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Atmospheric
Chemistry
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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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Here is in our final response. All comments are addressed.

Responses to referee 1 (referee's comments in italics).

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 (tO3) is defined as:

$tO3 = BO3 / FO3$ (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.

2) I am somewhat surprised at Figure 6a and the author's inter pretation. If I understand the authors correctly they interpret the convective mid-latitude upper troposphere and lower stratosphere ozone decrease as a global propagation of the tropical signal. The author's make a heuristic argument for this based on the height of convection, but do not show more definitely that the signal propagates from the tropics to the mid-latitudes. In fact there are some reasons to suspect it shouldn't: both Bowman and Carrie [2002] and Pierrehumber t and Yang [1993] suggest a barrier to mixing between the tropics and extra-tropics. The standard wisdom is that significant upper tropospheric transpor t across the subtropical jet only occurs in the N.H. in association with the summer monsoon (e.g., Chen,1995). Yet Figure 6 also shows strong transpor t to the Southern

Hemispere. Could the authors give more conclusive evidence that the signal they show is indeed due to a poleward propagation of the tropical signal?

We thank the referee for this comment. The annual-average large-scale mean flow pattern does suggest flow in the UT along the subtropical jet in the Northern hemisphere, but the large-scale flow does seem inhibited in the Southern Hemisphere. We have revised figure 1 (see response to referee ML comment 4) which shows mid-latitudinal convection at 200 hPa over some continental regions in the northern mid-latitudes and in the southern oceans. We have revised our text accordingly. Olivie et al., (2004) also suggest that the lower O3 in the tropical UT is transported to higher latitudes through latitudinal transport and transport downward along the subtropical front.

We have revised our text. We assume that convection in the mid-latitudes results in the mid-latitudinal UT ozone anomalies. We also suggest the possibility of some transport along the sub-tropical front in the Northern Hemisphere.

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. A new section 3.1 is added entitled "Evaluation of NOx, PAN and ozone". UT ozone is overestimated in the tropics (by 5-10ppbv) and mid-latitudes. However, the model results are within one standard deviation of the observations. Regarding precipitation see response referee ML comment 4. The model simulated precipitation compares favourably with the GPCP climatology.

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.

Yes, indeed. Annual-mean or 12 month running mean standard deviations have been added to Table 1. Significance has been assessed by use of a student T-test. Text is amended. We find all species and fluxes except stratospheric influx are significantly different between the control and convection off simulation at the 0.001 level. The revised table can be viewed at:

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

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 response to referee ML comment 6. We cannot say quantitatively what fraction of the NOx decrease can be explained by the PAN decrease, as due to data storage constraints we do not archive 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 (50

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

6) The authors have identified a number of competing factors whereby convection influences ozone: transport changes both the ozone and NOx concentrations; chemistr y decreases ozone production through PAN production, but increases the production through modifying the availability of HO2. The net result is obviously dependent on the cancellation of a number of processes. Bar-rring a more quantitative comparison, it would be helpful to make a table of these various processes (e.g., what processes are increasing ozone/decreasing ozone) and to indicate which the dominant processes are.

We have added a summary table as suggested. This can be viewed at:

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

7) Discussion of the N.H. convective influence (page 3756). It is not clear to me if the authors have made any additional cal-culations to ensure that their explanations are indeed correct? For example the annual importance of a convective influence extend-

ing up to 500 mbar during winter seems doubtful to me. Another explanation might be that summer time convection evacuates the boundary layer of NOx, an influence which is transported northwards in warm con-veyor belts. The authors need to make clear if their explanations are fully backed up by further analysis, or if they are simply hypotheses.

The largest changes in NOx in the northern polar latitudes occur in the winter months when surface and MT NOx concentrations are extremely high. The NOx changes at these latitudes are smallest in summer. We have revised text to make our explanation clearer.

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 ozone chemical production does increase in the northern mid-latitudes when convection is switched on. This is because the changes in NOx promotes ozone production in the mid-latitudes in the MT and UT, but there is less change in ozone destruction in the mid-latitudes as this is controlled by changes in the ozone distribution, We have expanded text in this section to include a discussions of changes in ozone chemistry in the extratropics.

Technical Comments

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

Global lightning NOx emissions are 7 TgN/yr. Text is added to section 2.

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 have expanded on the description in section 2. Although the convection scheme include downdrafts, these have not been implemented in the Collins et al., (2002) mixing scheme.

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. The main terms involved in ozone production and destruction (see below) have been added as a footnote to the table.

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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