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Atmospheric Chemistry and Physics Discussions

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 #4

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The manuscript presents measurements of stratospheric chlorine species by two instruments (MIPAS-B and TELIS) which flew on March 31st 2011 on a stratospheric balloon that was started from Esrange near Kiruna in northern Sweden. Thus, the measurements took place in the stratospheric polar vortex at the end of the chlorine deactivation period, after an Arctic winter with long-lasting low stratospheric temperatures and, as a result, strong ozone depletion. From the measured spectra, profiles of organic and inorganic chlorine species were determined.
Firstly, the chlorine partitioning is discussed and compared with model results from an EMAC simulation which is relaxed towards ERA-Interim and uses observations

and projections from WMO (2011) for the chlorine source gases. The meteorological dynamics and chemical composition in the simulation is thus assumed to correspond as good as possible to the real state of the atmosphere on the launching day. The agreement between model and measurements is quite good, except for some vertical regions where the HCl mixing ratio is underestimated by the model, and a strong underestimation of the $ClONO_2$ amount.
Secondly, the measurements are used to assess the total stratospheric chlorine mixing ratio in the Northern Hemisphere high latitude region, which is compared with satellite measurements and the already mentioned EMAC model results. The comparison of the total chlorine budget from measurements and model shows an underestimation of the total chlorine amount in the EMAC model which is mainly due to insufficient $ClONO_2$. However, the MIPAS-B and TELIS total chlorine budget agrees well with (extrapolated) satellite measurements and thus helps confirm the overall decrease of the stratospheric chlorine burden as a consequence of the Montreal Protocol and its amendments and adjustments.

The MIPAS-B and TELIS balloon measurements permit an assessment of nearly the total chlorine budget from two instruments flying on the same meteorological platform. They provide a (nearly) complete picture of the (vertically-resolved) chlorine partitioning on that special day without many inter-instrumental and other assumptions to be made. This makes these measurements very valuable even though they only cover a short time period. Such data are very important for the validation of satellites, but especially also for the validation of atmospheric chemistry models.

I recommend publication in ACP after the below mentioned points have been addressed respectively discussed.

1 General comments

- I think you should clarify more often that your chlorine species profiles are valid for the special day and the special atmospheric situation only.
- Concerning the budget, you should mention and argue/prove that you are able to calculate the budget from the species measured by MIPAS-B and TELIS because none of the species that you cannot measure is very important under the conditions of the flight day.
- In the conclusions, only the chlorine budget is discussed, but not the partitioning. I think you should include at least one or two sentences summarizing the findings of Sect. 3.

2 Specific comments

1. Page 5393, line 16: As you are not discussing changes in trends in the rest of the paper, you probably mean the decreasing chlorine amount here, not a decreasing chlorine trend?
2. Page 5393, line 25, to page 5394, line 6: I suggest to rewrite these sentences because they are slightly confusing and the second sentence is very long.
3. Page 5394, line 26: It is not clear which time range the trend you cite from Zander et al. (1996), 0.10 ppbv year^{-1}, is referring to. Please include this information.
4. Page 5396, line 1: The word "current" used for the status of the chlorine partitioning suggests (to me) that the results obtained are valid for a longer time range than they actually are (see also my first general comment). (In contrary, for the chlorine budget, the expression seems more appropriate to me.)

5. Page 5396, lines 11-17: These three sentences do not really fit under the heading "Instruments, data analysis and modelling", they are more a description of the meteorological situation of the preceding winter and on the launch day. Please consider moving them to the end of Sect. 1, more precisely for example onto page 5396, line 5, or changing the heading of Sect. 2 somehow ("Data description"?).
6. Page 5396, line 21: I suggest to mention the geographical coordinates of Kiruna/Esrange.
7. Page 5397, line 28: It is not clear how the vertical spacing of the retrieval grid increases with height between the balloon flight altitude and the top altitude at 100 km.
8. Page 5398, line 20: Do the a priori profiles of the target species vary with time of day? In this case, I think you should mention it here or when discussing the ClO results (e.g., page 5402, from line 21 on).
9. Page 5399, line 28 and following: If you retrieve the two isotopologues separately, it seems to me that you shouldn't need to take into account the relative abundances, just add your results, and maybe add some 0.01%, as H^{35}Cl and H^{37}Cl combine to only 99.99% of total HCl. You could change the sentence that starts in line 28 to just mentioning their relative abundances (and maybe whether you also find this relation).
10. Page 5401, line 18 to page 5402, line 7: In the introduction (page 5394, line 4), it sounds as if Cl_2 was the most important active chlorine species. So I think you should add an explanation why you may neglect it here in the budget calculations (please also see my second general comment in this context).
11. Page 5402, line 11: It is not clear what "This" refers to. Please specify explicitly which of the two conditions for measuring ClOOCl (activated chlorine and no PSC

signatures) were not fulfilled during the analysed balloon flight. If there were also PSCs (which I assume was not the case), maybe you should mention that when describing the meteorological situation.

12. Page 5402: Please add a citation to Eq. 6.

13. Page 5403, line 2: The word ”reproduced” sounds somehow strange to me when used for measurements. Please consider replacing it, e.g., by ”shown”.

14. Page 5403, lines 2-8, and Figure 2: Why are the time and magnitude of the maxima in MIPAS-B and TELIS ClO different? (Maybe because the instruments look into different directions? In this context, please see the comments on Figure 1.)
    And it is not completely clear from the statement here whether you use TELIS ClO data only above 26 km or for the whole altitude range.

15. Page 5403, line 25 to page 5404, line 4: It is not clear how the ”adapted” N_2O-Cl_y correlation was determined and which measurements of MIPAS-B were used for this, or whether maybe only the total (tropospheric) chlorine in the calculation was adapted to the time of the balloon flight? Please go a little more into detail here.

16. Page 5404, lines 6-9: Please try to rephrase the end of this sentence. The ”bias in the observations” confused me. As far as I understood, both the Cl_y and Cl_y^* values discussed here are observations respectively deduced from them.

17. Page 5404, lines 13-14: It is not clear whether ”increasingly” means a temporal or spatial change. I suggest to rewrite the beginning of the sentence for example like ”From about 17 km to the ground”, or similar.

18. Page 5404, line 18: Suggestion to add ”above 24 km” or ”in this altitude region” in this sentence.

19. Page 5404, line 22: I would prefer you to replace the word ”obvious” by ”visible” because in my feeling, ”obvious” is too strong in this context.

20. Page 5404, lines 25-26: Please give a rough estimate of the $ClONO_2$ deviation in % as well to make the large difference between model and measurement more obvious to the reader.

21. Page 5405, lines 8-10: I suggest to rewrite the sentence, for example like this: ”These higher N_2O values are connected with lower Cl_y values according to the compact N_2O-Cl_y relationship, resulting in an underestimation of the chlorine reservoir species (especially $ClONO_2$).”
    Furthermore, I suggest to add a concluding sentence like (with or without the part in brackets): ”So (at least part of) the $ClONO_2$ deficit in EMAC can be explained by the underestimation of the subsidence in the model.”

22. Page 5405, lines 11-12: Maybe move ”above 24 km” directly behind ”region”.

23. Page 5405, line 13: Connected with my comment concerning page 5404, lines 6-9, I suggest to rewrite ”simulated value of Cl_y^*” because in my understanding, it is a combination of measurements and EMAC model results in this case. Maybe change it to ”value of Cl_y^* deduced from the (EMAC) simulation”?

24. Page 5405, line 15: I suggest to start a new paragraph before ”The mean...”.

25. Page 5405, lines 16-23: I found the argumentation not very easy to follow. My final understanding is the following: Some minor CFCs and HCFCs (for example CFC-114, CFC-115, HCFC-141b, and HCFC-142b) are not included in the EMAC model and not measured directly with MIPAS-B/TELIS. However, in 24 km above ground, the largest part of the actually emitted amount has already been photolysed and the contained chlorine has therefore already reacted to become HCl (mainly). As a result, these minor species are indirectly contained in the

measurements of HCl (above 24 km), but of course not in the model calculations. So in case my understanding of your argumentation is right, I suggest to rewrite the "However" sentence so that the connection with the photolysis becomes clearer. And maybe it would be good to add some more clarifying explanations. However, I do not understand the connection to the very short-lived species which are mentioned in the middle of this paragraph. If there is no connection, please swap the sentences starting with "The remaining deficit" and "However, the chlorine amount". If there is one, please try to clarify this.

26. Page 5405, line 18: HCFC-141b is mentioned twice.

27. Page 5405, lines 23-24: Why is the percentage of inorganic chlorine above 24 km in EMAC larger than in the measurements (page 5404, line 18 says 95 %)? (Is this difference significant?)

28. Page 5406, line 10: Please mention the EMAC value of Cl_{total} explicitly again.

29. Page 5406, line 16: Please add a citation to the trend value -0.4% p.a.
For consistency and easier readability, I suggest you change "% p.a." to "% year^{-1}" (as on page 5394, line 26, and page 5395, line 28, for example).

30. Page 5406, lines 18-20: Please add again the information here that these species are not part of the MIPAS-B/TELIS chlorine budget.

31. Page 5406, line 22: To be more precise, I suggest to change "decreasing trend" to "decreasing amount" or "decreasing vmr" or "decrease of the stratospheric chlorine content" because it is not actually the trend that decreases (see comment concerning page 5393, line 16).

32. Page 5406, lines 5-9: I suggest to add "lower" between "hemispheric" and "stratosphere" (line 7). Furthermore, please rewrite/clarify/extend the last sentence.

Maybe you could move the sentence starting in line 3 ("We finally conclude...") to the very end of the text?

33. Page 5406, Sect. 4: I think somewhere in this section you should include one or two sentences with a conclusion concerning the stratospheric chlorine partitioning as measured by MIPAS-B/TELIS in comparison with the results from the EMAC model, i.e., summarize Sect. 3.

34. Table 3: If you cannot give an error estimate for the MIPAS-B/TELIS Cl_y^*, I would suggest to remove "$\pm$0.00".

35. Figure 1: Please improve the figure according to the suggestions of referee #3.

3 Technical corrections

1. Page 5395, line 2: I suggest to replace "occurred" (e.g., by "was performed" or "took place").

2. Page 5395, lines 16+17: You forgot the "Array" in the definition of CLAES: Cryogenic Limb Array Etalon Spectrometer.

3. Page 5398, line 8: I would find it better readable and understandable if you start this line with a lower-case letter in the word "where".

4. Page 5401, line 7: A hyphen is missing between "chlorine" and "containing".

5. Page 5402, line 25: Missing comma after "activation".

6. Page 5404, line 8: Suggest to change "to" to "towards".

7. Page 5404, line 25: Please add "by" after "deviates".

8. Page 5405, lines 19-20: A hyphen is missing between ”short” and ”lived” (at two other occassions as well). And please add ”to” between ”amount” and ”about”.

9. Page 5405, line 25: Please add ”of” between ”profiles” and ”ClO_x”.

10. Page 5406, line 7: Please remove the comma after ”reveals”.

11. Page 5406, line 15: Change ”this” to ”these”.

12. Page 5406, lines 22-26: Please add a comma after ”WMO (2011)”, add ”of air” after ”mean age”, and another comma before ”we”.

13. Figure 4, last sentence of the caption: I suggest to change ”Notice” to ”Note”.

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