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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.

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General Comments

This paper describes the global ozone budget calculated in a 3-D chemical transport model (TOMCAT), and presents ozone budgets over six different regions, comparing with aircraft measurements where available. It concludes, as other studies have done, that the troposphere is a net source of ozone, dominated by production over continental regions which outweighs loss over remote marine regions. The use of well-chosen diagnostic regions is a novel and valuable addition to the global budget analysis, allowing exploration of ozone behavior in different areas and better testing of model performance.

The paper is well written, logically organized, and gives full credit to previous work in the field. My principal concerns are that (1) there are sufficient questions about

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the model treatments of deposition, photolysis and stratospheric exchange to call into question the budgets calculated, and (2) there is little additional insight provided by the study over what is already available in the literature. These criticisms do not preclude publication - the budget calculation from this model is a new and valuable addition to previously published global chemical model studies. However, better justification of the calculated terms and their associated uncertainties is required if the authors are to reassure readers of the validity of their estimates. The regional budgets provide a new and original way of doing this by providing small windows in which to evaluate the model, but the present analysis (particularly for the ACTO flights) is weak. A better evaluation of the uncertainties involved would improve confidence in the budget presented, allow more substantial conclusions to be drawn, and strengthen the paper considerably.

Specific Comments

The ozone budget presented here is compared with that from previous model studies recently reviewed in IPCC 2001 and EC 2003. There is considerable variability in these previous studies, and recent convergence on the principal terms involved may reflect greater use of similar algorithms as much as any real improvement in model performance. New studies such as this using different algorithms are thus very welcome, and disagreement does not reduce their value. However, where there are discrepancies, they need to be explored in greater detail to reassure us that the new values are really valid and not merely a consequence of known model inadequacies. Determining the sensitivity of the budget terms to key processes would quantify these uncertainties, and make it clearer how much they influence the results. The deposition term should be the principal focus here, as this is the term that differs most greatly from previous studies - see suggestions below. In its present form, the paper does not inspire confidence in the calculated budgets, and thus appears weak.

Choice of the "UK and Ireland" region (Section 5.1) is fine, but comparison with ACTO flight data is inappropriate. The ozone budget in the region is strongly skewed by the

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presence of continental sources, while the flights are principally over marine regions, upwind of sources much of the time. These boundary regions (marine/continental, clean/polluted) are difficult to model correctly at relatively coarse resolution. The flight data is not a good test for the model, as the data is not representative of the diagnostic region chosen. The final paragraph of section 5.1 suggests that there is some discrepancy between net production over the region and net loss calculated for the ACTO flights, but these can of course both be correct. Comparison with a marine region northwest of the true flight tracks would be fairer, but is somewhat artificial.

The deposition code used is sensitive to meteorology and to surface type (page 998). However, strong deposition over the Sahara (page 1012 and Table 3) suggests a problem; deposition to sand should not be much greater than to the vegetated continental regions of the Midwest or Europe. A sensitivity study is called for here, perhaps varying the dependence of the deposition velocity on the diffusion coefficient, to explore the sensitivity. The scheme used here is based on Berntsen and Isaksen [1997], which gave the highest global deposition rates shown in the IPCC review; the present study has 30% greater deposition than this.

One thing missing from the conclusions is some assessment of the confidence in the global budget presented here against that from previous studies. How reliable are the calculated budgets? Given that a number of changes in model treatments are needed (deposition, photolysis and stratospheric input), how sensitive will the results be?

Abstract, lines 13-15: "...but this is not consistent..." As indicated above, comparison with marine flight data within a largely continental region is inappropriate, and the differences are not inconsistent. This sentence needs to be rephrased to make this clear, and "over the Atlantic/Irish Sea" added to the end (for example).

Page 992, line 21: Remove "Although only a trace gas". No need for an apology here - CO2 and CH4 are only trace gases too, yet dominate climate change.

Pages 992-996: The introduction provides a detailed overview of tropospheric ozone,

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but is longer and more basic than merited here. This review could easily be trimmed by removing some of the more elementary statements (such as reminding us that dry deposition is "a process by which certain trace species are transferred from the atmosphere to the Earth's Surface", page 995, lines 10-11). A more succinct introduction highlighting the key issues and processes addressed here would be beneficial.

Page 997, lines 2-3: it would be helpful to indicate which NMHCs are included, but avoiding a detailed listing. It appears later (line 22, for example) that isoprene isn't included - what else isn't covered?

Page 998, line 3: methylaldehyde is another name for formaldehyde (HCHO). MeCHO is usually referred to as acetaldehyde. IUPAC names avoid all this confusion, of course, but don't appear to have caught on in the atmospheric community yet.

Pages 998-1000: The evaluation of model performance here is very useful. However, the model-model comparisons provide less insight than the model-measurement comparisons. The last line of the section (page 1000, lines 2-4, "Each model...") is vague and should be removed.

Page 1000, line 27: What does the "residual" here actually represent - an artifact of disequilibrium/poor initialization, numerical drift or real trends? This needs to be stated if it is to be included in Table 1.

Page 1001, lines 11-16: The IPCC 2001 and EC 2003 reviews give different selections of results from published ozone budgets, so it would be better to consider them together rather than as separate assessments. Consideration of the minimum and maximum terms to define the range is also somewhat misleading, as the consensus value rarely lies midway between these outliers.

Page 1001, lines 21-22: "the net photochemical term being the sum of two large but oppositely signed terms" - this is a common but misleading statement. In most chemical schemes, the net photochemical term is integrated fully, while gross production

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and loss are approximated based on the NO + peroxy radical reactions under the assumption that ozone production is globally dominated by NOx cycling. In many cases the net term may be more reliable than gross production and loss. More important to state here is that chemistry, deposition and stratospheric input are closely coupled. The stratospheric impact is mentioned in the next sentence (lines 22-24), but it is also clear in the present case that if deposition were reduced then the net chemical term would also drop significantly, as chemical loss increases.

Page 1001, line 29: "is well within the range" - this is certainly the case, but comparison with these early model studies is not very reassuring! The best observation-based estimate of STE is about 550 +/- 140 Tg/yr [Olsen et al., 2001, JGR pp. 28771], and this fits well with the earlier statements in the validation section (page 999) that there is a small overestimation of ozone in the upper troposphere compared with measurements.

Page 1002, line 3: STE may or may not be sensitive to model resolution, depending on how meteorology is calculated or used. The calculation technique or boundary condition used (stratospheric chemistry vs. imposed fluxes or concentrations) is a larger source of error.

Page 1002, lines 15-19: it would be worth commenting on the location of STE as well as on the magnitude - Table 1 suggests that input in the southern hemisphere dominates, in contrast to available assessments (e.g., papers by Appenzeller et al.)

Page 1003, lines 12-15: "too well-mixed boundary layer" - this is possible, and easy to test (by scaling the vertical diffusion coefficient in the non-local PBL scheme by a factor of, e.g., 0.01 and leaving it unchanged in the deposition scheme). However, given that the ECMWF surface meteorology is relatively good, I suspect that the deposition algorithm is responsible for much of the overestimation.

Page 1005, lines 2-5: As noted above, the flight tracks were not representative of the region as a whole - this needs to made clear here.

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Page 1008, line 19: The difference between modeled and measured J(O1D) rates is important here. A climatological cloud field is not suitable for instantaneous comparisons, but given that this is all that can be used, some quantification is still required (mean profile along all flights?) Based on Fig 6a, better than a factor of 2?

Page 1009, line 14: "lack of non-methane hydrocarbons" - is this referring to isoprene, or to other NMHCs (see point made above for page 997). Note that maximum production rates will inevitable be underestimated due to model resolution (as noted) - median production rates make a better comparison. Note also that underestimated J-values would lead to underestimated production here through the impacts on NOx cycling and peroxy radical formation.

Page 1010, line 6: why are the regions compared during May - is this considered a representative time of year, or is this merely for convenience over the ACTO time period?

Page 1012, line 12: rapid dry deposition over the Sahara strongly suggests a problem in the treatment of deposition. The region is likely to be a sink of ozone, but the very efficient deposition seen here is unexpected. Justification for these findings are required here.

Page 1014, lines 10-13: As for the page 1003 comment above - the performance of the dry deposition scheme should be questioned, not just the boundary layer.

Page 1014, lines 23-24: a caveat on the ACTO comparison is required again here.

Figure 6: indication of the mean slope on these figures would be helpful, as it provides some quantification of model performance.

Figure 8: the individual days shown in the figure cannot be distinguished, and it would therefore be more clearer to present the mean profile and show the variability by 1-sigma error bars (the max and min can also be shown if desired).

Technical Corrections

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Interactive Comment

Two papers are included in the references but not cited in the text (Kentarchos and Roelofs [2003] and Wild et al. [2000]), and the "Y" entries come before the "W" entries!

Typos

Page 1007, line 27: tranporting –> transporting Page 1012, line 25: Isakesen –> Isaksen

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Interactive comment on Atmos. Chem. Phys. Discuss., 4, 991, 2004.