Atmospheric Chemistry and Physics Discussions

Interactive Comment

Interactive comment on "Modelling the global tropospheric ozone budget: exploring the variability in current models" *by* O. Wild

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I am very grateful to the reviewer for their encouraging comments and for their thorough reading of the paper. Many of the issues raised occur where I have attempted to be as succinct as possible in my description, but where clarity has been lost as a result. I have addressed each of the points raised, and have altered the text wherever appropriate to address the reviewers concerns. I believe that this has improved the paper, and I am grateful to the reviewer for their time and effort in pointing out where improvements could be made.

Response to Comments:

How large are the differences among models? For instance, later you give percent changes due to lightning NO_x and resolution – how large are these changes relative to

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the spread of models? The difference between models is addressed in a quantitative manner in the first section of the paper and in Table 1. However, the relative size of the sensitivity changes compared with the spread of the models was not clearly quantified, and this has now been addressed in the body of the paper.

You could cut "cross-tropopause transport...between models". There is some degree of repetition here, but I believe it is helpful to identify the processes that this study focuses on before quantifying the largest effects. However, I have removed some repetition by deleting "to the differences seen between models" at the end of this sentence.

By "model chemistry" are you referring just to the chemical mechanism? I have rephrased this sentence as "chemical mechanisms and dynamical schemes". I would note here that differences arise from solution techniques as well as from the chemical reaction sets and kinetic data used. Although differences in meteorology clearly make a significant contribution, it is important to note that how these data are used in models (e.g., advection and convection schemes) also contribute to differences.

Give some measure of the uncertainty or range in budget estimates. The 1σ variability is given explicitly in Table 1, but I have now also included it here.

Add "net" before "chemistry". I have replaced "chemistry" with "net production" to clarify this.

Why have NO_x emissions increased over time? What exactly was prescribed in AC-CENT? Higher estimates largely reflect improved understanding of sources and emission factors, and this is now stated explicitly. The real increase in the emission rate between 1990 and 2000 is only about 1–2 TgN/yr over the decade. Anthropogenic emissions were prescribed in the ACCENT studies; natural emissions were "recommended", but were permitted to differ where they were interactive with model vegetation and meteorology. The emissions are detailed in Stevenson et al. [2006] (and are

summarized in Table 1), but I have added a sentence here to explain the variability.

Typo, This. Corrected, thank you.

Explain the point you are making here about gross fluxes. Gross cross-tropopause fluxes cannot be adequately diagnosed in coarse resolution models, as pointed out by Hall and Holzer [2003]. "Upward" and "downward" fluxes are not restricted to particular regions, are variable in time, and may often be driven by horizontal rather than vertical fluxes, and estimation of cancellation between fluxes is therefore not meaningful. My intention here was to highlight differences in location (and perhaps mechanism?) between models, and I have altered the sentence to reflect this.

Is the 150 ppb ozone chemical tropopause definition still applicable in models using SYNOZ? The SYNOZ method is designed to constrain the net STE without requiring an good reproduction of stratospheric ozone levels, and is useful in models where the vertical resolution in the stratosphere is too poor to attempt to model O_3 explicitly. The location of STE and the position of the tropopause will both be affected by this approach, but I do not believe that the approach is any less suitable than application of fixed or flux boundary conditions for O_3 in the stratosphere.

Was the 150 ppb chemical tropopause used in constructing the ACCENT budgets in Table 1? Yes. I have now stated this in the text.

Describe what type of chemical mechanism is used in the model. Also, are there any other important details of the model implementation that are relevant here (e.g., convection scheme)? The model uses a simplified but well-tested chemical mechanism that is described in the paper cited. I believe that differences in chemical mechanism (including differences in kinetic and photolysis data, and in chemical solver) are a significant source of model differences, but more detailed analysis of this deserves a separate study dedicated to the topic. I have added a sentence to summarize the

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chemistry scheme used, but the reader is referred to the previous studies cited for more detailed description of advection, convection and boundary layer schemes and for the characteristics of the meteorological fields used.

What is done for isoprene emissions in the IIASA scenario? Isoprene emissions of 500 TgC/yr are used here with the IIASA scenario, and this is now stated explicitly in the text.

Explain why the lightning NO_x was different. The standard runs here use uniform emission profiles of lightning NO_x based on Price and Rind [1993] at a rate of 5 TgN/yr. Runs for the ACCENT study used a more realistic vertical emission distribution based on observed profiles of Pickering et al. [1998], and the emission total was increased to 6.5 TgN/yr following the original instructions for that study. I have now included this information at the end of this paragraph. Note that Fig 2 has been replotted because the ACCENT line on the 250 hPa panel in the Northern Hemisphere Tropics was incorrectly taken from the T21 version of the run.

How much of the NO_x emissions are distributed vertically? Lightning NO_x emissions are constant at 5 TgN/yr for all these runs, and this is now stated explicitly in the emissions scenario description. Higher emissions (6.5 TgN/yr) were only included for the ACCENT study run illustrated in Fig 2.

Add "gross" before "O_3 production". Added.

Why does O_3 lifetime decrease? Much of the additional O_3 production occurs in the lower troposphere, so the burden in the lower troposphere is enhanced and the net lifetime is reduced. The sentence has been altered to make this clearer.

Change "budget changes" to "budgets". This has now been corrected.

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What are the non-isoprene NMHC emissions? These emissions have now been included in an extra column in Table 3.

CH_4 lifetime clarification. I have rewritten this sentence to make it clear that the changes in lifetime refer to the difference between the BASE and IIASA scenarios.

This is an important point. Previous studies have indicated a large sensitivity of the ozone budget to the treatment of isoprene nitrates. I have added a sentence about the chemical scheme used in the model in the section introducing the model, but have not included an exhaustive list of all the reactions considered and/or neglected because I believe that too much detail would make the paper harder to follow. Clearly, a more thorough exploration of model chemistry schemes would be worthwhile in light of the results of these sensitivity studies.

Explain what simulations were done here. Also, were CH_4 emissions used in other simulations? This paragraph has been reworded to make the studies clearer, and the NMHC emissions are now detailed in Table 3. The initial sensitivity runs (Tables 3 and 4) were performed with a transported CH_4 tracer and emissions of about 550 Tg/yr using a spin-up of 8 months; this information has now been added in section 4. This was done to maintain consistency with earlier literature studies, but it is acknowledged that CH_4 will not be in steady state in these runs, and that the sensitivities derived therefore represent instantaneous rather than equilibrium responses. The effects of this on the O_3 budget have been verified to be small, and the change in the O_3 burden over a year deviates from the base run by less than 2 Tg for all runs except those involving complete removal of NMHC or NO_x emissions.

This is a strange way to do bookkeeping. What does it mean to say that increased influx of O_3 "is accounted for by decreased production"? I agree that the phraseology here is awkward. Stating that 90% of the O_3 influx is removed by chemistry is misleading, however, as it gives the impression that the source apportionment has not changed;

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in fact, the influx leads to tropospheric precursors making a smaller contribution to O_3. Effectively, 30% of the O_3 influx displaces O_3 that would have been formed by chemistry in the troposphere. I have now rephrased this sentence.

Explain which approach is used here to define the tropopause. A 150 ppbv ozone tropopause was used here as in all the sensitivity studies, but the conclusions for this particular sensitivity are highly dependent on the tropopause definition. I now make it clear which approach is used here.

Are you increasing the dry deposition velocities for all species? Yes, dry deposition velocities were increased for all species, and this is now stated explicitly. The O_3-OH link is now made clearer.

While these are both 50% changes, you could also say that the 50% change is a factor of 2 change while the 50% increase is a factor of 1.5 change. So maybe it is not surprising that the 50% decrease has a bigger effect on the O_3 budget. This is misleading, as a 50% reduction is a factor of 0.5 change, and all changes must be referenced to the same (base) case. If the response were linear, the impacts of these changes would be the same, and this is clearly not the case here.

Does the temperature change affect anything else in the model? The temperature and humidity changes applied here are allowed to affect the chemistry scheme only, and the text has been amended to reflect this.

Typo, ".." should be ".". Corrected - thank you.

Which processes did these studies change? These studies changed convective transport and convective washout only, as stated here. In hindsight this was unfortunate, as the previous studies considered transport changes only. I have now made this distinction clear.

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Transport of precursors explains part of the decreased production. This is certainly true, and the text here has been amended to reflect this.

Change "inferred" to "calculated". The STE fluxes here are inferred from the residual in the O_3 budget and the change in tropospheric burden over a year (generally close to zero), as this allows a cleaner comparison with previous studies such as Stevenson et al [2006] where a similar technique was used. I have added a sentence at the beginning of section 4 to describe this.

Does convection necessarily penetrate above the tropopause, or could it just change the vertical O_3 gradient near the tropopause? This is a very good point. Although convection inevitably penetrates the monthly-mean tropopause used here just due to day-to-day variations in height, it is highly likely that steepening of the O_3 gradient at the tropopause also plays a significant role. I have amended the text to include this as a contributing factor.

Explain what simulations were done here. I have added a sentence at the beginning of the section stating that the magnitude of lightning emissions was increased from 0 to 7.5 TgN/yr in steps of 2.5 TgN/yr.

How does the sensitivity of CH_4 lifetime to lightning NO_x that you find here compare with that from other studies? The Fiore et al. [2006] paper estimates a 0.11 year change for a 0.32 TgN/yr lightning change, or 0.34 yr/TgN/yr. Current sensitivity in the present study based on increases from 2.5 to 7.5 TgN/yr gives a 1.34 yr change for 5 TgN/yr, 0.27 yr/TgN/yr (or 0.32 yr/TgN/yr if applying the Pickering profiles where the lifetime response is 18% larger). The equivalent sensitivity for the Labrador et al. [2004] study is 0.23 yr/TgN/yr. The relationship between CH_4 lifetime and NO_x emissions is non-linear, but the agreement here appears reasonable.

Are you describing the effect of going from 0 TgN/yr to 5 TgN/yr with two different

vertical distributions? Clarify. This is correct; I have added the word "increase" to make it clear that the comparison is between 0 and 5 TgN/yr.

How much does the O_3 deposition velocity change? The deposition velocities from the standard Wesely scheme are dependent on meteorological and surface parameters, and I have not diagnosed annual mean values for direct comparison with the simpler Isaksen/Hough scheme; nevertheless it is clear that they are smaller. This scheme comparison is included here because the simpler deposition scheme has been used with the same Hough deposition velocities in a number of earlier studies in the literature.

How does the effects of clouds here and by Tie et al. compare with other studies? There have been a number of studies recently looking at the impacts of different cloud treatments (and overlap) on photolysis rates, and I present only a very simple simple clouds-on vs. clouds-off sensitivity here. Feng et al. [2004] and Yang and Levy [2004] do not integrate the global impacts on O_3, but I have included a comparison with Liu et al. [2006] to supplement the Tie et al. comparison. The results of the Liu et al. study are comparable to those found here, and are clearly smaller than those of Tie et al.

Change "Tg/yr" to "TgN/yr". Corrected - thank you.

It would be interesting also to examine the differences among various reanalyses for a given year, but this would be beyond the scope of this study. I agree that this would be very interesting to look at. I suspect that differences in the methodology for building reanalysis products (such as spin-up and forecast lengths for the ERA-40 fields, as considered by van Noije et al. [2006]) would also be a major source of differences.

Are the convective mass fluxes the same at all resolutions? For a given meteorological year the mass fluxes are the same for all resolutions. I have added a sentence in section 4.3 to make this clear.

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How large are these variations relative to the spread seen among ACCENT models? I have now quantified the variability seen in the ACCENT studies at the top of this paragraph to put the results into better perspective.

Add "in O_3 budgets" after "of the variability". Added.

Can you make any quantitative statements here? I have attempted to do this by using the standardization approach that is described in the final paragraph of this section.

Can you comment on how important dynamics might be based on your runs with 1997 and 2000 meteorology? What about model resolution? The 1997 and 2000 simulations provide a good indication of the effects of interannual variability, but do not provide a full measure of the sensitivity to meteorology, which would additionally be influenced by differences between reanalysis products (as highlighted above) and by different parameterizations/treatments of advective and convective processes, boundary layer mixing, etc. Two sentences have been added here to describe the effects of meteorology and resolution.

Underestimate versus what? This sentence has been rearranged as "Ozone loss and CH_4 lifetime are systematically lower in model studies omitting higher hydrocarbons."

The highest chemical lifetime shown in the plot is 12.5 years (not 15.2). This is correct (well spotted!), but the highest chemical lifetime is indeed 15.2 years; this model does not appear in the plot because they did not archive O_3 budget data and therefore we have no data for the O_3 chemical loss rate.

Note that you do at least consider the effects of clouds and aerosols on photolysis rates. I have considered the presence/absence of these variables, but have not explored how the use of different chemistry or photolysis schemes affects the budgets, so cannot make a stronger statement here.

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Mention that uncertainty in CH$_4$ lifetime implies an uncertainty in CH$_4$ emissions (for a fixed CH$_4$ burden). Also typo: "tracer gases" should be "trace gases". This is an important point, as it also explains our inability to correctly model recent trends in atmospheric CH$_4$. I have added a sentence to emphasise this point. The typo has been corrected.

Change "the terms are" to "the O$_3$ budget terms from each model are". Corrected - thank you.

Clarify: are all models scaled to the same O$_3$ dry deposition flux? Yes, all models are scaled to the same ensemble-mean budget fluxes.

Be more specific about the methods used here, or give an example of the scaling that is done. The standardization process described in this section has been simplified by applying linear correction terms only. This gives similar results but makes the scaling process easier to understand. As suggested, I have rewritten this section and now provided an example of how the scaling is done.

You could make this point more clearly by including in table 6 lines for the results from standardizing each variable individually. This is a good suggestion, and I have now included the individual terms in Table 6.

Explain that "loss" here refers to chemical loss only. The word "chemical" has been inserted infront of "loss" here.

Could other factors also be standardized (e.g., temperature, humidity)? It might be interesting to extend this analysis, but changes in temperature and humidity would be difficult to treat this way, as regional and seasonal variations would be important.

Why have emissions of NO$_x$ and isoprene increased? NO$_x$ emissions have been revised upwards over time; based on recent assessments [RETRO, POET, EDGAR] the

increase in emissions between 1990 and 2000 is thought to be about 1–2 TgN. Estimates of isoprene emissions have not changed, but the amounts used in models have increased due to a greater number of models including isoprene chemistry and to greater faith in the large emission estimates from the model community (related partly to improved oxidation schemes). I have added a sentence to state this explicitly.

Be more specific about the results of the Wu et al. study. The Wu et al. paper was not published until March, and hence a final version was not available before submission of the present paper. The paper examines the contribution of changing emissions and STE to changes in model ozone budgets, and reaches similar quantitative conclusions, as stated here. It reaches different conclusions regarding isoprene emissions, attributing a saturation in O_3 production to isoprene nitrate removal; this conclusion is also cited here, and provides further encouragement to study the sensitivity of the budget terms to chemical mechanism. Their other analyses are complementary to those presented here.

Is the choice of upper boundary condition (e.g., SYNOZ) also important? It is highly likely that different methods of treating ozone above the tropopause (fixed boundary, flux boundary, Synoz, Linoz, full stratospheric chemistry) affect the diagnosed tropopause height (and STE), but this is one sensitivity that hasn't been examined here in detail. I have added a sentence at the end of the paragraph to acknowledge this.

Comment here on which tropopause definitions are most robust. The 150 ppb tracer tropopause is simple to apply and consistent with model dynamics, but minimizes model differences and damps the response to applied changes. A dynamical PV tropopause would allow a more robust comparison between models using the same meteorology, but is less practical when different meteorological fields are used. I have added a statement supporting use of the 150 ppb tropopause here.

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Clarify that the tropopause is defined based on the "monthly mean" O_3 concentrations here. I have added "monthly-mean" here to emphasise this point. It is quite likely that use of a monthly-mean introduces some bias compared with daily or hourly data; it would be interesting to examine this further.

What fraction of the ACCENT variability is this? This has been addressed by adding "This accounts for about 10% of the variability in the O_3 budgets and almost 20% of the variability in the CH_4 lifetime."

Is the O_3 lifetime given here versus chemical loss and dry deposition? Also clarify how O_3 P and L are defined. The O_3 lifetimes here are against chemical loss and dry deposition, and this has now been added in the footnote to Table 3. The definition of P and L is now given in the text at the beginning of Section 4 where the model and diagnostics are described.

How is the CH_4 lifetime defined here? I have used the conventional chemical lifetime given by the global burden divided by tropospheric loss via OH; I have included this information in the footnote to Table 3.

Change "5 Tg NO_x" to "5 TgN/yr". Corrected - thank you.

Change "differing emissions, deposition" to "different lightning and surface emissions, dry deposition". The caption has been clarified as suggested.

Label endpoints for STE in panel (d). Endpoints have now been labelled.

Add "versus chemistry and deposition" after "Tropospheric O_3 removal rates". The caption has been amended as suggested.

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1995, 2007.