Review of “In situ observations of CH2Cl2 and CHCl3 show efficient transport pathways for very short-lived species into the lower stratosphere via the Asian and North American summer monsoons”

This study presents observations of very short-lived substances (VSLS), CH_2Cl_2 and $CHCl_3$, collected during the WISE aircraft campaign over the Northern Hemisphere Atlantic Ocean and Western Europe during fall 2017. These observations are analyzed using CLaMS simulations to identify source regions and regions with rapid convection that transport the VSLS to the extratropical lower most stratosphere (LMS) and the upper troposphere. The Asian summer monsoon anticyclone is identified as a significant pathway for transporting chlorinated VSLS into the LMS, including samples with elevated anthropogenic sources of CH_2Cl_2. The role of the North American monsoon in the rapid transport of VSLS to the extratropical upper troposphere/lower stratosphere is explored with hurricane Maria as a case study. Further refinement of the text is needed to clearly communicate the importance of this pathway to stratospheric composition.

Overall, the paper is well-written and thoroughly researched. However, at times the language is imprecise, specifically with regards to which observations are representative of stratospheric air and which transport time is being discussed.

Specific comments:

1. This distinction between tropospheric and stratospheric air is clear in the figures but does get lost in some of the text. For stratospherically relevant numbers reported, these should be calculated with only observations collected above the thermal tropopause. If that is already being done, it is not clearly communicated in the main text.

   For example, on line 19 in the abstract and in the main text it is not clear if the reported “1-5 weeks into the Ex-LS” was calculated only with observations collected above the thermal tropopause.

2. It is misleading to place N2O and month on the same axis in Figure 4. Measurements of CH2Cl2 at N2O mixing ratios of 325 ppb should not necessarily be compared to surface observations in June; some of the low mixing ratios of CH2Cl2 shown here are likely due to photochemical processing in the stratosphere, not seasonal surface trends.

   It should also be noted that biases between NOAA and Advanced Global Atmospheric Gas Experiment (AGAGE) records of CH2Cl2, particularly in the northern tropic station shown in Figure 4, have been identified, suggesting either calibration errors or longitudinal gradients in mixing ratios of CH2Cl2 (Engel et al., 2018).

   Additionally, the CH2Cl2-poor air is connected to the OH-driven seasonality based on ground-based observations at one AGAGE station without discussing other explanations. However, since CH2Cl2 is not well mixed in the troposphere, a contributing factor could be the uplift of air with less anthropogenic influence than measured at the ground-based station. This option is alluded to on line 402, but it is not clearly introduced as a separate mechanism for low CH2Cl2 mixing ratios.

3. Variations of "first study" statements appear throughout the paper. I would caution against that phrasing since it seems unnecessary for the importance of the paper and can be untrue if one of the qualifications in these statements are removed. From a literature search, at least Adcock et al. (2021) provides back-trajectory analysis and identifies source regions of measurements of CH2Cl2 in the UTLS. Additionally, Rotermund et al. (2021), which is properly referenced in the discussion section, should also be acknowledged in the introduction as a study that employed similar methods to investigate source regions of brominated VSLS during the WISE campaign.

4. It would be helpful to either place the results of the present study in the context of similar studies conducted with observations of other VSLS (e.g., Aschmann et al., 2009; Ashfold et al., 2012; Levine et al., 2007; Liang et al., 2014), or explain in the introduction why the results of those studies have limited applications for studying the transport of Cl-VSLS.

5. "…two most efficient and fast transport pathways from (sub-)tropical source regions into the extratropical lower stratosphere (Ex-LS)…" (Line 7)

   This study has identified the ASMA as an efficient pathway into the extratropical stratosphere and the North American monsoon as a fast transport pathway into the Northern Hemisphere upper troposphere/lower stratosphere. However, from the observations presented in this paper, it is not clear how efficiently Cl-VSLS lofted into the upper troposphere by hurricanes are mixed into the stratosphere. This sentence could be simplified to: "…two transport pathways from (sub-)tropical source regions into the extratropical lower stratosphere and upper troposphere…". Further detail of the difference between the two pathways is provided later in the abstract.

6. Line 132: Please state why earlier flights are not included in this study. Also, altitude and pressure ranges are not given for the measurements. Were any filtering criteria used to limit observations to the UTLS?

7. Section 3.1.3 would read more clearly if the "Analysis of transport pathways" is discussed before "Case study: convective uplift by hurricane Maria". As written, what is meant by fast transport time

8. Line 370: Please explain why the comparison to August and September ground-based observations is justified based on the transport time since maximum $\Delta\Theta_{18h}$ and not transport time since the boundary layer?

   Similarly, is the uplift described on line 373 recent or fast uplift of CH2Cl2-poor air (since max $\Delta\Theta_{18h}$ or since the boundary layer, respectively)?

9. In the discussion section, line 504, the authors state "…clearly benefits the use of CH2Cl2 observations to derive details about the different transport mechanisms and pathways…". It is an interesting and novel way to frame the paper, and if the authors would like to highlight that aspect, it should be introduced at the end of Section 1.

However, in the discussion please note that CH_2Cl_2 and CH_2Br_2 have similar atmospheric lifetimes, and regionally varying surface mixing ratios, not just the OH seasonal cycle, can influence the low CH_2Cl_2 observed in the UTLS. The use of the $CHCl_3$:CH_2Cl_2 ratio as a diagnostic tracer could also be better highlighted.

10. The discussion of the flight on 1 October (lines 542 – 556) could be moved to the hurricane case study section. Having this text at the end of Section 4 somewhat distracts from the big picture highlights given. Please note on line 553 and in similar text that while NAMA was observed to be a fast transport pathway to the UTLS, a low fraction of stratospheric air originates from central and western ITCZ (Figure 6).

Technical comments:

- "Regionally differing … $CHCl_3$ sources" (Lines 23 – 26): As written this sentence is confusing. Please clarify that a larger fraction of $CHCl_3$ than CH_2Cl_2 is emitted by natural sources, and, consequently, a lower $CHCl_3$:Cl_2Cl_2 is found in air parcels that originate from regions with significant anthropogenic influences than regions with weaker anthropogenic emissions.
- "Due to the sparseness of Cl-VSLS measurements in the stratosphere" (Line 36): This statement is true, but please reference some past studies that measured Cl-VSLS in the stratosphere, such as (Laube et al., 2008; Schauffler et al., 1993, 2003) in addition to some of the references already present in the paper.
- Paragraph beginning on Line 54: It's important for your analysis of the $CHCl_3$:CH_2Cl_2 ratio that the reader understands CH_2Cl_2 is largely emitted by anthropogenic sources, while a larger fraction of $CHCl_3$ is from natural sources. This should be directly stated for the reader here. Overall, the detail given for the different emissions sources for the two VSLS could be more concise and focused so that this point does not get missed.
- Line 77: It may be helpful to state the atmospheric lifetimes of CH_2Cl_2 and $CHCl_3$ at the beginning of this paragraph so that they are not lost in the discussion.
- Line 104: If there are modeling studies that explored source regions of Cl-VSLS in addition to Claxton et al. (2019), they should be referenced here.
- Lines 255 - 260: Some of the wording in this paragraph is hard to follow. A useful point that is missing here is N_2O, due to its long tropospheric lifetime is well mixed in the troposphere, while CH_2Cl_2 is not.
- "measurements of AGAGE" (Line 262): Here and elsewhere in the paper, please correct to read "ground-based measurements of CH_2Cl_2 from the AGAGE network"
- Line 299 – 304: The description of the different criteria is hard to follow. Please carefully reword. Also, readers may have to refer back to criteria (1) and (2), it could be helpful to format them as separate bullets.
- Figure 7: Could the transport time to the boundary layer be shown in a similar figure? The lines given in Figure 10 are hard to distinguish.
- Line 354: Locations of and transport times since maximum diabatic ascent rate

- Line 389: Please state how many of these air masses were collected above the thermal tropopause.
- "positively correlate with transport time" (Line 391): What is shown in Figure 8 is the time since max convection, not the overall transport time. Please clarify the discussion in this paragraph.
- Lines 411 and 434: Please clearly state that you are only performing back trajectories for observations collected above the thermal tropopause, otherwise here and elsewhere should read "into the UTLS"
- Line 447: State for the readers that a broader range of surface mixing ratios of CH2Cl2 (~10 – 70 ppt) are reported from surface stations than for CHCl3 (~5 – 15 ppt) (https://agage.mit.edu/data/agage-data).
- Line 476: Remind the reader here or earlier in the section that a larger fraction of CHCl3 emissions than CH2Cl2 are from natural sources and the two compounds have similar atmospheric lifetimes.
- Line 408: The ASMA pathway was also identified by Adcock et al. (2021) for CH2Cl2.

References

Adcock, K. E., Fraser, P. J., Hall, B. D., Langenfelds, R. L., Lee, G., Montzka, S. A., et al. (2021). Aircraft-Based Observations of Ozone-Depleting Substances in the Upper Troposphere and Lower Stratosphere in and Above the Asian Summer Monsoon. *Journal of Geophysical Research: Atmospheres*, *126*(1). https://doi.org/10.1029/2020JD033137

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