Response to the comments of Referee #2 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 #2 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):

General comments:

Introduction p2-3: To me there seemed to be a lot of detail here about the overall significance of the CL-VSLS, and about sources. This is important, of course, but it does not all seem to be essential to this paper. The key points, for this paper, are the general geographical characterisation for the source regions of the different species being considered. The reader would be able to focus more effectively on the major points of this paper if some of the detail was removed.

We agree. A similar remark was made by Reviewer #1 and we reduced the information given in the introduction to focus on the most essential details about Cl-VSLS. For details, we refer to the track change version of the manuscript.

p10-11: I think that it is a bit confusing to refer to the 'lower branch of the correlation' for the CH2Cl2-N2O relationship. It looks as though there are large number of parcels for which the N2O values are around 330ppb but the CH2Cl2 values are distributed across the range 30-45ppt. I think that in some ways it weakens your case if you call something a correlation when by conventional measures the correlation is rather weak. I am not disputing the fact that there are two 'families' in the plot at high N2O values – but can you find a more neutral term to describe them?

We agree that our use of the term "correlation" is rather based on lab jargon than on mathematics. We now use the general and more precise wording of a "CH_2Cl_2-N_2O relationship" instead of "CH_2Cl_2-N_2O correlation" if "correlation" is not specifically meant. We thus refer now to the "lower branch of the CH_2Cl_2-N_2O relationship" to describe the values of particularly low CH_2Cl_2 mixing ratios at high N_2O mixing ratios (similar for the upper branch). These modifications are made throughout the manuscript and for details we refer to the track change version of it.

p12: A similar comment applies to this approach of fitting a 'mean correlation curve' and using that as a basis for separating the upper and lower branches. The quadratic fit might be intended to seem quantitative – but there is really no reason to believe that the extrapolated values have any concrete relevance to the concentrations of CH2Cl2 measured in high N2O concentration parcels – the split between the two categories is essentially being made on the basis of the appearance of the plot. It seems very reasonable to make the split – but if the authors (or the readers) felt that it was more justifiable on the basis of a quadratic fit and an extrapolation then I would say that they were confused.

It is true, the split between the two categories of data was made on the basis of the appearance of the CH_2Cl_2-N_2O relationship and we do not claim otherwise. In order to decide on the "geometry" of the split we used the 'mean correlation curve' as a best-guess proxy of the annual mean CH_2Cl_2-N_2O relationship. Other methods can be thought of to realize a similar split of the data set. We changed the extrapolation of the shown 'mean correlation curve' in Figure 5 to a dashed line to reduce a possible misunderstanding about the origin of this curve.

p21: At this stage in the paper you use the term 'convective transport' quite frequently and in association with trajectories/CLaMS. I think that you should be a bit clearer about what transport is included in trajectories/CLaMS – which I believe is simply that in the ERA-I velocity fields, i.e. there is no inclusion of convective transport by parametrisation. I suspect that ERA-I velocities tend to be rapidly upward in regions of large-scale convection – and that serves as some kind of representation of convective transport, but it is unlikely that upward velocities are quantitatively correct. Certainly this sort of interpretation has been made by many authors who have used trajectory-based approaches, including myself, and I would not quarrel with it – but I do think that it needs to be clearly stated. The uncertainty perhaps becomes a bit more serious when considering tropical cyclones. How well is vertical transport in tropical cyclones represented by something like ERA-I? One imagines that maximum vertical velocities are significantly underestimated – but it could be, for example, that vertical transport is distributed over too large a region. You may not be able to resolve this uncertainty, but I think that you should at least say that it exists.

Many thanks for this helpful comment and we confirm that in CLaMS as used here there is no inclusion of convective transport by parametrisation. We added the following text to Sect. 2.3.

" In CLaMS, the diabatic approach was applied using the diabatic heating rate as the vertical velocity with contributions from radiative heating including the effects of clouds, latent heat release, mixing, and diffusion (for details, see Ploeger et al., 2010). CLaMS employs a hybrid vertical coordinate (ζ) which, in this study, transforms from a strictly isentropic coordinate (Θ) to a pressure-based orography-following coordinate system (σ coordinates) below a threshold of approximately 300 hPa(Pommrich et al., 2014). In both three-dimensional simulations as well as in trajectory calculations, the upward transport in CLaMS is driven by ERA-Interim reanalysis data in which changes are implemented to improve deep and mid-level convection compared to previous reanalysis data (Dee et al., 2011). However small-scale rapid uplift in convective cores is not included, therefore small-scale convection is most likely underestimated in CLaMS simulations driven by ERA-Interim. Nevertheless, upward transport in larger convective systems such as tropical cyclones is represented in CLaMS trajectory calculations driven by ERA-Interim (Li et al., 2017, 2020)."

CLaMS trajectory calculations using ECMWF's next-generation reanalysis ERA5 (Hersbach et al., 2020), demonstrate that diabatic trajectory calculations using ERA5 show much faster and stronger vertical transport than ERA-Interim primarily because of ERA5's higher spatial and temporal resolution, which likely resolves convective events more accurately (Li et al., 2020). Nonetheless, Li et al. (2020) demonstrate that the large scale convective uplift as represented in ERA-Interim does represent well the large scale uplift in convective systems.

Detailed comments:

l126: 'last accessed' info not needed in citation?

The last accessed info is now removed in the main text.

l164: 'essential' would be better than 'mandatory'. ('Mandatory' means 'required by some rule or regulation'.)

Good point, we changed "mandatory" to "essential".

l210-215: I found these sentences confusing. What is the distinction between 'pure CLaMS back trajectories', 'pure CLaMS trajectories' and 'back trajectories'? There seem to be two separate issues here – 'forward vs backward' and 'with CLaMS mixing vs without CLaMS mixing'.

We agree that this section was written a bit confusing and changed the text as follows:

"To support the interpretation of airborne measurements we use global three-dimensional simulations of the Chemical Lagrangian Model of the Stratosphere (CLaMS; McKenna, 2002a,b; Pommrich et al., 2014) as well as ~~pure~~ CLaMS back-trajectory calculations. ~~Pure~~ CLaMS back-trajectory calculations consider only the advective (reversible) transport, neglecting (irreversible) mixing processes entirely (e.g., Vogel et al., 2019; Hanumanthu et al., 2020)."

l221: 'lapsrate' should be 'lapse rate'.

Thanks, it is corrected in the text now.

l239-240: This seems to repeat some information on back trajectories what has been given early (which potentially causes more confusion – 'is this intended to be subtly different to what has been said previously').

No, it should not be different. We agree and removed in the revision in Section 2.3.2 the confusing statements. For details, we refer to the track change version of the manuscript.

l241: 'The spatial uncertainty of calculated back-trajectories increases with time because mixing processes occurring during transport are neglected' – the uncertainty doesn't just result from neglect mixing processes – the nature of chaotic advection is such that e.g. small errors in velocity fields convert into increasingly large errors in particle position. The idea that one could accurately calculate a trajectory for, e.g. 50 days, simply doesn't make sense (whether or not one accounts for mixing). The key point for back-trajectory calculations on this sort of time scale is that ensembles of particles are used – so one is essentially calculation probabilities of location of origin rather than 'the' location of origin'.

We agree and changed the respective text in Section 2.3.2 as follows:

"~~The spatial uncertainty of calculated back-trajectories increases with time because mixing processes occurring during transport are neglected. However, the back-trajectory analysis is used here in a statistical way (ensembles of about 100 to 200 trajectories) and not to consider single trajectories. In addition, t~~In general, trajectory calculations have limitations caused by trajectory dispersion increasing with the trajectory length, therefore ensembles of trajectories (of about 100 to 200 trajectories) are used here."

l250: 'Thereby mainly air parcels of ... branch are from below the thermal TP' – do you mean this – i.e. that most of the parcels from below the thermal tropopause are from the lower branch, or do you mean something slightly different, that most of the parcels from the lower branch are from below the thermal tropopause – in which case the 'mainly' should before 'from below'.

Actually both versions are true. However, we wrote the sentence as intended thus no corrections are made here.

Figure 15: You use the term 'convective updraft' in this Figure. There seems to be potentially an unfortunate confusion with the use of 'updrafts' and 'downdrafts' as description of rather small-scale features (perhaps 1km) of convective clouds. 'Convective transport' might be a better term (and a better fit to the fact that, as noted above, your model calculations are incorporating some kind of global re-analysis scale representation of large-scale transport by convective systems.

Many thanks for this interesting remark! The labeling in Figure 15 is updated now as suggested.

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