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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.

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This is an interesting manuscript which adds to the literature on the effects of deep convection on tropospheric chemistry by essentially repeating the analyses done in Lelieveld and Crutzen (1994) [LC94] and Lawrence et al. (2003) [L03], focusing on the net effects of deep convective transport on tropospheric ozone. The authors use a contemporary global chemistry-transport model to perform two runs, a base run (with "normal" convective transport) and a sensitivity run with convective transport of all tracers turned off. They arrive at the overall result that deep convective transport of O3 and

its precursors causes a 15% decrease in total global O3, which is in strong contrast to the 12% increase that we computed in L03 (which was, in turn, the opposite of the 20% decrease computed earlier with a simpler model in LC94). The authors extend the previous analyses in a couple of points, e.g., by doing 20-year (rather than 1-year) runs, and by breaking down the results into land and sea regions. I strongly support the eventual publication of these results, as I think they will contribute to an interesting, long-term discussion on this topic; in particular, the authors have the opportunity to set the stage for what could become an important future process-oriented model intercomparison (i.e., how chemistry-transport models respond to deep convection or possible future changes in its intensity and distribution). However, I feel that the manuscript needs considerable development before it will be of a quality appropriate for ACP. In particular, the analysis needs to be deepened in order to allow a proper interpretation of the computations and the reasons behind the differences with earlier (and likely follow-up) studies. Overall, there are two major shortcomings: 1) the evaluation of the model (base run) results versus observations is insufficient; and 2) a few simple sensitivity runs are needed to test at least some of the speculation presented in the current discussion. My specific comments and suggestions for improving the manuscript are outlined below.

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

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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.

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 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 part 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

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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).

3) The authors should not refrain from really pointing out just how large the differences between these results and L03 really are - this makes a substantial statement about the state of contemporary models for such studies, especially if the deepened analysis can help point out why these differences exist. One example that makes the contrast very clear is that only about 5% of all points in this model show an increase in column ozone (and only over limited biomass burning regions, not over the industrial polluted regios), whereas in L03 *all* of the points showed an increase in column ozone, especially over the outflow of populated regions.

4) The distribution of deep convection in the model seems a bit puzzling; from experience and observations, deep convection very 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)?

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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.

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.

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.

8) In the abstract: before “The combined effects ofĚ” need to add “We examine this with a 3D CTM and findĚ”

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

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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)

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)?

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.

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.

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 worth explicitly mentioning this comparability; I believe the same applies to LC94 as well)

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

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

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individual points represent (monthly means at a location averaged over 20 years, or otherwise)?

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?

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

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

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.

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