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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 #1

1) I really don't understand the global ozone budget with and without convection. The tendency equation for ozone consists of transport, production and loss. In a global integral the transport of ozone cancels out (due to mass balance considerations). The

result is the globally integrated ozone can only change through net ozone production (neglecting dry deposition). Global net ozone production increases in the author's model, but global ozone decreases. The authors attribute this inconsistency to the fact the ozone lifetime decreases (e.g., page 3759). I do not think this explanation is correct. The problem is the lifetime (defined, I presume as the inverse of the ozone loss) does not take the production of ozone into account. In the author's analysis the production and loss increase with convection, but the production increases more. Thus, ozone should increase even though the lifetime decreases. This suggests that either: a) there is some problem in the analysis, or b) transport does not cancel because their global budget is not really global (e.g., the budget is over the troposphere so that the ozone decrease is due to differences in the stratosphere-troposphere exchange). If the result can be attributed to differences in stratosphere-troposphere exchange, this suggests a large sensitivity to convection. It is not at all clear to me that a 9-level model can accurately simulate this sensitivity.

We agree that the interpretation of the ozone budget is somewhat counter-intuitive; however we stand by our original explanation. In addition, we offer some further clarification. The tropospheric ozone budget can be expressed as: d[BO3]/dt = P + S - L - D (1) where the left-hand term is the rate of change of the tropospheric ozone burden (BO3), which, over long time periods, such as an annual cycle, is small so we set this to zero for illustration (but see below). P is the chemical production; S is the influx from the stratosphere; L is the chemical loss; and D is the surface deposition flux. All have units of Tg(O3)/yr. Net chemical production (NCP) is simply defined as: NCP = P - L (2) From simple inspection of equation (1) it is apparent that NCP is also equal to (D - S). In effect, this means that NCP is wholly defined by the fluxes of ozone at the upper and lower boundaries of the troposphere. As such it is not a useful quantity to define the chemical activity of the troposphere - the absolute production and loss fluxes are much more useful. The ozone lifetime (O3) is defined as: O3 = BO3 / FO3 (3) where FO3 is the 'overturning flux' of ozone, given by: FO3 = L + D (= P + S) (4) These simple equations show that there is absolutely no inherent direct relationship between NCP and BO3. Rearranging equation (3), it is apparent that a higher (lower) burden reflects a longer (shorter) lifetime and/or a faster (slower) overturning flux. Comparing the convection on and convection off experiments (Table 1), the lower burden in the base convection on case is accompanied by a shorter lifetime, but a faster overturning flux. Clearly, the shorter lifetime is the dominant change. The shorter mean lifetime can be understood in terms of a redistribution of the mean ozone profile (e.g. Figure 6a) - decreasing UT O3 (where the lifetime is longer), and increasing LT O3 (where the lifetime is shorter). We don't believe that the minor changes in stratosphere-troposphere exchange are important.

Although the term d[BO3]/dt is small and can in general be ignored, we found that by including this small term (less than 10 Tg(O3)/yr) in our budget analysis, using interannual variations in burden, slightly more accurate results were achieved. Therefore we will use this term in our revised Table 1.

3) I'm somewhat disturbed by the lack of observations in this paper. The authors should show, or give references to the fact that the model does a reasonable job at simulating upper tropospheric ozone. How well does the model predict precipitation?

See referee ML comment 1. 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. For precipitation, the distributions compare favourably, see referee ML comment 4 for more detail.

4) The authors made extended simulations (20 years) with convection on and off. This should allow them to include the interannual variability in Table 1. They also should be able to make an assessment of whether the differences between the two cases are significant or not.

These will be added to Table 1. We find that all the changes are significant, except the changes in stratospheric influx.

5) The authors show qualitatively that convection decreases upper tropospheric NOx through conversion to PAN. Can the authors make this argument more quantitative: what fraction of the NOx decrease can be explained by the PAN increase?

See referee ML comment 6. We cannot say quantitatively what fraction of the NOx decrease can be explained by the PAN decrease, as we do not have a suitable tracer species. However, comparing changes in NOx and PAN in the UT (350-150hPa) we find UT NOx decreases by 0.032 Tg N and a UT PAN increases by 0.028 TgN. Therefore, in terms of N conservation, it seems fairly clear that the main process occurring is that NOx is being converted to PAN.

8) While the authors extensively discuss on the interplay between convection and chemistry in the tropics, they do not really touch on this subject in the mid-latitudes. One might think that transporting NOx to the middle and upper troposphere would increase ozone net production. Could the authors comment a bit more on the importance of chemistry in the mid-latidudes (it seems to be mentioned in passing on the bottom of 3757, the top of 3758)?

Net chemical production does increase slightly in the northern mid-latitudes when convection is switched on. We will add text about chemical production and destruction in the mid-latitudes to the discussion.

Technical Comments

1) I could not find information on the global production of NOx from lightning.

Global lightning NOx emissions are 7 TgN/yr.

2) Due to the importance of the convective scheme in this paper, it would be appropriate if the author's could briefly describe its characteristics. For example, does the convection scheme include downdrafts?

We will expand on the description in section 2. This convection scheme does not include downdrafts.

3) How is ozone production and loss computed? These quantities depend on the definition of odd oxygen.

The ozone budget is defined below. The scheme was devised by Colin Johnson; the basis of it is that: O3, O3P, O1D, PAN and NO2 are ’worth’ 1 O3 molecule; N2O5, NO3, HNO3 are worth nothing.

Ozone chemical production terms: NO+HO2 NO+CH3O2 NO+RO2 (several terms)

Ozone chemical destruction terms: O1D+H2O O3+OH O3+HO2 O3+hydrocarbons (C2H4, C3H6(2 routes), C5H8, MVK)

Other ’net’ O3 losses (some, somewhat confusingly, are sources (negative ones)): -1 x HNO3+hv = NO2 + OH -2 x NO3+hv = NO2 + O3P -1 x N2O5+hv = NO2 + NO3 +2 x NO2+O3 = NO3 + O2 -2 x NO+NO3 = 2NO2 +2 x NO2+O3P = NO + O2 +1 x NO2+NO3 = N2O5 +1 x NO2+OH = HNO3 -2 x NO3+NO3 = 2NO2 -1 x N2O5 = NO2 + NO3 -1 x NO3+HO2 = NO2 + OH +1 x PAN+OH = NO3 + HCHO -1 x RNC2H4+OH = HCHO + NO2 -1 x RNC3H6+OH = CH3CHO + NO2 -1 x RNC5H8+OH = CH3CHO + NO2 +1 x NO2 dry deposition +1 x PAN dry deposition

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