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Interactive comment on “Partitioning and budget of inorganic and organic chlorine species observed by MIPAS-B and TELIS in the Arctic in March 2011” by G. Wetzel et al.

Anonymous Referee #3

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OVERVIEW:

The focus of this paper is on a detailed analysis/comparison of measurements of numerous chlorine species by two instruments on a balloon flight in the Arctic vortex on 31 March 2011. The chlorine budget derived from measurements on this flight is compared with that from a chemistry climate model and with previous chlorine budget results. The paper is reasonably well-written and the analysis appears to be thorough and should be of interest to ACP readers. I recommend publication in ACP if the concerns and questions listed below are addressed.

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GENERAL COMMENTS:

1. The most important overall revision that I think is needed to support the analysis and the conclusions drawn is to include some discussion of how representative the chlorine budget derived from one balloon flight in the polar vortex near the end of one very exceptional Arctic winter is of the "current" budget overall. The authors conclude that their results "confirm" a slightly decreasing chlorine trend in the stratosphere, but this is only the case if they can show/argue that the chlorine budget derived under these quite unusual conditions is representative of the current budget in general.

2. Have there been any comparisons between the MIPAS-B and TELIS measurements described here with other instruments? In particular, are there not ACE-FTS measurements in 2011 at a similar time of year that could be (or perhaps have been) used to construct a chlorine budget?

3. There are numerous places, noted below in the specific comments, where references need to be added to support points that are made on the basis of previous work.

SPECIFIC COMMENTS (in order of appearance, not importance):

page 5394, lines 1–6: It would be good to include Solomon (1999, Revgeo) here.

page 5394, line 10: Rather than bring in the concept of age of air (which is somewhat abstract and not otherwise important for this paper), it would be better to simply say something that conveys that it takes several years for changes in surface emissions to propagate to the stratosphere.

page 5394–5395: In the discussion of trends in stratospheric chlorine, can you give some indication of the degree of uncertainty implied by the results of the previous studies described?

page 5395, lines 18–20: It isn't clear how the neural networks are used, why they were needed, or whether those details are even important to this paper.

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page 5396, lines 1–2: As per general comment 1, this seems to be a lot to expect from analysis of measurements from one flight, unless it can be shown/argued that those measurements are representative of the broader CI distribution.

page 5396, line 12: The vortex in the lower stratosphere in 2011 was intact, well-defined and represented a strong transport barrier until approximately mid-April (e.g., Manney et al., 2011; also see any readily available potential vorticity maps in the lower stratosphere, e.g., those at http://mls.jpl.nasa.gov/plots/met/met_plot_locator.php).

page 5396, lines 19–20: Please provide citations/support for this statement about the timing of the chlorine deactivation. The primary instrument measuring active chlorine throughout the vortex on a daily basis at that time was Aura MLS, which had an anomaly and was temporarily shut down between 27 March and 19 April 2011; chlorine was still substantially activated on the first date, and completely deactivated on the second date (Manney et al., 2011). Are there other observations that pin down the timing of deactivation more specifically to that described here?

page 5397, line 14, and ensuing discussion: The material on the reliability of the MIPAS-B data relies heavily on the results of Friedl-Vallon et al. (2004). Kleinert (Appl. Optics, 2006) discusses a nonlinear effect in the MIPAS-B2 detectors that is not accounted for in the methods of Friedl-Vallon et al. If a correction for this effect is used here it should be noted and the Kleinert paper cited; if not, an explanation should be given as to why it was not necessary.

page 5398, line 23: Why was the updated HITRAN database (Rothman et al., 2013, JQSRT) not used? How can material in a *2003* paper be used to "update" a *2009* database?

page 5399, line 21: See comment on page 5398, line 23.

page 5400, lines 25–27: How has the PSC scheme in the model been validated?

page 5401, lines 1–3: Can you give a reference for the implementation of the nudging

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to ERA-Interim, and/or describe how closely it "ties" the model to realistic meteorology?

page 5402, lines 5–7: Can you give a reference or other supporting evidence that these species are indeed negligible?

page 5403, line 6 and line 20: I'm guessing that "noise error" means error due to spectral noise?

page 5403, lines 5–7: Although the time evolution of MIPAS-B ClO near 30km does look problematic, the maximum values there look similar to what I'd expect – for instance, similar to those typically seen by MLS at high latitudes in the 30–40-km altitude region in late March – while the values from TELIS appear lower. Have these values (or values measured by these two instruments at other times) been compared with other measurements?

page 5403, line 14: Please give a/some reference/s supporting the statement that these ClONO₂ values are typical.

page 5403, lines 14–16: This sentence is not very clear. I think you are pointing to a reduction in the vertical ClONO₂ gradients as evidence that ClONO₂ is lower than it would be if chlorine was completely deactivated, but you don't explicitly make the connection between lower ClONO₂ *gradients* and lower ClONO₂ *values*. Perhaps this could be reworded to clarify.

page 5403, line 25 (first reference), and following text: Since you are using N₂O from MIPAS-B, the uncertainties and retrievals for N₂O should be discussed in section 2 (and included in Table 1).

page 5404, equation (7), and discussion of N₂O-Cl_y correlation: What is this proxy relationship based on (i.e., how was it derived)? Is this the same method discussed by Strahan et al. (2014, JGR)?

page 5404, line 6, "...supposed to be vertically constant...": What work/theory shows that it is "supposed to be" vertically constant? Can you give a/some reference/s.

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page 5404, lines 8–9: How do you know the bias is in the observations rather than in the calculations?

page 5404, lines 10–11: What is "the reference"? Can't you check the N₂O values observed at this altitude on this flight to test whether this statement is the case?

page 5404, lines 23–24: To my eye, this is only the case between about 15 and 20km, not everywhere "below 20km".

page 5405, line 2: Can you give reasons for the Cly deficit in EMAC and/or references to support that there is such a deficit?

page 5405, line 11: Please specify where the "quasi-altitude constant" region is.

page 5405, lines 22–23: It is not entirely clear to me how the missing chlorine species are included in the observations via HCl, and what "(after being photolyzed)" refers to? After what is photolyzed?

page 5405, lines 8–9: The "cold period", defined as having minimum temperatures below the NAT PSC threshold lingered into the first several days of April 2011 (e.g., Manney et al., 2011).

page 5405, lines 7–9: MLS measurements on 26 March 2011 (publicly available maps on MLS website) show ClO values up to about 0.9 ppbv in the core of the vortex, and a very non-uniform distribution with highest values in the core and near zero values near the edge of the vortex. The flight in question sampled air towards the outer part of the vortex (which would be apparent if you revise Figure 1 as I suggest below), where ClO would be expected to be lower than the maximum values. While significant deactivation undoubtedly occurred between 26 and 31 March, the non-uniformity of the ClO distribution in the vortex means that you cannot make a generalized statement about the overall degree of deactivation from these very localized measurements.

page 5405, lines 16–18: What about ACE-FTS chlorine measurements in the Arctic in 2011? Can't you compare with these?

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page 5405, line 22: Per general comment (1) above, and other comments on the localized (in space and time) nature of the measurements analyzed here, I think "confirms" is too strong. I would suggest something like "is consistent with".

page 5406, lines 8–9: I don't understand what this final sentence is getting at, please clarify.

FIGURES:

Figure 1: It is impossible to distinguish which dots are from which instrument on this figure. It is also difficult to get the context of where these measurements are within the vortex because too small a geographic area is shown. I would suggest a panel showing a larger latitude/longitude range, with just the boundaries of the area of the measurements shown, along with a panel showing a much smaller geographic region around the measurement locations with clearly distinguishable symbols for each instrument.

Figure 2: Needs to be larger (this may only be an issue in the ACPD format).

Figures 4 and 5: The yellow used for Cly and Cly* is quite hard to see – I'd suggest choosing a different (more intense) color.

WORDING SUGGESTIONS, TYPOS:

page 5393, line 8, "is" should be "has been".

page 5393, line 15: suggest changing "previously carried out" to "previous".

page 5393, lines 23–24, replace "the seventies of the last century" with "the 1970s".

page 5394: using "cold" (or "warm") to modify "temperatures" is incorrect, since "cold" by itself means "low temperature". Rather, "low" (or "high") should be used.

page 5394: line 18, change "has been" to "was".

page 5395: line 10, change "of" to "from".

page 5396, line 15, "which" should be "that". (Note that "which" and "that" are often,

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though not consistently, misused throughout the paper, either one used when the other should be or "that" used and preceded by a comma, or "which" used and not preceded by a comma. A review of restrictive and non-restrictive clauses might help.)

page 5396, line 21, suggest adding "Locations of" before "Recorded".

page 5398, lines 5, 26; page 5403, line 14; page 5404. line 14; page 5405, line 26; page 5406, line 5; Figure 5 caption, 2nd to last line: "which" should be "that".

page 5398, line 26, add a comma after "error".

page 5399, line 3, add a comma after "retrieval".

page 5400, line 3, suggest changing "corresponding to" to "resulting in".

page 5400, line 4, change "like" to "such as".

page 5400, line 11, change "on" to "of".

page 5400, line 24, add commas before and after "among others".

page 5401, line 14, "was" should be "were".

page 5402, lines 15-16, I think you mean "correspond to noon maximum and minimum values, respectively".

page 5403, line 4, "was" should be "were" page 5404, line 21, move "in detail" to after "some differences" and delete comma after "detail".

page 5405, line 1, add a comma after "2011".

page 5405, line 20, add "to" between "amount" and "about".

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 5391, 2015.

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