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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A Tropical West Pacific OH minimum and implications for stratospheric composition" *by* M. Rex et al.

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General comments

This paper is directly related to a growing recognition that better understanding of troposphere – stratosphere couplings is required in atmospheric chemistry. It is also relevant for the major questions that still exist about how OH will change in response to increasing concentrations of the reduced species, and the potential for competing influences on it to vary from one region to another. While global average values for tropospheric OH are reasonably well known, there still remain systematic discrepancies between models and observations for the distribution of OH, particularly between the northern and southern hemispheres [e.g. Bousquet, et al. Atmospheric Chemistry

and Physics 5, 2635-2656, 2005; Naik, et al. Atmospheric Chemistry and Physics 13, 5277–5298, 2013].

As noted in the paper, observations of very low tropospheric O3, and so presumably also OH, were seen 20 years ago in the boundary layer over the Pacific ocean [Kley, et al. Science 274, 230-233, 1996]. Also some models, such as OsloCTM2, have indicated a zonal variability by a factor of $\sim$4 in boundary layer OH across the tropics at some times of the year [Berglen, et al. Journal of Geophysical Research 109, D19310, 2004]. However, this paper now presents a strong case for O3 and OH to both be exceptionally low from the surface to the tropopause in the region that has a predominant role in troposphere to stratosphere transport.

Given that the context for this paper is very important, I would recommend that it be revised to provide more details on the different types of data being used, and on the level of detail that has been used in the model based analyses. Also while the authors will be very familiar with what is covered in several other related papers about atmospheric chemistry in the Western Pacific region, it would be helpful for a broader audience if some key points from other papers were also summarised here to set the context for this work more clearly.

More specific comments

P 28870, L 25: Additional references that would be relevant here are Lelieveld, et al. Atmospheric Chemistry and Physics 4:2337-2334, 2004, and Berglen, et al. Journal of Geophysical Research 109, D19310. Both of these clearly show tropospheric OH distributions that are low in the Western Pacific, but that is quite specifically only for the boundary layer.

P 28871, L 9: After equation (R3), I would recommend adding a reference to Naik, et al. Atmospheric Chemistry and Physics 13, 5277–5298, 2013, and, to set the general context for this paper, to note that there are still some systematic discrepancies between models and observations for the spatial distributions of OH. Then to set up the

regional context for this paper it would be helpful to say specifically that Fueglistaler et al, 2004 had shown that about 80% of the cross tropopause flux is occurring in the region that is being covered in this work.

P 28871, L 10: "2. Measurements and chemistry ..." This section would be easier to follow if it had a clearer separation between coverage of the measurements and then of the chemistry analysis. Also it refers to 'Appendix' four times but never says which one, and there are five appendices.

P 28871, L 20: This is making a point about the context that I think would be better set out towards the end of the Introduction, however, if it stays here it should be made clearer for many readers by noting the relative magnitude of cross tropopause transport occurring in this key region, e.g. similarly to the way it is summarised in Fueglistaler et al, 2004.

P 28872, L 1: Presumably the appendix referred to here is Appendix A.

P 28872, L 9-10: The effective lifetime of ozone is being mentioned here but the specific value that is given as 4 days is for odd oxygen. For many readers it would be helpful to say something briefly about the relationship between these two lifetimes. Also which Appendix is the one being referred to here.

P 28872, L 11-15: This is presumably a general summary of the processes, but it could be clearer. For example, it is not being made clear to what extent low O3/OH concentrations right up to the tropopause are being attributed to strong vertical mixing combined with rapid removal within the boundary layer, or whether significant removal must also be occurring above the boundary layer.

P 28872, L 16-28: This paragraph is dealing with transport by advection to the tropopause level for the case shown in Fig 2 but the links between this and the previous paragraph are not being made clear. E.g while Appendix B notes that the STRAT/ER-2 measurements used here are for 1995-1996 it would be better if the main text

also explicitly noted that data from different time periods was being brought together in this analysis.

P 28872, L 28: Presumably a key point being made by this use of the ER-2 data is to provide evidence that the depletion of O3/OH can persist when the air mass becomes transported to higher latitudes and eastward. But it is not clear to what extent there are data supporting that point. Is it just for a single case shown by the outlier points in Fig 2 and the air trajectory in Fig A3? Also the extent to which entrainment of air originating from different latitudes over the 12 day back trajectory could alter the OH value depends on the latitudinal gradient for OH. This has a significant seasonal cycle at latitudes around 24°N, so the time of year when the data were collected can be significant. This is not to argue that the data being shown in Fig 2 is irrelevant, but rather to suggest that more details need to be provided.

P 28873, L1-11: These two paragraphs alternate between talking about the GEOS-Chemistry model and the data that is being used. It would be clearer if the text was reorganised to separate these parts out more clearly and to also integrate the points that are being made here with those at the start of the next section 3.

P 28873, L 12-13: "3. Model calculations ..." An important point to make here, for those who have not read (and remembered) Ridder et al (2012), is that the GEOS-Chemistry model is a 3-D chemistry and transport model and that the analysis being cited there was done specifically for the same region and time period as is covered in Fig 3.

P 28874, L 6-17: It would be helpful to say why the ATLAS Lagrangian transport model is being used here and the extent to which changes in chemical composition, as treated in that model, are consistent with the GEOS-Chem model. The last sentence in this paragraph is making a key point and so setting out some more of the details that lie behind it is important.

P 28874, L 12-17: This seems to be implying that advection of OH can be significant

for stratospheric chemistry but, for this to be the case, OH would also need to have a much longer lifetime than its normal one of a few seconds. So isn't the issue here really about the atmospheric composition of the air masses that are ascending into the stratosphere not having the normal balance between O3, NOx, and the more reactive reduced species, that produces OH.

P 28874, L 13: Presumably a new sentence should start here at "Combining the . . ."

P 28874, L 21: Which model is being used to produce the results shown in Fig 4a?

P 28875, L 8-12: This paragraph would be better if it was reworded so as to introduce the additional analyses which are then summarised in the next two paragraphs. Starting by referring to 'our findings' implies that it is about what has already been covered in the earlier parts of the paper, whereas it is actually now introducing the use of the AER model and that is described below.

P 28875, L 11-12: A number of different explanations have now been published for a 'decrease' in global warming following the extreme El Niño year of 1998 (e.g. see the IPCC Working Group I report released in September) and it is a bit misleading to imply that the Solomon et al 2011 paper suggested aerosols were a 'key' element in this, particularly as that paper was focussed on stratospheric aerosols being 'persistently variable'. So I think it would be better to say something like '. . . a key element for stratospheric chemistry and potentially contributing to the recently observed decrease in global warming rates'

P 28876, L 1-8: This paragraph starts by talking about 'these background conditions' and that does not seem to connect with the previous paragraph which ends by talking of increasing SO2 emissions together with a long term increase in aerosol loading. In fact this paragraph seems to be repeating points that were made in the last two paragraphs. Is it necessary?

P 28876, L 9-14: This does not seem to be a very useful final paragraph. Whether a

different form of atmospheric chemistry in this region has a significant effect on tropospheric concentrations of HCFCs or HFCs seems debatable because of its relatively small volume. However, given that it is being identified as an anomalous situation, and the potential for non-linear responses to occur in atmospheric chemistry, it certainly deserves a lot more analysis. But the bigger issue seems to be whether this OH minimum has been formed recently, e.g. by increased surface temperatures in the Western Pacific ocean leading to more rapid convection and more cross tropopause transport as is generally suggested in climate models, or by the very large relative increases in atmospheric hydrocarbons compared to the preindustrial era, or whether it is natural feature that has actually existed for a long time. So I think that the authors would be justified to finish by making the case that this has opened up a new question about atmospheric chemistry which requires more analysis.

Appendix A: This is a very clear description of the ozonesonde measurements and a useful summary of the ways in which uncertainties have been dealt with.

Appendix B: As noted earlier, the time of year when the OH measurements were made can be significant. Also has any model based interpretation of the OH data that is being used here been covered in a previous publication. E.g. is it part of what was covered by Wennberg, et al, 1998, Science 279, 49-53.

Appendix C: No reference is being given here to a paper that would provide more details about the Tropospheric Emission Sounder (TES) data and, while this paper mentions selection of 'Level 2 profile data' and the 'master quality flag', there is no way of finding out what that means. I think that a reference should be given to cover this or else more specific details about the data quality be provided in this Appendix. Also the TES data shown in Fig 3 is being used in the main part of this paper whereas Fig A4 seems to be providing some more background information for that over the previous years. But that data and its implications for some interannual variability are not being mentioned in the main text. So what are we meant to learn from Fig A4?

P 28879, L 21: In Appendix D, this sentence seems to have a mistake in it. Should it say that the vertical winds are calculated from the horizontal winds and heating rates?

Appendix E: Is a clear summary of the process used in Figs 3 and 4.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 28869, 2013.