

## Reply to reviewer #1

We thank anonymous referee #1 for his/her constructive review that helped to improve the contents of our paper. The review comments by anonymous referee #1 are numbered and repeated below as *in italic letters*, followed by our answers.

### **General comments:**

(1). *The title is not really representing what actually has been done in this study. In the title only the FTIR measurements from Syowa are mentioned, but also the satellite observations and model simulations comprise a major part of this study. This should be reflected in the title. I would suggest to simply change the title to: “Temporal evolution of chlorine and minor species related to ozone depletion at Syowa station, Antarctic during austral fall to spring in 2007 and 2011” to be more general or to “Temporal evolution of chlorine and minor species related to ozone depletion from observations and model simulations at Syowa station, Antarctic during austral fall to spring in 2007 and 2011” to be more specific.*

The title was modified to “Temporal evolution of chlorine and minor species related to ozone depletion at Syowa Station, Antarctica during austral fall to spring in 2007 and 2011” as suggested.

(2). *The content of the Appendices are not that unimportant and therefore I would suggest that these parts are moved to the main text while rather other figures, as e.g. parts of Fig 13-16 could be put into the Appendix (see my comment on these figures below for more details).*

Old Figures 13-16 were combined into one new figure, as well as old Figures 2-4, to reduce the number of figures. We keep Appendices A and B as appendices, because the contents of MIROC 3.2 CCM is not the main focus of this paper, nor the direct comparison of model results and observations.

(3). *The discussion section is rather a continuation of the result section. These two subsections should be divided into subsections with subsections header that describe what actually is analysed (time series, correlation etc.)*

The contents of first part (old Figure 10 and 11) which previously appeared in the Discussion section was now moved to Result section. Now, the contents of result section deal with the facts which were read from observations, while the contents of discussion section deal with model results and ClO/ClONO<sub>2</sub> correlations.

(4). *There are too many tables and figures in the current version of the manuscript. These tables/figures could be combined. This would not only reduce the number of figures, but also make the manuscript more concise (see also my specific comments below).*

The old Tables 4-9 are now combined into new Table 4, and some unimportant contents are omitted.

Moreover, we combined old Figures 2-4, and old Figures 13-16 into new one figures.

***Specific comments:***

(5). P2, L6: *...similar to the case in the Arctic. . . .This needs more explanation and maybe also some references.*

There was a comment by another reviewer that the term “super-recovery” is not good. We rephrased to “additional increase of ClONO<sub>2</sub> from initial value”. We think showing reference in the abstract is not suited. The reference (Webster et al., 1993) is cited in the introduction.

(6). P2, L13: *“..great year-to-year variability.” I think this only holds for the Arctic and not for the Antarctic. The distinction between Arctic and Antarctic should be made here and the differences between both regions should be discussed.*

We meant normal year-to-year variability of ozone depletion in the Antarctic. We deleted the adjective “great” here. The difference in ozone destruction between Arctic and Antarctic is now described in the Introduction.

(7). P2, L18: *References should be added.*

We added (Solomon et al., 1986) in the reference, after reactions (1) and (2).

(8). P3, L3: *Please add which temperatures are needed. Do you mean with the term “inactivation” the “deactivation”?*

Temperatures warmer than NAT PSC saturation temperature are needed. The description is added. “Deactivation” was correct. It was corrected.

(9). P3, L15-16: *This sentence should be rephrased and maybe even better split into two sentences. It is not clear what you mean here. Also in Antarctica PSCs should occur within the polar vortex. Or is there a shift between the cold area and the vortex? If yes this should be said more clearly. However, you discuss here the temperature, so I guess you just mean that there are less PSCs in the Arctic due to the generally higher temperatures there.*

Yes, we just meant that there are less PSCs in the Arctic compared with Antarctic, due to the difference in mean winter temperature. We rephrased this part in the text.

(10). P3, L30ff: *Section 3 describes the validation of the FTIR measurements, but in the introduction, nothing is mentioned about this validation.*

At the last part of the introduction, content of each Section is now described.

*(11). P4, Section2: I would suggest to make two subsections, one for FTIR and one for the satellites and to also extend the description of the satellite data sets since these data sets are not that unimportant for this study.*

We divided Section 2 to two subsections. Description of the satellite data sets was extended.

*(12). P4, L5: I would suggest to discuss the discovery of the ozone hole based on the typical papers as e.g. Farman et al., 1985, Nature, first and then to discuss the measurements of the ozone hole over Syowa.*

The initial idea to show Chubachi 1984 is to show the topic of ozone measurements at Syowa Station. However, we added description of ozone hole measurement at Halley Bay (Farman et al., 1985) here, as well.

*(13). P4, L23ff: The abbreviations FASATM, CIRA-86, ILAS-II have not been introduced yet. The abbreviations of Aura/MLS and Envisat/MIPAS have been introduced in the abstract, but should be introduced once again here in the main text of the manuscript.*

There was a mistake of the name. FSCATM was correct, not FASATM. When I looked at the original document (Gallery et al., 1983), there was no abbreviation for FSCATM. It looks like a name of a FORTRAN program. The abbreviations of CIRA-86, ILAS-II, Aura/MLS, and Envisat/MIPAS was added in the main text.

*(14). P4, L27ff: O<sub>3</sub>, HNO<sub>3</sub>, HCl were used as a priori. What about ClO and ClONO<sub>2</sub>. Were these also used as a priori?*

No. For FTIR vertical profile fitting of O<sub>3</sub>, HNO<sub>3</sub>, and HCl, we need a priori profiles. Since ClO and ClONO<sub>2</sub> are satellite observation products, they don't need a priori profiles.

*(15). P5, L5: The abbreviation CALIPSO has not been introduced yet.*

The abbreviation CALIPSO is now introduced in the text.

*(16). P4-5. General: Different satellite instruments were used for different trace gases. It should be motivated why different instruments have been used and it also should be discussed if instrumental differences influence the results. What is the quality of the used data products?*

ClO is only measured by Aura/MLS, and frequent ClONO<sub>2</sub> measurement is only made by Envisat/MIPAS at the time of analyzed year. In order to show the quality of the used data, some references for validating the data products are added in the "2.2 Satellite measurements" section.

(17). P7, L12ff: *A motivation is missing why two years and two altitudes have been considered. So far I could not see that the results are that much different so that it is justified to show two different altitudes and two different years. The differences between these two years and two altitudes need to be more clearly described.*

There are distinct differences between 18 and 22 km as is discussed in Section 5. At 18 km, the O<sub>3</sub> amount was finally depleted more than 80%, and the recovery of ClO mainly went to HCl. At 22 km, the O<sub>3</sub> amount remained about half of the initial value even at the most depleted period, then the recovery of ClO went to ClONO<sub>2</sub>. We showed two years (2007 and 2011) in order to show typical Antarctic chlorine behavior which is not a special case for a single year. The reason for selecting two altitudes are described in the text.

(18). P8, L1ff: *As mentioned above the discussion is rather a continuation of the result section. This section should be divided into subsections that describe what actually is analysed (time series, correlations etc.)*

We moved the contents of the first part (Figure 10 and 11) which previously appeared in the Discussion section to the Result section. Now, the Result section deals with the result of observations, while the Discussion section deals with model results and ClO/ClONO<sub>2</sub> correlations.

(19). P9, L7ff: *The comparison to model simulations is not really a comparison. It's rather a different way of analyses of the winter using a different data set than the ones before.*

You are right. The comparison between model simulations and observations are only shown in Appendix B. In the main text, we added a sentence "Hereafter, the result of MIROC3.2 CCM is discussed".

(20). P10, L21-22: *"...as there was little NO<sub>2</sub> available due to the depleted O<sub>3</sub> amount here." This needs definitely to be explained. What is the relation between these two species?*

There was little explanation about this in the previous text. We added detailed explanation of this issue by adding two more reactions (R14 and R15) in the introduction part. We explained this issue by referring (Douglass et al, 1995) and Section 1 here.

(21). P11, 3-4: *"This study was (! were) the first continuous measurement of chlorine species related to the ozone hole from the ground in Antarctica" If this is really the case, then this should be also mentioned in the abstract.*

It was also mentioned in the abstract now.

(22). P11, L25-26: *"...like the case in the Arctic ! like it is the case in the Arctic". That needs more explanation. It has not been stated what the usual case in the Arctic is and what generally the differences between Arctic*

*and Antarctic are.*

The difference of deactivation pathways of active chlorine into reservoir species (HCl/ClONO<sub>2</sub>) between Antarctic and Arctic is now explained in the Introduction (Section 1) in detail around (R12, R13, R14, and R15). The pathway is mainly controlled by the availability of ambient O<sub>3</sub> (>0.5 ppmv or not). We deleted description of NO<sub>x</sub> here, because it might confuse the readers.

*(23). P11, L26ff: Conclusions should be given.*

We thought Section 6 (Summary) is somewhat equivalent to Conclusions. We now used term ‘Conclusions’ instead of ‘Summary’.

*(24). P11, L26ff: References should be given. The processes and interplay between the species are already known and documented. At statement what actually are the new findings of this specific study is missing.*

Douglass et al. (1995) was cited here. The new finding of this study is that even in the Antarctic, the recovery to ClONO<sub>2</sub> can occur in an altitude (22 km in our case) where there was relatively high ozone amount, which was not apparently documented in Douglass et al. (1995). The altitude differences are now described in the Abstract and in Conclusions.

*(25). Appendix A and B: Move this to these parts to the main text.*

As was described in the reply to comment (2), we decided to keep Appendices A and B as they are.

*(26). Tables: Too many tables! I would suggest to combine Table 4 to 9, thus creating a long table. Add a column for the species and then put the current tables under each other.*

We combined old Tables 4 to 9 into new Table 4, as suggested.

*(27). Figures 2-4: There are also too many figures. Figures 2-4 could be combined by putting these under each other.*

Old Figures 2 to 4 are now combined into new Figure 2, as suggested.

*(28). Figure 8: Is it really necessary to show here two years? Couldn't one of the years put into the appendix or a supplement to the paper since the results do not seem to be that much different.*

Since there were some differences between 2007 and 2011 (early ClO enhancement and temporal ozone loss around day 150-170, enhancement of ClONO<sub>2</sub> at 18 km around day 270-300 are only seen in 2011), we decided

to keep these two years in the main part of the paper.

(29). *Figure 10: I would suggest to use lines instead of symbols. The interplay between the trace gases would then become much better visible (the same holds for Figure 6-9).*

Since FTIR and satellite measurements are not available on everyday bases, using lines instead of symbols will mislead the conclusions. Also, there are some days when Syowa Station was outside the polar vortex (shaded days). Therefore, we think keeping symbols is better than using lines.

(30). *Figures 13-16: Why is a complete different representation of the model results used than for the measurement data? How are these comparable? I would suggest to combine Figures 13-16 and not to show all trace gases, but a selection. Put the gases in the rows and the dates in the columns. The remaining gases could be put into the appendix or a supplement to the paper.*

Until old Figure 12, we discussed phenomena only above Syowa Station. From old Figure 13, we tried to discuss more-like vortex-wide phenomena. In order for such a purpose, we used MIROC3.2 CCM model results. As is suggested, we combined old Figures 13 to 16 into new one Figure.

(31). *Figures B1 and B2: These figures should be put into the manuscript instead of the appendix.*

As was described in the reply to comment (2), we decided to keep Appendices A and B as it are.

**Technical corrections:**

(32). *P1, L14: please add “the”, so that it reads “understanding of the mechanisms ...”.*

’the’ was added as suggested.

(33). *P1, L29: change sentence to: “by reaction of ClO with NO<sub>2</sub>”.*

The sentence was changed as suggested.

(34). *P2, L19ff: The numbering of the reactions. Doing this differently than just using number as e.g. using R1 to R13 would be more concise. However, how to do the numbering of equations should be checked with the ACP guidelines.*

I checked the ACP guideline, and found that chemical reactions should be numbered as R1, R2, ... etc. Therefore, chemical reactions are now numbered from R1 to R15, whereas other formulae are numbered as (1), (2), and (3).

(35). P3, L3: *when stratospheric temperature gets warmer ! when stratospheric temperatures get warmer*

Corrected as suggested.

(36). P3, L4: *mainly occur ! mainly occurs*

Corrected as suggested.

(37). P3, L9: *between in ! either “between” or “in”, but not both.*

This part was rephrased. The new sentence is: ‘... are different between typical conditions in the Antarctic and those in the Arctic.’

(38). P3, L23: *Use a different beginning for the sentence as it is done now. Maybe it is even easier to combine this sentence with the one before so that it reads: “. . . . .discovery of the ozone hole by direct observations from high-altitude aircraft.”*

We rephrased the sentences as follows: ‘... have been monitored by several techniques since the discovery of ozone hole. These measurements consist of ...’

(39). P3, L23ff: *If a not complete references list is given because there are too many references, these should be referenced with “e.g.” in front and not with “etc.” at the end.*

‘e.g., ...’ was now used instead of ‘... etc.’

(40). P3, L28: *trace components ! trace gas components or better to write trace gases*

Now, the term ‘trace gas species’ is used throughout the manuscript.

(41). P3, L30: *trace species ! trace gas species or trace gases*

Same as above.

(42). P3, L32: *“during the course of the winter” or “during the ozone hole formation and dissipation period”, at least “in the” should be replaced by “during”.*

Now it was rephrased as: ‘during the ozone hole formation and dissipation period.’

(43). P3, L33: rephrase sentence to “...Finally, distributions of minor species simulated with the MIROC3.2 chemistry-climate model . . . . .”.

Now it was rephrased as: ‘Finally, in Section 5, distributions of minor species simulated with the MIROC3.2 chemistry-climate model ...’ as suggested.

(44). P4, L30: *Figures 1(a), (b), and (c) ! Figures 1 (a)-(c)* .

Because we corresponded O<sub>3</sub>, HNO<sub>3</sub>, and HCl with Figures 1(a), (b), and (c), respectively, we kept these figure letters as it is.

(45). P5, L11: *Same here for Figure 2.*

It was now rephrased as: ‘Figures 2(a)-(b)’ as suggested.

(46). P5, L16ff: *Instead of just “D value” it should read “relative difference D” or just “relative difference”.*

It was now rephrased as: ‘relative difference D’ as suggested.

(47). P5, L26: *Figures 4(a) and 4(b) show ! Figure 4 shows*

Now, old three Figures 2-4 are combined into one new Figure 2 (a)-(f)

(48). P5, L29: *the maximum ! a maximum*

Two ‘with the minimum of -14.6% and the maximum of -3.0%’ was now replaces as ‘with a minimum of -14.6% and a maximum of -3.0%’ as suggested.

(49). P6, L5: *Rephrase sentence to ”Figure 5 shows the temporal variation of temperature and chlorine species. . . . .”*

Old Figure 5 (new Figure 3) shows only temporal variation of temperature, not chlorine species.

(50). P6, L7: *on the figures ! in the figures*

Corrected as suggested.

(51). P6, L12: *temperature ! temperatures*



Corrected as suggested.

(52). P6, L12: *are ! were*

Corrected as suggested.

(53). P6, L16: *Figures 6,7,8 and 9 ! Figures 6-9*

Now figure numbers are changed. They are now: 'Figures 4-7'.

(54). P6, L18: *MLS in parentheses obsolete since it is written "...observed by Aura/MLS..."*

They are deleted as suggested.

(55). P6; L21: *correlation ! equation. More correctly it should be written "polynomial equation derived from the correlation of  $Cl_y$  and  $N_2O$ ".*

It was corrected as suggested.

(56). P6, L22: *Skip "of" and replace the comma by a colon*

It was corrected as suggested.

(57). P7, L12: *is ! was*

It was corrected as suggested.

(58). P7, L25 and 28: *ppb ! ppbv*

'ppb' and 'ppm' were corrected to 'ppbv' and 'ppmv' throughout the manuscript.

(59). P8, L5: *Rephrase sentence to "Figures 10 and 11 show the temporal variation of the ratios of the chlorine species to  $Cl_y^*$ " or "Figures 10 and 11 show the temporal variation of the ratios between the chlorine species and  $Cl_y^*$ ".*

It was now rephrased as: '... show the time series of the ratios of each chlorine species with respect to  $Cl_y^*$  ...'

(60). P8, L25: *include "the" so that it reads "...with HCl increase, then the  $ClONO_2$  amount gradually. ..."*

It was corrected as suggested.

(61). P9, L7: *Figures 13, 14, 15, 15 ! Figures 13-16*

They are now combined into (new) Figure 11.

(62). P9, L7: *(a), (b), (c), (d), (e), (f) are obsolete.*

They are deleted.

(63). P9, L16: *reaction (12) ! Reaction 12*

It was corrected to 'reaction (R12)'.

(64). P9, L18: *(1), (2), (3), (4) ! (1)-(4)*

It was corrected to 'reactions (R1), (R2), (R3), and (R4)'.

(65). P9, L34: *simplify this sentence with listing the trace gases and then adding Figure 16 in parentheses.*

This sentence was simplified. We omitted to show the panel names ((a), (b), etc.) of (new) Figure 11 throughout the manuscript.

(66). P10, L8: *comma before "are" obsolete.*

We deleted the comma before 'are'.

(67). P10, L12: *comma before through obsolete*

We deleted the comma before 'through'.

(68). P10, L13: *At higher latitudes ! "At this latitude" or "At latitudes > 87.9"*

We corrected to 'At 87.9°S, ...'.

(69). P10, L14: *after ! in*

It was corrected to 'in winter'.

(70). P10, L15: *enhancement ! enhancements*

It was corrected to ‘enhancements’.

(71). P11, L6: *altitude ! altitudes*

It was corrected to ‘altitudes’.

(72). *Figure 6 caption: remove (FTIR) and (MLS) and (sonde) since it is enough if this suffix is given in the figure legend. Change last three sentences to “See text for details. The unit of O<sub>3</sub> is ppmv and of the other gases ppbv. The light and dark shaded areas are the days when Syowa station was at the boundary region and outside the polar vortex, respectively.*

Since there are two datasets for HCl, O<sub>3</sub>, and HNO<sub>3</sub> measured by FTIR, MLS, and sonde, (FTIR), (MLS), and (sonde) are needed for these species. The last three sentences are corrected as suggested.

(73). *Figure 8 and 9: The months should be added here as it done for figures 6 and 7.*

The months of (old) Figures 6 and 7 ((new) Figures 4 and 5) are now added as (new) Figures 6 and 7.

(74). *Figure 12: “day number 220 – 260” ! add time period as dates*

Dates (August 8 – September 17) are now added.