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Interactive comment on "Midlatitude ClO during the maximum atmospheric chlorine burden: in situ balloon measurements and model simulations" *by* B. Vogel et al.

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Review of Vogel et al. "Midlatitude ClO during the maximum atmospheric chlorine burden: in situ balloon measurements and model simulations", manuscript ACPD, 5, 875-909, 2005.

Overall this is a very nice analysis of two balloon profiles of ClO obtained at mid-latitudes during 1996 and 1999. The measurements are shown to exhibit overall good agreement with model calculations. There are a few discrepancies between model and measured ClO that lie outside the range of measurement uncertainty, but taken as a

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whole, the analysis shown in Figures 6 and 7 supports the view of very good understanding of ClO photochemistry. There are a few aspects of this paper that should be improved, upon revision, but these should be straightforward to address. The first aspect is the need to state that the observations were obtained not only at a time of maximum chlorine loading, but also during a time of low aerosol levels. The aerosol profile used in the analysis should be described and added to certain tables. Finally, even though the paper does an admirable job of providing documentation of the calculations, a few more items should be added to the tables and an extra figure should be shown, as described below.

Major comments:

1. The aerosol levels that prevailed in 1996 were fairly low, and in 1999 were even lower. This aspect of the "condition of the measurements" should be mentioned in the Introduction, with some appropriate references to aerosol climatology.

The value of stratospheric sulfate aerosol loading used in the calculations should be added to Tables 2, 3, and 4. The source of the aerosol data should be indicated in Table 1. If the aerosol surface area information comes from some source other than SAGE II, then the model profile of surface area should be compared to SAGE II, and results of this comparison should be presented, either as a brief discussion or as a brief discussion accompanied by a new figure.

2. Tables 2, 3, and 4 provide a detailed level of documentation of the calculations. However, certain important parameters are not given. These include: temperature (or pressure), H2O, CO, and, as noted above, surface area. This information should be added to these tables.

3. Since I have access to HALOE data, and have conducted many simulations using a similar approach, I have checked the profiles of CH4, Cly, and NOx used in this analysis versus my version of the HALOE data and of tracer-tracer relations.

Overall, the model input specification used here is fine. The Leon flight poses a special challenge, given the large variability in CH4 for air inside the vortex compared to extra vortex air. The authors have pursued a reasonable approach given the unfortunate loss of data from the whole air sampler system for the Leon flight.

It looks like the level of NOx given in Tables 3 and 4 is less than NOx reported by HALOE. I suspect the reason for this is the nature of the calculation: even though the model is initialized by HALOE NOx, the value of NOx is allowed to change over the 9 cycles of iteration.

It would be quite helpful to add a new figure, similar to Figure 4, but for the Leon flight at 650 K (region of largest apparent discrepancy between measured and modeled ClO). Indeed, a two panel version of this new figure, one for "vortex" and one for "mid-latitude", would be helpful. Finally, for the two new panels as well as the present version of Figure 4, it would be helpful to denote, perhaps as an asterisk or circle tied to the Cycle 000 curve, the initial values of all calculated species (I know this info is already contained in the figures, but it would be easier to read if a symbol were used to draw attention to the initial values).

The time evolution of NOx shown in the 7th panel of Figure 4, as well as what I infer will be the time evolution of NOx that will be seen at 650 K and higher THETA levels for the Leon flight, is somewhat of a concern for the interpretation of the data. For the region of largest disagreement, the 650 K level of the Leon flight, it would be helpful if the authors could comment on whether the discrepancy gets worse (or better) if the HALOE measurement of NOx is correct: e.g., how would modeled ClO change if the model were forced to go through HALOE NOx, as well as forced to satisfy constraints for Cly, O3, and NOy? This comment ties into the importance of surface area, since the NOx/NOy ratio at 650 K over Leon is controlled largely by heterogeneous reactions. I infer from Case 1 vs Case 3 for the Leon flight that basically, if HALOE data for NOx are assumed to be correct, that the discrepancy between measured and modeled ClO largely is explained. But, I am not sure I am interpreting Case 1 versus Case 3 correctly.

Regardless, this point should be clearly explained upon revision.

Minor comments:

1. Several places it is stated that the discrepancy between modeled and measured ClO, at 650 K over Leon, is "up to a factor of two approximately".

However, on page 888, it is stated "all other ratios ... are in the interval 1.0 +/- 0.5 including the discrepancy found in the ClO daylight profile at around 650 K for the Leon flight.

It does not appear that the mean difference between modeled ClO (about 100 pptv) and measured ClO (about 175 K) is quite large enough to be described as "about a factor of 2".

2. Somewhere in the paper, the authors should comment on the odd result that calculated ClO using "vortex profiles" over Leon at 650 K is less than calculated ClO using "mid-latitude" profiles, despite more Cly in the vortex profiles. Presumably, this difference is driven by the higher levels of NOx in the vortex profile.

3. The last three sentences of the abstract are confusing. 3d to last says the model "substantially overestimates" measured ClO. The 2nd to last says "no indication ... [of] ... substantial uncertainties". The last sentence again returns to a thought of "substantial overestimation".

The last part of this abstract will be very confusing until colleagues read the paper.

I believe the glass is much more than "half full": e.g., I think the comparisons show overall "excellent" , or perhaps "very good" agreement. I suggest some modification of the abstract. In my view, I think the overall good agreement should be emphasized and then the exceptions can be noted. Regardless, the abstract will confuse many if the overall message of the last few sentences is so self-contradictory.

4. page 877, line 16, "was" should be "were"

5. page 877, line 19, perhaps a paper reporting recent measurements of ClO from the POLARIS mission (e.g., Stimpfle et al., 1999 or some other paper) can be added to the two citations already given. This Stimpfle paper is the same one that is cited elsewhere.

6. page 878, lines 20 to 27: perhaps the findings of Sen et al. (JGR, 104, 26653, 1999), who examined ClONO2/HCl and ClONO2/Cly using balloon

7. page 880, line 6: it is not clear what "the Island" means.

8. page 880, lines 10 and 11: need a comma after "consequence". Most importantly, nothing is obvious to me from Figure 1 unless color is used. Black and white for these PV maps really doesn't work. Is there a cost associated with use of color for an on-line journal ?!?

9. page 880, line 13: need a comma after "ClO mixing ratios"

10. page 880, lines 14-15: this sentence is awkward and should be revised to read something like: During descent, both a nighttime and sunrise profile of ClO were measured. During ascent, only a daytime profile was derived.

11. page 881, lines 9 and 10: perhaps state "so ozone measurements could not be obtained for all altitudes".

12. The variation of the O3 abundances for the Aire flight is a bit confusing. Perhaps a new table, giving values of O3 from the sonde, from HALOE, from the 2D model, and for the two simulations (e.g., case a and case b) could be added for the 14 trajectories already detailed for this flight. Or, somehow, some other indication of the ozone differences should be provided. I did not fully understand the discussion on page 883, lines 13 to 26. Also, I don't see any indication in the figures or tables (but perhaps I have missed something) that shows the difference between ozone for case a compared to case b.

13. page 884, line 18: should add a semi-colon after "simulations".

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14. page 885, line 24: there are two 600 and 650 K points "during daylight", so perhaps state something like: except between 600 and 650 K during the most sunlit portion of the flight (SZA about 67 deg).

15. page 886, lines 1 to 6. First sentence says "ClO is sensitive to O3". Last sentence says "sensitivity of ClO ... on O3 ... determined this way ... is negligible". This is a confusing paragraph. Figure 6 shows some sensitivity of ClO to ozone. The paragraph should better reflect the figure.

16. One problem with the speculation of the cause of the discrepancy between measured and modeled ClO at 600 to 650 K for the Leon flight is that, if the explanation is caused by a process such as pressure dependence of J_clono2, then this should affect comparisons of ClO at 600 to 650 K for the Aire flight.

The only robust result common to both flights is the apparent tendency for the model to overestimate ClO during twilight. Even this is tricky, as the Air flight at 87 deg shows a different result than the Leon flight at 87 deg.

I think the discussion of j_clono2 should include a brief statement that the Aire comparisons, for 600 to 650 K, agree with standard photochemistry.

Also, while I am not suggesting any change to Figure 9, this figure can be misleading in that the amount of extra ozone loss implied by the model at SZAs between 87 and 90 deg, compared to that implied by the data for the same SZAs, is tiny compared to 24 hr average ozone loss. The reference to Riviere et al. [2003] at the top of page 889 suggests a larger type of discrepancy. Riviere et al.'s observations challenge conventional thinking. I would say the problems noted by Figure 9 are in a different class of modifications to our thinking; e.g, they suggest perhaps a fine refinement. Anyway, it is OK to cite Riviere's work, but I feel the paper overall tends to focus a bit too much on the minor discrepancies and should, upon revision, perhaps present a more balanced view of (in my view) the overall excellent agreement between modeled and measured ClO. On the other hand, I realize progress is made by searching for disceprancies, and then understanding them. The authors should take this comment into consideration and proceed with a presentation they are completely comfortable with.

17. Captions of Figures 6 and 7 should include description which ClO uncertainties are shown (e.g., are they precision, or accuracy, or both) and whether these are 1-sigma, 2-sigma, etc.

18. Nice job with this paper. Sincerely, Ross Salawitch

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 875, 2005.