Re-review of "Comparison of Inorganic Chlorine in the Antarctic and Arctic lowermost stratosphere by separate Late Winter aircraft measurements" by Jesswein et al.

The manuscript has been substantially revised in response to the comments of the two referees. In general, the authors have done a good job in responding to the points raised by the reviewers, and the manuscript has been much improved. However, some of the concerns from the original reviews have not been adequately addressed, and new issues have been introduced through the revision process. Thus I feel that further corrections and clarifications, as detailed below, are necessary before the paper can be accepted for publication.

As before, both major substantive issues and minor points of clarification, wording suggestions, and grammar / typo corrections are listed together for each Section (including Supporting Information) in sequential order through the manuscript.

Respectfully,
Michelle Santee

Abstract & Section 1:

- L1: extend --> extent
- L12-13: could be --> was (in all three places in these two lines)
- L50: delete "results"

Section 2:

- L78: No need to define "RGA" here as the airport codes are given in L84-85
- L90: type --> types
- L98: ERA-5 data --> ERA-5
- L99: dynamic --> dynamical
- L100: the 2 PVU --> 2 PVU
- L101: potential temperature … as tropopause --> the potential temperature … as the tropopause
- L110: the spelling of "ionisation" here is inconsistent with that in L111 and L144
- L122: precision … measurement were --> precision … measurements was
- L128: 0.2% and 0.64% --> 0.2% and 0.64%, respectively

Section 3:

- L146: region --> region of origin
- L150: reanalysis --> reanalyses
- L153: theta has already been defined in L53 and is again defined in L174, so it does not need to be defined here
- L154-160: Much of this discussion repeats concepts that have already been covered adequately in the Introduction (L47-54). Since the audience for this paper is likely to be quite familiar with this background material, it is arguably not necessary to include it at all, but in any case it certainly does not need to be reiterated. I suggest merging and streamlining the description in these two places. In addition, it would be better not to have two sentences in one paragraph starting with "A tracer like N_2O …".

- L158: a subsidence --> subsidence
- L160-161: a small variability --> small variability
- L162-163: "the vertical gradient in mid-latitude N_2O" – is vertical gradient really meant here, or horizontal gradient?
- L163: it would be clearer to say "both tropical and polar air"
- L171-172: affected by the diabatic decent --> affected by diabatic descent [typo: "decent"]
- L176-202: Overall, the description of the reference profile derivation has been improved in the revised manuscript. However, additional points of clarification are needed:
  - L183: I am confused about the binning in θ (or Δθ). It is stated here that the measurements are binned in 5 K intervals. But according to the Appendix, that is true only for the vortex reference; for mid-latitudes, a 2 K bin was used. But then Figure S2 and its caption suggest that both 2 K and 5 K bins are used for both vortex and midlatitude profiles at different stages of the process. Please clarify.
  - L189: the descending stops --> descent generally stops
  - L192: I do not think that "calculated" is the right word here. Measurement points are being categorized to segregate them into the N_2O_{vor} and N_2O_{mid} data sets, but saying that these data sets are "calculated" gives the impression that some other manipulation is being done.
  - L194-199: These sentences are particularly badly written and the English is very confusing. Words like "added" are misused, and, unless I have misunderstood, the N_2O data sets are being mixed up with the fit functions. Assuming this is an accurate description, I suggest rewriting these lines as: "The following then applies for each N_2O measurement: if the mixing ratio is below the vortex fit function plus the prescribed cutoff value, then it is assigned to the N_2O_{vor} data set. Otherwise, if the mixing ratio is above the mid-latitude fit function minus the associated variability, then it is assigned to the N_2O_{mid} data set. Mixing ratios above the vortex fit function plus the prescribed cutoff value and below the mid-latitude fit function minus the associated variability are assigned to the boundary region. Measurements for which the vortex fit function plus the prescribed cutoff value and the mid-latitude fit function minus the associated variability overlap cannot be uniquely classified and are assigned to both the vortex and the mid-latitudes in later analysis."
  - L200: the prescribed cutoff value --> the prescribed vortex cutoff
  - L201: variability N_2O --> variability in N_2O
  - L201-202: The reader is referred to the supporting information "and reference therein". However, no references are given in the SI, and in fact this discussion has been moved from the SI to the Appendix. But rather than refer to the Appendix, it would probably be better to simply cite Strahan et al. (1999) here for this point.
  - Figure 2 caption: Cutoff criterion --> The vortex cutoff criterion (see main text for details); where cutoff and mid-latitude variability crosses --> where the vortex cutoff and mid-latitude variability cross
- L213: Vortex and boundary region --> The vortex and boundary regions
- L214: The first phase contains some flights that have predominantly sampled vortex --> The first phase includes some flights that predominantly sampled the vortex
- L216: vortex boundary layer --> vortex boundary air

Section 4:

- L222: took --> take; this means sampling air along --> this means that air is sampled along
- L228: both channels --> the two channels
- Figure 4 caption: measured on … and on --> measured in … and in
- L230: measurements on --> measurements in
- L235: show a higher --> show higher
- L238: up-sampled --> up-sampled data
- Figure 5 caption: The first sentence of the caption needs to be re-written to make it clear what the comparison is with. Also, measured on --> measured in
- L244: Air enters predominantly --> Air enters the stratosphere predominantly
- Section 4.2: The discussion of the semi-direct and indirect calculations of Cl_y remains quite confusing.
  - Unless I am mistaken, what is referred to here as the "semi-direct" calculation – that is, Eqn. (1) – is performed twice: once with CCl_y obtained from up-sampled GhOST-MS measurements, and a second time with CCl_y calculated from CFC-12 alone. This is not immediately clear from the current text, but it should be spelled out explicitly. For example, the distinction can be drawn between the calculations mentioned in L257-258 and those mentioned in L282-284, with Eqn. (1) referenced in both places.
  - L275-277: These lines talk about comparing the semi-direct Cl_y values from SouthTRAC to "indirectly determined" values, but Figure 6 shows only the correlations of various species with CFC-12, not Cl_y. Also, indirectly determine --> indirectly determined
  - L280: form --> from
  - L284: delete "hereafter"
  - Then, again assuming that I have understood the process, once CCl_y has been derived just from CFC-12 data (the second step above), a curve is fit to those values to obtain the coefficients for the "indirect" method of Eqn. (2). Is that correct? The problem is, at this point the reader has no idea why this extra step is needed. So far only SouthTRAC measurements have been discussed, and for them Cl_y can be obtained semi-directly from Eqn. (1) with the in situ CCl_y data (the first step above). The justification of the need for the indirect approach should come *before* it is presented, not after.
  - In addition, the paragraph in L259-294 is extremely long and tiring to read. Thus I suggest inserting a paragraph break in L284, after "between Cl_{total} and CCl_y". The new paragraph should begin with a brief sentence of explanation as to why it is necessary to go through the effort of implementing the indirect approach, maybe something along the lines of: "With the good agreement between the two semi-direct Cl_y calculations, we explore whether Cl_y can be successfully estimated from CFC-12 for situations in which measurements of CCl_y are not available. That is, a correlation function …".
  - L288: correlation to --> correlation with
  - L291: Just to be really clear: CFC-12 … is used for --> CFC-12 … is used as the reference in Eqn. (2) for
- L297: Very young air also shows larger differences between the two Cl_y estimates.
- L298: the suggested lifetime --> its suggested lifetime
- L301: I think it would be clearer to say "… age tracers. The fundamental picture does not change, however, hence we use the uncorrected mean age of air."
- L314: delete "of θ"

- L315: errorbars --> error bars
- L316-317: Cl_y is given for all measurements --> the Cl_y is estimated based on all measurements
- L317-318: Cl_y is given according to the region --> Cl_y is estimated separately in each region
- L320-322: First, Figure 8 should be referenced again at the beginning of this paragraph. Second, it is stated that the SouthTRAC measurements of long-lived chlorinated substances are consistent with the Cl_{total} from AGAGE, but I'm not sure exactly what that means. Is this statement referring specifically to the troposphere?
- Figure 8 caption: -90° to -40° --> 40°-90° S
- L329: at highest --> at the highest
- L330: that in 2018 mid-September --> that in mid-September 2018
- L344-345: provide comparable --> provide values comparable
- L352-353: With a mean PV-based tropopause at 306 K, during PGS, it is only slightly lower than during SouthTRAC with 308 K --> The mean PV-based tropopause was at 306 K during PGS, only slightly lower than that during SouthTRAC at 308 K
- L355-357: First, I'm not sure what "considering the long-lived chlorinated substances" means. Does this statement refer to WMO (2018)? More importantly, the difference between the rate of decline seen in this study and that reported in WMO (2018) (~16 vs. 12.7 ppt/yr) is nonnegligible (~25%). Can the authors speculate on what is giving rise to this discrepancy? Does it imply something about the accuracy of their estimated Cl_y values?
- L362-364: This sentence is very difficult to read and confusing. Assuming that I have interpreted it correctly, I suggest re-writing as: "The maximum fraction of total chlorine in the form of Cl_y during PGS at the same distance from the local tropopause as the maximum SouthTRAC Cl_y fraction is about 20% in the mid-latitudes (not shown) and about 40% inside the vortex (Fig. 9b)." Note that the original sentence suggested that the comparison was being made at the same $\Delta\theta$ as the largest values *inside the vortex* during SouthTRAC, but I assume that that was not what was actually done for the mid-latitudes. Also, if the NH mid-latitude result has been shown in this paper, then a specific pointer to it should be added.
- Figure 9 caption: It would be better to turn the fourth sentence around: PV tropopauses for PGS (black) and SouthTRAC (green) are displayed as dashed horizontal lines with the 1σ variability as shaded areas.
- L367-368: of each respective hemisphere --> of the two hemispheres
- L376: For maximum clarity, add "(Fig. 9)" after "measurements". Also, rather than "vortex and outer vortex", it would be better to say "vortex core and vortex edge".
- L376-379: It doesn't make sense that two sentences about the mid-latitudes (L377-378) are interposed between sentences (L376 and L379) talking about the vortex comparisons. Thus the sentence "It must be noted that … transport barrier." should be moved up and edited/merged with the sentence in L376.
- L380: The comparison on equivalent latitude is therefore only possible to a limited extent --> Therefore performing the comparison in equivalent latitude / potential temperature coordinates removes only some of the sources of discrepancy
- L380-395: The entire discussion of the interhemispheric differences in Fig. 10 is poorly written and not well thought-through. The authors have placed on the reader the burden of figuring out how previously reported BDC and age of air differences may be related to the

differences in Cl_y that they observe. The linkage between differences in age of air (and the trends therein) and Cl_y differences must be drawn much more explicitly in the paper. The discussion of how their results relate to previous mid-latitude trends is also muddled.

- L382: Konopka et al. (2015) showed, that north of 60 °N, age of air is always younger than south of 60 °S in the same season --> Konopka et al. (2015) showed that the age of air is always younger north of 60 °N than south of 60 °S in the corresponding season
- Since air is younger north of 60N than it is south of 60S, it seems to me that lower values of Cl_y are expected at NH high latitudes than at SH high latitudes, as seen in Figs. 9 and 10. I do not believe that such a statement is made in the manuscript.
- How do the differences in Cl_{total} between the two campaigns – which are not well explained as noted above – interact with / affect the interhemispheric Cly differences?
- The authors mention recent work quantifying interhemispheric differences in the trends in AoA, but again the connection to their Cl_y results is not made explicitly. If there is a positive trend in AoA in the NH (i.e., air is getting older) and a negative trend in AoA in the SH (i.e., air is getting younger), then wouldn't that mean that Cl_y should be getting slightly larger in NH mid-latitudes and smaller in SH mid-latitudes? But surely these trends are very small and could not be expected to be evident above interannual variability over the three-year interval between the campaigns. Thus I am not convinced that AoA trends have any relevance for the observed Cl_y differences.
- L385-390: The discussion of long-term trends in mid-latitude HCl and $ClONO_2$ has been largely lifted from WMO (2018) without attribution. While I applaud their desire to go back to original sources, the authors have been sloppy and careless in presenting this material. For example, they state that Mahieu et al. (2014) reported data "through the end of 2016". In fact, Mahieu et al. (2014) only show data through 2011; the plots are updated through 2016 in the WMO Report. Then it is stated that GOZCARDS lower stratospheric HCl shows "larger" decreases at SH latitudes – larger than what? Than the increases seen in the NH? The citation for this statement is Froidevaux et al. (2015), but a more recent paper by Froidevaux et al. published in 2019 would be a more up-to-date reference. Most importantly, nowhere in these lines in the manuscript are the actual trends for HCl and $ClONO_2$ quoted. According to WMO (2018), for the period 1997–2016, total column HCl decreased by $0.42\pm0.23\%yr^{-1}$ and total column $ClONO_2$ decreased by $0.60\pm0.39\%yr^{-1}$. It doesn't seem to me that trends of that magnitude could account for the >200 ppt difference in estimated mid-latitude Cl_y between the PGS and SouthTRAC campaigns seen in Fig. 10. Therefore the statement that "higher values of Cl_y in the mid-latitudes during PGS [seem] to be plausible" is not supported.

- L391-392: It is not clear to me what is meant by "the lowest 20 K above the local tropopause show weak impact of the stronger Antarctic". Is this sentence referring to the Antarctic vortex? Even if it is, its meaning is unclear. Moreover, in Fig. 10 bins in the lowest 20 K above the tropopause are not filled at the highest EqLs, so the relevance of the vortex is not clear.
- L392: Two exceptions are singled out for discussion, but other bins in the vicinity show differences nearly as large.
- L394: this range --> this θ range; almost zero --> generally small

Section 5:

- L411: Since a reference is included for the Arctic winter of 2015/2016, it would be appropriate to include one here for the Antarctic winter of 2019 as well.
- L422: can be --> was (twice)
- L423: vortex --> vortices
- L425: Elsewhere “inter annual” has been written as “inter-annual” (L18) and “interannual” (L37). Please be consistent.
- L431: can be --> were (twice)
- L432-433: The statement “These hemispheric differences can also be found in simulations based on reanalysis, e.g., Konopka et al. (2015)” is a bit misleading here because the preceding sentence was about Cl_y, whereas Konopka et al. (2015) did not discuss Cl_y. Again, the connection between AoA and the Cl_y calculated in this work needs to be made directly.
- L434: should be subject to --> should be the subject of
- L435: which are not only used to --> which are used to not only
- L436: reveal --> reflect

Appendix, References, & Supplemental Material:

- L455: this corresponds --> 6 ppb corresponds
- L460: outlier --> outliers
- L628: implivations --> implications
- L632: Arctiv --> Arctic
- SI, L4: requieres --> requires
- SI, L5: Figure S 5 --> Figure S 4
- SI, L10: Fig. S 7 --> Figure S 6
- Figure S 7: It needs to be made clear in the caption that the displayed curves were derived from CFC-12 from GhOST-ECD and N_2O from UMAQS.