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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influence of convective transport on tropospheric ozone andits precursors in a chemistry-climate model" *by* R. M. Doherty et al.

R. M. Doherty et al.

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Continued response to referee Mark Lawrence.

5) I suspect that a major possible cause for par t of the differences to earlier studies is the use of an ozone threshold (O3=150 ppbv) for the tropopause. This is a fine and useful definition for many studies, but particularly for this study it will move considerably between the base and sensitivity runs, which may substantially affect the tropospheric ozone burden. A physical tropopause was used in both LC94 and L03 (in the latter we used the WMO lapse rate definition, diagnosed each time step, so that it was the same for all runs). I would suggest that the authors examine the budgets using both

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definitions. A physical tropopause (whichever definition is preferred) which stays the same between all runs shows how O3 changes within the volume defined as the troposphere. The additional information based on the ozone-threshold-tropopause then shows of the secondary effect of convection on the area that would be defined as the troposphere in other studies.

We accidentally omitted from our paper text that we used the same tropopause for the base and sensitivity runs. This tropopause is shown in Figure 6 in our paper. We define the tropopause as any grid-cell with monthly- mean (over the 20-year period) ozone in either the base or sensitivity run as exceeding 150 ppbv. We have also calculated the ozone budget for a 100 hPa limit and for a physical tropopause as suggested above. We obtain the same direction and similar magnitudes of change in the ozone budget between the base and sensitivity runs for all tropopause definitions. The budgets for both calculations can be viewed at:

http://www.met.ed.ac.uk/~dstevens/convection_paper/tables.doc

6) The finding of a negligible role of lofting of NOx is puzzling, and is in strong contrast to a number of earlier studies, dating all the way back to Ehhalt et al. (1992) and the series of studies by Pickering and Dickerson and colleagues. If the authors are right in their speculation and this is really due to the binding of NOx into PAN by lofted hydrocarbons, then it is quite an interesting possibility, but needs to be substantiated by comparing PAN to observations, and by matching up the NOx and PAN budgets (or better yet, following the Lagrangian parcels) to make sure this is what is really happening.

Unfortunately, due to storage constraints we did not archive a suitable tracer species to be able to quantify what fraction of the NOx decrease can be explained by the PAN decrease. Simulated PAN profiles show agreement within one standard deviation with observations over the tropics. There is some overestimation of PAN in parts of the UT, but also some overestimation of UT NOx (see text for comment 1). Therefore, the

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binding of NOx into PAN seems plausible process, since it acts to bring simulated NOx concentrations into better agreement with observations. However, other mechanisms such as excessive lightning NOx emissions or errors in their vertical distribution could also explain the UT NOx overestimates in the tropics. Comparing changes in NOx and PAN in the UT (350-150hPa) we find UT NOx decreases by 0.0318 Tg N (50% decrease) and a UT PAN increases by 0.0281 TgN (65% increase). In terms of N conservation we expect that a substantial fraction of the NOx decrease arises from the PAN increase.

We have added this text to section 3.2 (model results for the tropics) and the discussion.

7) The analysis would benefit greatly from the addition of a few pressure-level figures to help demonstrate exactly where increases and decreases are occurring.

We have produced pressure-level figures for O3, NOx, O3 Production and destruction. We will include some of these figures if we can find space in this already rather lengthy paper.

8) In the abstract: before "The combined effects of" need to add "We examine this with a 3D CTM and find"

Text amended.

9) I think the statement that "Convection redistributes lightning NOx emissions downwards at the expense of the UT" is an overstatement, at least it doesn't fit with the many observations of enhanced NOx in convective anvils, or the strong peaks in UT NOx emissions in the Pickering et al vertical profiles (some is indeed transported in downdrafts, but this is probably more at the expense of the MT)

We agree this statement is confusing. In our model set-up we use the Pickering et al., vertical profiles to distribute lightning NOx emissions, and these are then also subject to convective mixing once they enter the model transport and mixing schemes.. We did not make clear that the Pickering profile is applied in the convection off experiment

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as well as the convection on experiment.

We have added this clarification to the text.

10) The stratospheric influx is at the very low end of observational and other model estimates - how much will this influence the results? Also, Murphy and Fahey (1994) gave different N:O3 ratios for the tropics and extratropics, why is only one used here and is it possible that this could influence the results (I doubt it but wouldn't be able to rule it out without a short sensitivity run)?

We have slightly revised our ozone budget calculation (see response to referee 1 comment 1) - the annual mean stratospheric influx (plus or minus 1 standard deviation) is 394+/-15 Tg O3. This is towards the low end of the range of observational estimates. Although the stratospheric influx changes between our two runs, the changes are not significant. We don't think the relatively low stratospheric input will strongly influence the sensitivity of ozone to convection that we calculate.

Table entry and text is revised.

11) How is the precipitation scavenging of soluble gases (especially HNO3 and the isoprene oxidation intermediates) treated? Are any scavenged by ice? Also, how is their transport in deep convection differentiated from insoluble gases? There is a lot of free room for choosing how to do these, given the large uncertainty, but the specifics of the treatment will have substantial effects on the results, and this is very difficult to assess without at least a good description (or pointer to where it is available in other literature) of exactly what has been done.

Scavenging is described in Stevenson et al (2003). We should clarify that in the convection off experiment, scavenging still operates - only the mixing is switched off.

12) For the citation to Labrador et al. (2004), it would be better to refer to the Labrador et al. (2005) study which just appeared in ACP.

Text amended.

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13) In answer to one of the questions posed by one of the other referees, in L03 we kept water vapor distributions and lightning NOx emissions the same for all runs, exactly as is done in the runs for this study (it might be wor th explicitly mentioning this comparability; I believe the same applies to LC94 as well)

We have added this text to the discussion.

14) Why would convection "flatten the C-shaped profile [of NOx] over land"? - this is opposite of what one usually expects from convection, which is known to produce C-shaped profiles in short-lived tracers with surface sources

This statement refers only to the tropics where surface NOx emissions (that arise mainly from biomass burning) and UT NOx emissions (from lightning) result in surface and UT NOx concentrations that are similar in magnitude as shown in the new Figure 2a see comment 1 referee ML).

Text is clarified.

15) Figure 5 represents and interesting approach, but it needs to be described in more detail: how is the UT O3 defined (what altitude?)? Is the TC "total" or "tropospheric" column (and if the latter, is O3=150 ppbv also used for the tropopause)? What do the individual points represent (monthly means at a location averaged over 20 years, or otherwise)?

Text is amended so that this figure is clearer. The UT is 150-350 hPa, TC should read TTC - tropical tropospheric column - as defined in the introduction. Individual points are grid-box 20-year annual average data.

16) Why does the stratospheric influx decrease with convective mixing in these runs? The UT O3 mixing ratio decreases nearly everywhere, so one would expect the strat-trop gradient to be larger and, if anything, for the STE source to increase; also, convective mixing itself should, if anything, increase the source due to mixing in the UTLS region. Is this an artifact of using the O3-tropopause?

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To calculate stratospheric ozone influx, we use an ozone climatology at 100 hPa, and vertical winds at 100 hPa. Neither of these change between experiments. We therefore expected no change in STE between experiments. However, there are some minor differences because in our Lagrangian scheme, convection changes the distribution of air parcels, and hence the sampling of the stratospheric influx. In our re-analysis of the ozone budgets (see referee 1 comment 1), the STE values now change by less between the two runs, and when we analyse interannual variability, we find that the changes are not significant. The change in the STE term is similar using a chemical and a physical tropopause - it is not an artifact of the masking technique.

17) In the comparison of Lagrangian vs. Eulerian resolution, we used a model version with 28 (not 20) ver tical levels (although approximately 5-10 are in the stratosphere, depending on latitude, so perhaps this is what was intended)

Yes, we were just concerned with levels in the troposphere, so we estimated the mean number of model levels in the troposphere in MATCH-MPIC to be 20. We have clarified the text and revised calculations for the numbers 18-23 given above.

18) In Fig. 3, it would be helpful to add a third column with the change in net O3 chemistry tendency

We have done this and included text in section 3.2. Figure 3 nor Figure 4 can be viewed at: http://www.met.ed.ac.uk/~dstevens/convection_paper/figure4new.pdf

19) Finally, if the authors would find it helpful to have access to any of the output from the MATCH-MPIC runs used in L03, they are welcome to it.

Many thanks! We have compared convective 3-d mass flux fields and included text in the discussion (see comment 4, referee ML).

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3747, 2005.