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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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We would like to thank all the referees for their detailed and thoughtful comments on the paper. Here we present our initial responses to most of the referees' comments. Not all points raised by the referees are dealt with here. We will include a full response to all comments in our final response.

Some responses to referee #3 Mark Lawrence.

1) It is difficult to know if there are any overall model deficiencies that may affect these results without a decent evaluation of the distributions of the main trace gases being available. I have brought along copies of all the Stevenson et al. and Collins et al.

papers cited herein and gone through them looking for comparisons with observations. The model was originally evaluated for O3 in Collins et al., 1997, which was comparable to other evaluations at that time period (e.g., Roelofs and Lelieveld, 1995). In Collins et al. (1999), several key VOCs were compared to a limited set of observations (e.g., MLOPEX), showing a tendency to overestimate several gases such as HCHO, CH3OOH, and CH3COCH3 (acetone). This may be indicative of excessive photochemical activity, which would of course influence the response to convection, but it is hard to judge without a more extensive evaluation. Unfortunately, both of these studies were done with the old deep convection parameterization; a new parameterization was introduced in Collins et al. (2002) and evaluated for radon, but the only evaluation of ozone-related trace gases which I could find after this was in Stevenson et al. (2004 ,in which there were some very clear problems with ozone (presumably in the MT and UT) being underestimated at a few locations (Wallops Island and Ascension Island), which may be related to vertical transport. Since this study is being done with the new convection scheme from Collins et al. (2002), and is focused on the effects of convection, a better evaluation is really needed. Although it is not reasonable to expect this paper to perform a full-blown evaluation, it should at least 1) connect in carefully to any other evaluation work which has been done (I think there was some for the IPCC, too, but do not have it with me), and 2) include comparisons to the Emmons et al. composites for at least O3 and NOx (which look roughly okay based on figure 2, although the O3 mixing ratios look probably too low in the subtropical MT), and especially PAN, which is critical to the discussion of the results in this study.

Model evaluation for ozone with the current convection scheme was performed in Stevenson et al., (2004) and Dentener et al., (2005). These results were very briefly mentioned in section 4 (p. 3760). We will add a new section after section 2 to expand on this text and perform a NOx and PAN evaluation.

2) The analysis needs to be extended. Although a considerable computational effort goes into two 20-year runs, the only substantial results which are really added to the

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literature at the moment are that the results of L03 do not hold up in another contemporary model, that this is robust for that model for a 20-year comparison, and that there are considerable differences in the mean responses over land and sea. The analysis should be extended in two ways. First, it would benefit from very simple sensitivity studies to support some of the speculation about the differences in the results. In particular, a run (or runs) should be done with lower isoprene and lightning NOx emissions, similar to those used in L03. This may already be enough to explain the differences in the two studies, or at least a large par t of the difference, but it might also yield a surprise (I had expected the NMHC chemistry to be the main difference between L03 and LC94, and was really surprised at the results of our sensitivity runs). These runs do not need to be for 20 years; 1 year would even be sufficient with forced SSTs, given that the anticipated signals are very large (5 years would definitely be plenty). Second, the analysis of these main two runs also could be deepened. In particular, no analysis of the interannual variability, e.g., the dependence on ENSO, was given based on the 20-year runs. Also, a number of statements are made without direct support from the model (only circumstantial evidence), e.g., "most of the UT NOx arises from advection of UT land lightning NOx emissions", many of which could be better substantiated and quantified (e.g., what fraction is "most"), especially given that the study uses a Lagrangian model (which makes some issues like this much easier to quantify than in Eulerian models).

We have performed further sensitivity runs to lightning NOx emission magnitudes and these are the subject of a further paper. We will mention any results briefly in this paper where relevant. Further sensitivity runs involving isoprene would undoubtedly be interesting, but we feel are beyond the scope of an already lengthy paper. It is an extremely interesting question to understand the exact reason for inter-model differences between our study and L03 but in one paper we cannot explicitly answer this question since we believe there are many complex factors at work. We indicate that we believe that the impact of convection on ozone is influenced by both model convection and model chemistry schemes and differences in these can produce very different results.

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We have discussed whether potential reasons for differences between L03 and LC94 could explain differences between this paper and L03.

We will expand Table 1 to include standard deviations associated with interannual variability and significance as suggested by referee 1 (see referee 1 comment 4). ENSO is the subject of another paper currently being written.

We will make the text clearer regarding any speculative comments. From our further lightning sensitivity experiments we can verify that lightning NOx emissions are the main source of the UT NOx over the tropical oceans. We infer that advection from the UT over land is probably the main source of the oceanic UT NOx, based on the large ratio of UT lightning NOx over land compared to ocean of in the tropics.

4) The distribution of deep convection in the model seems a bit puzzling; from experience and observations, deep convection ver y often reaches the tropopause (about 200 hPa) in the mid-latitudes (and some, e.g., Folkins, might even argue that it reaches actually reaches the real tropical thermal tropopause less often than in the midlatitudes); there seems to be an excess of detrainment in the MT of the mid-latitudes, at least with respect to what I am familiar with from observations and cloud resolving models; if only shallow convection is present in the mid-latitudes in the model, but not much deep convection, this is likely a major cause for differences with the MATCH results in L03; do the authors have any observational evidence in support of the distribution of convection (also with respect to some of the other oddities I pointed out in my access review, such as the enormous fluxes north of the Himalayas, which will transport a lot of biofuel pollutants, and the missing convection over Mexico and the Gulf of Mexico, which one normally sees vividly in precipitation climatologies)?

Figure 1 in our paper displays the convective mass flux field for only one level: ˜600 hPa. Deep convection does occur in some mid-latitudes regions at up to ˜200 hPa. To address the referees question regarding convective precipitation we have compared the HadAM3 total precipitation field with the GPCP precipitation climatology (Rudolf et

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al., 2005). The distributions compare favourably. Precipitation peak amounts in the Atlantic and Pacific oceans between 30-60N are slightly underestimated in HadAM3. HadAM3 precipitation has been evaluated by Pope et al.,(2000). We will add a fuller discussion of this in the text. Note that Figure 1 in our paper is convective mass flux at ˜600HPa and not convective precipitation. For an analysis of differences in convection height (i.e. shallow and deep convection) we will compare our 3-D convective mass flux fields with those in Lawrence et al., (2003) (L03), so that we can add text to the discussion. The anomalies over the Himalayas are non-precipitating convection and are a consequence of model orography - it remains to be seen if these are anomalous to HadAM3, or present in other models.

5) I suspect that a major possible cause for part 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 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

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between the base and sensitivity runs for all tropopause definitions.

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, we did not archive sufficient 3-D flux terms to calculate a full NOx or PAN budget. But we will compare our simulated PAN to observations (see question 1 this referee) and include this in the text. Nevertheless, the magnitude of UT NOx decrease is similar to PAN increase, and we think that this is the main explanation for the model behaviour.

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 as well as the convection on experiment. We will clarify 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

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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.

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 will add 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. This will be clarified in the text.

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 will be amended to make this figure clearer. The UT is 150-350hPa for this fig-

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ure, TC should read TTC - tropical tropospheric column - as defined in the abstract. 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?

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. We will add text to clarify this.

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 would like to compare convective 3-d mass flux fields.

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