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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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Anonymous Review of "Modelling the global tropospheric ozone budget: exploring the variability in current models" by O. Wild

This paper analyzes the contributors to the differences in tropospheric ozone budgets among current and recent chemistry-transport models. In particular, the paper first seeks to explain the increase in estimated ozone productions rates from earlier assessments to more recent model intercomparisons. The author concludes that this increase is largely due to increased precursor emissions in the newer models. Even when similar emissions are used, however, models still show a wide range of estimates for the ozone budget. The paper investigates the contribution to this range from processes including: cross-tropopause transport, humidity, wet and dry deposition, and lightning NOx. This well-written paper is a highly useful assessment of the current

state of chemical transport modelling (and its uncertainties). I recommend the paper for publication in ACP with the many minor revisions I detail below.

Comments

Abstract

Page 1996, lines 3-6 - How large are the differences among models? For instance, later you give percent changes due to lightning NOx and resolution - how large are these changes relative to the spread of models?

p.1996, l.14-15 - You could cut “cross-tropopause transport ... between models.” Saying that these processes all make “large contributions” here is less informative than the quantitative statements made in the next few sentences.

p.1997, l.1 - By “model chemistry”, are you referring just to the chemical mechanism, or also to other aspects (e.g., emissions)? Similarly, what do you mean by “dynamics” here - just wind fields, or other meteorological fields (temperature, convection)?

1. Introduction

p.1997, l.3-6 - Give some measure of the uncertainty or range in budget estimates here.

2. Tropospheric ozone budgets in global models

p.2000, l.4 - Add “net” before “chemistry”.

p.2000, l.23-28 - Why have NOx emissions increased over time (Fig. 1)? Also, explain why there is still a range of NOx and isoprene emissions if ACCENT constrained emissions. What exactly was prescribed in ACCENT?

3. Constraints from observations

p.2001, l.15 - Typo, “this” should be lowercase.

p.2001, l.15-17 - Explain the point you are making here about gross fluxes. How much

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cancellation is there typically between upward flux (in the tropics) and downward flux (in the extratropics)?

p.2002, l.18-21 - Is the 150 ppb ozone “chemical tropopause” definition still applicable in models using SYNOZ? They typically have much too low O3 values in the tropopause region (and stratosphere). Thus, the 150 ppb tropopause would be unrealistically high.

p.2003, l.4-8 - Was the 150 ppb chemical tropopause used in constructing the ACCENT budgets in Table 1 (based on Stevenson et al. [2006])?

4. CTM sensitivity studies

p.2003,l.26 - p.2004,l.1 - 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)?

p.2004, l.17-21 - What is done for isoprene emissions in the IIASA scenario? The total emissions are 500 TgC/yr from Table 3. Are they distributed the same as in the BASE scenario?

p.2005, l.12-14 - Explain why the lightning NOx was different. Was a different parameterization used in the ACCENT run, or did the different horizontal/vertical resolution and different meteorology produce the different lightning NOx? What are the lightning NOx totals for the two runs?

4.1 Sensitivity to precursor emissions

p.2006, l.7 - How much of the NOx emissions are vertically distributed (e.g., lightning NOx) versus emitted at the surface in the two runs?

p.2006, l.8 - Add “gross” before “O3 production”.

p.2006, l.14-16 - Why does O3 lifetime decrease? Because the excess O3 is mostly in the lower troposphere?

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p.2006, l.23 - Change “budget changes” to “budgets”.

p.2006, l.26-27 - What are the non-isoprene NMHC emissions? (Not shown in Table.)

p.2007, l.9-11 - Does this comment about CH4 lifetime remaining little affected refer to going from the BASE run to the ACCENT run? Clarify.

p.2007, l.13-21 - This is an important point. Previous studies have indicated a large sensitivity of the ozone budget to the treatment of isoprene nitrates. Be sure to indicate in the model description section pp. 2003-4 some description of the chemical scheme used here.

p.2007, l.22-29 - Explain what simulations were done here (listed in bottom section of Table 3). Also, were CH4 emissions used in the other simulations? If so, how was CH4 spinup handled if you only range for one year (using 1996 meteorology)?

4.2 Sensitivity to physical processes

p.2008, l.16-18 - This is a strange way to do the bookkeeping. What does it mean to say that increased influx of O3 Ĕ “is accounted for by decreased production”?

p.2008, l.18-27 - Explain which approach is used here to define the tropopause. Also, mention that increased STE improves the ozone simulation (Table 4).

p.2009, l.1-3 - Are you increasing the dry deposition velocities for all species? Explain how O3 chemical loss is related to the tropospheric OH source. You shouldn’t assume that this connection is obvious.

p.2009, l.15-17 - While these are both 50% changes, you could also say that the 50% decrease 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 O3 budget.

p.2009, l.20-23 - Does this temperature change affect anything else in the model, or just kinetic reaction rates?

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p.2010, l.8 - Typo, ".." should be ".".

p.2010, l.11-16 - Which processes did these studies change? Just convective transport, or also convective washout, lightning NOx, etc? What about in this study (l.16-17)?

p.2010, l.18-20 - Transport of precursors from the lower troposphere to the upper troposphere (presumably) explains part of the decreased production in the lower troposphere and of the increased production in the upper troposphere.

p.2010, l.20 - Change "inferred" to "calculated". Or, are the fluxes across the tropopause actually inferred as a residual in the ozone budget?

p.2010, l.21 - Does convection necessarily penetrate above the tropopause, or could it just change the vertical O3 gradient near the tropopause?

p.2011, l.2-18 - Explain what simulations were done here. For instance, lightning NOx was set zero, 50% (2.5 TgN/yr), and 150% (7.5 TgN/yr).

p.2011, l.8-9 - How does the sensitivity of CH4 lifetime to lightning NOx that you find here compare with that from other studies (e.g., the Fiore et al., 2006 paper you cite)?

p.2011, l.14-18 - Are you describing the effect of going from 0 TgN/yr to 5 TgN/yr with two different vertical distributions? Clarify.

p.2011, l.23-24 - How much does the O3 deposition velocity change? Is the change linear with the deposition flux? How much do deposition velocities of other species change?

p.2011, l.11-16 - How does the effect of clouds calculated here and by Tie et al. compare with other studies? For instance, H. Liu et al. (JGR, 2006), Feng et al. (JGR, 2004), Yang and Levy (JGR, 2004).

4.3 Sensitivity to meteorology and resolution

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p.2012, l.28 - Change “Tg/yr” to “TgN/yr”.

p.2013, l.9-11 - 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.

p.2013, l.12-14 - Are the convective mass fluxes the same at all model resolutions?

4.4 Examining inter-model variability

p.2014, l.21-24 - How large are these variations relative to the spread seen among ACCENT models?

p.2015, l.2 - Add “in O3 budgets” after “of the variability”.

p.2015, l.2-3 - Can you make any quantitative statements here? It is hard to tell how all of the factors listed add up.

p.2015, l.3-4 - Can you comment how important dynamics might be based on your runs with 1997 and 2000 meteorology? What about model resolution?

p.2015, l.13-14 - Underestimate versus what?

p.2015, l.16 - The highest CH4 lifetime shown in the plot is 12.5 years (not 15.2).

p.2015, l.18-20 - Note that you do at least consider the effects of clouds and aerosols on photolysis rates.

p.2015, l.20-23 - Mention that uncertainty in CH4 lifetime implies and uncertainty in CH4 emissions (for a fixed CH4 burden). Also, typo: “tracer gases” should be “trace gases”.

p.2015, l.25 - Change “the terms are” to “the O3 budget terms from each model are”.

p.2015, l.28 - Clarify: are all models scaled to the same O3 dry deposition flux (vs. deposition velocity)?

p.2015, l.27 - p.2016, l.1 - Be more specific about the methods used here, or give an

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example of the scaling that is done.

p.2016, l.4-6 - You could make this point more clear by including in Table 6 lines for the results from standardizing each variable individually.

p.2016, l.6 - Explain that “loss” here refers to chemical loss only. As dry deposition increases, chemical loss decreases.

p.2016, l.8-11 - Could other factors also be standardized (e.g., temperature, humidity)?

5. Conclusions

p.2016, l.17-19 - Why have emissions of NOx and isoprene increased? Have NOx emissions actually increased over the past decade (i.e., using different base emission years), or have estimates just been revised upwards?

p.2016, l.22 - Be more specific about the results (and methods) of the Wu et al. study. That study and the current one have many similarities. It would be useful to comment further about the similarities and differences between the two studies and their findings.

p.2017, l.1-3 - Is the choice of O3 upper boundary condition (e.g., SYNOZ) also important?

p.2017, l.1-9 - Comment here on which tropopause definitions are most robust, based on your analysis.

p.2017, l.8 - Clarify that the tropopause is defined based on the *monthly mean* O3 concentrations here.

p.2017, l.12-15 - What fraction of the ACCENT variability is this?

Table 3

- Is the O3 lifetime given here versus chemical loss and dry deposition? Also clarify how O3 P & L are defined? As production & loss of odd oxygen (definition)?

- How is the CH4 lifetime defined here? Do you use the usual definition of total atmo-

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spheric burden divided by loss via tropospheric OH?

Table 4

- Change “5 Tg NOx” to “5 TgN/yr”.

Table 6

- Change “differing emissions, deposition” to “different lightning and surface emissions, dry deposition”.

Figure 5

- Label endpoints for STE in panel (d).

- Add “versus chemistry and deposition” after “Tropopspheric O3 removal rates”.

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