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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric ozone budget: regional and global calculations" *by* F. M. O'Connor et al.

Anonymous Referee #2

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General Comments

The paper presents and evaluates the ozone budget of the global chemical tracer model TOMCAT. Global and also regional budgets are presented and compared to other studies and in some cases evaluated with available observational evidence. The paper is well written and clearly stuctured.

I believe that there is great benefit to be obtained from a study that focusses on these derived parameters (ozone production and loss) and which therefore can go into more depth in the evaluation of issues related to the ozone budget than the common model/observation comparisons of ozone mixing ratios can provide. However, in its present form, the study does not fully take advantage of this opportunity. From a paper focussing on the budget terms of ozone I would expect more information on how well

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one can trust these new figures and not just a statement that they “are within the range” of previous studies. This is also a problem with citing model intercomparison studies: Not all of budget numbers are equally trustworthy, as in some papers the original authors do find indication that they calculated figure are biased (e.g. STE in the KNMI and MATCH studies). Also, there are additional observational constraints other than from the simulated ozone distribution (e.g. [5]). Sometimes the evaluation is a bit shallow, mostly relying on citing other modeling studies, but not giving sufficient observational constraints, or in a less direct that possible way. Also, more information on calculated fields than given in other model/observation comparison studies in the model should be given to faciliate the discussion and future comparisons (e.g. plots + brief discussion of net-PO3 during January and July at the surface, deposition velocities for ozone).

Some of the comments given below will require further analyses and thus modification of the conclusions, and some only minor additions. If the points raised can be sufficiently addressed, however, the paper should be fit for publication in ACP.

Specific Comments

- Abstract, l.13 and discussion in Section 5.1: It needs to be mentioned that most of the flight data in the UK+Ireland region are taken over the ocean, and probably mostly up-wind of the land. It is therefore not surprising that the chemical regime is not representative of the sampled model grid-cells which most likely contain parts of the land (and thus direct emissions) as well. The conclusion was reached based on the comparison to NO data and this should be stated.
- p.997, l.3ff: More information on the chemical scheme are needed, e.g. which parent hydrocarbons are included, and which aren’t (alkenes, isoprene, aromatics ?).
- Section 3 should be renamed "Previous model validation(s)" as no new valida-

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tions are performed in this section.

- p.999 l.13: Another possible explanation is too low OH in the Southern Hemisphere (possibly linked to the general underestimate of PAN in the model).
- As these are not provided in the other studies validating TOMCAT plots of surface O_3 and the deposition velocity (preverably at 1 m) for January and July would be helpfull in the discussion and for comparison. The latter is especially needed in view of the very high calculated global deposition flux. Additional analysis of this term would be very helpfull.
- l.27: Perhaps it is interesting for the discussion of e.g. the Tropical Pacific region to mention that TOMCAT seems to overestimate HNO_3 in the Southern Hemisphere and underestimate PAN everywhere. This should influence net-PO3 in some remote regions.
- p.1000 l.19: The issue with the sampling frequency is unclear. Any budget calculation must be performed at every model time step. This should be clarified.
- Table 1: It should be explained which processes the "Residual" budgets include. I assume it must be convection, sub-grid diffusion, movements of the tropopause, and perhaps a small net tendency term. Thus the "Transport" category should be renamed to "Resolved transport" or "Resolved advection". Since TOMCAT is using a flux-form advection scheme and is driven by external wind fields, there will also be a significant contribution from a “pressure missmatch” term (see [4]). [3] have recently quantified this error and found it to be quite significant (187 Tg/yr, 87 Tg/yr of which in the troposphere). It is well possible that TOMCAT’s too strong mixing in the UT/LS region can be partially attributed to this problem.
- p.1001 l.28: From the comparisons cited in the last section it should be concluded here that the 850 Tg/yr STE is probably an upper limit estimate. The use of other studies not cited in IPCC could strengthen this discussion.

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- p.1002 l.15: It is unclear how the statement of downward transport of ozone in the tropics is derived. It is much more likely that this is transport from the extratropics.

- Throughout Section 4: The tropical budget term can (and should be) compared with values calculated by [3]. As a suggestion, it might be usefull to also take the ozone budgets recently published (and after the IPCC-report) into account (e.g. [3, 9, 7, 2]).

- p.1003 l.12: In the cited reference only one surface comparison and five at 700 hPa are shown. A different reference should be used here. In fact, also the evaluation with MOZAIC data is too sparse to make firm statements on the general surface abundance of ozone in the model. Information on sites influence by biomass burning and/or strong deposition (like tropical rain forests) would support this point. Unfortunately, no comparison with the standard ozone sonde data seems to be published for TOMCAT. In [1], an overestimate of O3 (SONEX data) of TOMCAT in the lower troposphere is visible. The point that vertical mixing is too strong is thus not well supported. The fact that this would only be consistent with an agreement of surface ozone mixing ratios if at the same time the deposition velocity was too high should also encourage some investigation on the deposition velocities applied. As mentioned above for this purpose a figure depicting these for 2 months is advised. This could be compared to similar plots in other CTM publications. Even better, although perhaps too much to ask, would be an evaluation with measured deposition velocities.

- p.1003 l.26: Other good analyses of the springtime ozone maximum stem from models carrying a stratospheric ozone tracer (e.g. [6, 9]) which can also give a better time resolution of the phenomenon.

- p.1005 l.19: I disagree with the attribution of the discrepancy to continental European sources. More local sources e.g., from around Glasgow which is very

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close to most of the flight positions, is probably sufficient to explain the overestimates of NO. The important point that probably most data are taken upwind of the European source is neglected in the discussion and should be added. An analysis of the wind directions during the different flights would strengthen the analysis. It may also be interesting to analyze the north-western-most part of flight 20000514 separately or to sample the model one or two grid cells farther to the west. Overall, this region seems to be problematic for a comparison with a coarse grid model.

- Figure 3b+c: I think that for the analysis a scatter plot of modelled vs. observed NO colored with altitude would perhaps be clearer. In the altitude plots the direct relation of each model/measurement pair is lost.

- p.1008 l.19: Information on the degree of the underestimate of j(O1D) and its height dependence (not necessarily in a plot) should be added. Is it sufficient to explain the underestimate of ozone loss during the EXPORT campaign?

- p.1008 l.22: The discussion of j(O1D) for the ACTO campaign is interesting and should be added to the dicussion in that section to provide more quantitative information. This is the real in-depth analysis of modelling of the net PO3!

- p.1008 l.25: The sentence with the absence of clouds is unclear: I thought that the 2D model includes a climatological cloud effect. Was this effect switched off in a sensitivity simulation?

- p.1008 l.27: Another uncertainty factor that may be briefly mentioned is $\pm$20% uncertainty in the rate constant of OH + O1D ([8]).

- p.1009 l.1: I assume the answer is “no”, but has perhaps HO_2 also been measured? This would of course then be an even more important term in the ozone budget. Also, only **some** NMHCs are missing in the chemical scheme, right?

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- p.1009 l.17: Does calling for potentially missing NMHCs in TOMCAT (such as short-lived alkene and aromatic species) as an aditional factor imply that the underestimate of highest NO values cannot fully account for the underestimated O3 production rate over central Europe?

- p.1011 l.27: The statement is unclear: Wouldn't this depend on the altitude at which the pollution is imported into the region? If this is above the boundary layer it is not related to the boundary layer height.

- p.1012 1st paragraph: Here is an example where a plot of net-PO3 would be usefull. In general this would give a wider overlap with evaluations in other studies and mitigate the somewhat arbitrary choice of regions being discussed in Section 5.3

- Conclusions, p.1014 l.10ff: Again, as far as I understand this, the argument of a too efficient boundary layer mixing only holds if also the deposition velocity were too high. More precisely these are actually only higher than in other models included in the IPCC summary, since no direct information on the deposition velocities of ozone is included in this study. It is noted that for example an overestimate of dry deposition rates or an overstimate of ozone mixing ratios over the tropical rain forest (e.g. due to the lack of the ozonolysis of isoprene) could strongly affect the global dry deposition term.

- p.1014 l.24: The generall caveats of the comparison in this region does not allow such a statement. If the analysis is extended this can be rephrased, otherwise mentioning the NO disagreement and its likely cause is as far as one can go.

- p.1014 l.29: An attribution to one input parameter cannot be made unless the evaluation is more quantitative. Strictly, this would require a constrained box model, but perhaps some indication can be given with back-of-the-envelop calculations of simple steady state considerations.

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Technical corrections

None, as far as I can see.

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