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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Using Satellite Measurements of N_2O to remove dynamical variability from HCl measurements" *by* Richard Stolarski et al.

Anonymous Referee #2

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General overview

This paper presents a nice perspective on observed variability and trends in northern mid-latitude stratospheric HCl. It describes an approach whereby dynamical influences on that variability can be accounted for through consideration of a trace gas such as N2O, which shares many of the dynamical influences as HCl but experiences different chemical processes.

While I recognize the value of the method described, and am keen to see it appear in the literature, I am concerned the uncertainties ascribed to some of the numbers found/used by the authors are on the optimistic side. I wonder if more complete as-

sessment of these uncertainties might lead to a reduction in the reported "significance" of the result and this assessment might then suggest that a softening of some of the wording is merited. I also have a concern as to whether the ordering of the operations in their method is appropriate, and whether more robust results might be obtained if it were reversed. Both of these topics are expanded upon below.

The standard of English is reasonably high, but it would clearly have benefited from a more careful read through by the authors as there are several parts that are erroneously and/or ambiguously worded. I've endeavored to identify some of these, but fear I may have overlooked some others.

Major concerns

My concern about the ordering of steps in the method is as follows. I would have thought that it would have been better to "correct" the N2O for both the likely MLS drift and the surface growth rate before using it as an explanatory variable in the HCl analysis rather than, as appears to be the case, after. My sense is that this would lead to a corrected N2O variable that would do a better job of explaining the dynamical influences on the HCl, enabling a clearer trend to be obtained. The results may be little different in the end, but my sense is that the study would be better expressed in that manner. If nothing else, the authors would do well to enact that alternative formulation and comment on the difference it makes to the result (even if they chose not to show it in the end). It might make sense to include an actual algebraic expression for the fit and the various corrections. This would make for an easier description for the various terms involved and their uncertainties.

My more major concern relates to the uncertainties quoted for some of the results. This is particularly important given the extent to which many of them are only just statistically significant (using the authors' 2-sigma threshold). Firstly, it is clear that the level-to-level variations in the bottom line results are mostly driven by the reported N2O 190/640 drift (r=0.75 between it and the result) rather than by the observed HCl trend (r=-0.24). That is to say, the results are affected more by the "correction" than by the actual input (the latter being the HCl trend with the N2O fit term included). Accordingly, this correction deserves particular scrutiny. The degree of level-to-level changes in this drift term is large compared to the uncertainty quoted on many of the individual drifts. Arguably, the standard deviation (1-sigma=1.5%/decade) of these different estimates would be just as valid a measure of the uncertainty in any or all of them. Indeed it might have been just as valid to chose to use the multi-level-mean drift as the value for all levels, given the uncertainty introduced by the inherent assumptions being made. Foremost among those assumptions is the one that the N2O drifts seen in the first part of the MLS mission are the same as those expected in the post-2013 period, when the 640 GHz N2O product is unavailable. I would have thought that the uncertainties derived here might need to be inflated in some way to account for this. Might more information be gained through consideration of other MLS products measured in the same period? Fundamentally, I think more information is needed here (including from the MLS team) on these uncertainties and their validity.

My second concern on the uncertainty relates to the 0.05%/decade (2-sigma) uncertainty quoted on the impact of N2O emissions. Firstly, the use of a constant 2.8%/decade trend at all altitudes here strikes me as highly simplistic. There are factors such as changes in age of air (and its spectrum) that surely come into play and might lead to variations. Similarly, the use of a 3-year lag at all altitudes seems overly simplistic. I grant that these issues may only have a small impact, and they may be very hard to quantify from the measurements available. Thus, the use of a constant value may well be justified in that light. However, I find it hard to believe that, in the face of those issues, the 0.05%/decade 2-sigma uncertainty estimate is an appropriate one. If nothing else, I would urge the authors to validate this number through, for example, examination of CCM runs (to which this team has ready access). Quantifying the degree to which the modeled 45N N2O timeseries at different pressure levels tracks the surface trend would provide a useful measure of this uncertainty. This issue is perhaps tied up with the ordering one discussed above, as the use of N2O as an

explanatory variable for the sought-after HCl trends may absorb these factors to some extent (though I haven't thought this through fully). In my mind all these issues argue that a more complete exploration of their methods, their inherent assumptions, and the uncertainties therein should be included in the manuscript.

More minor points

— Page 1

Line 19: "Statistically" -> "Statistical"

Line 25: "altitude" -> "vertical" (as you're using pressures rather than altitudes in words that follow).

Line 27: "... amount of inorganic stratospheric chlorine. This marker can be ..." to avoid the ambiguity about whether it is the HCl or the inorganic chlorine that "can be measured from the ground and from satellites".

Line 33: Commas needed after "showed" and "measurements"

Line 34: "Inorganic chlorine" is more than just HCl and ClONO2, though granted the others may be minor. Or is the point that Rinsland et al. only measured those two species and argued that they are the bulk inorganic chlorine. Please clarify.

Line 37: Jungfraujoch misspelt

Line 38: "during the early 2000s. This was followed by an increase in the HCl column over Jungfraujoch from ..." to avoid the ambiguity about whether it is the HCl or the source gases (the most recent things being discussed) being referred to.

Lines 39-43: The way this is worded, it seemingly ignores the fact that Mahieu et al. also looked at this signal in MLS data (as embodied in the GOZCARDS dataset). Please reword accordingly.

— Page 2

Line 3: Quote the latitude of Jungfraujoch in the caption. Also, some redundancy, as you say the MLS data is a 100-10hPa column in one sentence and then talk about it being a partial column (without the numbers) later on.

Lines 10-15: Again, please be sure your wording is consistent with the use to which Mahieu et al. put MLS data.

Line 10: "results from simulations using the SLIMCAT model driven by..."

Line 21: July 2004 doesn't sound like "late 2004" to me.

Line 23: "altitude" -> "vertically resolved", given that the vertical coordinate is pressure.

Line 26: "has little change since" -> "shows little change from"

Line 30: Perhaps put "band 14" in quotes as it's jargon that's not explained earlier (and is presumably covered in the references given earlier in the paragraph).

Lines 32-34: Please clarify, has the N2O product been "redefined" since the release of v4.2, or was the redefinition part of v4.2 from the outset?

Line 34: Unless I've misunderstood, it's part of MLS that has "deteriorated" is it not? Starting at some point during the mission. The way this is worded it sounds like the MLS data files are somehow deteriorating with time (like food going off in the refrigerator) regardless of the time at which the observations were made. Please reword more precisely.

— Page 3

Line 1: "next" -> "following" sounds better to me.

Line 2: Are the "640 channel" measurements also from the v4.2 dataset or from some earlier version?

Lines 1-6: This would presumably be a good place to have a discussion about the validity of assuming that the pre-2013 drifts are representative of the post-2013 obser-

vations. (Or possibly on page 6, see later).

Figure 2: The way you've drawn this, with the shaded envelope being narrow at the left hand edge is not an accurate depiction of the manner in which the regression is capturing in the uncertainty in the fit. The way it's shown it implies that the regression is constrained to have a fixed value at t=t0, which is not the case (unless you specifically performed such a fit, which I doubt). I suggest you leave the envelope off to avoid this potential for confusion (I don't see a more accurate but clear way to depict this uncertainty graphically). The caption will need to be updated to match.

Line 9: Actually isn't this "mixing ratio" rather than "concentration"? (sorry to be picky)

Line 10: Actually the dashed line doesn't look that "heavy" to me.

Line 18/19: "...are shown as a percentage deviation..." sounds better to me.

Line 19: Define "seasonal mean", is it three-monthly averages (DJF, MAM etc.) or monthly averages?

Line 22: "look at" -> "examine" sounds more scientific to me.

— Page 4

Line 7: Perhaps "effects" -> "cycles"?

Lines 23-26: Add "MLS" before "HCl" (line 23) and "N2O" (line 24) and then delete "from MLS measurements of each constituent."

Line 24: add "a" before "de seasonalized"?

— Page 5

Line 2: "determined by" -> "that due to"

Figure 4: As with figure 2, I suggest you remove the "flared" red shading (and update caption accordingly).

Line 30 - Page 6 Line 1. The point about the "raw" and "Trend with N2O fit" being similar at the higher altitudes is a good one and makes geophysical sense to me. However, this then exposes a weakness in the authors' arguments and methods, in that the N2O drift and surface N2O trend terms add significantly to the "final" result, moving it far from the "raw" original. If dynamical variability is indeed "relatively small" at these altitudes then why do these modifying terms get the same "weight" at these upper levels as they do lower down where dynamical variability is significant? There seems to be some kind of inconsistency here that needs thought.

— Page 6

Lines 5-13: This is the other place where it would be good to talk about the validity of assuming pre- and post-2013 N2O drifts are consistent.

Table 1 caption: Suggest that you delete "with 2-sigma uncertainties" on line 18 and instead say at the end of the caption something like: "All uncertainties are quoted at 2-sigma".

Lines 25-30: This is where some discussion of age-of-air and related issues would clearly go.

— Page 8

Lines 12-14: Again, this point is seemingly at odds with the "final" results for the higher altitudes.

Line 29: "kkm" typo.

Lines 34-41: Doesn't the age-of-air spectrum come into this issue too? In any case, it would be best to "show your working" as to how the -4.9% estimate is arrived at here.

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