Review of "Global tropospheric hydroxyl distribution, budget and reactivity", by J. Lelieveld et al.

General comments

This interesting and well written paper describes sources and recycling of OH and HO_2 in the EMAC model. In a few places, I found the text could benefit from clarification, and I think some additions/revisions are needed. A full budget of OH is not presented, and the three 'OH recycling' mechanisms are not well defined. I also found it a bit surprising that no comparison with measured values was included. The modelled methane lifetime seems quite short compared to the accepted value – this is a common feature of models, but it goes unmentioned (and I think the comparison with the real world gets worse as more recycling mechanisms are added). Direct comparison to OH/HO_2 measurements is also not evident – we are directed to other papers and the model web-site (p4 l22-23), but these papers seem (from their titles) to evaluate other aspects of the model, not HOx, and I couldn't find any mention of the EMAC model version on the web-site. Some statements in the text appear unsubstantiated or a bit over-blown (see specific comments below). Having said all that, I think if these comments and those listed below are adequately addressed, this paper will make a very useful addition to the literature and should be accepted for publication.

Specific comments

P1 l26 Spivakovsky

P2 l4 combine, not recombine

P2 l7 This sentence implies RH features in the previous equation(s), but it doesn't?

P2 l23 I find the definition of 'r' (= 1 - P/G) a bit obscure. Isn't r = S/G clearer?

P3 Paragraph from l29 onwards. This paragraph is perhaps not immediately comprehensible to most readers. I suggest you try and make it a bit less technical/more accessible.

P4 l12 …notably FOR carbon…

P4 l30 …in SOME previous… (Not all previous atmospheric chemistry-transport models have had to artificially reduce natural VOC emissions.)

P5 l11 may be -> are

P7 l13 Plane -> Plain

P7 l25 "…tropospheric production of HO_2 – and thus HOx…" I don't think this follows. Isn't most HO_2 production associated with OH destruction (i.e. HOx recycling, e.g., R3 and R4)? Production of HOx is thus only primary production of either OH or HO_2 (i.e. from R1/R2 and HCHO photolysis), whereas production of HO_2 is dominated by conversion of OH to HO_2. Thus production of HOx and production of HO_2 are quite different.

P8 l11 Dividing better than 'relating'.

P8 l23 I wondered what "strongly underestimated" meant here (it is rather non-specific). I would say normally something that is strongly underestimated is 50% or less of its correct value. From your budget in Figure 6, if the VOC reactions were simplified/not included, I don't think OH reactivity would be underestimated by as much as 50%. So I think you are being over-dramatic and non-quantitative, which is unhelpful.

P8 l29 '…indicates that air masses that traverse the TTL into the stratosphere have been largely cleansed from compounds that react with OH'. This seems like overstatement – aren't CO and CH_4 the two main compounds that react with OH in the troposphere? I don't think TTL air is 'largely cleansed' of these two gases?

P8 l30 onwards. The 'side note' about SO_2, OH, halocarbons and stratospheric O_3 seems a bit odd.

P9 l3 onwards. The discussion of the methane lifetime (to oxidation by OH) of 8.5 years in the model should also be compared to observational estimates (e.g., Prather et al., 2012: 11.2 ± 1.3 yr). Pretty much all models, and EMAC with MOM seems not to be an exception, apparently underestimate the methane lifetime. Do we have any idea why this is? It seems that we need less OH in model's atmospheres, but by adding new OH sources from recycling this discrepancy gets worse. Doesn't this suggest that models are missing something fundamental about OH?

P9 l31 '…over the oceans G is the same as over the continents.' G is defined earlier (p2 l23) as 'gross OH formation'. I am unclear whether you mean G over the oceans as a whole compared to G over the continents as a whole, or if you mean per unit area. Obviously this makes a big difference.

P10 l7 '…S is also the same over the oceans and continents…' Same query as previous.

P10 l11 '…P declines steeply with solar radiation and water vapor.' Figure 7 show that P declines steeply with increasing altitude (ignoring the stratosphere) and latitude. Water vapor declines with increasing latitude and altitude (so that's OK). Solar radiation declines with increasing latitude, but increases with increasing altitude. So the relationship of P with solar radiation seems more complex than stated.

P10 l30 'is subordinate to' -> is less than?

P11 l1-2 Isn't 'r' larger in the extra-tropics mainly just because P is small?

P11 l2 The last sentence is true for the MBL but not the CBL, so it is incorrect for the BL as a whole.

P11 l4-13 Some clarification of what is exactly meant by the NOx, O_3 and OVOC 'mechanisms' of OH recycling is needed. The earlier reaction equations and discussion is very good and useful, but I am not completely clear on which reactions make up each mechanism.

P11 l13 Do you mean from the FT to the BL (rather than 'transport in the FT')?

P11 l17 'The complementarity of the three mechanisms is remarkable.' Is it? Don't they have to add up to 100% by definition? Figure 10 is certainly interesting, but I am not sure it is 'remarkable'. As suggested earlier, clearer definitions of the three mechanisms would help the discussion.

P12 l4 Have you demonstrated in this paper that including MOM 'increases OH reactivity'? I can believe this is the case, but I don't think you present evidence of what the OH reactivity was in the model before you included MOM.

P12 l13 I note your reference to 'measurement campaigns'. There is no comparison with observations in this paper, which seems like an oversight. Can you demonstrate that modelled OH is improved and compares well to reality?

P18 Table 1. The caption doesn't adequately describe the table – which contains fluxes for HOx primary production (O^1D+H_2O), recycling (NO+HO_2, O_3+HO_2, photolysis reactions) and loss (H_2O_2 deposition). This table could be more comprehensive, and describe the full OH and HO_2 budgets, i.e. include all the primary sources, OH to HO_2 inter-conversions, and sinks (e.g., Derwent, 1996). The sources and sinks should balance (this is not obvious from the current table). If this were done, it could also clarify the definitions of the three mechanisms, as suggested earlier.

P19 Figure 1 (and all zonal mean plots). It looks like surface pressures go up to 1000 hPa everywhere, but that can't be the case over Antarctica (etc.). Is the vertical scale really pressure?

P21 Figure 6. Related to my comments on Table 1 – I note the caption says 'Main' production terms of OH… Wouldn't a figure that shows *all* the OH sources be more useful?

P24 Figure 9. The text defines recycling efficiency as (S-P)/G. This allows it to take on negative values (where P>S). I find this a bit confusing, as efficiency normally refers to a number between 0-100%. The recycling probability (S/G) does just go from 0-100%. I'm not sure you need both quantities?

P25 Figure 10. Again, clear definitions of the three mechanisms are needed.

References

Derwent, R. G. (1996) The Influence of Human Activities on the Distribution of Hydroxyl Radicals in the Troposphere, Phil. Trans. R. Soc. Lond. A, 354, 501-531; DOI: 10.1098/rsta.1996.0018

Prather, M. J., C. D. Holmes, and J. Hsu (2012), Reactive greenhouse gas scenarios: Systematic exploration of uncertainties and the role of atmospheric chemistry, Geophys. Res. Lett., 39, L09803, doi:10.1029/2012GL051440