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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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Here is our final response. All comments are addressed. The referee's original comments are in italics.

Responses to referee Mark Lawrence. (Part 1).

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 too briefly mentioned in section 4 (p. 3760). We have added a new section 3.1 entitled “Evaluation of modelled NOx, PAN and O3 against observations”. This shows that the model simulates the observations with some success. Figure 2 can be seen at:

http://www.met.ed.ac.uk/~dstevens/convection_paper/figure2.pdf

2) The analysis needs to be extended. Although a considerable computational effort

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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 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 parallel paper. We have mentioned the results of these experiments in this paper twice 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

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model convection and model chemistry schemes and differences in these can produce very different results.

We have expanded Table 1 to include standard deviations associated with interannual variability and significance as suggested by referee 1 (see referee 1 comment 4). See http://www.met.ed.ac.uk/~dstevens/convection_paper/tables.doc

ENSO is the subject of another paper currently being written.

We have made the text clearer regarding where we make definitive and speculative comments. From our further lightning sensitivity experiments we can verify that lightning NOx emissions are the source of the UT NOx concentrations above 50 ppt over the tropical oceans. However we cannot breakdown the contribution of land and ocean lightning to UT NOx concentrations over the ocean. We infer that advection from the UT over land is the main source of the oceanic UT NOx, based on the large ratio of UT lightning NOx over land 40:1 compared to ocean of in the tropics.

This text has been included.

*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.*

Text amended in section 6 to read:

“Differences in the patterns of TCO change are large with decreases almost everywhere in our study and increases everywhere in Lawrence et al., (2003).

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It is important to ascertain the root cause of the contrasting results concerning the impact of convective mixing on tropospheric ozone between our study and that of Lawrence et al., (2003) in order to provide a robust uncertainty estimate of the effect of deep convection and future changes therein. Below we discuss potential differences between the two models. . . "

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)?

A comparison of annual-average precipitation in HadAM3 and in the GPCP precipitation climatology (Rudolf et al., 2005) can be seen at : http://www.met.ed.ac.uk/~dstevens/convection_paper/compareprecip.doc. The distributions compare favourably. Precipitation peak amounts in the Atlantic and Pacific oceans between 30-60N are slightly underestimated in HadAM3.

Our original figure 1 was misleading, partly because of the scale. There is deep convection in the mid-latitudes in HadAM3, in fact HadAM3 tends to overestimate convection. We have revised Figure 1 and included an evaluation with ERA-40 convective mass fluxes. A preliminary figure can be seen at: http://www.met.ed.ac.uk/~dstevens/convection_paper/figure1new.pdf We have in-

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cluded the following text at the beginning of section 3:

"Figure 1a depicts average convective mass updraught mass fluxes for northern hemisphere summer (JJA) from HadAM3. The main region of deep convection, with strong up-draughts that reach the tropopause, occurs in the tropics. The inter-tropical convergence zone (ITCZ) and South Pacific Convergence Zones are clearly delineated (Figure 1a,b). Substantial convection at 200hPa can also be seen over the S.E.Asian continent, southern North America, and eastern Siberia/Japan (Figure 1a). Convection that extends up to the mid-troposphere also occurs in the tropics, and in mid-latitudes (particularly in the southern oceans and northern continents), typically associated with frontal activity (Figure 1b). The anomalies over the Himalayas are non-precipitating convection and may be a consequence of model orography. HadAM3 convection is compared to ERA-40 renanalyses (courtesy of P. van Velthoven Pers. Comm.) in Figures 1c and d. Note that the HadAM3 fluxes are displayed on eta (P/Surface pressure)x1000 levels, as opposed to actual pressure depicted in the ERA-40 renalyses, therefore there will be some differences near the surface. HadAM3 overestimates the strength (by up to a factor of 2) and height of deep convection in the UT. It also overestimates convection that reaches the MT in the southern latitudes. Therefore, HadAM3 has a tendency towards over-vigourous and deeper convection compared to the ERA-40 reanalyses."

In addition, thanks to the referee we have compared convective mass fluxes between HadAM3-STOCHEM and the MATCH-MPIC model.

We have included a new Figure 8 which can be viewed at: http://www.met.ed.ac.uk/~dstevens/convection_paper/figure8.pdf We have included the following text in the discussion section 4:

"Differences in the convection schemes themselves (Prather et al., 2001) may be an important, if not the most important cause, of the different effects of convective mixing on the O3 burden in the two studies. In particular, differences in the height and

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strength of convection (Olivieé et al., 2004), efficiency of vertical transport of NOx and O3 out of the boundary layer (Beekman et al., 2003), as well as differences in representing the relevant convective processes (e.g updraughts, entrainment/detrainment, mass-balance subsidence) may be crucial. Figure 8 depicts JJA convective updraught fluxes for the two models for 1998. The STOCHEM-HADAM3 updraught fluxes include both shallow and deep convection whilst the MATCH-MPIC updraught fluxes represent deep convection alone. This explains the lack of strong shallow convection in the lower part of the LT in the MATCH-MPIC model (Figure 8b). The strength of the convective updraught flux in the tropics and mid-latitudes is typically a factor of 2-3 stronger everywhere in STOCHEM-HadAM3 compared to MATCH-MPIC. Whilst HadAM3 overestimates the strength and height of convection in the UT (Figure 1), the MATCH-MPIC underestimates the strength and height of deep convection in the UT (compare 5-10 g/m2/s contours in Figure 8a,b and Figure 1c,d). In particular, there is much less convection at 200 hPa in MATCH-MPIC. This implies more vigourous and extensive deep convection in the STOCHEM-HadAM3 model. The spatial location of convection in the MT and UT are displayed in figure 8 c-f. There is generally agreement in the location of convective updraughts in the MT (600hPa), except over the Eastern Pacific branch of the ITCZ, and the mid-latitude oceans. The large amounts of deep convection over the Himalayas in STOCHEM-HadAM3 (not seen in MATCH-MPIC) are non-precipitating and may be a feature of model orography. In the UT (200hPa) there are much greater differences between the simulated convective updraught fluxes. STOCHEM-HadAM3 exhibits considerably larger mass fluxes across the ITCZ and shows areas of deep convection in the mid-latitudes, in contrast to MATCH-MPIC. Olivié et al., (2004) compared the impact of using convective mass fluxes derived from two different methods on tropospheric ozone distribution. One source of convective mass fluxes was from ERA-40 reanalyses, the other used a convective parametrisation based on wind,temperature,evaporation and humidity from ERA-15 reanalyses and derived convective mass fluxes “offline”. Both derived mass fluxes are based on the same underlying Tiedke (1989) parametrisation, but have dif-

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ferent modifications to their scheme. The datasets also have different temporal averaging properties (time-averaged vs, instanteous). One of the main differences reported by Olivié et la., (2004) was higher convective mass fluxes in the ERA-40 reanalyses compared to those diagnosed offline.

We suspect that the derivation of convective mass fluxes in HadAM3 is not too dissimilar to that in ERA-40. In L03 the MATCH-MPIC model used reanalyses fields to derive convective mass fluxes, which.suggests their derived convective mass fluxes are more similar to the offline fluxes calculated from ERA-15 fields. Thus, the difference in convection height are in agreement with the findings of Olivié et al., (2004).

Comparing the two schemes Olivié et al. find lower O3 and NOx concentrations in the UT and higher concentrations in the free troposphere as a result of the higher convection in the ERA-40 scheme. This promoted greater transport of O3-poor air from the surface to the UT, as well as more subsidence to the MT. Therefore, we expect that it is the higher and larger amount of convection that leads to the large reduction of ozone in the UT in STOCHEM in the tropics which is not seen in the MATCH-MPIC simulations. Olivié et al., (2004) further suggest that the lower O3 in the tropical UT is transported to higher latitudes through latitudinal transport and transport downward along the subtropical front. Their global decreases in O3 using the ERA-40 reanalyses are in qualitative agreement with the patterns of change shown in Figure 6a. In the mid-latitudes, larger ozone concentrations in the MT in the simulations using the ERA-40 reanalyses arise from enhanced downward transport in the higher latitudes. Despite enhanced convection in the midlatitudes in STOCHEM relative to MATCH, the MT increases are smaller. Therefore, we suspect differences other than those arising from the convections schemes, are likely to be contributing to the constrasting effects of convective mixing displayed by the STOCHEM and MATCH-MPIC models."

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