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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.

Anonymous Referee #1

Received and published: 31 August 2009

General Comments

This paper attempts to answer an important question posed in the Scientific Assessment of Ozone Depletion 2006 (WMO, 2007), particularly the contribution to stratospheric chlorine from Very Short-Lived Substances (VSLS), using in situ observations from SPIRALE. The topic is clearly within the scope of ACP and the rarity of in situ observations of chlorine species in the TTL or above make the work novel, unique and relevant. The paper is generally well-written, well-structured and concise, although some specific disagreements that I have are stated in the "Specific Comments" and

"Technical Corrections" section.

While I do believe that the paper can be made suitable for full publication given some important changes, it remains difficult to draw firm conclusions on the contribution of VSLS to stratospheric chlorine from this work. The paper concludes that 85±35 pptv of stratospheric chlorine comes from VSLS, which is in agreement with the WMO 2007 Report's estimate of 50-100 pptv. Their value comes from 49±6 from VSLS source gases, a maximum of 45 pptv from phosgene and 25±25 pptv from SPIRALE measurements, giving a range of 50-120 pptv, also stated as 85±35 pptv by taking a simple average. However, HCl in the TTL was below the SPIRALE detection limit (20-30 pptv) and the authors are forced to work with and interpret this value as the only contribution to the 85±35 pptv of stratospheric chlorine coming directly from their observations.

A significant part of their argument that I did not find convincing is that the HCl in the TTL was entirely the result of the breakdown of VSLS, which was based on two 7-day back trajectories. Although I wholly agree that these back trajectories do not indicate any contribution from stratospheric air, I do not think that two 7-day back trajectories are sufficient for the assumption that the HCl in the TTL is entirely the result of the breakdown of VSLS. TTL transport is a complicated problem and there is much evidence that the tropical troposphere consists of isolated regions of convective ascent among weaker large-scale descent, which together give net ascent. I believe that some more work needs to be done to show that the measurements have not been exposed to any air carrying HCl from the stratosphere, because at present, I am not entirely convinced that this is the case. Longer back-trajectories would at least make a stronger case, unfortunately it is well-known that the accuracy of back-trajectories rapidly decreases with time, so this would not add that much to the discussion. Can the authors show some evidence that this is argument is sound for the bulk of the TTL? It should also be acknowledged that chemical transport modeling work indicates that essentially all meteorological fields (ECMWF included) have difficulty accurately representing stratosphere-troposphere exchange.

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COCl2 values in the WMO report come from the work of Toon et al. (2001). The original work must be cited to give credit appropriately. That being said, these values may be seriously outdated for an analysis of 2005 and 2008 observations. The recent work of Fu et al. (2007, Global phosgene observations from the Atmospheric Chemistry Experiment (ACE) mission, Geophys. Res. Lett., 34, L17815, doi:10.1029/2007GL029942) must also be cited and the more extensive measurements in that work must be considered for the interpretation of the COCl2 contribution.

Specific Comments

I would personally prefer expansion of the acronym VSLS in the title, but I think this is an editorial decision, not mine.

Figure 7 in Froidevaux et al. (2008) describes the difference between MLS HCl versions 1.5 and 2.2 in detail. Please explain why two different versions of MLS data were used in this work, instead of just the newer version 2.2. There are some significant differences between MLS HCl and ACE-FTS v2.2 HCl, although agreement with ACE-FTS v2.2 is better for MLS v2.2, especially in the upper stratosphere. Please also restate the MLS version numbers in section 3.3.

Could the values given in the second last paragraph of section 2.1 be shown as a table or figure for improved clarity?

It is not clear to me at all what the thickness of the lines in Figure 4 below 19 km represents. is this representing variability which is not shown for higher altitudes? Please explain this in the text and caption.

On page 16178 where MkIV, FIRS-2 and ALIAS-II are mentioned, a distinction must be made between remote and in situ measurements. The balloon-borne remote sensing observations are in many ways more like satellite observations than balloon-borne in situ measurements.

On page 16179 the discussion of the ACE and MLS comparison “ACE HCl values have

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been shown to be 2 to 10% larger than MLS ones from 50 to 10 hPa (∼21–31 km) and from 1 to 0.2 hPa (∼48–60 km); the agreement was better than 3% in the range 10–1 hPa" is a bit sloppy and possibly misleading. In the lower altitude range (21-31 km), the agreement is very good, but the values are low whereas in the higher altitude range (48-60 km), the two datasets actually diverge but since HCl VMR is much higher, as a percentage, the difference is coincidentally similar to the lower range.

The sentence "As a conclusion, the present paper answers the WMO (2007, p. 2.42) requirement for accurate HCl measurements in the TTL," in the final paragraph is too strong of a statement since HCl was actually below the detection limit, so accurate and quantitative measurements are still elusive.

Technical Comments

page 16165, line 1: "Chlorine active" to "Active Chlorine"

page 16165, line 8: "essentially methane" to "especially methane"

page 16166, lines 25-30: COCl2 work of Fu et al. (2007, GRL)

page 16167, line 13: "Envisat satellite" to "ENVISAT"

page 16167, line 21: check grammar

page 16168, line 2: Omission of the expanded SPIRALE acronym seems common for SPIRALE papers. I would like to see it expanded even though it is French. Surprisingly, this was done for another French acronym MIMOSA, which is much less central to the work.

page 16168, line 19: "with a high frequency" to "with high frequency"

page 16168, line 26: "trace gases vmr" to "trace gas vmr"

page 16168, line 27: "meters" (US spelling) to "metres" (International)

page 16174, line 18: "combined to" to "combined with"

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page 16175, line 26: “on 2005” to “in 2005”

page 16176, line 15: “of ACE” to “from ACE”

page 16178, line 15-16: HALOE and ACE-FTS are instruments on satellites, but are not satellites

page 16179, line 15: “lead to conclude” to “lead to the conclusion”

page 16180, line 4: MLS-Aura is an instrument, Aura is the satellite

Figure 1: Why are the zeros on the left and right axes not aligned. What is the significance (if any) of this offset? Also, the fonts for the exponents are too small and difficult to read.

Figure 2: Could the LZRH also be shown?

Figure 3: It would be helpful to label the location of Teresina.

Figure 4: Explain the line thickness below 19 km.

Figures 5&6: Black background is not standard for journal figures and makes for poor quality printing.

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

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