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Atmospheric Chemistry and Physics Discussions

Interactive comment on "More evidence for very short-lived substance contribution to stratospheric chlorine inferred from HCl balloon-borne in situ measurements in the tropics" *by* Y. Mébarki et al.

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General comments:

This manuscript presents balloon-borne in situ measurements of HCl from 2005 and 2008, above Teresina, Brazil. Such measurements in the tropics might help constrain the abundance of Very Short Lived Substances (VSLS), in terms of their contribution to total chlorine abundance (VSLS degradation products leading to Cl and ultimately, HCl) at the base of the stratosphere. Very low HCl should imply a very low contribution

from VSLS... This is relevant to the total chlorine budget, which is uncertain in part because of this unknown contribution (estimated at 50 to 100 pptv in the past).

Given that very low abundances are indeed measured, which supports past tropical aircraft data, one would like to know what the constraint really is. This depends on other measurements and previous estimates mentioned in the manuscript, as other temporary reservoirs are possible. It would be nice to have a clearer statement regarding the uncertainties, as the level of significance for the in situ data appears to be masked by random errors. Showing average values at the lowest altitudes might help the readers in this respect. If transport from above does affect the low altitude measurements, the estimated values are likely to represent upper limits for a contribution from VSLS; this may be a useful statement to make as well.

Some consistency with satellite data is discussed, although this argument is fairly weak, given the error bars in total chlorine implied from upper stratospheric satellite data.

It would probably be better if the VSLS contribution was arrived at from UT data on VSLS themselves, given the uncertainties and assumptions surrounding HCl, but this is a difficult question to answer well. The current manuscript is a worthwhile study and, if nothing else, an indication of an upper limit. However, the authors should attempt to clarify certain aspects relating to the uncertainties and the discussion (and the satellite comparisons). More specifics are given below.

Specific comments:

- One should understand that, regarding the issue of satellite data agreement versus models, including the previous (rough) estimates of about 100 pptv for VSLS, such comparisons do not carry significance at the better than 200 pptv level, given the possible systematic errors in MLS (or other satellite) measurements in the upper stratosphere; of course, the model estimates have additional un-

certainties. Therefore, measurements of VSLS in the UT/LS and measurements such as the SPIRALE HCl data are likely to carry the "burden of significance" in terms of possible VSLS contributions to total chlorine in the stratosphere. If the current manuscript (coupled with other evidence) can convincingly imply 85±35 pptv for this VSLS contribution [see comments on uncertainties below], the consistency with satellite data is only a mild connection, given the larger satellite data uncertainties.

- Some comments on uncertainty values:

> If this is not more clearly discussed or demonstrated, readers may have some trouble with the numbers you arrive at, in terms of "global" [total] uncertainties for the VSLS contribution.

> There are comments regarding the random uncertainties for the SPIRALE data, but can you state that systematic errors are most likely lower than 20 (or 10) pptv, so that the total error estimate arises mostly from the random errors? Further comments on this issue would be desirable, for clarification of the error bars. This is important because the total uncertainty will be a key number to associate with the likely VSLS contribution. The note (e.g., pg. 16173) on experimental scatter being 30 or 20 pptv at the 1 sigma level would seem to imply that this study is almost dealing with an upper limit... It may be difficult to state that

90 pptv HCl is not present, unless you can use some averaging arguments over a wider height range, possibly (why not?). In this respect, why not show actual average values as a solid line especially at the lower altitudes?

> It is also worth noting that 2 balloon flights may not suffice to "nail down" the upper limit for VSLS contribution to chlorine, so additional data may be useful as further confirmation in the future. This could mean that the error bars are somewhat too low, although this is difficult to estimate... A cautionary note about this would still be worth-

while.

- Pg. 16167, lines 19/20 (L19/20) and pg. 16172: It would be useful to have (or point to) a good (quantitative) argument, regarding the possible direct (and local, if one can really ignore transported HCl) contribution to measured HCl from the (longer-lived) CFC's? The local photolysis rate should be very small at low "enough" altitude in the tropics, but is this really a zero pptv contribution or possibly a small number approaching the 20 pptv that you measure? Without quantification (or a reference to this), one cannot have a really firm conclusion (with zero error). Only if this number (and associated uncertainty) provides less than 5-10 pptv can that source of error be ignored, in terms of assigning tropical HCl values to VSLS only.

Along these lines , the discussion on pg. 16174 mentions the possibility (and ref. to Laube et al., 2008) that up to 56 pptv inorganic Cl could come from CH3Cl (or maybe a similar SG)... So how much of this might (or might not) end up in the measured HCl? In the limit, could there not be zero VSLS contribution, with all the (25 pptv) measured HCl [from SPIRALE] really coming from longer-lived product degradation? Again, some clarification and a clearer argument need to be provided, assuming that the 25 pptv HCl value from SPIRALE (with 25 pptv total uncertainty) is a fair estimate.

Finally, the point is made elsewhere that aircraft data (Marcy et al.) have indicated very low HCl values in the tropics before. This is worth some emphasis and possibly further comments, as your data appear to confirm this previous information (correct?).

- Pg. 16189, L6-10: A shorter summary sentence is suggested, possibly as follows: "Our result regarding a VSLS contribution of X±Ypptv to stratospheric chlorine supports the previous agreement between MLS-inferred upper stratospheric total chlorine and model chlorine, taking into account about 100 pptv from VSLS,

although satellite results do not constrain the VSLS contribution to better than 200 pptv.

- Regarding the MLS data in Table 1 and Fig. 7: it is not clear that the version 1.5 data should be included, given that the latest data version (v2.2) should typically be viewed as a replacement (unless otherwise stated). If there is no obvious reason to do this (please state if there is), it would be best to simply include v2.2 data (and the Table does not mention what version is used). Fig. 7 does not make it clear which symbols refer to what version anyway. Showing the average MLS data might be sufficient (with error bars). Finally, it is not obvious why some heights (shown in Table 1) have 3 "selected points" and one has 6 points – some clarification would be useful.

Technical comments/corrections

Here are a number of suggested smaller changes:

Pg. 16164 (Abstract)

- L5: can safely delete "at three year interval", as years are given. . .
- L8: delete "globally" (not sure what this really means or adds)
- L16: you could delete the parentheses for "85 $\pm$ 35 pptv" (and similarly elsewhere in this manuscript) - but this is a detail.
- L19: change "and MLS-Aura" to "and the Aura MLS"

Pg. 16165

- L19: correct spelling for "Sounder"

- L27: change "generally" to "the generally"
- L28: change "perfect" to "very good", as nothing is really perfect, and there are uncertainties on the models and the data, so perfect would most likely be fortuitous agreement

Pg. 16166

- L7: change "in volume" to "by volume"
- L8: I suggest that you add "or a more direct determination of the abundance of VSLS in the upper troposphere and lower stratosphere" after "(WMO, 2007)".
- L16: add a comma after "stratosphere"
- L21: check that the value from Laube et al. is really 49$\pm$6 pptv at 15.2 km.

Pg. 16167

- L2: why not state "about 50 and 100 pptv" as there are error bars here also.
- L8: I suggest a new paragraph starting at "Our in situ. . . "
- L12: change "frame" to "framework"
- L14: "at three year intervals" is not needed (this is easy enough to count)
- L17: change "allowing for indicating" to "which indicate"
- L21/22: change "MLS Aura" to "the Aura MLS"

- Last sentence is poorly worded. Something like the following would be an improvement: " The result of such comparisons can lead to an assessment of the validity of previous estimates regarding a potential contribution of about 100 pptv from VSLS to total stratospheric chlorine."

Pg. 16168

- L9/10: change "An alternative... applying" to "An adopted alternative method consists of applying..."
- L20: vmr has already been defined in the Abstract (minor detail)
- L22: I suggest "total" uncertainty rather than "global" in general here, as "global" can often imply "relating to the globe", whereas your measurements are tropical only...
- L23: can safely delete "on the vmr".
- L27: change "line-width" to "linewidth".
- L29: change "parameters" to "parameter" [uncertainties]

Pg. 16171

- L24: delete "located"

Pg. 16172

- L13: change "from south-west and north-west" to "southwest and northwest"

- L19: change "by those from layers below..." to "by layers below..."
- L27: change "from west" to "west" and change "around" to "about".

Pg. 16173

- L1/2: I suggest a semi-colon for "altitudes; no downward..."

- L8: change "perfectly agree to" to "agree perfectly with" .

Pg. 16174

- L18: change "combined to" to "combined with".
- L20: add a comma after "contribution".
- L23/24: change "; as a result" to "; this can be explained as a result of the reaction... following photolysis and ..." [although one has no reference given here, and this is qualitative].
- L25: change "rising the" to "rising into the".

Pg. 16175

- L27/28:I suggest "Based on the study of Chen et al. (2005), for pressures larger than 50 hPa ..."

Pg. 16176

- L4: change "in these" to "at these".
- L6: change "atmosphere" to "atmospheric"

- L10: change “vmr” to “vmr values”.
- L11/12: change “over 2005 and 2008 years” to “for 2005 and 2008” and change “on average” to “, on average”.
- L13: can delete “of HCl”.
- L14/15: I suggest “with ACE-FTS measuring about 20% more than SPIRALE at 30 km”
- The reference to ATMOS is indeed too difficult to use as a comparison point, given the time difference, so I would simply delete this.
- L23: change “this chlorine content slow decay” to “a slow decay in chlorine content”.
- L27: change “in coincidence with” to “on”.

Pg. 16177

- L3/4: Change to “The spatial and temporal differences were smaller on 9 June 2008, as. . . ”
- L5: can delete “from its location”.
- L15: change “over six” to “out of six”.
- L27: delete “for comparison”.

Pg. 16178

- L1: I suggest “Despite the major differences between the remote and in situ methods and the geographical . . . in the tropical middle stratosphere.”
- L4: “for pressures larger than 46 hPa”
- L6: A comparison of MLS . . . ”
- L9: change “have yielded a good. . . ” to “yielded good. . . ”.
- L10/11: “where HCl abundances are larger, but again, poorer agreement. . . ”
- L22: Change “On the other side, a” to “A”.
- L28: Change “So, to summarize, ” to “In summary, . . . are larger. . . than SPIRALE and MLS values between. . . and than MLS values between. . . 48 and 55 km”. [Published satellite comparisons do not go as high as 60 km].

Pg. 16179

- L1/2: “Among these . . . measurements at. . . excellent agreement. . . ”
- L4: “The very good agreement. . . ”
- L6: you should credit other studies and assumptions here again, not just SPIRALE, for the 50 to 120 pptv estimate [if these are indeed the final numbers]. or simply say “in this analysis” instead of “from SPIRALE”.
- L12/13: I suggest “These measurements allowed for a study of the HCl content. . . , as well as an investigation of the total. . . ”
- L16: change “nor” to “or”.

- L17: might be better to write “source gas” rather than SG here [some people may only have time for or interest in the Conclusions. . .].
- L22: change “lead to” to “lead us to”.
- L24: I suggest “approximately correct” [unless you can really demonstrate that the 85 pptv number with error bar is the final word on this. . . and I suggest it probably will not be].
- L25: change “has sampled” to “sampled”.

Pg. 16180

- L1: spelling error / typo in “biennial”.
- L4: change “of MLS Aura” to “from Aura MLS, acquired. . . ” and better to start a new sentence “This consolidates the reliability. . . of tropical HCl in the altitude range” [can delete “amounts”].

Fig. 5: The white crosses are somewhat hard to see (a thicker/larger symbol would help); brighter labels would also help (and black background is less desirable than white, but other colors would also need to change, e.g. white symbols, in this case).

Fig. 6: Same as for Fig. 5; also, the black symbols for MLS measurement locations are not visible enough, so white might be useful for these symbols also (with a dot or other symbol rather than a cross).

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 16163, 2009.