do feel that Synoz is adequate for the purposes at hand. More sophisticated approaches will be used in forthcoming work.

C: The model results are scattered through the paper. In Section 3, they focus on average ozone north of 30 N; ozone for 50-90 N would make more sense, in the context of the data used.

R: Perhaps, although we show (from model data) that the stations analyzed (mostly above 50 N) capture the averaged signal from 30-90 N.

C: A method of tagging NO<sub>x</sub> (and hence ozone produced in the troposphere) is used to back out the stratospheric contribution to tropospheric ozone, and unfortunately, the model results are only shown as the 12 month running mean anomalies. The almost monotonic increase in ozone from 1990 to 1999 (Figure 1) suggests that the model has some issues with initialization, and certainly, the model does not show the relatively high values in 1990-91 seen in the observations.

R: We checked the initialization carefully. The simulation was initialized in 1987, over two years before the results are shown and from a previous simulation. Ozone in the troposphere has a rather short equilibration time and will easily equilibrate on this timescale. If there were issues with initialization then stratospheric ozone would keep

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drifting even if we cycle the meteorology (i.e., repeatedly use the same meteorology for multiple years). We checked this extensively (Figure 2) and show no sensitivity to initialization. Thus we conclude are not likely to be initialization issues.

C: The authors need to explain why the ozone produced from tropospheric chemistry (deduced from NO<sub>x</sub> tagging) has the time dependence shown in Figure 1 (an increase until 1998, then a decrease), even though emissions are constant.

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. 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. Nevertheless we will comment on this downward trend in more detail in the revised paper.

C: It is hard to know how well this model simulates extra-tropical ozone when the only comparisons shown (in Section 5) are with 12 month running mean anomalies. Does the model get the amplitude of the seasonal cycle right at 500 hPa? Many models do not.

R: We show the seasonal cycle at 500 hPa in the supplement (see Figures below). The model trends to simulate the 500 hPa ozone maximum about a month after the observed maximum.

C: The discussion of the tests of the "Synoz" tracer (which isn't really SYNOZ in the McLinden et al. definition) is confusing. It is well known that the age of air in the lower stratosphere is ~4 years, so how can the model equilibrate at ~150 hPa in a year or

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two, unless its circulation is too fast?

R: Please see the detailed explanation above with regards to Synoz. The initial Synoz distribution is taken from many years of model simulation and is thus fully equilibrated. Stratospheric ozone will equilibrate within the troposphere on the tropospheric photochemical timescale.

C: The circulation in a CTM with NCEP winds will not necessarily be the same as that in the parent GCM.

R: It should be extremely close. We use the same winds, model levels and other meteorological parameters.

C: The statement is made in the Conclusions (p. 22746, 110-12) that "Stratospheric ozone is parameterized in these simulations assuming no interannual variability in the stratospheric ozone concentrations." Is this really true? Surely it is true only in the source region (30 N-30 S)? Clarify.

R: Thank you. This is poorly worded. There is no variability due to chemistry. The input concentration of Synoz changes on a monthly timescale in the source region, but not interannually. Outside the source region Synoz will change interannually.

Model/data comparisons

C: In Section 5 the model and data are compared only in terms of the AAMD time series, and only for 1996 onwards for correlations. The model does badly prior to 1996. Comparisons to data (absolute values) are made only in Table 2. Biases are generally small, but the reader should be shown whether the model matches the observed seasonal cycle.

R: The seasonal cycle will be given in the new supplement. Please see Figures at the end of this reply.

C: Figure 7 that compares the time series (as AAMD) is so small it is hard to discern

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details. The panels should be expanded lengthwise (say 4 stacked panels, page width).

R: Thank you. We will do this.

C: Given my concerns about the model treatment of STE, I do not have much confidence in the fraction of the variability in the troposphere attributed to the stratosphere. For the data, the fraction is appropriate for highly averaged and highly smoothed mid-high latitude data as noted above.

R: We agree that the paper examines large time-scales and large-spatial scales. We intend to repeat the simulation with a model that simulates stratospheric dynamics and chemistry to obtain a better-simulated estimate of the interannual variability due to the stratosphere. That said, the current model simulation significantly underestimates the measured interannual variability – we do not simulate full extent of the ozone increase during 1999 and we do not simulate the ozone minimum associated with Pinatubo.

C: The model is also compared to 4 surface sites that vary considerably (Figure 5 and 8), so it would make more sense to show them individually in Figure 8.

R: Thank you for the suggestion. This makes sense.

C: Figure 7 shows that the model does best at capturing the behavior of ozone from 1997 to 2000, namely the relatively large increase from 1997 to 1999 followed by lower ozone in 2000 in Canada, northern Europe and the eastern U.S. Since this is the largest signal in IAV from 1996 to 2008, it likely that this period drives most of the correlation between the model and data. It is certainly of great interest to know what caused this signal in the atmosphere, which is apparent from Mace Head to the high Arctic sites, so I would encourage the authors to explore this further, including a seasonal analysis.

R: We do not disagree with this statement. We are currently working to understand this anomaly in more detail and its global distribution in a more sophisticated model that includes a more resolved stratospheric circulation and stratospheric chemistry.

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C: I think the authors have quite a bit of work to do before this paper is ready for publication, but I urge the authors to do it. They must address the reviewers' concerns (including mine) about their treatment of "Synoz".

R: The treatment of Synoz is described in greater detail above

Other comments:

General:

C: The paper is written so it jumps between data analysis and model results in a confusing way. It would make more sense to present all the data analysis first, then the model results and evaluation with data.

R: We agree with this and will rewrite the paper to present the data first.

C: The nomenclature "annual average monthly deviations" (AAMD) is confusing, as there are not 12 annual averages in a year.

R: Thanks. We will change the notation.

C: Using the AAMD, instead of say the real annual average, increases "n" in the correlation analysis by a factor of 12. It will thus increase the significance of any correlation present in the annual means.

R: We assumed one degree of freedom per year so that the correlations will be equivalent to those using a simple annual average instead of a smoothed average.

C: In terms of presentation, it would make more sense to show and discuss Figure 4-5 before Figure 3.

R: Agreed. We will swap the order.

C: What are the units on Figures 4-8? ppb? Figure 1 is in standard deviations, most others are not labeled.

R: These figures are also plotted in terms of standard deviation. We will make sure to

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label the axis.

C: Keep a consistent color for the model in all plots. I am not repeating comments by other reviewers on correct names of stations etc.

R: Thank you.

C: 22725, 13. This study used only BM data at the central European sites. This means that it does not use the generally more reliable ECC data at Payerne. Jeannot et al. (2007) showed that the Payerne BM data are unreliable in the early 1990s. I recently submitted a paper to J. Geophys. Res. (September, 2011, copy sent to Hess) showing problem with the Payerne and Hohenpeissenberg BM data in the mid-1990s (and with the latter in the early 1990s), and showing that the data after 1998 are more reliable, based on comparisons with high altitude sites and MOZAIC data. This work was presented at the workshop in early April 2011 in Toulouse, "Tropospheric ozone changes: Observations, state of understanding, and model performances", attended by Zbinden and Hess.

R: We thank Jennifer Logan for sending us a copy of her submitted paper and for helping to sort out the rather confusing record of long-term ozone measurements over Europe. The Logan et al. submitted paper is very relevant for this work and we will certainly reference. As stated in the text we used the BM data at the central European sites because we wanted to analyze consistent records at each station. Switching from BC to ECC can clearly introduce an inconsistency in the data. The ECC data do not cover a sufficiently long period to use in the long-term analysis presented in this paper.

C: 22726, 3-10. Give the range of MOZAIC profiles for all 3 clusters, and note that the data are extremely sparse (often less than a few days per month) in 2005-2007 for the eastern U.S. and for some periods for Japan.

R: We are not sure what you mean by the range of the MOZAIC profiles. However, we will note that during some periods the data is extremely sparse.

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C: 22726, 22. Some analyses are done with normalized AAMD, in which the time series are normalized by "the standard deviation of the time series". Do you mean the standard deviation of the 12 month running mean anomalies, or that of the actual monthly mean time series? Clarify.

R: We will clarify this in the text. The normalization is with the standard deviation of the 12 month running mean anomalies.

C: 22726, 13-14. Do you mean that the AAMD for the sites have to be correlated with each other? This section should refer to the Tables S1 and S2, and make clear if all pairs have to be significantly correlated with each other in a cluster.

R: Yes we mean the AAMD must be correlated. Thanks for the suggestions. We will incorporate them.

C: 22732. Discussion of Japanese sites. It is not surprising that these sites are so different from each other, they span from mid-latitudes to sub-tropics, while all other sites are in mid to high latitudes.

R: We will make sure to mention this. Thank you.

C: 22733, 19. Be specific. The increase is clearly after 1993, after the post-Pinatubo minimum in the winter of 1992/93.

R: Will do.

C: 22734, 19. The pronounced dip is in 1992-94, not 1991-95. Why is the Boulder record not used? It is available from the NOAA/ESRL web-site.

R: The Boulder record is incomplete on the WDOUC website. We obtained ozonesonde data from WDOUC because of the common file formats from WDOUC and the fact that data is fairly well described. We thank Jennifer for pointing out that the NOAA/ESRL site does have more data from Boulder, although frankly the data on the site is poorly described.

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C: 22735, 1: Utilized is just a long word for used in this paper. Please change “utilize” to “use” throughout the paper.

R: Thank you.

C: 22735-22726. See my comments above on problems with the European sonde data in the early 1990s. Also, note that the Zugspitze data seem more reliable in the early 1990s than the Jungfraujoch data.

R: We will reference this work.

C: 22739, 14-6. Does this refer to the model? to the correlation among regions at 500 hPa? to the correlation between 150 and 500 hPa?  $r=0.4-0.5$  means  $r^2 = 0.2-0.25$ , pretty small.

R: These low correlations refer to the correlation of the 500 hPa ozone signal between different regions (Eastern US, Canadian and Northern European). The simulated correlations in the 500 hPa signal between these regions is 0.79-0.95. We intend to put much of this information in tables.

C: 22740, 1 5-6. Explain why you omit 1991-1995, and why you think there was a large effect on ozone at 500 hPa. The effect of Pinatubo in the stratosphere was first evident in extra-tropical ozone in the stratosphere in the winter of 1992-93, according to the WMO assessments. Figure 7 shows the model does not do well in 1990-1992, so including these years would reduce the correlations.

R: We did include 1990 in the correlations. We agree that we should also include 1991 in the revised version. There is a clear dip in the simulated 500 hPa ozone signal in the model during this period with a strong correlation to the 150 hPa ozone signal. While we have not proved this is due to Pinatubo we believe it is likely. Attributing the tropospheric changes to Pinatubo also concurs with the analysis of Oltmans et al. (1998)

C: 22740, 1. 12-13. Of course the low values in 1992-2009 will impact trends that start  
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in 1990 and are for periods as short as 10 and 20 years.

R: We will assess the importance of this in the revised paper.

C: Table 2. What does standard deviation refer to in this table? Does “Annually averaged ozone” have one value per year or 12? Similarly, is the correlation based on one value per year or 12? There cannot be trends for the eastern US starting in 1990, as you show only data from 1995.

R: We will clarify these points in the table. We should have said “12-month smoothed ozone” was used to construct the statistics. Standard deviation, correlations (with 1 degree of freedom/year) and trends use the smoothed ozone. We will note the trends in the Eastern US begin in 1995 (Thank you).

#### REFERENCES FOR REPLIES

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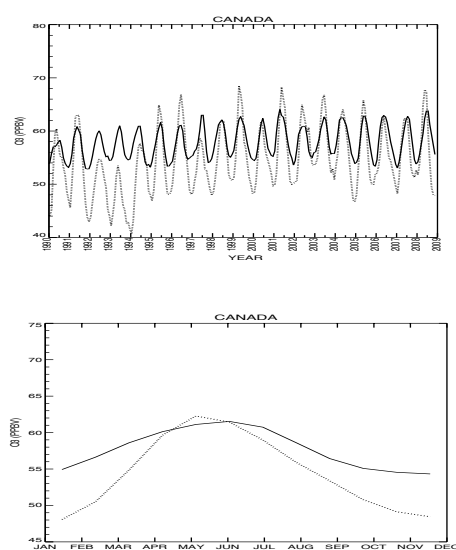


Figure 1. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the six Canadian ozonesonde stations. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 1.

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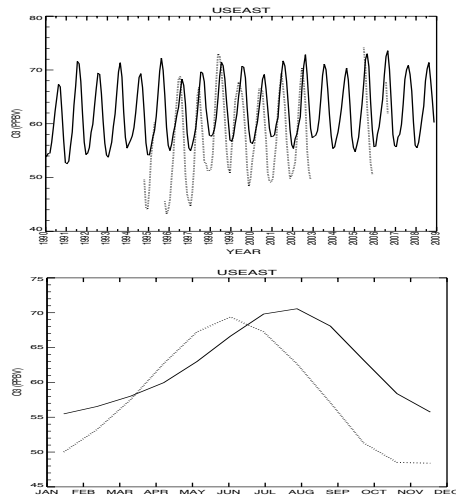


Figure 2. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the MOZAIC cluster and ozonesonde soundings for the Eastern US. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 2.

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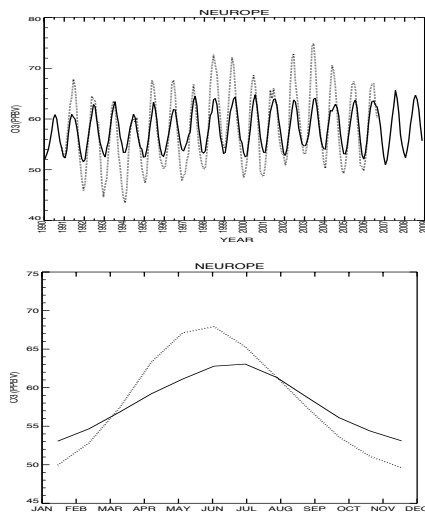


Figure 3. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the three ozonesonde soundings over Northern Europe. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 3.

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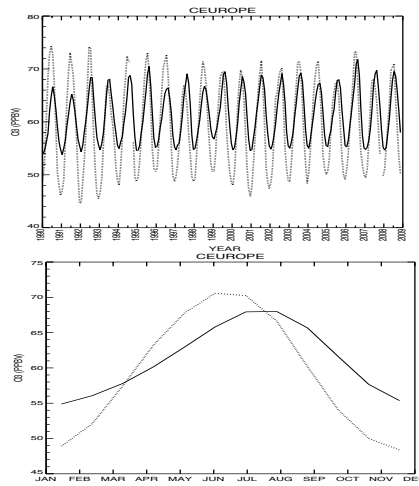


Figure 4. a) Simulated (solid) and measured (dashed) ozone (ppbv) at 500 hPa averaged over the five ozonesonde soundings and MOZAIC cluster over Central Europe. Ozone is smoothed over 3 months; b) Average seasonal cycle of simulated (solid) and measured (dashed) ozone from (a).

Fig. 4.

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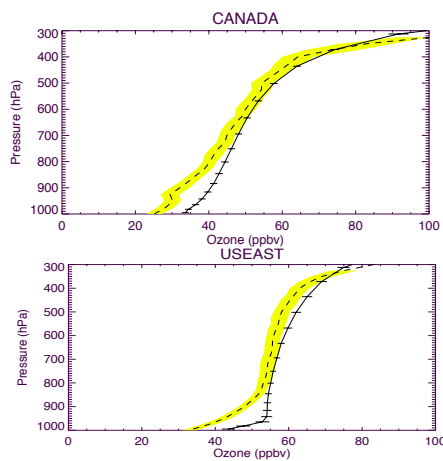


Figure 5. a) Average (1990 and 2009) simulated (dashed) and measured (dashed) vertical profiles of ozone (ppbv) over a) the six Canadian ozonesonde stations and b) the Eastern US ozonesonde station and MOZAIC cluster. Measured standard deviation (width of yellow shading) and simulated standard deviation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

Fig. 5.

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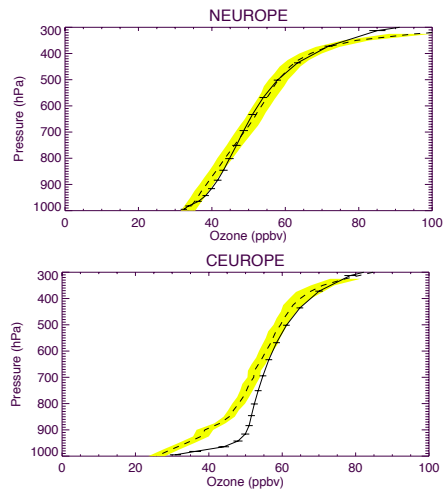


Figure 6. a) Average (1990 and 2009) simulated (dashed) and measured (dashed) vertical profiles of ozone (ppbv) over a) the three Northern European ozonesonde stations and b) the over the five ozonesonde soundings and MOZAIC over Central Europe. Measured standard deviation (width of yellow shading) and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

**Fig. 6.**