

Response to comments by three referees

(Response in **bold**, and comments in *italic*)

Response to Referee #1

We would like to thank the anonymous referee #1 for reading this manuscript and offering valuable suggestions. We hope to have addressed all the comments in the reply below.

General Comments:

Wordy. Be more concise. Limit the use of parentheses.

I have restricted the use of the auxiliary verb as much as possible. I have deleted sentences using parentheses, for example, p.6740, L.12-13 and p.6742, L.23-24. Furthermore, I will receive the English editing service before the publication of the paper for AMT.

Figures:

Include approximate altitude axes on the right side of every figure that has PT as the y-axis.

I have revised figures from 2 to 5 and A1 as suggested. This is also mentioned in each figure caption.

Text:

Abstract:

Mention that SMILES is on the ISS.

I have added "on the International Space Station (ISS)" in the first sentence.

Line 16: change "smaller" to "lower"

I have revised it as suggested.

Maintain present tense, i.e., change "was" to "is" in multiple places.

I have changed them as suggested.

Provide differences for HCl and ClO in both concentration and percent.

I have added differences for HCl and ClO in both ppbv and percent in Abstract.

Include quantitative agreement between SMILES, MLS, and ACE HCl/Cly ratios.

I have also added information on HCl/Cly ratios in Abstract. Correspondingly, I have added information on derivation of the quantitative analysis in Sect. 4.3.

"Taking the average of the four HCl/Cly profiles shown in Figure 5, the difference (X – average) with regard to the average ranges from –5% in the 650–675 K bin for SMILES-NICT to 8% in the 625–675 K bins for ACE-FTS, where X is one of SMILES-JAXA, -NICT, MLS, and ACE-FTS."

Reword last sentence. Something like “The high HCl values and HCl/Cly ratios observed by the three instruments in the lower stratospheric Antarctic vortex is consistent with previous observations in late austral spring.”

I have revised the sentence as suggested.

1. Introduction:

Add a paragraph outlining the paper at the end.

I have added an outline of the paper at the end of Introduction.

2. Measurements of SMILES – consider changing to “Satellite Measurements”

Add Section 2.1 SMILES, 2.2 MLS, 2.3 ACE-FTS. Move text from beginning of Methods section (page 10) back into the Measurement sections.

I have changed the construction of Sect. 2 as suggested. Descriptions about MLS and ACE-FTS measurements in Sect. 3 have moved to the new Sect. 2.2 and 2.3, respectively.

3. Method

Begin this section talking about the vortex and Derived Meteorological Products.

According to the above revision, this section begins with the vortex situation and the utilization of DMPs.

Page 12, Last Paragraph: Elaborate on the effect of differences in LST on HCl and ClO comparisons.

The last paragraph of Section 3 is now re-organized. The first sentence has moved to the 2nd paragraph of the new Sect. 2.1: "Because the ISS is in a non sun-synchronous circular orbit with an inclination angle of 51.6° to the equator, the SMILES measurement at each tangent point occurs at various local solar times (LST)." Some

results have moved to Sect. 4.2: "Although the number of data points is small, the SMILES ClO values in the SZA range between 85° and 95° are zero to 0.05 ppbv between 400 K and 700 K levels. For measurements with SZAs larger than 95°, ClO reveal values around zero."

To elaborate on this, I have changed the last paragraph of Sect. 3.

"Figure 1 shows measurement latitudes as a function of LST used in this study (data taken inside the vortex). As mentioned in Sect. 2.1, the SMILES measurements occur at various LST. Therefore, it is crucial to consider the diurnal cycle of ClO and ClONO₂ in comparisons between SMILES, MLS, and ACE-FTS. In the following section, we consider SMILES and MLS ClO data only for solar zenith angles (SZAs) less than 85° representing daytime measurements. The daytime measurements from SMILES occurred at LSTs between 03:00 and 09:00 LST, whereas the daytime measurements from MLS occurred at LSTs of 14:00–15:00 LST. These LST differences are carefully treated in the following discussion. Data with SZAs larger than 85° are color-coded in black for each symbol. For ACE-FTS measurements, all of the occultations occurred at a SZA of 90° from the satellite sunset for this time and location."

Changes by authors in proof-read.

1. In Sect. 1, I added a reference describing the increased amount of HCl in the Antarctic.
2. In Sect. 4.3, I changed from 'measured' or 'observed' Cly to 'composite' Cly, since SMILES and MLS do not measure ClONO₂.

Response to Referee #2

We would like to thank the anonymous referee #2 for reading this manuscript and offering valuable suggestions. We hope to have addressed all the comments in the reply below.

One general question which came to my mind is, if the authors have considered other satellite data sets for their comparison as well? In particular, I was wondering if Envisat MIPAS data are available for the time period considered in this study? Such additional data may be helpful regarding a more thorough analysis of the Cl partitioning.

An HCl product from Envisat/MIPAS is not available. However, I have re-organized a paragraph (p.6746, L.10-22) adding information from the MIPAS measurements of ClONO₂. I have analyzed the latest IMK/IAA ClONO₂ data product in the period of 19-24 November, 2009, and found that there was a difference of 0.2 ppbv in the 675-700 K potential temperature bin between daytime and nighttime measurements. This is added in Sect. 4.3 as one paragraph.

"In addition, we further analyzed data taken by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard Envisat (von Clarmann et al., 2009, 2013). We used the Institute for Meteorology and Climate Research (IMK) / Instituto de Astrofísica de Andalucía (IAA) ClONO₂ data product, version V5R_CLONO2_220. The data were extracted for latitudes between 60°S and 66°S and longitudes between 60°W and 120°W, which correspond to the vortex interior during the period of 19–24 November 2009. Since the MIPAS measurements occurred at both day (09:48 LST) and night (22:30 LST) times, the diurnal change in ClONO₂ can be seen. For instance, the average and its one sigma standard deviation was 0.91±0.07 ppbv (averaged over 11 data points) for the daytime and 1.13±0.14 ppbv (averaged over 10 data points) for the nighttime in the 675–700 K PT bin. The difference of 0.22 ppbv is comparable to the value of SMILES ClO (0.21 ppbv) in the same PT bin, supporting the conclusion that the diurnal cycle of ClO is caused by the photolysis of ClONO₂."

I also put a sentence at the end of Sect. 4.2: "A further discussion of the diurnal changes in ClO and ClONO₂ is provided in the next subsection."

Specific Comments

p6732, 123-29: At this point it might not be clear to the reader how low ozone values relate to the Cl+CH₄ or ClO+NO₂+M reactions. This is explained in section 4.3 in more

detail and a forward reference might be okay to clarify this.

I have added "(see Sect. 4.3)" in the sentence.

p6733, 14-12: What is causing the differences in Cly partitioning in the Arctic and the Antarctic?

The cause of the difference is added referring Douglass et al. (JGR, 1995). An exception in the 2002 Antarctic is also added.

"Very low ozone values in the Antarctic play a central role in the difference in Cly partitioning between the Arctic and the Antarctic (e.g., Douglass et al., 1995), except in the unusual Antarctic winter 2002, when some deactivation into ClONO₂ occurred (e.g., Grooß et al., 2005; Höpfner et al., 2004)."

p6733, 123-25: Could you explain a bit more in detail which changes in the ISS observation geometry have to occurred that facilitated measurements at southern high latitudes? Was the time period 19-24 November the only one where measurements at southern high latitudes are available?

I have added one paragraph in new Sect. 2.1: "Because the ISS is in a non sun-synchronous circular orbit with an inclination angle of 51.6° to the equator, the SMILES measurements at each tangent point occur at various local solar times (LST). For several specific periods, including 19–24 November 2009, the ISS rotated 180° around its yaw axis, and thus the observation latitude range was shifted to southern high latitudes up to 66°S. A more detailed description of the observation latitudes and periods is found in Figure 1 of Kasai et al. (2013). There were also southern high latitude measurements on 10–19 February and 8–17 April, 2010."

p6734, 15...: I would suggest to add a paragraph to section 2 that describes the observation geometry of SMILES and the ISS in more detail. The information can be found in different sections of the paper, but it might be more clear to have it in one place.

As mentioned above, I have added the information in new Sect. 2.1.

The paragraph at p6734, 115-22 describes the spectral measurements and ends with just one sentence on vertical resolution. I did not found any information on the vertical range of the measurements. The ISS orbit is first explained in the method section (at p6740); I think this should also be introduced much earlier.

I have added the information: "vertical range of the measurements is from ~ 12km to

96 km." As mentioned before, the ISS orbit is now explained in new Sect. 2.1.

p6735, 126: The optimal estimation retrieval is based on an a priori mean state x_a and a corresponding covariance matrix S_a . How was S_a chosen in the two retrieval schemes? Is a diagonal matrix used or are there smoothing constraints included?

For the JAXA's retrieval, a priori errors for HCl and ClO are about 40% and 100%, respectively, which are used in diagonal elements of S_a . A smoothing constraint is also included as a correlative length of 10 km for both HCl and ClO. For the NICT's retrieval, a priori errors for HCl and ClO are about 50% and 60%, respectively, and a correlative length is 6 km for both HCl and ClO.

We have not added this to the text, because we feel that it is a supplementary information.

p6736, 113-22: I have no idea how these differences in line frequency, air broadening, and temperature dependence would affect the simulated spectra. Are these differences in forward modelling causing significant retrieval errors?

I have added the effect of the difference in new Sect. 2.1: "There are also differences in spectroscopic parameters used in each forward model; however, the impact of the differences on the retrieved data products seems to be small in the lower stratosphere, as shown in Sagawa et al. (2013) for ClO and Yokoyama et al. (to be submitted to J. Geophys. Res.) for HCl."

I have made Table 1 describing the spectroscopic parameters used for each SMILES retrieval, as Referee #3 also pointed this out.

p6737, 18-9: The NICT precision is 1-2% which seems much smaller than the JAXA error ("less than 10%")? What is the reason for this, if measurements are the same?

Indeed, there is no large difference between JAXA and NICT HCl precisions. I have reported the JAXA's precisions more precisely as 10% at 15 km and 1% at 30 km in new Sect. 2.1.

p6738, 15-7: It is stated that NICT profiles with a chi-square value larger than 0.6 are discarded. However, a chi-square of one would be expected on average (Rodgers, 2000). This could mean that there is a problem with the errors or the a priori uncertainties in the retrieval or that you are over-pessimistic and throwing away good measurements? Please clarify.

First of all, I apologize my mistake not to have revised the number as 0.8 (also the

measurement response as 0.8). All of the analysis in this paper are indeed based on this data screening, in line with the other studies such as Sagawa et al. (AMTD, 2013). Now, I just revised the numbers as 0.8. For the NICT retrieval, chi-square is the summation of the squared and variance weighted residuals in the measurement space and the null space after they are normalized by the number of measurements and retrieval parameters. Typical chi-square values are smaller than unity because of the overestimation of the measurement noise (Baron et al., AMT, 2011). Thus, no good retrievals have been discarded by this data selection. This is added in new Sect. 2.1 (moved from Sect. 3, as suggested by Referee #1). In practical, profiles with chi-square values larger than 0.6 are a small number, so that most of the profiles used in this study have chi-square values less than 0.6 for both HCl and ClO.

p6738, 115-21: It is difficult to keep all the different precision, accuracy, and resolution estimates for ACE-FTS, MLS, and SMILES in mind, which are found in different parts of the paper. It might be helpful to include a summary table.

As suggested also by Referee #1, now all descriptions of satellite sensors are moved to Sect. 2. So that, I hope this makes it easier to read.

p6741, 125: I think it would be more accurate to say that the differences are due to different forward models (and retrievals schemes) rather than "retrieval approaches" only? (In fact you mention in the same paragraph that you found no significant differences due to the a priori used in the retrievals, but that the differences in modelling of continuum absorption are a possible source of differences.)

The difference arises from different approaches including both forward models and retrieval schemes. So that, more precisely, I have revised it as "different approaches used in the forward models and retrieval schemes". Why the impact of a priori difference on the HCl retrieval is small is now mentioned in new Sect. 2.1: "However, as stated in Sect. 4.1, the impact of the difference on the retrieved HCl values is insignificant because of the high sensitivity of the measurements studied here."

p6744, 16: Negative ClO volume mixing ratios in the MLS data may be a mathematically correct solution of the inverse problem, but they are physically unrealistic. I am always curious how these negative values are treated in the forward model?

We do not use any non-negative constraints in either MLS or SMILES retrievals for

CIO. Generally speaking, when the spectral signal is weak, a negative value would occur in the retrieved result because of the spectral noise or inadequate corrections of the baseline. For MLS, there seem to be interferences from CH₃Cl, which has lines in two wing channels of the band used to measure CIO, and CH₃OH, which has a cluster of lines in the image sideband with an intermediate frequency nearly the same as that of CIO (Santee et al., JGR, 2008). This may result in larger negative values in MLS than in SMILES. As stated in Livesey et al. (2011), the negative bias in CIO is largely mitigated, primarily through retrieval of CH₃Cl, in v3.3. Such a bias should be corrected as a function of geophysical variables such as ozone and temperature, most likely to be giving rise to the spectral features that induce the bias (Livesey et al., 2011).

We have not added this to the text, because we feel that it is a supplementary information.

p6745, 113-15: Here you mention a case where observations in 2004/2005 contradict the hypothesis of "low ozone leads to large HCl". What is the meaning of this for your study? Is this an unusual, random event or part of inter-annual variability? Are there any problems with the observations?

The reasons for this difference may not be fully understood. I have deleted these two sentences.

p6746, 110-12: It is stated that CIONO₂ values for the local solar times of the SMILES and MLS observations would be needed for a precise calculation of Cly. How large are the errors quantitatively when using ACE-FTS data as a proxy?

As I stated at the beginning of this reply, there is a 0.2 ppbv difference between daytime and nighttime measurements from MIPAS. Thus, such a difference (at most) in the estimate of Cly would be plausible for the error from using the ACE-FTS CIONO₂ value as a proxy. I have added a sentence: "As mentioned above, the Cly value calculated from Equations (1) and (2) may have a positive bias up to ~ 0.2 ppbv, amounting to ~ 6% for a Cly value of 3.2 ppbv." in Sect. 4.3.

p6747, 117: I think you do not need to mention again in the summary that measurements are made with a 4-K mechanical cooler and SIS mixers.

I have deleted it as suggested.

p6747, 125: It seems 15-20% would be a more appropriate value for the NICT HCl

errors rather than 10% (looking at Fig. 2 and 3)?

I have put more accurate numbers in Abstract and Conclusions. I have revised two sentences: "The SMILES-NICT HCl agrees within 10% with the MLS HCl between 450 and 575 K and with the ACE-FTS HCl between 425 and 575 K. Above 575 K, the SMILES-NICT HCl values are, however, 11–19% smaller than those from MLS and 16–22% smaller than those from ACE-FTS." in Conclusions.

Appendix: It seems there is no link to the appendix in the main text of the paper. Perhaps add a reference to the appendix in section 4.3?

Indeed, I had been added such a reference on p.6746, L.22.

Fig. 2: Do the precision curves show the mean of the precision values of the individual measurements or are these scaled by $1/\sqrt{n}$?

This is an average value not scaled by $1/\sqrt{n}$.

Fig. 2: What do the vertical lines at each square indicate? Do they illustrate the bin width? Perhaps better remove these to make the plot less busy?

I have removed them as suggested.

Fig. 2: There are only few data points for MLS at the 500K and 650K levels, resp. Is the 3.4 ppb maximum value for MLS at 500K reliable or is it just cause by the small number of data points (6 in this case)?

For the 475-500 K bin (ave. = 3.4 ppbv, N = 6), the values range from 3.0 ppbv to 3.6 ppbv. For the 625-650 K bin (ave. = 2.1 ppbv, N = 8), the values range from 1.8 ppbv to 2.4 ppbv. I think both two averaged data are not influenced by some outliers in the respective bins. Thus, I think these are reliable.

We have not added this to the text, because we feel that it is a supplementary information.

Fig. 3: You may consider showing the differences between ACE-FTS and MLS in this plot as well. There seems to be very good agreement above 550-600K, while both SMILES retrievals are about 20% lower?

I have added one paragraph at the end of Sect. 4.1: "The HCl values from MLS and ACE-FTS agree to within 10% above 550 K (not shown). In general, both SMILES retrievals are a maximum of 23% smaller than MLS and ACE-FTS at these levels."

For reference, a plot showing differences between MLS and ACE-FTS HCl is below:

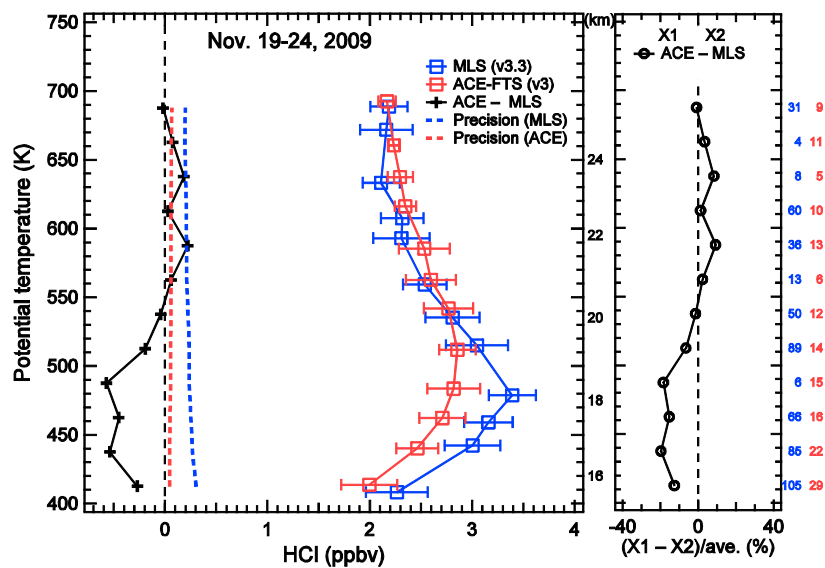


Fig. 4: The x-axis label in the middle panel should be replaced by " $(X1 - X2)$ (ppbv)" for consistency with the right panel.

I have revised it as suggested.

Fig. 5: "SMILES-JAXA ClO" and "ACE-FTS ClONO₂" should have different line styles.

I have changed symbols for ACE-FTS ClONO₂ to X-marks.

Technical Corrections

p6732, 11: as the sum of the volume mixing ratios of Cl,...

I have added it as suggested.

p6732, 118: from the Atmospheric

I have revised it as suggested.

p6732, 119: Space Shuttle were conducted

I have revised it as suggested.

p6733, 12: remove "At that time" (?)

I have removed it as suggested.

p6740, 12-3: change "grid" -> "grid point"

I have added it as suggested.

p6740, l3: remove "above"

I have removed it as suggested.

Fig. A1, caption: deduced_from_the N2O values

I have added it as suggested.

Changes by authors in proof-read.

- 1. In Sect. 1, I added a reference describing the increased amount of HCl in the Antarctic.**
- 2. In Sect. 4.3, I changed from 'measured' or 'observed' Cly to 'composite' Cly, since SMILES and MLS do not measure ClONO₂.**

References:

- Baron, P., Urban, J., Sagawa, H., Möller, J., Murtagh, D. P., Mendrok, J., Dupuy, E., Sato, T. O., Ochiai, S., Suzuki, K., Manabe, T., Nishibori, T., Kikuchi, K., Sato, R., Takayanagi, M., Murayama, Y., Shiotani, M., and Kasai, Y.: The Level 2 research product algorithms for the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), *Atmos. Meas. Tech.*, 4, 2105–2124, doi:10.5194/amt-4-2105-2011, 2011.
- Douglass, A. R., Schoeberl, M. R., Stolarski, R. S., Waters, J. W., Russell, J. M., Roche, A. E., and Massie, S. T.: Interhemispheric differences in springtime production of HCl and ClONO₂ in the polar vortices, *J. Geophys. Res.*, 100, 13967–13978, doi:10.1029/95JD00698, 1995.
- Grooß, J.-U., Paul, K., and Müller, R.: Ozone chemistry during the 2002 Antarctic vortex split, *J. Atmos. Sci.*, 62, 860–870, doi:10.1175/JAS-3330.1, 2005.
- Höpfner, M., von Clarmann, T., Fischer, H., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., Mengistu Tsidu, G., Milz, M., Steck, T., Stiller, G. P., Wang, D. Y., and Funke, B.: First spaceborne observations of Antarctic stratospheric ClONO₂ recovery: Austral spring 2002, *J. Geophys. Res.*, 109, D11308, doi:10.1029/2004JD004609, 2004.
- Kasai, Y., et al.: Validation of stratospheric and mesospheric ozone observed by SMILES from International Space Station, *Atmos. Meas. Tech.*, 6, 2311–2338, doi:10.5194/amt-6-2311-2013, 2013.
- Livesey, N. J., Read, W. G., Froidevaux, L., Lambert, A., Manney, G. L., Pumphrey, H. C., Santee, M. L., Schwartz, M. J., Wang, S., Cofield, R. E., Cuddy, D. T., Fuller, R. A., Jarnot, R. F., Jiang, J. H.,

- Knosp, B. W., Stek, P. C., Wagner, P. A., and Wu, D. L.: Earth Observing System (EOS), Aura Microwave Limb Sounder (MLS), Version 3.3 Level 2 data quality and description document, D-33509, Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California, 2011.
- Sagawa, H., Sato, T. O., Baron, P., Dupuy, E., Livesey, N., Urban, J., von Clarmann, T., de Lange, A., Wetzel, G., Kagawa, A., Murtagh, D., and Kasai, Y.: Comparison of SMILES ClO profiles with other satellite and balloon-based measurements, *Atmos. Meas. Tech. Discuss.*, 6, 613–663, doi:10.5194/amtd-6-613-2013, 2013.
- Santee, M. L., et al.: Validation of the Aura Microwave Limb Sounder ClO measurements, *J. Geophys. Res.*, 113, D15S22, doi:10.1029/2007JD008762, 2008.
- Yokoyama, K., et al.: First observation of HCl in the upper stratosphere and mesosphere measured by Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), to be submitted to *J. Geophys. Res.*

Response to Referee #3

We would like to thank the anonymous referee #3 for reading this manuscript and offering valuable suggestions. We hope to have addressed all the comments in the reply below.

p. 6739 L 25 The equivalent latitudes are -71 for SMILES and -76 and -75 for MLS and ACE-FTS. I would have thought that might be enough difference for SMILES to be nearer the Antarctic 'collar'. MLS should have enough data to discern if this much difference in equivalent latitude might affect some of the comparisons.

I have analyzed MLS data only for regions between -65 and -73 equivalent latitudes inside the Antarctic. However, the HCl values at those region (close to the vortex edge) are very close (mostly less than 2%) to HCl values derived from the whole data as listed in Table below.

| PT bin | whole | | edge | |
|---------|---------------|-----|---------------|----|
| | HCl (ppbv) | N | HCl (ppbv) | N |
| 400-425 | 2.26 | 105 | 2.28 | 61 |
| 425-450 | 3.00 | 85 | 2.98 | 17 |
| 450-475 | 3.16 | 66 | 3.16 | 25 |
| 475-500 | 3.39 | 6 | 3.23 | 3 |
| 500-525 | 3.05 | 89 | 2.98 | 18 |
| 525-550 | 2.81 | 50 | 2.84 | 17 |
| 550-575 | 2.54 | 13 | 2.54 | 13 |

We have not added this to the text, because we feel that it is a supplementary information.

I have trouble figuring out which of the possible causes for difference in retrieval are important. For example:

First discuss differences in approach - although I have no sense of the magnitude of the differences, the physical argument (that the larger spectral interval gives more information) at least make sense intuitively.

I have added the magnitude of the difference on the retrieved results, as listed below.

The paragraph on 6736 lines 13-23 about differences in the spectroscopic data is difficult to read, and there is no sense of how such differences might contribute to differences in the final product. Also, a table might be more readable.

I have added the effect of the difference in new Sect. 2.1: "There are also differences in spectroscopic parameters used in each forward model; however, the impact of the differences on the retrieved data products seems to be small in the lower stratosphere, as shown in Sagawa et al. (2013) for ClO and Yokoyama et al. (to be submitted to J. Geophys. Res.) for HCl."

I have made Table 1 describing the spectroscopic parameters used for each SMILES retrieval, as Referee #2 also pointed this out.

In the discussion of the differences in the retrievals - raise the possibility of the a priori (p. 6735, l 24) and then say later that it isn't important (p. 6741 L 26)

I have added a sentence in new Sect. 2.1: "However, as stated in Sect. 4.1, the impact of the difference on the retrieved HCl values is insignificant because of the high sensitivity of the measurements studied here."

On P. 6740 call out the uncertainty in the meteorological input (up to 1K) but again no sense as to how large an error might come from this.

I have put the degree in the sentence: "..., which has a small impact on the calculation of PT."

I looked at Livesey et al. and could not find anything to suggest the high HCl bias for values greater than ~ 3ppbv. So this must come from somewhere other than the summary table 3.9.1 which mentions bias only at low latitudes 147 hPa.

Figure 3.9.1 of Livesey et al. (2011) shows a positive bias of 0.2 ppbv in the band 14 retrieval in the upper stratosphere where the HCl value is greater than 3 ppbv. It is possible that the difference between SMILES and MLS HCl could be partly explained by this bias, as mentioned in p.6742, L.21-23. I have mentioned this feature.

Grammar p 6731 L 6 it is suggested that HCl IS (rather than was)

I have revised it as suggested.

P. 6732 L 19 measurements from the Atmospheric Trace MOlecult Spectroscopy on the Space Shuttle WERE conducted (rather than was)

I have revised it as suggested.

p. 6736 While SMILES-NICT ff - this is not a sentence. Delete 'While'.

I have deleted it as suggested.

p. 6744 l 29 Since this high HCl happens every spring in the Antarctic vortex (now observed as a regular feature for about a decade), I would delete 'anomalous'.

I have deleted it as suggested.

p. 6745 L 9 Since the conversion is quite rapid for a least a week or two (derivatives of plots in Santee et al.) and far more rapid than Cl + CH₄ would suggest for Cl in any other part of the lower atmosphere, I would delete 'gradually'.

I have deleted it as suggested. Further, I have added a reference regarding this point: "A recent theoretical study suggests that this conversion is quite rapid in the Antarctic vortex through the reaction (R4) and a reaction Cl + CH₂O (Groß et al., 2011)."

p. 6747 I don't know what is meant by "This strongly suggests that even in the late November period HCl dominates Cly inside the Antarctic vortex in the lower stratosphere, when the vortex situation was somewhat distinct compared to other years."

I have deleted words starting with "when the vortex...".

p. 6748 L 25 'dedicated' is the wrong verb. Do you mean it is strictly speaking appropriate only for the Arctic? It seems like that is where you are heading with the later discussion of older air in the Antarctic than in the Arctic.

I do not think it is only for the Arctic. I changed it to "calculated for".

Changes by authors in proof-read.

- 1. In Sect. 1, I added a reference describing the increased amount of HCl in the Antarctic.**
- 2. In Sect. 4.3, I changed from 'measured' or 'observed' Cly to 'composite' Cly, since SMILES and MLS do not measure ClONO₂.**

References:

- Groß, J.-U., Brauttsch, K., Pommrich, R., Solomon, S., and Müller, R.: Stratospheric ozone chemistry in the Antarctic: what determines the lowest ozone values reached and their recovery?, *Atmos. Chem. Phys.*, 11, 12217–12226, doi:10.5194/acp-11-12217-2011, 2011.
- Livesey, N. J., Read, W. G., Froidevaux, L., Lambert, A., Manney, G. L., Pumphrey, H. C., Santee, M. L., Schwartz, M. J., Wang, S., Cofield, R. E., Cuddy, D. T., Fuller, R. A., Jarnot, R. F., Jiang, J. H., Knosp, B. W., Stek, P. C., Wagner, P. A., and Wu, D. L.: Earth Observing System (EOS), Aura Microwave Limb Sounder (MLS), Version 3.3 Level 2 data quality and description document, D-33509, Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California, 2011.
- Sagawa, H., Sato, T. O., Baron, P., Dupuy, E., Livesey, N., Urban, J., von Clarmann, T., de Lange, A., Wetzel, G., Kagawa, A., Murtagh, D., and Kasai, Y.: Comparison of SMILES ClO profiles with other satellite and balloon-based measurements, *Atmos. Meas. Tech. Discuss.*, 6, 613–663, doi:10.5194/amtd-6-613-2013, 2013.
- Yokoyama, K., et al.: First observation of HCl in the upper stratosphere and mesosphere measured by Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), to be submitted to *J. Geophys. Res.*