Reply to the comments of Referee #1 on the paper

In situ observations of CH_2Cl_2 and $CHCl_3$ show efficient transport pathways for very short-lived species into the lower stratosphere via the Asian and North American summer monsoons

by V. Lauther et al.

We thank Referee #1 for the thorough reading of and the very helpful remarks on our manuscript. The comments have been considered carefully while revising the draft and resulting modifications are addressed point-py-point in the following (Referee's comments are cited in bold face):

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.

This is a very good remark! We agree and have now specified the given transport times at the respective locations in the paper. In addition, we have added an overview table with median transport times derived from back-trajectories of different lengths (from the model boundary layer and from the location of maximum convection) for all observations in the UTLS and either only for those in the UT or only for those in the LS. The used data set in the new table (Table 2) is also filtered to include only measurements of CH_2Cl_2-poor air linked to the NAM as well as measurements of CH_2Cl_2-rich air linked to the ASM. We consistently use median transport times now and provide ranges of transport times derived from the 25. and 75. percentile of the respective data set. For details, we refer to the track change version of the manuscript.

In addition, we noticed that the separation between measurements in the UT and the LS shown in the figures was based on ECMWF's ERA5 reanalysis data (Hersbach et al., 2020). There are but a few differences regarding the discrimination between UT and LS of individual data points when using ERA5 instead of ERA-Interim data. However, to be consistent, we changed the displayed separation between UT and LS in all relevant figures (3, 12 (now 13), 13 (now 14), B1 (now C1); no changes were necessary in Fig. 9 (now 10)) to the calculations based on ERA-Interim.

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.

This is true and we thought about that while carefully choosing the wording of this section (i.e. "Although this simplified view ignores the impact of mixing processes and chemical reduction of CH_2Cl_2 it qualitatively explains the lower branch of the correlation curve for air parcels younger than a few months."). However, we think Figure 4 is an interesting qualitative comparison between two different data sets providing the reader a good first impression of a plausible explanation for one part of the rather complex CH_2Cl_2-N_2O relationship. Nevertheless, we agree that Figure 4 should be interpreted with care and not be used out of context. In the caption of Figure 4 we thus refer to the similarities between the CH_2Cl_2-N_2O relationship and the ground-based CH_2Cl_2 observations from the AGAGE network focusing only on the lower branch of the correlation — implying a comparison of the correlation with AGAGE data from about July to September. This range for a plausible comparison is also stated in the main text ("The observed decrease of low CH_2Cl_2 mixing ratios for increasing N_2O mixing ratios (from older to younger air) agrees well with the decreasing tropical monthly averaged CH_2Cl_2 mixing ratios from about July to September

2017, as observed by AGAGE."). However, we agree that the figure might be misleading to some readers and therefore we have now included vertical lines highlighting the comparison between July and September to clarify the plausible range of the comparison. In addition, we have specified the respective text in the Figure 4 caption as follows:

"The gradient of the CH_2Cl_2-N_2O relationship's lower branch (low CH_2Cl_2 mixing ratios between vertical lines) qualitatively fits the temporal variation of the ground-based CH_2Cl_2 measurements."

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

These are good points and we have included them in the text of Section 3.1 as follows:

"It is thus very likely that the lower branch of the CH_2Cl_2-N_2O ~~correlation~~relationship is caused by the tropical Atlantic CH_2Cl_2 surface seasonality. However, the low CH_2Cl_2 mixing ratios observed during WISE could also be impacted by air uplifted from regions less influenced by CH_2Cl_2 sources than the observations from the AGAGE network at Barbados. Ground-based observations of CH_2Cl_2 surface mixing ratios from the AGAGE and the NOAA network show strong regional differences particularly in the NH tropics. However, it is unclear if these differences are caused by calibration biases or are of a natural origin (Engel et al., 2018)."

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.

We agree and have removed the respective statements of "first study" from the paper. Also Adcock et al. (2021) is discussed in the paper. For details, we refer to the track change version of the manuscript.

Rotermund et al. (2021) is now acknowledged in the introduction of the paper in the following way:

A study by Rotermund et al. (2021) employed similar methods to identify source regions and the impact on the Ex-LS of Br-VSLS using measurements from the same aircraft campaign as the measurements used in the present paper and is compared to our results in Section 4.

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.

The results of similar transport studies focused on other VSLS (usually Br-VSLS with mainly natural, oceanic sources) cannot necessarily be used to describe the transport of Cl-VSLS (mainly anthropogenic, land-based sources) because of their different

source distribution in the troposphere. Therefore it is necessary to describe transport pathways into the stratosphere for VSLS (particularly for those with strongly varying mixing ratios in the troposphere) specifically for the respective VSLS or group of VSLS with similar characteristics (e.g., photochemical lifetime and source distribution). To point this out, we have now added the following explanations to the introduction of the paper:

"Observational evidence for Cl-VSLS being transported into the stratosphere is extremely rare ~~and their main transport pathways into the stratosphere have not been described on an observational basis~~ (e.g., Schauffler et al., 1993; Woodbridge et al., 1995; Schauffler et al., 2003; Laube et al., 2008; Park et al., 2010; Adcock et al., 2021). Transport pathways into the stratosphere for VSLS have been derived from observations of bominated VSLS (Br-VSLS; e.g., Sturges et al., 2000; Ashfold et al., 2012; Wales et al., 2018; Filus et al., 2020; Keber et al., 2020; Rotermund et al., 2021) or modeled specifically for Br-VSLS (e.g., Levine et al., 2007; Aschmann et al., 2009; Ashfold et al., 2012; Liang et al., 2014) which have mainly natural emission sources (Engel et al., 2018). However, the only Br-VSLS with a photochemical lifetime comparable to those of CH_2Cl_2 and $CHCl_3$ is CH_2Br_2 (150 days; WMO, 2018) which is mostly emitted by the oceans and, consequently, is differently distributed in the troposphere than the mainly anthropogenically (land-based) emitted CH_2Cl_2 and most Cl-VSLS (e.g., Engel et al., 2018). Thus, transport studies of Br-VSLS focus on transport into the stratosphere from likely different source regions than those of Cl-VSLS and their results might not necessarily be directly applicable to the transport into the stratosphere of CH_2Cl_2 and $CHCl_3$. In addition, in order to specifically study transport into the stratosphere via the ASM it is beneficial to observe VSLS with their strongest sources being located in the core region of the ASM. This is the case for CH_2Cl_2 while most Asian Br-VSLS sources are located only in adjacent regions of the ASM."

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.

This is true. We changed the sentence as suggested.

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?

We have reconsidered the wording of Line 132 and added the full ranges of the used data set regarding potential temperature, altitude, and pressure. No explicit filtering of the data set was used to confine the presented observations to the UTLS region. However, due to a malfunctioning water trap of the HAGAR-V instrument, we could perform CH_2Cl_2 and $CHCl_3$ measurements only at ambient water vapor levels below about 100 ppm (measuring mainly at around 6 ppm), thus "naturally" confining the observations to the upper part of the troposphere and above. This is briefly explained in Section 2.2.1 to which we now refer in the introduction to the used data set (Sect. 2.1). We also added to Section 2.2.1 why we were able to measure only during the last ten flights of the WISE campaign. The respective parts of the manuscript are modified as follows (sections 2.1 and 2.2.1, respectively):

"In this study we present UTLS measurements ~~up to~~between a potential temperature of 315 K and 404 K (i.e., 7.4 – 14.5 km altitude; 388 – 130 hPa pressure) of the last ten WISE flights, i.e., from 28 September to 21 October 2017 (Figure 1). Due to technical issues of the instrument, CH_2Cl_2 and $CHCl_3$ measurements below the given range and during earlier flights of the WISE campaign were not performed (cf. Section 2.2.1)."

"However, during WISE the dehydration system of HAGAR-V was malfunctioning. ~~could not be used. Consequently,~~ For the last 10 WISE flights that system was bypassed and the MS module measured only at low ambient water vapor levels (mainly

at $H_2O < 100$ ppm; median: 5.6 ppm), i.e., in the UTLS region. ~~With this solution the MS module measured~~ thus yielding measurements during about 90% of a typical ~~WISE~~ flight's duration (i.e., about 7.6 h per flight). MS measurements of WISE flights before 28 September could not be used for analysis due to the malfunctioning sample dehydration unit."

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

We agree and have rearranged the respective paragraphs and figures. Note, that this is not directly marked in the track changes version of the manuscript to keep changes within the text highlighted.

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)?

This is a good point and by providing the different calculated transport times (Table 2; cf. response to specific comment "1.") this should be clear now. We additionally included the information about the transport time since the boundary layer to the sentence (see quote below). For CH_2Cl_2-poor air parcels with maximum convection above Central America the difference between median transport times from the location of maximum convection and from the model boundary layer is only about one week. Thus 'recent transport' can also be considered 'fast transport'. We agree that this was not clear before we added Table 2 to the paper. In regard to specific comment "8.", the text of Sect. 3.1.3 - "Locations of maximum diabatic ascent rate and transport times" was modified as follows:

"The transport times to the UTLS since the ascent above Central America mainly range between ~~1~~2 – 5 weeks ~~and are much shorter than for those air parcels lifted up above Asia~~ (cf. Table 2). The main uplift of CH_2Cl_2-poor air above Central America thus falls in the time period of late August and throughout the entire September. ~~T~~With transport times from the BL being only about one week longer (Table 2), this result supports the comparison of CH_2Cl_2-poor air with the seasonal minimum CH_2Cl_2 mixing ratios observed by AGAGE at Barbados"

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 CH2Cl2 and CH2Br2 have similar atmospheric lifetimes, and regionally varying surface mixing ratios, not just the OH seasonal cycle, can influence the low CH2Cl2 observed in the UTLS. The use of the CHCl3:CH2Cl2 ratio as a diagnostic tracer could also be better highlighted.

These are good points. With our revision of the introduction regarding the specific comment number "4." we also highlighted the beneficial use of a VSL tracer like CH_2Cl_2 with its main sources located in the core region of the ASM to analyze transport into the stratosphere via the ASM.

We also changed the wording regarding the advantage of CH_2Cl_2 over the Br-VSLS analyzed by Rotermund et al. (2021) due to its very strong Asian emission sources and very low mixing ratios in other regions of strong convection. Thereby we highlight the more general fact that we could derive the two transport pathways so clearly due to the strong contrast of tropospheric CH_2Cl_2 mixing ratios in the regions of significant convection. The possible reasons for the low CH_2Cl_2 mixing ratios are already discussed in Section 3.1 (see our response to the specific comment nr. "2."). However, we wanted to keep the phrasing "[...] a longer lifetime [...]" when comparing CH_2Cl_2 to the Br-VSLS analyzed by Rotermund et al. (2021) without specifically commenting on the comparable atmospheric lifetime of CH_2Br_2 (this is already mentioned in the introduction, cf. answer to specific comment nr. "4.") because of two reasons: (1) our statement is correct (CH_2Cl_2 and CH_2Br_2 have similar

atmospheric lifetimes, but on average that of CH_2Cl_2 is up to one month longer; WMO, 2018); (2) we write that not a single but the combination of attributes (lifetime and source distribution) is the reason why CH_2Cl_2 is the preferred tracer for the analysis of details on the transport pathways into the Ex-UTLS in NH late summer. The respective part of the discussion is now modified as follows:

"Nevertheless, compared to the very short-lived bromine species analyzed by Rotermund et al. (2021), the combination of a longer lifetime, highly significant Asian emission sources~~, and a strong seasonal cycle~~ and very low mixing ratios in other regions of strong convection clearly benefits the use of CH_2Cl_2 observations to derive details about the different transport mechanisms and pathways from the source region into the NH summertime UTLS. In addition, using the $CHCl_3$:CH_2Cl_2 ratio to support the analysis of air mass origin is a unique and helpful tool in the analysis of transport pathways."

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).

We agree and moved this paragraph and Figure 14 (now Fig. 11) to the hurricane case study section which itself has also been moved further below in the text (cf. specific comment number "7."). Note, that the moving of figures and paragraphs is not directly marked in the track changes version of the manuscript to keep changes within the text highlighted.

When discussing the transport pathway into the Ex-LS via the NAMA we now added the information about the relatively low fractions of central and western ITCZ air mass origin in stratospheric air as well as the note that we actually observed only 25 % of the air parcels transported by the NAMA within the Ex-LS. We modified the text at three different locations (Section 3.1.3 - Analysis of transport pathways, Section 4 - Discussion (now Section 3.1.3 - Case study: convective uplift by hurricane Maria), and Section 5 - Conclusion) as follows:

"Another aspect adding to the different transport times is the longer transport pathway from Asia because simulations indicated air masses ~~were always observed to reach~~ reached the location of measurement always from the west. Nevertheless, air parcels observed in the Ex-LS are impacted more strongly by air masses transported via the ASMA than via the NAMA (cf. figures 6 and 14)."

"We have thereby shown that tropical surface mixing ratios of VSLS from the region of Central America and the Atlantic Ocean can be efficiently transported into the Ex-LS during the late North American monsoon season. For instance, this is of particular importance for brominated short-lived substances (e.g., CH_2Br_2 and $CHBr_3$) that have a high ODP and some of their largest emission sources located in tropical oceans (e.g., Hepach et al., 2015; Rotermund et al., 2021). However, it has to be noted that only 25 % of air parcels transported by this pathway were observed in the Ex-LS and stratospheric air masses showed relatively low fractions of air originating in the region of central and western ITCZ compared to those originating in southern and eastern Asia (cf. Figure 14)."

Eventually, these air masses mixed into the LS by Rossby wave breaking and influenced the chemical composition of the NH Ex-LS $10 - 20$ K below the air masses dominated by transport via the ASMA. However, only 25 % of air parcels transported via this pathway were observed above the thermal TP with transport times from the BL to the location of measurement ranging from 5 to 9 weeks.

Technical comments:

- "Regionally differing … CHCl3 sources" (Lines 23 – 26): As written this sentence is confusing. Please clarify that a larger fraction of CHCl3 than CH2Cl2 is emitted by natural sources, and, consequently, a lower CHCl3:Cl2Cl2 is found in air parcels that originate from regions with significant anthropogenic influences than regions with weaker

anthropogenic emissions.

We changed the wording of the mentioned sentence in the abstract as follows:

"~~Regionally differing $CHCl_3$:CH_2Cl_2 emission ratios derived from our UTLS measurements suggest a clear similarity between $CHCl_3$ and CH_2Cl_2 when emitted by anthropogenic sources and differences between the two species mainly caused by additional, likely biogenic, $CHCl_3$ sources.~~ CH_2Cl_2 and $CHCl_3$ have similar atmospheric sinks and lifetimes but the fraction of biogenic emissions is clearly higher for $CHCl_3$ than for the mainly anthropogenically emitted CH_2Cl_2, consequently lower $CHCl_3$:CH_2Cl_2 ratios are expected in air parcels showing a higher impact of anthropogenic emissions. The observed $CHCl_3$:CH_2Cl_2 ratio suggests clearly stronger anthropogenic emissions in the region of southern and eastern Asia compared to those in the region of Central America and the tropical Atlantic."

- "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.

References to other studies presenting Cl-VSLS measurements in the stratosphere are now given: "~~Due~~Owing to the sparseness of Cl-VSLS measurements in the stratosphere (e.g., Schauffler et al., 1993, 2003; Laube et al., 2008; Park et al., 2010; Adcock et al., 2021 the impact of changes in Cl-VSLS surface emissions on their distribution in the stratosphere has yet to be fully characterized on an observational basis."

- Paragraph beginning on Line 54: It's important for your analysis of the CHCl3:CH2Cl2 ratio that the reader understands CH2Cl2 is largely emitted by anthropogenic sources, while a larger fraction of CHCl3 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.

We agree and the phrase " Nevertheless, on a global scale $CHCl_3$ has a significant fraction of biogenic emission sources in contrast to CH_2Cl_2 which is almost exclusively emitted by anthropogenic sources." was added to the mentioned paragraph to highlight this difference between the two analyzed species.

In addition and in agreement with a comment of Referee #2 we shortened the information given in the introduction to focus more on things directly relevant to the paper.

- Line 77: It may be helpful to state the atmospheric lifetimes of CH2Cl2 and CHCl3 at the beginning of this paragraph so that they are not lost in the discussion.

We agree and the mentioned paragraph is now rearranged accordingly:

"For CH_2Cl_2 Hossaini et al. (2019) suggest an average tropospheric lifetime of 168 days (about 6 months) and a stratospheric lifetime of $1-2$ years (outside the poles) was estimated by Hossaini et al. (2017). The main atmospheric sink of both CH_2Cl_2 and $CHCl_3$ is the reaction with hydroxyl radicals (OH) in the troposphere. Both species have similar reaction rates with OH implying similar photochemical lifetimes for both Cl-VSLS (Hsu and DeMore, 1994). Time series of background mixing ratios of both species are anticorrelated to the seasonal cycle of OH (Cox et al., 2003). In the NH seasonal anthropogenic use of products releasing $CHCl_3$ to the atmosphere (e.g., landfill and chlorination of water) have been observed to have a small local impact on the seasonality of $CHCl_3$ (Gentner et al., 2010). In addition, the global distribution of OH shows significant regional differences (Spivakovsky et al., 2000; Hanisco et al., 2001; Lelieveld et al., 2016). Therefore, also the photochemical lifetimes of CH_2Cl_2 and $CHCl_3$ are regionally different. ~~For CH_2Cl_2 Hossaini et al. (2019) suggest an average tropospheric lifetime of 168 days (about 6 months) and a stratospheric lifetime of $1-2$ years (outside the poles) was estimated by Hossaini et al. (2017). Both species have similar reaction rates with OH implying similar photochemical lifetimes for both~~

~~Cl-VSLS (Hsu and DeMore, 1994).~~"

- 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.

This paragraph highlights the regional dependency of the Cl-VSLS's ODP which is, to the best of our knowledge, presented only by Claxton et al. (2019).

- Lines 255 - 260: Some of the wording in this paragraph is hard to follow. A useful point that is missing here is N2O, due to its long tropospheric lifetime is well mixed in the troposphere, while CH2Cl2 is not.

We considered the comment and added the proposed additional information to the text as follows:

"With a photochemical lifetime of 123 years (Ko et al., 2013) N_2O is well mixed in the troposphere and has a much longer lifetime than CH_2Cl_2 which exhibits strongly varying mixing ratios throughout the boundary layer (e.g., Simmonds et al., 2006)."

- "measurements of AGAGE" (Line 262): Here and elsewhere in the paper, please correct to read "ground-based measurements of CH2Cl2 from the AGAGE network"

The text is now changed to the proposed phrasing at all convenient places in the manuscript. For details, we refer to the track change version of the manuscript.

- 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.

We agree and reconsidered the wording and structure of the paragraph modifying it as follows:

"Further, to work out differences of air mass origin between CH_2Cl_2-rich and -poor air, ~~for each surface origin tracer the median fraction~~ the median fraction of a surface origin tracer in CH_2Cl_2-rich air parcels is compared to that in CH_2Cl_2-poor air parcels. To combine regions of air mass origin with a particularly high relative impact on either CH_2Cl_2-rich or -poor air, the ratio of these median surface origin tracer fractions in CH_2Cl_2-rich and -poor air is analyzed. Surface origin tracers with particularly high relative median fractions in either CH_2Cl_2-rich or -poor air are combined following these two criteria:

(1) considered are only surface origin tracers with median fractions $\geq 1\,\%$ in CH_2Cl_2-rich or -poor air parcels, and

(2) the ~~median~~ ratio of a median surface origin tracer fraction (CH_2Cl_2-rich/CH_2Cl_2-poor air respectively CH_2Cl_2-poor/CH_2Cl_2-rich air) must be > 1.8.

- 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.

Yes, we added Figure B1 to the appendix and refer to it in Section 3.1.3 as follows:

" The locations of trajectory end points at the model boundary layer color coded with transport time are given in the appendix (Figure B1)."

- Line 354: Locations of and transport times since maximum diabatic ascent rate

We have now included also transport times from the boundary layer (Table 2; see reply to specific comment number "1.") and will thus keep the title of the subsection as it is.

- Line 389: Please state how many of these air masses were collected above the thermal tropopause.

It were 5 out of 27. The sentence was modified as follows:

"This analysis directly links 27 WISE measurements (five were observed above the thermal TP) to the convection of hurricane Maria with transport times since convection ranging between one week and one month."

- "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.

We focus on the transport since the main convection by hurricane Maria because this gives a better impression on the time scale for air parcels lofted by a major hurricane to reach the Ex-UTLS and we analyze impacts on these air parcels induced in the UTLS. However, all trajectories linked to air parcels lifted up by hurricane Maria reach the model boundary layer in September and most do so above the Atlantic Ocean, i.e., at the time of seasonal surface minimum CH_2Cl_2 mixing ratios and in a region of weak CH_2Cl_2 emission sources. We added the respective information to the text and included a note to the relationship of CH_2Cl_2 and transport time since the boundary layer as follows:

"Interestingly, CH_2Cl_2 mixing ratios of measurements linked to hurricane Maria positively correlate with transport time since maximum convection ($R_{Pearson} = 0.85$; Figure 10, top right. Note, these CH_2Cl_2 mixing ratios also correlate with transport time since the model BL but with a lower $R_{Pearson} = 0.64$ and transport times between 9 and 48 days. However, here we focus on the transport since convection by hurricane Maria to derive impacts on the air parcels induced by processes in the UTLS region). Those air samples related to short transport times contain the lowest CH_2Cl_2 mixing ratios at $N_2O > 325$ ppb measured during WISE (Figure 10, top left). According to the back-trajectories, most of the air parcels lifted up by hurricane Maria left the model boundary layer above the tropical Atlantic in September where CH_2Cl_2 sources are small (Figure 10, bottom). In addition, in that region the seasonal minimum of CH_2Cl_2 mixing ratios is in September (cf. Section 3.1)."

- 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"

We analyze the trajectories for observations in the UTLS region and corrected the wording accordingly.

- 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).

We have considered this statement carefully and concluded that it is not necessarily helpful to the readers. The much clearer split of the CH_2Cl_2-N_2O relationship than that of the $CHCl_3$-N_2O relationship is likely not based on the different global absolute surface mixing ratio ranges of the two Cl-VSLS. For air parcels measured during WISE we identified significant regions of air mass origin only around the summertime ITCZ (cf. Figure 2). The surface mixing ratio ranges within these regions between July and September could provide a supporting argument to the discussion. However, using the monthly averaged surface mixing ratios ignores the daily variations at the given location. Including the standard deviation or any other measure of the mean surface mixing ratio scatter range within the relevant regions would actually provide a useful argument for why we can clearly resolve the two branches in the CH_2Cl_2-N_2O relationship but not so clearly in the $CHCl_3$-N_2O relationship ($CHCl_3$ has stronger relative variations of mixing ratios within the source regions than CH_2Cl_2). However, the sparseness of ground-based Cl-VSLS observations in these regions prevents us from using such an argument. In any case, we think it is not helpful if we provide the global range of monthly averaged mixing ratios of the two Cl-VSLS here.

- 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.

This is a good suggestion and it was included in the text of Section 3.2 as follows:

"The $CHCl_3$-CH_2Cl_2 correlation slope thus flattens with increasing entry of air masses originating from southern and eastern Asia. ~~T~~ Knowing that both species have similar sinks and photochemical lifetimes but $CHCl_3$ has a larger fraction of emissions from biogenic sources than CH_2Cl_2, this suggests larger $CHCl_3$:CH_2Cl_2 emission ratios in the region of the central and western ITCZ region (with presumably mostly biogenic sources) than in southern and eastern Asia (where anthropogenic sources likely dominate)."

- Line 408: The ASMA pathway was also identified by Adcock et al. (2021) for CH2Cl2.

We partly agree on this point. In this paragraph we highlight the transport pathway from Asia via the ASM including eastward outflow of the ASMA into the Ex-LS derived from observations above western Europe/Atlantic. Adcock et al. (2021) present a transport pathway into the LS based on measurements within the ASMA (StratoClim 2017) and observations of westward ASMA outflow (without back-trajectory analysis) above eastern Europe and the Mediterranean (StratoClim 2016). We do not argue the point that the observations by Adcock et al. (2021) are an important contribution to observational studies regarding Cl-VSLS transport into the LS by the ASMA (and we refer to the paper elsewhere in our manuscript) but quoting their paper at line 508 would be inaccurate to the content of this paragraph.

References

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