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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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.

G. Wetzel et al.

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Response to referee #4:

First of all we thank the referee for his/her effort to carefully read the manuscript and for all comments.

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.

We now mention more clear the measurement situation of a late winter Arctic vortex.

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.

Minor CFCs (e.g. CFC-114 and CFC-115) and HCFCs (e.g. HCFC-141b and HCFC142b) which are not measurable by MIPAS-B/TELIS contribute to Cltotal not more than 1% above 24 km (Brown et al., Atmos. Chem. Phys. Discuss., 13, 23491–23548, 2013). Further species (above 24 km) which are not measurable by the balloon instruments are very short lived chlorine species which altogether amount to about 0.1 ppbv (Mébarki et al., Atmos. Chem. Phys., 10, 397-409, 2010; WMO, Report No. 52, 516 pp., 2011). However, the chlorine amount of these missing species is implicitly contained in the HCl measurement (since the short lived chlorine species are converted to HCl after being photolyzed) and hence included in the observed chlorine budget. We made some changes in the last paragraph of section 3 to make this more clear.

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.

There is already some discussion on the chlorine partitioning at the beginning of the conclusions. We extended this discussion.

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?

Yes, that's true. We changed the text accordingly.

2. Page 5393, line 25, to page 5394, line 6: I suggest to rewrite these sentences

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because they are slightly confusing and the second sentence is very long.

We only slightly changed these introductory sentences since the main reason for the complexity is the large number of (necessary) citations.

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, is referring to. Please include this information.

We included this information in the text.

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.)

We modified this sentence to make things more clear.

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"?).

We moved these sentences to the last paragraph of section 1.

6. Page 5396, line 21: I suggest to mention the geographical coordinates of Kiruna/Esrange.

We included the coordinates in the text.

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.

It increases gradually. This is written in the text now.

8. Page 5398, line 20: Do the a priori profiles of the target species vary with time of

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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).

No, the a priori profiles don't vary.

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 H35Cl and H37Cl 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).

The natural abundances for the isotopes have been accounted for during the retrieval process and the volume mixing ratio was scaled with the individual isotope abundance. Hence, the combined HCl was deduced by calculating the sum of the isotopes weighted with their abundances. We slightly modified the text.

10. Page 5401, line 18 to page 5402, line 7: In the introduction (page 5394, line 4), it sounds as if Cl2 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).

The species Cl2 is not important in the case of our measurement since the chlorine activation was only small by the end of March. Cl2 values in EMAC are no larger than 0.03 ppbv during day and zero during night. Here we only define the chlorine families. In the following discussion of the results we also give explanations on the impact of missing chlorine gases to the budget.

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.

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ClOOCl concentrations were below the detection limit. We explain this in the text now.

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

It was explained by Wetzel et al. (Atmos. Chem. Phys., 12, 6581-6592, 2012). We included this citation in the text.

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".

Changed.

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 Figure1.) 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.

The instruments look in the same direction (we explicitly mention this now in Fig. caption 1). MIPAS-B ClO is quite difficult to retrieve since spectral line intensities of ClO are comparatively weak in the mid-infrared. Above 30 km, the signal to noise ratio is low and hence the maxima shown by MIPAS-B in this altitude regions are characterized by large error bars. However, at 19 km, the signal to noise ratio is higher such that ClO can be retrieved here with better accuracy. Anyhow, the overall accuracy of TELIS is better than the one of MIPAS-B and therefore we used the TELIS ClO data in the whole altitude range for calculating the chlorine budget (now written in the text).

15. Page 5403, line 25 to page 5404, line 4: It is not clear how the "adapted" N2O-Cly 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.

The word "adapted" is a mistake in the text since the correlation was directly applied to the MIPAS-B N2O measurement. The text has been changed accordingly together

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with a further explanation.

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 Cly and Cly values discussed here are observations respectively deduced from them.*

We rephrased the text to make this issue more clear.

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.

Of course a spatial change (since no temporal change is shown here). We modified the text to make this distinct.

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

We followed the reviewer's suggestion.

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.

Changed.

20. Page 5404, lines 25-26: Please give a rough estimate of the ClONO2 deviation in % as well to make the large difference between model and measurement more obvious to the reader.

We added this information to the text.

21. Page 5405, lines 8-10: I suggest to rewrite the sentence, for example like this: "These higher N2O values are connected with lower Cly values according to the compact N2O-Cly relationship, resulting in an underestimation of the chlorine reservoir species (especially ClONO2)." Furthermore, I suggest to add a concluding sentence

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like (with or without the part in brackets): "So (at least part of) the ClONO2 deficit in EMAC can be explained by the underestimation of the subsidence in the model."

We followed the reviewer's suggestion.

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

Changed.

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

We changed the text for better understanding.

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

Okay.

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 photolyzed 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.

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We modified the text to describe this issue more clear.

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

The second one is HCFC-142b. We changed this in the text.

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?)

Taking into account the error bars of the measurement, this difference is clearly not significant.

28. Page 5406, line 10: Please mention the EMAC value of Cltotal explicitly again.

Okay.

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).

The citation belonging to these observations occurs already before this sentence. We changed a "the" in "these" to make it clear that trend and observations belong together.

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.

Okay.

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).

Changed.

32. Page 5407, lines 5-9: I suggest to add "lower" between "hemispheric" and "strato-

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sphere" (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?

Modified.

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.

We added some more text at the beginning of section 4.

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

The estimate is given and it is ±0.002 ppbv.

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

We did improve this figure.

Technical corrections:

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

Changed.

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

Okay.

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".

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Changed.

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

Okay.

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

Okay.

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

Changed.

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

Okay.

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

Changed.

9. Page 5405, line 25: Please add "of" between "profiles" and "ClOx".

Okay.

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

Okay.

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

Changed.

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

Changed.

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

Okay.

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