nature portfolio

Peer Review File

Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality

24th Aug 22

Dear Dr Vogel,

Please accept our apologies for the delay in obtaining reports for your manuscript titled "Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region". Your manuscript has now been seen by 3 reviewers, and I include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. In particular, we ask that you address the points raised regarding model uncertainty and validation, back trajectory, and clearly contextualise the advance beyond previous work. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the completed checklist:
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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Clare

Clare Davis, PhD
Senior Editor
Communications Earth & Environment

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Review of “Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region” by Vogel et al.

Vogel et al. present an approach to reconstruct vertical CO2 profiles over the Asian monsoon region from ground-based observations and GOSAT-L4B satellite data employing a Lagrangian transport model in backward mode. They validate their approach by reconstructing highly temporally and vertically resolved in-situ CO2 profiles observed during a flight campaign (HAGAR instrument) in July 2017. Furthermore, in-situ N2O profiles taken in the same flight campaign are shown and the cavities of the presented approach concerning N2O are briefly discussed.

Major claim
Vertical CO2 and N2O profiles can be reconstructed accurately from ground-based observations and supplemented by satellite observations employing Lagrangian transport models in backward mode (cavities for N2O due to non-linear chemistry, if I understand correctly).

Novelty of claim
Since I don’t have a very good overview on the related literature, I am not sure I can evaluate the novelty of this approach definitely, but such an approach is the first I see, it’s well explained and seems sound.

Interest to others

Definitely.

Claim convincing?
Yes, I am convinced that the presented approach can be used to accurately reconstruct vertical CO_2 profiles. The authors also argue that there is a lack of ground-based observations covering this highly populated region in order to apply their approach which I am also supporting. We need more observations in this region.

Other experiments necessary?
It would be nice to see a simulation with even longer back-trajectory length, as to me Fig. S1 seems to indicate that this might further improve the case S2a (ground-based observations only). If the authors don't want to do this, I would highly appreciate more discussion (also in the main text) on why the chosen back-trajectory length is the optimal choice. This is not obvious to me, also after looking at the provided supplement material.

Claims discussed appropriately in context of previous literature
Yes

The paper is well written and clearly explained, for most parts. However, I have a few questions and suggestions for extended discussion to hopefully further improve the manuscript. The authors argue that there are two main limitations to their approach to reconstruct vertical profiles: 1) lack of observations 2) transport errors, which I agree in principle.

• I am not sure I would argue that the transport model error is a severe constraint, I see the by far bigger problem in the lack of observations. Nevertheless, I would recommend to discuss the transport errors a bit more in the paper, this might also help to understand why you think it's a severe constraint.

• Furthermore, I highly recommend to add more discussion, also in the main text, why the chosen back-trajectory length is the optimal one, this is not obvious to me.

• Combination of the two point above. Wouldn't an even longer back period maybe improve the results? (especially from Fig. S1, I get this impression). Maybe this would be something to check with an additional experiment. If the authors don't want to do this, I at least would appreciate a more detailed discussion on how they concluded that the chosen period is the optimal one.

• Vertical coordinates: For the authors the relationship between potential temperature and altitude in meters or pressure levels might be obvious, but I would highly recommend adding a second y-axis giving altitude in meters in addition to potential temperature. And also give more references between potential temperature and altitude in meters in the text, as potential temperature is not very intuitive if one does not deal with this on a daily basis. I think this would improve readability of the manuscript.

After the authors address these few points above (and the more detailed comment below), in any way they seem fit, I highly recommand this mansucript for publication. I enjoyed reading this very nice paper. Hopefully my comments can help to improve it further. All the best. Please find my specific comments below.

L40: Which transport errors are you referring to here? Do you mean that individual trajectories get more uncertain if you go further back in time from the release time? How is your study helping to address these transport errors? Since you use a Lagrangian transport model for your reconstructions. Please add some more explanation.

L87: Again, it would be good if you could explain the transport error a bit better and how you are dealing with this error since you also use a transport model. I see the bigger issue in the lack of observations, not sure how severe the transport error is (assuming using a sufficient number of particles in your simulations to get robust sampling).

L124-176: In principle, I like overview maps with all mentioned sites. However, this might be tricky here, since your sites basically cover the whole hemisphere. Maybe it would be good to refer to Fig. 2 already early in this paragraph to show the location of the main sites in your study area. Furthermore, I would suggest to add some indication on where a mentioned site is located when you mention it the first time, e.g., Cape Matutala (Samoa). You have it in Fig. 1, but it would be better to also have some indication in the text, for people which are not so familiar with these sites.

Figure 1:

You might consider to add an explanation of the areas highlighted in gray (pre-monsoon, but especially the StratoClim period) also to the legend or elsewhere in the plot (e.g. on top), so it is immediately clear to the reader what these areas mean. You have it in the caption, but it would be nice to have it visually as well.

Figure 2:

Caption: “high altitude” instead of “high-flying”? like in L245.

Figure: Please highlight the three main locations (Naintal, Kathmandu, Comilla) more prominately, e.g., bold, larger font, white buffer around the letters. The font size should be larger in general. You might also consider to add an indication in which direction Mt. Waliguan is located, e.g., with an arrow or similar.

Figure 3:

Caption: You might consider not to use the BL abbreviation in the caption to avoid any confusion (or define a second time in the caption). You define it in the text, but the figure might show up in the typeset manuscript before the reader gets to the definition in the text, like it does in this review version here. Also, the figure might be looked at individually.

L219: “... air parcels that were released at the model BL before 1 June 2016 (aged air) are marked in black.” In the beginning, I was not sure why all these air parcels are still in the BL, until I saw that there is also some further up. Especially the black dots in the upper levels in panel (a) are not very well visible. Could you please highlight them in some way? Maybe use another marker, e.g., a cross, for them. Also, please add a legend for these "aged particles".

By the way, I don't understand why most "aged particles" are still in the BL, while the other older particles (e.g., 428 days) all appear to be in the highest layer. Maybe there are also a lot in the lower layer, but they are covered by the younger particles? If so, you should consider to work with transparencies to show also particles which overlap.

It would be nice to have also the observed ground level CO2 and N2O concentrations in Fig. 3 somehow, maybe as vertical lines (mean over the campaign period), or just marked on the x-axis? The ones you refer to in L253-255.

Figure: Could you please add a second y-axis showing the altitude in meters? Please especially add a rough tropopause altitude in your study region.

Please consider to add also altitude in meters to the discussion, as potential temperature is not very inuitive and many readers might not have a good feeling for the relationship between potential temperature and altitude in meters.

L238-242: I am not particularly familiar with this satellite product, but are all vertical levels sampled in a similar quality? In the methods, 17 vertical levels are mentioned for the GOSAT-L4B product, couldn't you show a vertical profile then as well, not just the column-averaged CO2? Is it the GOSAT interpolated in Fig. S2, this is not really clear to me?

L286-287: Unfortunately, until here it is not clear to me how old your "aged particles" are. Release before 1 June 2016, but how long are your simulations for these "aged particles"? In L272 you mention something about one year. How old are your oldest particles, i.e., how long does it take the "oldest" particles to reach the BL in the backward mode? Maybe add a remark what age range your "aged particles" cover? Does the "aged air" include particles followed backward from all time periods? So the age range is between 3 months (Sept. 2016 back to June 2016) and 15 months (Sept. 2017 back to June 2016)? What happens to the particles which did not reach the BL in the backward runs until 1 June 2016? Please make this more clear in the text.

Figure 4: Ok, now I see that you actually show all locations on a map, maybe add an overview map after all (without any data plotted on top; maybe combined with Tab. 2)? Also please highlight the labels in Fig. 4, they are partly unreadable (e.g., bold + white buffer around the edge of the letters)

L365-405: How exactly did you use the data in Fig. 4 to optimize the regions shown in Fig. 5?

Figure 5: Where are the regions mNH and cNH in Fig. 5? It took me a while to realize that you separate between land and ocean in the North, but not in the South. Maybe you could add the labels for mNH, cNH, tSH, and Wpool also in the figure? E.g., in slightly darker colors as the colors used to mark the regions, to set them apart from the measurement sites.

L452-453: Maybe these two sentences can be combined, e.g., "In Fig.7, CO2 mixing ratios reconstructed in this way are shown as median of 1 K intervals for several measurement sites."

L541-545: Here you discuss the sensitivity on the trajectory length and refer to a discussion in the supplement. However, looking at Fig. S1 and reading this discussion, it is not obvious to me why the length from 1 Dec 2016 is the best. The case S2a seems to improve with increasing length in the higher altitudes and for the 1 Jun 2016 case the fit between simulated and observed profiles is already quite good. Also the uncertainties in the higher altitudes are still smaller then when adding the GOSAT data. What happens if you go back even further? How many particles are you using for your simulations?

L655: Please add an explanatory sentence on this AirCore sampling. You give a reference, but it would be nice to give one sentences on what makes it cheaper than other methods. Is it used operationally anywhere?

Availability of data and materials: The used HAGAR CO2 and N2O data and the ground-based observations should be made openly and permanently available by uploading it to a repository like https://zenodo.org/ to receive a DOI.

The ClaMS settings used to simulate the back-trajectories (at least examples for a few simulations to be able to reproduce) should be made openly and permanently available by uploading it to a repository like https://zenodo.org/ to receive a DOI.

https://www.nature.com/nature-portfolio/editorial-policies/reporting-standards

Reviewer #2 (Remarks to the Author):

Review of "Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region" submitted to Communications Earth & Environment by Bärbel Vogel et al.

Major comments

The subject of this paper is the creation of high vertical resolution data by using the aircraft in situ measurement data in the Asian monsoon region, which has few ground stations but is important as a source of CO2 emissions.

The motivation for the research, the originality of the data used, and the quality of the data are guaranteed. It is expected that high-vertical resolution data will be effectively used for top-down research on CO2 emission data.

However, this reviewer shows a few points to consider which should be cleared before publish.

1) Representativeness : First, how representative is the high vertical resolution data on a

limited number of observation points? Subsequent analysis (an inversion method for estimating emissions) using data constructed from data from a few points in a region where CO_2 has high variation may cause miss-leading.
2) Uncertainty of model : The discussion of the uncertainty of the numerical model should be added. While the introduction shows errors and uncertainties in the numerical model, an important part of this dataset is the use of the Lagrange model. Although using more realistic data than traditional transport models, the uncertainty is likely to be high even in transport processes using diabatic heating, especially in the Asian troposphere, where water vapor and cloud variability are severe. Especially in the UTLS (upper troposphere/lower stratosphere) region, the transport process across the tropopause, including the convective parameterization scheme, has a large variation between reanalysis.
In addition, normally, the back trajectory in the troposphere is limited to one week, but in this study, it is calculated for one year or more. Even though aged air is more than a year old in the stratosphere, transport within the middle world is weeks to months, especially in the summer of the Asian monsoon region. Therefore it is doubtful how realistic the reconstructed data are especially near the tropopause (higher aircraft observation).
3) Validation : Third point, which is related with second point, is how to validate reconstructed data.
Since the reconstructed data have already high quality, it is key how to verify the bias/error of Lagrangian transport and the upper layer data, although the verification work is done by only GOSAT-L4B data as the sensitivity experiment. (Please check vertical transport tendency of NIES-TM driven by JRA-25, which is old version in JRA.) It is not enough to me because there is a large difference depending on the reanalysis data that drives the numerical model, especially around the tropopause, so it is recommended that you to carry out verification using other reanalysis data, JRA55 and MERRA2. If the tendency is the same, the reader will be satisfied the results.
Further, is it possible to apply it to other aircraft observation data, like CONTRAIL and CARIBIC?

Minor comments:
L223-226, N_2O in Figure1: I couldn't get the seasonality that the authors said. Please explain a little more carefully.
L227-229: related with major comment #1, is it okay to say the representativeness of a vast area with only two points?
L240-242 : Please add the explanation why the amplitude of GOSAT-L4B is smaller than in situ measurement for general readers.
L324-347, Figure4: Please add the distribution of age of air from BL at the flight altitude/lower stratosphere, which is helpful to understand the transported length and distance of source deeply.
L355: why the latitude gird is 0.6 degree, not 0.5 degree?
L430: Add the “Asian monsoon” words to make it easier for the reader to read.
L507-524, Figure 7: It is difficult to divide the similar color, HAGAR and B. Kototabang.

Reviewer #3 (Remarks to the Author):

This is a creative analysis of a unique dataset over the poorly observed Indian subcontinent that provides insight into surface influences on air in the mid/upper-troposphere and lower stratosphere during the Indian monsoon season. The success of the CLaMS model reconstruction of the profiles up to ~380/400K is remarkable. However, the paper lacks important context from previous work, and so I am recommending a significant revision. I think this can be a truly excellent paper with just a bit of reframing and potentially some minor but important additional scientific analysis.

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Overarching comments/concerns:

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The paper presents a model analysis of CO2 measurements over the Indian subcontinent during the Asian monsoon season. Aircraft vertical profiles were obtained in the Northern subtropics between 20N and 30N. This is outside of the "deep tropical upwelling region", and near the southern border of what has been referred NH extratropical surf zone. The profiles extend from near the surface and well into the stratosphere (potential temperature 300K - 480K). There is a large body of work on upper troposphere lower stratosphere (UTLS) transport that provides important context for interpetting the observed vertical structure, but the current manuscript lacks any significant discussion of this prior work.

Global-scale stratospheric transport includes two regimes, the stratospheric "overworld" corresponding to potential temperature surfaces greater than ~380K and the "lowermost extratropical stratosphere", which is also known as the stratospheric "middleworld" (Holton et al., 1995). In the overworld, isentropes are contained wholly within the stratosphere, whereas in the lowermost stratosphere the isentropes are above the tropopause in the extratropics and below the tropopause in the deep tropics. Thus, in the lowermost stratosphere (potential temperature < ~380-400K) rapid mixing can occur between airmasses characteristic of the upper tropical troposphere and of the mid-latitude lower stratosphere. In the overworld, no such rapid mixing of tropospheric and stratosphere can occur. The boundary between the overworld and the lowermost stratosphere shows up clearly in Fig 6a and in Fig 8a.

The CLaMS model has been carefully evaluated and seems to do a credible job of capturing air mass transport in these two transport regimes, and panels 6 b-d provide interesting insight into how tropospheric from the NH, SH and Warmpool areas airmasses propagate into the stratospheric overworld. The paper would benefit from some discussion along these lines (e.g. NH and SH contributions are roughly equal; the Warm Pool signature can be seen in panels 6c-3, but is it's contribution commensurate with it's area or does the warm pool account for a disproportionately large fraction of the air in the overworld because of the strong convection in that region?)

Also lacking from the current paper is any discussion of stratospheric transport in the stratospheric overworld. There are many relevant papers about this, a few of which are listed below. The rapid decrease of N2O above 380-400K is indicative of mixing of relatively young air with aged stratospheric air that has descended from much higher altitudes where the photochemical lifetime of N2O is short (photochemical loss at altitudes corresponding to

400-470 K is too slow to explain rapid decrease with altitude). The Boering et al, 1996 (Science) and both Andrews et al. 2001 (JGR) papers below show that CO2 and N2O relationships are remarkably consistent throughout the lower stratospheric overworld, once the tropospheric growth of both species is taken into account and if the CO2 stratospheric boundary time series is convolved with a reasonable representation of the age spectrum (e.g. Andrews et al., 2001a, b; Ray et al. 2017). The discrepancy between the modeled an observed CO2 profiles in Figure 8a is clearly due to the mixture of aged stratospheric air with younger air. Age spectra in the lower stratosphere have been convincingly shown to be asymmetric with a strong peak indicating young air (consistent with Fig 6 in the current manuscript!) and a tail indicating the presence of older air.

It would be very interesting to explore the extent to which the upper portion of the HAGAR profiles in Fig 8a could be reconstructed as follows:
(1)computing the Age:N2O relationship from equation 3 of Andrews et al. 2001 (https://doi.org/10.1029/2001JD000465) after scaling N2O to account for the difference between tropospheric avg N2O summer 2017 (329.9 ppb) in 1997 (313ppb) and in 2017 (i.e. N2O[1997] = N2O[HAGAR]*(313ppb/330ppb)

(2) Use a method like Ray et al., 2017 to convolve a timeseries of CO2 representative of the stratospheric boundary condition at the tropical tropopause (which can be approximated using the average of Mauna Loa and Samoa data lagged by 2 months) with a set of reasonable age spectra having mean ages corresponding to the age profile obtained in step 1 (i.e. this is applying the method described in Ray et al. 2017 but inverted to solve for CO2 as a function of Age instead of Age as a function of CO2). The resulting computed CO2 profile could be directly compared to the Hagar CO2 observations above 400 K and would provide insight into Age:CO2:N2O relationships in the stratospheric overworld circa 2017 compared with the previous work.

Finally, it would be *very* useful to show how CarbonTracker and other available CO2 models (there are many e.g. from the OCO-2 MIP) that have been optimized against observations (e.g. The EU Copernicus CAMS product and the suite of "OCO2 MIP models" presented by Crowell et al., 2019, Atmos. Chem. Phys., doi: 10.5194/acp-19-9797-2019) compare with the HAGAR profiles. The HAGAR profiles are in an especially important and poorly sampled region. I would not necessarily expect these models to perform well at all in this severely under-constrained region, and that is really important to highlight in the literature to strengthen the argument for more vertical profile data in undersampled regions. I also expect these models may perform poorly in the stratosphere because they have mostly been optimized for tropospheric transport (i.e. native model levels left out at high altitudes to speed computation, etc), but if they are performing poorly in the stratosphere then that has *major* implications for possible biases when assimilating satellite data. CO2 is a terrific tracer of stratospheric transport and so even a simple model evaluation could provide a lot of insight. It would be nice to include a figure in the supplement or the main paper showing how well the individual OCO2 MIP model simulations do or do not agree with the HAGAR profiles.

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Relevant publications cited above and a few others about CO_2 in the UTLS that should perhaps be considered for inclusion in the intro or discussion:
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Holton et al., 1995, Stratosphere-troposphere exchange, Reviews of Geophysics, https://doi.org/10.1029/95RG02097

Xueref et al., 2004, Combining a receptor-oriented framework for tracer distributions with a cloud-resolving model to study transport in deep convective clouds: Application to the NASA CRYSTAL-FACE campaign, GRL, 2004, 10.1029/2004GL019811

Boering et al., 1995, MEASUREMENTS OF STRATOSPHERIC CARBON-DIOXIDE AND WATER-VAPOR AT NORTHERN MIDLATITUDES - IMPLICATIONS FOR TROPOSPHERE-TO-STRATOSPHERE TRANSPORT, JGR, DOI: 10.1029/95GL02337,

Boering et al., 1996, Stratospheric mean ages and transport rates from observations of carbon dioxide and nitrous oxide, Science, 10.1126/science.274.5291.1340

Park, S et al., 2007, The CO_2 tracer clock for the Tropical Tropopause Layer, ACP, DOI10.5194/acp-7-3989-2007

Andrews A et al., 2001, Empirical age spectra for the midlatitude lower stratosphere from in situ observations of CO_2: Quantitative evidence for a subtropical "barrier" to horizontal transport, JGR, 10.1029/2000JD900703

Andrews et al., 2001, Mean ages of stratospheric air derived from in situ observations of CO_2, CH_4, and N_2O, JGR, 10.1029/2001JD000465

Ray et al., 2017, Quantification of the SF_6 lifetime based on mesospheric loss measured in the stratospheric polar vortex, JGR, doi:10.1002/2016JD026198

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Specific Comments:

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Suggestion to remove or greatly reduce verbiage about N_2O from abstract and intro, since there is actually very little analysis of the N_2O profiles in the current version of the paper.

CLaMS driven by ERA-5 seems to be ideally formulated for reproducing mid/upper troposphere profiles in the NH subtropics. Is it expected to work as well elsewhere?

The abstract and intro include a lot of verbiage about N_2O emissions, but the paper is really about CO_2 as reflected in the title. I suggest removing mention N_2O from the abstract and limiting discussion in the intro, since it's really just functioning as a tracer of stratospheric air.
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Although reframing as suggested above might lead to other changes in these sections such that N2O has a larger role in interpreting the stratospheric overworld data.

Line 640: I agree that the rapid ventilation up to 14 km is consistent with significant influence from India and Tibet, but I think stating that the influence of asia and india can be discerned up to 22km is an overstatement. At those highest altitudes, the signature of surface emissions is tho

Figure 1 shows that BKT has lower values than MLO, WLG and other stations, suggesting that BKT is impacted by local CO2 uptake by vegetation. Some explanation about why CO2 at BKT is so much lower than the other sites is needed. In contrast, MLO, WLG, and SMO are regionally representative records. Due to it's high elevation (nearly 4km above sea level), MLO is representative of the free troposphere rather than the marine boundary layer. I recommend evaluating the extent to which BKT is regionally representative by considering the NIES CONTRAIL dataset of measurement from commercial aircraft flights. There are many vertical profile data in the vicinity of Indonesia. However, the contribution of the Warm Pool region is rather small and likely does not contribute significantly to the reconstructed profile. I would venture to guess that the BKT timeseries could be replaced with either SMO or MLO or the average of the two with minimal impact on the reconstructed CO2 profile. I'm not sure the success of the method is dependent on treating the Warm Pool as a separate region. If so, then that would be an interesting result worth highlighting a bit more.

Regarding the use of surface CO2 observations from the World Data Center for Greenhouse Gases. I think they include a fair use statement and metadata requesting that the actual data providers be contacted about how they should be acknowledged in publications. The WDCGG functions as a clearinghouse for datasets, they do not directly provide observations. Please look at the metadata and consult the actual data providers about how they would like to be acknowledged. If the data are from NOAA's Global Greenhouse Gas Reference Network as I suspect, then please contact Xin.Lan@noaa.gov. She may also have some advice about the represtativeness of BKT data.

Figure 3:There are a lot of black points at low altitudes that I would assume are the youngest particles. This is confusing since the caption says that aged parcels > 1 yr are shown in black. Are both the oldest and the youngest particles shown in black? Maybe the figure should be reworked so that the oldest parcels are shown in grey/black and the youngest are shown in color. The trajectories corresponding to the oldest air are likely the least reliable, so maybe not useful to show different colors for e.g. air > 365 days. This would allow color scale to be used to full effect in the portion of the profiles between 350-410K where the CLaMS reconstruction is most successful.

Line 227: N2O emissions are found... What is the source of this information about the geographic distribution of N2O emissions? Inventory estimates?

Line 257: "CO2 is chemically intet in the troposphere and stratosphere." CO2 has a small source from CH4 oxidation in the stratosphere/mesosphere that is not completely negligible in the lower stratosphere. The contribution of CO_2 from this source to the observed profile can easily be estimated from coincident measurements of CH_4 (i.e. it is equivalent to the observed CH_4 minus CH_4 at ~400 K).

Line 297/298: I'm not sure what is meant by "most of the trajectories were released at the model BL much earlier". Perhaps it could be reworded something like: "Air parcel trajectories were calculated backward in time until June 2016 by which time approximately X% of the air parcels reached the model BL"

Line 313: summery -> summary

I would omit most of the N_2O paragraph starting line 472 unless you add the analog of Figure 8a. It makes sense that the N_2O profile above ~400K cannot be reconstructed due to chemical loss, but what about the tropospheric profile? I understand that the profile is nearly vertical, but do you get the right tropospheric N_2O value based on these sites? Or are all the tropospheric values essentially the same?

Figure 6 warrants more discussion, in particular, the progression back through the seasons across the panels is interesting in that for panels d and e show the surface influences above 410 K are about equally distributed between tSH and mNH,with a large fraction of older air. The region above ~400K is the stratospheric overworld where isentropes are contained wholly within the stratosphere. The difference between the overworld transport regime and the subtropical UT/lowermost stratosphere corresponding to ~400 K.

The distribution of influences is consistent with papers using NASA ER-2 CO_2 measurements in the 1990's through the early 2000's showing that the stratospheric overworld boundary condition can be well-represented by the average of MLO and SMO data lagged by 2 months (references). It may also be possible to compare the relative contributions of surface air and aged stratospheric air in the lower stratospheric overworld region (i.e. above ~380/400K) with the empirical age spectra derived in Andrews et al. I think the label "free atmos" and corresponding references in the text should simply be changed to "aged stratospheric air". When I think of free atmospheric air, I usually think of free troposphere above the boundary layer. Irene Xeuref has a relevant paper on CO_2 as a clock tracer in the UT/LS that seems highly relevant. Kristie Boering had a paper on using CO_2:H_2O relationships to estimate transit times from the tropical tropopause to the lower mid-latitude stratosphere that is also relevant.

I don't think the inclusion of the GOSAT-L4B product to improve the profile reconstruction at the highest altitudes adds much to the paper. The error bars are very large, and a mismatch of several ppm in the stratosphere cannot be considered a good fit to the observations. Why not also include CAMS and CarbonTracker in the main paper along with GOSAT L4B and make a stronger point about the poor performance of the available model products? Figure S2B is underwhelming. How does the GOSAT-L4B compare directly with the HAGAR data? My understanding is that the GOSAT-L4B trajectories are sampled at their endpoints and that extrapolation is necessary above 10hPa. This doesn't seem at all compelling. I'm just not sure what we are learning from this part of the analysis. Maybe I am missing something?

Line 621: "Thus not surprisingly, the CO2 distribution at the ground nor the vertical distribution of CO2 over South Asia during summer 2017 is not well represented in CarbonTracker (35)." I think maybe this is supposed to be Ref #34 (Jacobson et al) rather than Ref #35 (Konopka et al, submitted).

In the discussion of Figure 7 the agreement between the observed profile and the CO2 reconstruction based on Nainital >410 K is described as remarkable. I would instead describe it as coincidental. It's not really that surprising given the range of observed surface CO2 across the cases that one of them falls on top of the obs. I don't think there is any physical significance to this result.

Author Comment to Referee #1

Communications Earth & Environment (COMMSENV-22-0506-T), 'Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region' by B. Vogel et al.

We thank Referee #1 for the positive review and for further guidance on how to revise our manuscript. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics. Passages from the revised version of the manuscript are shown in blue.

Review of "Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region" by Vogel et al.

Vogel et al. present an approach to reconstruct vertical CO2 profiles over the Asian monsoon region from ground-based observations and GOSAT-L4B satellite data employing a Lagrangian transport model in backward mode. They validate their approach by reconstructing highly temporally and vertically resolved in-situ CO2 profiles observed during a flight campaign (HAGAR instrument) in July 2017. Furthermore, in-situ N2O profiles taken in the same flight campaign are shown and the cavities of the presented approach concerning N2O are briefly discussed.

Major claim Vertical CO2 and N2O profiles can be reconstructed accurately from ground-based observations and supplemented by satellite observations employing Lagrangian transport models in backward mode (cavities for N2O due to non-linear chemistry, if I understand correctly).

Novelty of claim Since I don't have a very good overview on the related literature, I am not sure I can evaluate the novelty of this approach definitely, but such an approach is the first I see, it's well explained and seems sound.

Interest to others Definitely.

Claim convincing? Yes, I am convinced that the presented approach can be used to accurately reconstruct vertical CO2 profiles. The authors also argue that there is a lack of ground-based observations covering this highly populated region in order to apply their approach which I am also supporting. We need more obser-

vations in this region.

Other experiments necessary? It would be nice to see a simulation with even longer back-trajectory length, as to me Fig. S1 seems to indicate that this might further improve the case S2a (ground-based observations only). If the authors don't want to do this, I would highly appreciate more discussion (also in the main text) on why the chosen back-trajectory length is the optimal choice. This is not obvious to me, also after looking at the provided supplement material.

Claims discussed appropriately in context of previous literature Yes

We thank Referee #1 for this very positive review. A detailed discussion about the trajectory length and the optimal choice of trajectories follows below.

General Comments:

The paper is well written and clearly explained, for most parts. However, I have a few questions and suggestions for extended discussion to hopefully further improve the manuscript. The authors argue that there are two main limitations to their approach to reconstruct vertical profiles: 1) lack of observations 2) transport errors, which I agree in principle.

- *I am not sure I would argue that the transport model error is a severe constraint, I see the by far bigger problem in the lack of observations. Nevertheless, I would recommend to discuss the transport errors a bit more in the paper, this might also help to understand why you think it's a severe constraint.*

  Many thanks for this comment. We agree that the expression 'transport model error' is too vague and we have to be more precise. Therefore we added the following paragraph in the revised version of the paper:

  In state-of-the-art chemistry transport models, the transport of air parcels differs because different methods (Eulerian, Lagrangian), different vertical velocities (kinematic, diabatic) and different meteorological reanalyses (e.g. ERA5, ERA-Interim, JRA-55) are used to drive the models (e.g. Bergman et al., 2013; Ploeger et al., 2019). Further, the implementation of convection and irreversible mixing differs from model to model.

The results of Lagrangian trajectory calculations with CLaMS used in our study, are analysed in a statistical sense, because mixing processes between different air parcels are neglected. We added some further explanations on this issue in the revised version of the manuscript (details see below). Having said all this, we agree with the reviewer that the 'lack of observations is a very severe issue. We hope this point comes across clearly in the revised version.

- *Furthermore, I highly recommend to add more discussion, also in the main text, why the chosen back-trajectory length is the optimal one, this is not obvious to me.*

  Many thanks for this comment. We agree that our explanation was too short. We changes this paragraph in the revised version of the manuscript (details see below under Specific Comment No.18).

- *Combination of the two point above. Wouldn't an even longer back period maybe improve the results? (especially from Fig. S1, I get this impression). Maybe this would be something to check with an additional experiment. If the authors don't want to do this, I at least would appreciate a more detailed discussion on how they concluded that the chosen period is the optimal one.*

  We agree with this comment, further details see below under Specific Comment No.18.

- *Vertical coordinates: For the authors the relationship between potential temperature and altitude in meters or pressure levels might be obvious, but I would highly recommend adding a second y-axis giving altitude in meters in addition to potential temperature. And also give more references between potential temperature and altitude in meters in the text, as potential temperature is not very intuitive if one does not deal with this on a daily basis. I think this would improve readability of the manuscript.*

  Many thanks for this comment. We added pressure and altitude levels to Fig. 3 of the revised version of the paper (shown as in Fig. 2 of this reply) for better clarity.

After the authors address these few points above (and the more detailed comment below), in any way they seem fit, I highly recommand this mansucript for publication. I enjoyed reading this very nice paper. Hopefully my comments can help to *improve it further. All the best. Please find my specific comments below.*

Many thanks, we were very pleased with Referee #1 review. We are confident that your comments helped to further improve our paper.

Specific Comments:

1. *L40: Which transport errors are you referring to here? Do you mean that individual trajectories get more uncertain if you go further back in time from the release time? How is your study helping to address these transport errors? Since you use a Lagrangian transport model for your reconstructions. Please add some more explanation.*

   We revised the sentence 'Also the transport error of the employed models is a severe constraint.' as follows to be more precise.

   Also differences in transport of the employed models cause a severe uncertainty.

2. *L87: Again, it would be good if you could explain the transport error a bit better and how you are dealing with this error since you also use a transport model. I see the bigger issue in the lack of observations, not sure how severe the transport error is (assuming using a sufficient number of particles in your simulations to get robust sampling).*

   We replaced the sentence 'Further, the transport error of the employed models is a severe constraint.' as follows for clarification (see above).

   In state-of-the-art chemistry transport models, the transport of air parcels differs because different methods (Eulerian, Lagrangian), different vertical velocities (kinematic, diabatic) and different meteorological reanalyses (e.g. ERA5, ERA-Interim, JRA-55) are used to drive the models (e.g. Bergman et al., 2013; Ploeger et al., 2019). Further, the implementation of convection and irreversible mixing differs from model to model.

3. *L124-176: In principle, I like overview maps with all mentioned sites. However, this might be tricky here, since your sites basically cover the whole hemisphere. Maybe it would be good to refer to Fig. 2 already early in this paragraph to show the location of the main sites in your study area. Furthermore, I would suggest to add some indication on where a mentioned site*

is located when you mention it the first time, e.g., Cape Matutala (Samoa). You have it in Fig. 1, but it would be better to also have some indication in the text, for people which are not so familiar with these sites.

We agree and introduced a map in Fig. 2 of the the revised manuscript showing the geographical positions of all sites used (see Fig. 1 of this reply).

4. *Figure 1: You might consider to add an explanation of the areas highlighted in gray (pre-monsoon, but especially the StratoClim period) also to the legend or elsewhere in the plot (e.g. on top), so it is immediately clear to the reader what these areas mean. You have it in the caption, but it would be nice to have it visually as well.*

   We considered the comment but we think that Figure 1 would be to overloaded including explanation for the greyish areas into the figure.

5. *Figure 2: Caption: "high altitude" instead of "high-flying"? like in L245. Figure: Please highlight the three main locations (Naintal, Kathmandu, Comilla) more prominately, e.g., bold, larger font, white buffer around the letters. The font size should be larger in general. You might also consider to add an indication in which direction Mt. Waliguan is located, e.g., with an arrow or similar.*

   We agree and revised Fig. 2 of the manuscript as shown in Fig. 1 of this reply.

6. *Figure 3: Caption: You might consider not to use the BL abbreviation in the caption to avoid any confusion (or define a second time in the caption). You define it in the text, but the figure might show up in the typeset manuscript before the reader gets to the definition in the text, like it does in this review version here. Also, the figure might be looked at individually.*

   Thanks, done.

7. *L219: "... air parcels that were released at the model BL before 1 June 2016 (aged air) are marked in black." In the beginning, I was not sure why all these air parcels are still in the BL, until I saw that there is also some further up. Especially the black dots in the upper levels in panel (a) are not very well visible. Could you please highlight them in some way? Maybe use another marker, e.g., a cross, for them. Also, please add a legend for these "aged particles".*

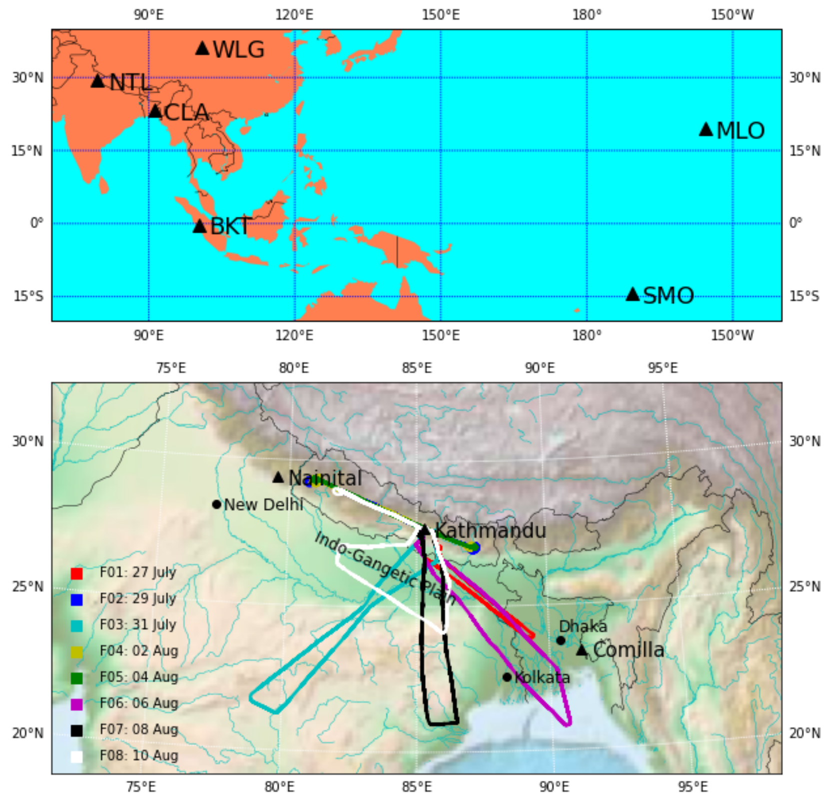

Figure 1: **Regional map of the measurement sites for Greenhouse gases and of the aircraft measurements on the Indian subcontinent.** The locations of the measurement sites for Greenhouse gases in Nainital (NTL, India) Comilla (CLA, Bangladesh), Mt. Waliguan (WLG, China), Bukit Kototabang (BKT, Indonesia), Mauna Loa (MLO, Hawaii) and Samoa (SMO, Cape Matatula) (top) and the flight paths of the eight local scientific flights (F01-F08) by the high altitude research aircraft Geophysica (bottom) are shown. The scientific flights were carried out every second day from Kathmandu (Nepal) between 27 July and 10 August 2017.

Many thanks for this comment. We agree, that Fig. 3 needs some modifications and revised the figure as shown in Fig. 2 of this reply.

8. *By the way, I don't understand why most "aged particles" are still in the BL, while the other older particles (e.g., 428 days) all appear to be in the highest layer. Maybe there are also a lot in the lower layer, but they are covered by the younger particles? If so, you should consider to work with transparencies to show also particles which overlap.*

   There is a misunderstanding. There are no 'aged particles' in the model boundary layer (see Fig. 2 of this reply).

9. *It would be nice to have also the observed ground level CO2 and N2O concentrations in Fig. 3 somehow, maybe as vertical lines (mean over the campaign period), or just marked on the x-axis? The ones you refer to in L253-255.*

   A detailed comparison of ground level CO_2 and N_2O with HAGAR measurements is show in Fig. 7 of the revised version of the manuscript.

10. *Figure: Could you please add a second y-axis showing the altitude in meters? Please especially add a rough tropopause altitude in your study region.*

    We added the WMO tropopause as well as some altitude levels (see Fig. 2 of this reply).

11. *Please consider to add also altitude in meters to the discussion, as potential temperature is not very inuitive and many readers might not have a good feeling for the relationship between potential temperature and altitude in meters.*

    Some pressure and altitude levels are added to Fig. 3 of the revised version of the manuscript (see Fig. 2 of this reply).

12. *L238-242: I am not particularly familiar with this satellite product, but are all vertical levels sampled in a similar quality? In the methods, 17 vertical levels are mentioned for the GOSAT-L4B product, couldn't you show a vertical profile then as well, not just the column-averaged CO2? Is it the GOSAT interpolated in Fig. S2, this is not really clear to me?*

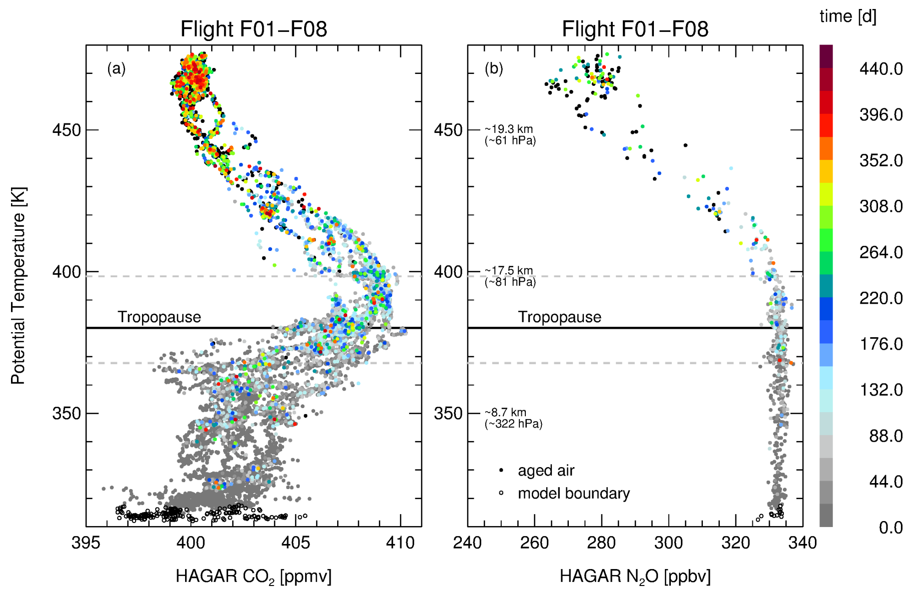

Figure 2: **Airborne CO_2 and N_2O measurements from the StratoClim campaign in Kathmandu (Nepal) during July and August 2017.** Each air parcel is coloured by the transport time from the model boundary layer (BL) to the time of measurements inferred by Lagrangian back-trajectory calculations. Air parcels located in the model BL as well as aged air (air located in the free atmosphere on 1 June 2016) are marked. The number of air parcels is determined by the different temporal resolution of the CO_2 **(a)** and N_2O **(b)** measurements (details see Methods). In addition, the mean WMO tropopause (Hoffmann and Spang, 2022) as well as the lowest and highest tropopause (grey dashed lines) over Kathmandu during the flight days are shown.

We revised the old sentence: 'The seasonal variability of CO_2 over the Indian subcontinent (mean value between 10–35°N and 65–95°E) of the lowest model level at 975 hPa of the GOSAT-L4B product (details see Methods) inferred from column-averaged satellite measurements is compared to ground-based CO_2 measurements in Fig. 1a,b.' as follows for better clarity.

The seasonal variability of CO_2 over the Indian subcontinent (mean value between 10–35°N and 65–95°E) at the ground (the lowest model level at 975 hPa) as estimated by the GOSAT-L4B product (details see Methods) is compared to ground-based CO_2 measurements in Fig. 1a,b. The GOSAT-L4B product is a model simulation using CO_2 surface fluxes inferred from column-averaged satellite measurements (details see Methods); The lowest model level of GOSAT-L4B is closest to the inferred CO_2 surface fluxes and is not strongly influenced by the tracer transport of the underlying transport model.

Fig. S2 (black dots) shows simulated CO_2 from GOSAT-L4B model simulations interpolated to the time and location of the HAGAR measurements. Measured CO_2 profiles from GOSAT are not available, there are only column-averaged measurements or 3-dimensional GOSAT-L4B model simulations. However, we added a comparison of HAGAR CO_2 profiles with GOSAT-L4B profiles for each research flight in the revised version of the supplement (see Fig. S3).

13. *L286-287: Unfortunately, until here it is not clear to me how old your "aged particles" are. Release before 1 June 2016, but how long are your simulations for these "aged particles"? In L272 you mention something about one year. How old are your oldest particles, i.e., how long does it take the "oldest" particles to reach the BL in the backward mode? Maybe add a remark what age range your "aged particles" cover? Does the "aged air" include particles followed backward from all time periods? So the age range is between 3 months (Sept. 2016 back to June 2016) and 15 months (Sept. 2017 back to June 2016)? What happens to the particles which did not reach the BL in the backward runs until 1 June 2016? Please make this more clear in the text.*

    We agree that the formulation is confusing therefore we revised the caption of Tab. 1 'The analysis of CLaMS back-trajectories (see Methods) is performed back until the start time of each season. Air parcels that were released at the model BL before 1 June 2016 are considered as aged air.' as follows:

The analysis of CLaMS back-trajectories (see Methods) is performed back until the start time of each season. For each season air parcels that were released at the model boundary layer (BL) are analysed. The longest simulation time is back until 1 June 2016 ($\sim$ one year). Air parcels that are located in the free atmosphere on 1 June 2016 are considered as aged air.

14. *Figure 4: Ok, now I see that you actually show all locations on a map, maybe add an overview map after all (without any data plotted on top; maybe combined with Tab. 2)? Also please highlight the labels in Fig. 4, they are partly unreadable (e.g., bold + white buffer around the edge of the letters)*

    We introduced a regional map with all used measurement sites in Fig. 2 of the revised version of the paper (as shown in Fig. 1 of this reply).

15. *L365-405: How exactly did you use the data in Fig. 4 to optimize the regions shown in Fig. 5?*

    We removed 'optimize' from the sentence 'Based on the frequency distribution shown in Fig. 4 and on the availability of CO_2 ground-based measurements in the region of the Asian monsoon and in the tropics in 2016 to 2017 an optimised regional mask was developed where different BL regions (Fig. 5) are defined.'

    Based on the frequency distribution shown in Fig. 4 and on the limited availability of CO_2 ground-based measurements in the region of the Asian monsoon and in the tropics in 2016 to 2017 a regional mask was developed where different BL regions (Fig. 5) are defined.

16. *Figure 5: Where are the regions mNH and cNH in Fig. 5? It took me a while to realize that you separate between land and ocean in the North, but not in the South. Maybe you could add the labels for mNH, cNH, tSH, and Wpool also in the figure? E.g., in slightly darker colors as the colors used to mark the regions, to set them apart from the measurement sites.*

    We agree and and add a legend to Fig. 5 in the revised version of the paper (as shown in Fig. 3 of this reply).

17. *L452-453: Maybe these two sentences can be combined, e.g., "In Fig.7, CO2 mixing ratios reconstructed in this way are shown as median of 1 K intervals for several measurement sites."*

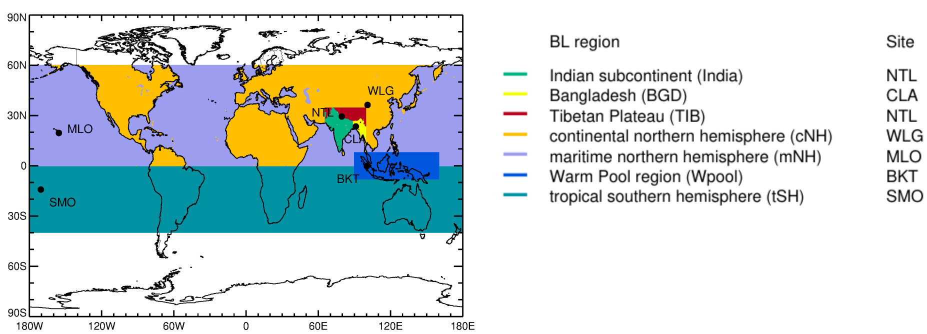

Figure 3: **Regional mask to reconstruction CO_2.** Regional mask to reconstruction CO_2 using CO_2 ground-based measurements at different sites in Asia and the Pacific. In each model boundary layer (BL) region (marked by different colours) CO_2 is prescribed from one specific measurement site: tropical southern hemisphere (tSH) by Samoa (SMO), Indian subcontinent (India) by Nainital (NTL), Bangladesh (BGD) by Comilla (CLA), Tibetan Plateau (TIB) by Nainital (NTL), maritime northern hemisphere (mNH) by Mauna Loa (MLO), continental northern hemisphere (cNH) by Mt. Waliguan (WLG) and Warm Pool region (Wpool) by Bukit Kototabang (BKT).

Thanks, done.

18. *L541-545: Here you discuss the sensitivity on the trajectory length and refer to a discussion in the supplement. However, looking at Fig. S1 and reading this discussion, it is not obvious to me why the length from 1 Dec 2016 is the best. The case S2a seems to improve with increasing length in the higher altitudes and for the 1 Jun 2016 case the fit between simulated and observed profiles is already quite good. Also the uncertainties in the higher altitudes are still smaller then when adding the GOSAT data. What happens if you go back even further? How many particles are you using for your simulations?*

    Many thanks for this question. The selection of the trajectory length is an important point. Therefore, we revised the Supplementary material as well as the paragraph (L541-545) in the main paper as follows:

    The sensitivity of the quality of the reconstruction of CO_2 (case S2b) on the employed trajectory length was tested. They can be too short (and thus miss contributions from the model BL) or too long (resulting in higher uncertainties). The longer the back-trajectory calculations the higher the altitudes of the end points of the trajectories from the free atmosphere. Based on the latter trajectories CO_2 is reconstructed from GOSAT-L4B data that are providing CO_2 values up to 10 hPa. The longer the trajectories the more the altitudes of the end points exceeds the altitude of the pressure level of 10 hPa and the CO_2 values are here extrapolated to higher pressure levels which increases the uncertainties of reconstructed CO_2 (see Supplementary (Fig. S1) for a detailed discussion on this issue). We decided to show back-trajectories to 1 December 2016, because for this date up to 410 K reconstructed CO_2 is determined solely by CO_2 prescribed at the model BL and by the transport of air parcels along the back-trajectories. Here, the uncertainties regarding the CO_2 extrapolation to higher pressure levels are negligible.

    If we would expand the CLaMS back-trajectory calculations to longer than a year the uncertainties in the CO_2 reconstruction for the free atmosphere from GOSAT-L4B data would probably increase further. To avoid this increase, we prefer to do global 3-dimensional CLaMS simulations driven by ERA5 including irreversible mixing (i.e. forward calculations) over several years ($\sim$ 4-6 years) using CO_2 ground-based measurements and a initial global CO_2 distribution. This would have the advantage that we could simulate the entire CO_2 profile without using GOSAT-L4B for the stratospheric background. This kind of simulation would be however, beyond the scope our the manuscript submitted to Communications Earth & Environment, but such a simulation will be the next step of our work.

Finally, the number of trajectories is added under Methods in the revised version of the manuscript as follows: Overall ∼110000 back-trajectories are calculated between 9000 and 16000 per flight depending on the flight lengths.

19. *L655: Please add an explanatory sentence on this AirCore sampling. You give a reference, but it would be nice to give one sentences on what makes it cheaper than other methods. Is it used operationally anywhere?*

    We added an explanation to AirCore measurements:

    We conclude, the quantification of CO_2 and other GHG surface fluxes and their temporal changes would highly benefit from an expansion of the ground-based GHG measuring network in South Asia complemented by regular vertical CO_2 soundings, which could be achieved at comparatively low cost by AirCore sampling and subsequent laboratory analysis (a method requiring only moderate instrumentation collecting air in a very long lightweight stainless-steel tube, usable on a variety of platforms including small balloons) (Karion et al., 2010).

    The AirCore sampling technique is used by different groups (e.g. Karion et al., 2010; Engel et al., 2017; Laube et al., 2020), but to our knowledge not operationally so far.

20. *Availability of data and materials: The used HAGAR CO2 and N2O data and the ground-based observations should be made openly and permanently available by uploading it to a repository like https://zenodo.org/ to receive a DOI.*

    Many thanks for this reminder. We are aware of this data policy and will upload the final HAGAR data on a public repository before the paper will be published.

21. *The ClaMS settings used to simulate the back-trajectories (at least examples for a few simulations to be able to reproduce) should be made openly and permanently available by uploading it to a repository like https://zenodo.org/ to receive a DOI.*

    https://www.nature.com/nature-portfolio/editorial-policies/reporting-standards

Many thanks for this reminder. We are aware of this data policy and will upload the final CLaMS data on a public repository before the paper will be published.

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Author Comment to Referee #2

Communications Earth & Environment (COMMSENV-22-0506-T), 'Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region' by B. Vogel et al.

We thank Referee #2 for the positive review and for further guidance on how to revise our manuscript. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics. Passages from the revised version of the manuscript are shown in blue.

Major Comments:

The subject of this paper is the creation of high vertical resolution data by using the aircraft in situ measurement data in the Asian monsoon region, which has few ground stations but is important as a source of CO2 emissions. The motivation for the research, the originality of the data used, and the quality of the data are guaranteed. It is expected that high-vertical resolution data will be effectively used for top-down research on CO2 emission data. However, this reviewer shows a few points to consider which should be cleared before publish.

1. *Representativeness : First, how representative is the high vertical resolution data on a limited number of observation points? Subsequent analysis (an inversion method for estimating emissions) using data constructed from data from a few points in a region where CO2 has high variation may cause miss-leading.*

   First of all, we agree that the number of data used in our study is limited which is caused by the fact that CO_2 and N_2O high-resolution profiles measured during StratoClim are the only available in situ measurements up to 20 km altitude in the region of the Asian monsoon up to now. Further, in addition the number of ground-based observation sites of CO_2 and N_2O over the northern Indian subcontinent is rather limited, therefore we conclude, that the quantification of CO_2 and other GHG surface fluxes and their temporal changes would highly benefit from an expansion of the ground-based GHG measuring network in South Asia. We agree, that a CO_2 reconstruction using a few observational sites could cause uncertainties, however we demonstrate, that if measurements in Nainital and Comilla were operationally available, it would be possible to reconstruct CO_2 profiles over the northern Indian subcontinent much more reliably. This is a conclusion of our paper, which is in agreement of the suggestion of the reviewer.

2. *Uncertainty of model : The discussion of the uncertainty of the numerical model should be added. While the introduction shows errors and uncertainties in the numerical model, an important part of this dataset is the use of the Lagrange model. Although using more realistic data than traditional transport models, the uncertainty is likely to be high even in transport processes using diabatic heating, especially in the Asian troposphere, where water vapor and cloud variability are severe. Especially in the UTLS (upper troposphere/lower stratosphere) region, the transport process across the tropopause, including the convective parameterization scheme, has a large variation between reanalysis. In addition, normally, the back trajectory in the troposphere is limited to one week, but in this study, it is calculated for one year or more. Even though aged air is more than a year old in the stratosphere, transport within the middle world is weeks to months, especially in the summer of the Asian monsoon region. Therefore it is doubtful how realistic the reconstructed data are especially near the tropopause (higher aircraft observation).*

   Many thanks for this comment. We agree that the uncertainties of the numerical models should be better discussed. We added the following paragraph to the introduction of the revised version of the manuscript:

   In state-of-the-art chemistry transport models, the transport of air parcels differs because different methods (Eulerian, Lagrangian), different vertical velocities (kinematic, diabatic) and different meteorological reanalyses (e.g. ERA5, ERA-Interim, JRA-55) are used to drive the models (e.g. Bergman et al., 2013; Ploeger et al., 2019). Further, the implementation of convection and irreversible mixing differs from model to model.

   We agree that in general, trajectory calculations have limitations due to trajectory dispersion depending on the trajectory length. However, the frequently employed trajectory length to study transport processes in the Asian monsoon region ranges from a couple of weeks to several months depending on the transport times from Earth's surface to atmospheric altitudes (e.g. Chen et al., 2012; Bergman et al., 2013; Vogel et al., 2014; Garny and Randel, 2016; Li et al., 2017; Hanumanthu et al., 2020). Further, CLaMS diabatic Lagrangian back-trajectory calculations driven by ERA5 reanalysis used in our study, are assessed statistically, which means the reconstruction of CO_2 is done for a set of trajectories in potential temperature intervals of 1 K.

3. *Validation : Third point, which is related with second point, is how to validate reconstructed data. Since the reconstructed data have already high quality, it is key how to verify the bias/error of Lagrangian transport and the upper layer data, although the verification work is done by only GOSAT-L4B data as the sensitivity experiment. (Please check vertical transport tendency of NIES-TM driven by JRA-25, which is old version in JRA.) It is not enough to me because there is a large difference depending on the reanalysis data that drives the numerical model, especially around the tropopause, so it is recommended that you to carry out verification using other reanalysis data, JRA55 and MERRA2. If the tendency is the same, the reader will be satisfied the results. Further, is it possible to apply it to other aircraft observation data, like CONTRAIL and CARIBIC?*

   Many thanks for this comment. To reconstruct the upper part of the HAGAR CO_2 profile, we need a global CO_2 distribution of the stratospheric background (otherwise we had to calculated CLaMS back-trajectories over several years using our approach) including a good representation of CO_2 over Asia. The data availability of global CO_2 data suitable for our study, is also very limited. We are aware that the transport in GOSAT-L4B data is not optimal, however, this was the best global CO_2 information we could find. We agree with Referee #2's comment, that it would be better to use JRA55 reanalysis to drive NIES-TM instead of JRA-25. However, that is a task for the Japan Aerospace Exploration Agency (JAXA) providing GOSAT-L4B data. We can not perform this work.

   We added a comparison of HAGAR CO_2 profiles with CarbonTracker and GOSAT-L4B data in the revised version of the supplement (see Fig. S3). We demonstrate that CarbonTracker performs poorly over the Northern Indian Subcontinent. GOSAT-L4B data are somewhat better as CarbonTracker, but still worse than the reconstruction with CLaMS back-trajectories presented in our paper. This supports that CO_2 profiles over Asia are purely described in global 3-dim data sets in particular in Eulerian model calculations.

   Regular commercial airliner measurements programs such as CONTRAIL and IAGOS-CARIBIC are also very interesting, however, their maximum altitude is only up to ∼12 km (∼180 hPa) (e.g. Schuck et al., 2010; Patra et al., 2011; Sawa et al., 2012; Umezawa et al., 2018) compared to the 20 km altitude of the StratoClim data that we are using here (see introduction of our manuscript). Further, CONTRAIL measurement are focused on the region around Japan and the western Pacific and only a few IAGOS-CARIBIC flight paths from Europe to South Asia cross the northern Indian subcontinent.

Minor Comments:

- *L223-226, N2O in Figure1: I couldn't get the seasonality that the authors said. Please explain a little more carefully.*

  We agree with Referee #2's comment and revise the sentence 'N_2O mixing ratios in Nainital and Comilla are higher in summer (June–August) and winter (November–February) compared to sites in the Pacific (Mauna Loa, Cape Matutala) which coincides with the application of nitrogen fertiliser, biomass burning and change in monsoonal/trade winds (Nomura et al., 2021; Patra et al., 2022).'

  The seasonal variability of N_2O on the northern Indian subcontinent is consistent with the application of nitrogen fertiliser, biomass burning and change in monsoonal/trade winds (Nomura et al., 2021; Patra et al., 2022). Therefore, the N_2O mixing ratios in Nainital and Comilla are in general higher compared to sites in the Pacific (Mauna Loa, Cape Matutala; see Fig. 1c).

- *L227-229: related with major comment #1, is it okay to say the representativeness of a vast area with only two points?*

  We agree with Referee #2's comment and revise the sentence 'Higher N_2O emissions are found in the eastern Indo-Gangetic Plain, including Comilla, compared to the western Indo-Gangetic Plain influencing Nainital (Nomura et al., 2021).' as follows:

  Higher N_2O values are found in Comilla located in the eastern Indo-Gangetic Plain compared to Nainital located in the western Indo-Gangetic Plain (Fig. 1c).

- *L240-242 : Please add the explanation why the amplitude of GOSAT-L4B is smaller than in situ measurement for general readers.*

  Many thanks for this comment. We revised the paragraph as follows:

The GOSAT-L4B product is a model simulation using CO_2 surface fluxes inferred from column-averaged satellite measurements (details see Methods); the lowest model level of GOSAT-L4B is closest to the inferred CO_2 surface fluxes and is not strongly influenced by the tracer transport of the underlying transport model. The GOSAT-L4B mean value has a similar seasonal variability as other CO_2 ground-based measurements on the northern hemisphere (Mt. Waliguan, Nainital, Mauna Loa), however its amplitude is lower than for the ground-based measurements in Nainital demonstrating the limitations of GOSAT-L4B data compared to in situ measurements.

- *L324-347, Figure4: Please add the distribution of age of air from BL at the flight altitude/lower stratosphere, which is helpful to understand the transported length and distance of source deeply.*

  We are not completely sure if we understand Referee #2's comment right. In Fig. 4 of our manuscript, in each grid box the number of the air parcels originating in the model boundary layer in this grid box are shown. We could replace the number of air parcels in each box with the mean age of air (inferred of the trajectories from the model boundary layer), however that would be a completed different additional figure. And of course a certain information on mean age of air is given through the different panels of Fig. 4 giving different time periods. Maybe there is a misunderstanding? However, to give the reader an additional time information about the age of air for each panel in Fig. 4 of our manuscript, we added the age of air in Tab. 1 in the revised version of our manuscript (Tab. 1 of this reply).

- *L355: why the latitude gird is 0.6 degree, not 0.5 degree?*

  Many thanks for this comment. In Fig. 4 the ratio between longitude (x-axis) and latitude (y-axis) is not 1:1, therefore the grid is adjusted. Further we found a typo in the grid size and revised the sentence as follows:

  The frequency distribution is calculated in longitude-latitude bins of $2.0° \times 1.5°$.

- *L430: Add the 'Asian monsoon' words to make it easier for the reader to read.*

  done: At the top of the Asian monsoon anticyclone ...

season	time period	start time	age of air
monsoon 2017	June–September 2017	1 June 2017	$\sim$ 2 months
pre-monsoon 2017	March–May 2017	1 March 2017	$\sim$ 2–5 months
winter 16/17	December 2016 – February 2017	1 Dec 2016	$\sim$ 5–8 months
post-monsoon 2016	October–November 2016	1 Oct 2016	$\sim$ 8–10 months
monsoon 2016	June–September 2016	1 June 2016	$\sim$ 10–14 months
aged air	older than 1 June 2016		$>$ 14 months

Table 1: **Time periods and age of air of considered seasons on Indian subcontinent.** The analysis of CLaMS back-trajectories (see Methods) is performed back until the start time of each season. For each season air parcels that were released at the model boundary layer (BL) are analysed. The longest simulation time is back until 1 June 2016 ($\sim$ one year). Air parcels that are in the free atmosphere on 1 June 2016 are considered as aged air.

- *L507-524, Figure 7: It is difficult to divide the similar color, HAGAR and B. Kototabang.*

  We changed the color for the data from B. Kototabang in Fig. 7 as well as in Fig. 1 of the revised version of the paper.

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Author Comment to Referee #3

Communications Earth & Environment (COMMSNV-22-0506-T), 'Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region' by B. Vogel et al.

We thank Referee #3 for the positive review and for further guidance on how to revise our manuscript. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics. Passages from the revised version of the manuscript are shown in blue.

This is a creative analysis of a unique dataset over the poorly observed Indian subcontinent that provides insight into surface influences on air in the mid/upper-troposphere and lower stratosphere during the Indian monsoon season. The success of the CLaMS model reconstruction of the profiles up to 380/400K is remarkable. However, the paper lacks important context from previous work, and so I am recommending a significant revision. I think this can be a truly excellent paper with just a bit of reframing and potentially some minor but important additional scientific analysis.

We thank Referee #3 for the positive review. We certainly have improved the context of previous work in the revised version of our manuscript.

Overarching comments/concerns:

The paper presents a model analysis of CO2 measurements over the Indian subcontinent during the Asian monsoon season. Aircraft vertical profiles were obtained in the Northern subtropics between 20N and 30N. This is outside of the "deep tropical upwelling region", and near the southern border of what has been referred NH extratropical surf zone. The profiles extend from near the surface and well into the stratosphere (potential temperature 300K - 480K). There is a large body of work on upper troposphere lower stratosphere (UTLS) transport that provides important context for interpetting the observed vertical structure, but the current manuscript lacks any significant discussion of this prior work.

Global-scale stratospheric transport includes two regimes, the stratospheric "overworld" corresponding to potential temperature surfaces greater than 380K and *the "lowermost extratropical stratosphere", which is also known as the stratospheric "middleworld" (Holton et al., 1995). In the overworld, isentropes are contained wholly within the stratosphere, whereas in the lowermost stratosphere the isentropes are above the tropopause in the extratropics and below the tropopause in the deep tropics. Thus, in the lowermost stratosphere (potential temperature < ∼380-400K) rapid mixing can occur between airmasses characteristic of the upper tropical troposphere and of the mid-latitude lower stratosphere. In the overworld, no such rapid mixing of tropospheric and stratosphere can occur. The boundary between the overworld and the lowermost stratosphere shows up clearly in Fig 6a and in Fig 8a.*

We agree with Referee #3 that there is a large number of publications related to upper troposphere lower stratosphere (UTLS) transport, however the number of references in Communications Earth & Environment is restricted to 70. Therefore, we have added the context of previous work, but we had to select carefully the references cited in our publication to not exceed the number of 70 (more details see below).

The paper by Holton et al. (1995) is very famous regarding upper troposphere lower stratosphere transport. They discuss the transport from the troposphere into the stratosphere in the framework of the general circulation – in a more conceptual way – not including the transport from the troposphere into the stratosphere via the large monsoon systems (e.g. Asian and American monsoon). However, we agree with Referee #3 that the transport from the troposphere into the stratosphere via the Asian summer monsoon is poorly described in our submitted manuscript and therefore we added the following paragraph (to the introduction) in the revised version of the manuscript for further clarification:

From about June to September, the Asian summer monsoon constitutes a seasonally persistent zonally restricted circulation pattern transporting climate-relevant emissions rapidly from the surface boundary layer to greater altitudes, i.e. to the lower stratosphere (e.g. Mason and Anderson, 1963; Randel and Park, 2006; Park et al., 2007; Vogel et al., 2015, 2019). The Asian summer monsoon is associated with deep convection over the Indian subcontinent and an anticyclonic flow in the upper troposphere and lower stratosphere (UTLS) over the Asian monsoon region spanning from northeast Africa to the Pacific (e.g. Park et al., 2007). Air parcels are uplifted quickly by convection followed by slow diabatic uplift in the UTLS superimposed by the anticyclonic flow, while in other regions within the tropical transition layer the heating rates are in general smaller during boreal summer (Vogel et al., 2019). The higher the air parcels are located above the level of maximum convective outflow ($\approx 360\,\mathrm{K} \approx 13\,\mathrm{km}$), the larger the contribution of air masses is from outside the Asian monsoon anticyclone (i.e. from the stratospheric background) to the upward spiraling flow (Vogel et al., 2019).

The CLaMS model has been carefully evaluated and seems to do a credible job of capturing air mass transport in these two transport regimes, and panels 6 b-d provide interesting insight into how tropospheric from the NH, SH and Warmpool areas airmasses propagate into the stratospheric overworld. The paper would benefit from some discussion along these lines (e.g. NH and SH contributions are roughly equal; the Warm Pool signature can be seen in panels 6c-3, but is it's contribution commensurate with it's area or does the warm pool account for a disproportionately large fraction of the air in the overworld because of the strong convection in that region?)

We agree that CLaMS driven by high-resolution ERA5 reanalysis does a credible job of capturing air mass transport in the region of the Asia monsoon anticyclone. We added a short discussion related to the NH and SH contributions in the revised version of our manuscript as follows:

After a simulation period of $\sim$14 months (until 1 June 2016) the contributions from the tropical southern hemisphere and the maritime northern hemisphere are roughly equal in the lower stratosphere.

Also lacking from the current paper is any discussion of stratospheric transport in the stratospheric overworld. There are many relevant papers about this, a few of which are listed below. The rapid decrease of N2O above 380-400K is indicative of mixing of relatively young air with aged stratospheric air that has descended from much higher altitudes where the photochemical lifetime of N2O is short (photochemical loss at altitudes corresponding to 400-470 K is too slow to explain rapid decrease with altitude). The Boering et al, 1996 (Science) and both Andrews et al. 2001 (JGR) papers below show that CO2 and N2O relationships are remarkably consistent throughout the lower stratospheric overworld, once the tropospheric growth of both species is taken into account and if the CO2 stratospheric boundary time series is convolved with a reasonable representation of the age spectrum (e.g. Andrews et al., 2001a, b; Ray et al. 2017). The discrepancy between the modeled an observed CO2 profiles in Figure 8a is clearly due *to the mixture of aged stratospheric air with younger air. Age spectra in the lower stratosphere have been convincingly shown to be asymmetric with a strong peak indicating young air (consistent with Fig 6 in the current manuscript!) and a tail indicating the presence of older air.*

We agree that CO_2 is a useful tracer to estimate stratospheric mean ages as discussed in the literature. We thank Referee #3 for the proposed references and added several of them:

CO_2 is chemically inert in the troposphere and stratosphere and can be used as an age tracer considering time periods of several months (e.g. Boering et al., 1996; Andrews et al., 2001; Ray et al., 2022).

We agree that the rapid decrease of N_2O above 380-400K is indicative of mixing of relatively young air with aged stratospheric air that has descended from much higher altitudes where the photochemical lifetime of N_2O is shorter. We added the following selected refrences to the revised version of the manuscript.

N_2O is essentially inert in the troposphere and has no significant sinks at the surface of the Earth. The reduction of N_2O in the lower stratosphere occurs via photolysis and reaction with excited atomic oxygen ($O(^1D)$). The decrease of measured N_2O profiles above 400 K potential temperature (Fig. 3) indicates mixing with older stratospheric air that has descended from higher altitudes (Boering et al., 1996; Andrews et al., 2001). The high-resolution CO_2 and N_2O vertical profiles up to 20 km altitude presented here yield a unique insight into their altitude dependency in the region of the Asian monsoon.

It would be very interesting to explore the extent to which the upper portion of the HAGAR profiles in Fig 8a could be reconstructed as follows:

(1) computing the Age:N2O relationship from equation 3 of Andrews et al. 2001 (https://doi.org/10.1029/2001JD000465) after scaling N2O to account for the difference between tropospheric avg N2O summer 2017 (329.9 ppb) in 1997 (313ppb) and in 2017 (i.e. N2O[1997] = N2O[HAGAR](313ppb/330ppb))*

We computed the mean age of N_2O using equation 3 by Andrews et al. (2001) :

$$mean\_age = 0.0566 * (313. - N2O[1997]) - 0.000195 * (313 - N2O[1997])^2.$$

a shown in Fig. 1 of this reply. Further, the correlation by Andrews et al. (2001) is shown for N_2O mixing ratios adapted to 2017 using

$$N2O[1997] = N2O[HAGAR] * (313ppb/335ppb).$$

In addition, the mean age is calculated using a correlation by Engel et al. (2002) which is based on measurement from 1997 and 2000.

$$mean\_age = 6.03 - 0.0136 * N2O[1997] + 8.5892 * 10^{-5} * N2O[1997]^2 - 3.376968 * 10^{-7} * N2O[1997]^3$$

The correlation by Engel et al. (2002) is also adapted to N_2O mixing ratios from 2017. Further the age for each N_2O measurement is shown derived from CLaMS back trajectory calculations driven by ERA5 in Fig. 1 of this reply.

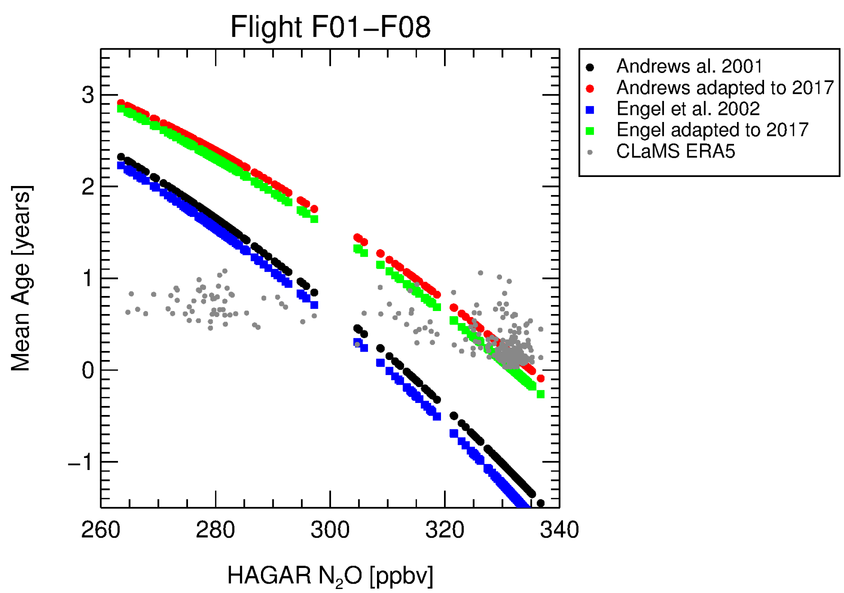

Figure 1: Mean age from Andrews et al. (2001) and Engel et al. (2002) and age derived from CLaMS backward trajectories using ERA5. Only trajectories reaching the model boundary layer until 1 June 2016 are considered here and only N_2O measurements above 380 K are shown.

(2) Use a method like Ray et al., 2017 to convolve a timeseries of CO2 representative of the stratospheric boundary condition at the tropical tropopause (which

can be approximated using the average of Mauna Loa and Samoa data lagged by 2 months) with a set of reasonable age spectra having mean ages corresponding to the age profile obtained in step 1 (i.e. this is applying the method described in Ray et al. 2017 but inverted to solve for CO2 as a function of Age instead of Age as a function of CO2). The resulting computed CO2 profile could be directly compared to the Hagar CO2 observations above 400 K and would provide insight into Age:CO2:N2O relationships in the stratospheric overworld circa 2017 compared with the previous work.

We have considered the suggestion in the review in depth and we agree such an analysis would contribute substantially to the understanding of the N_2O/age/CO_2 world. We show in Fig. 1 of this reply both the Age/N_2O relation by Andrews et al. (2001) (as suggested in the review) and the Age/N_2O relation reported by by Engel et al. (2002). We show both the original (1997) data and the data shifted to 2017 (as suggested in the review). The relations are very similar, but differ by about a month in age in the lower stratosphere. In addition, we show the age inferred from CLaMS backward trajectory calculations driven be ERA5 demonstrating the large variability of the age in the lower stratosphere of a several months in the lower stratosphere.

Assuming a CO_2 growth rate of 2.4 ppm/year, a difference of a few month in age corresponds to a difference in CO_2 which is not insignificant. Given further uncertainties in an CO_2/age relation (which is difficult to find – could not easily find literature on the subject), it is not straightforward to deduce a CO_2 profile from such an analysis (which could be compared with the StratoClim observations). In summary, while we agree that the suggestion in the review is very promising, we have to defer work in this direction to a future project and cannot include such an analysis in the present paper.

*Finally, it would be *very* useful to show how CarbonTracker and other available CO2 models (there are many e.g. from the OCO-2 MIP) that have been optimized against observations (e.g. The EU Copernicus CAMS product and the suite of "OCO2 MIP models" presented by Crowell et al., 2019, Atmos. Chem. Phys., doi: 10.5194/acp-19-9797-2019) compare with the HAGAR profiles. The HAGAR profiles are in an especially important and poorly sampled region. I would not necessarily expect these models to perform well at all in this severely underconstrained region, and that is really important to highlight in the literature to strengthen the argument for more vertical profile data in undersampled regions. I also expect these models may perform poorly in the stratosphere because they* *have mostly been optimized for tropospheric transport (i.e. native model levels left out at high altitudes to speed computation, etc), but if they are performing poorly in the stratosphere then that has *major* implications for possible biases when assimilating satellite data. CO2 is a terrific tracer of stratospheric transport and so even a simple model evaluation could provide a lot of insight. It would be nice to include a figure in the supplement or the main paper showing how well the individual OCO2 MIP model simulations do or do not agree with the HAGAR profiles.*

Many thanks for this comment. We agree that the HAGAR profiles are especially important in the poorly sampled region over the Indian subcontinent. We added a comparison of HAGAR CO_2 profiles with CarbonTracker and GOSAT-L4B data in the revised version of the supplement (see Fig. S3). We demonstrate that CarbonTracker performs poorly over the Northern Indian Subcontinent. This result supports our main message of the paper, that we need more CO_2 measurements over the Indian Subcontinent. Further, we would like to point here out that the HAGAR measurements will be made available following the publication of this paper. Hopping that the data will be of help for model evaluation and future model developments.

Relevant publications cited above and a few others about CO2 in the UTLS that should perhaps be considered for inclusion in the intro or discussion:

- *Holton et al., 1995, Stratosphere-troposphere exchange, Reviews of Geophysics, https://doi.org/10.1029/95RG02097*
- *Xueref et al., 2004, Combining a receptor-oriented framework for tracer distributions with a cloud-resolving model to study transport in deep convective clouds: Application to the NASA CRYSTAL-FACE campaign, GRL, 2004, 10.1029/2004GL019811*
- *Boering et al., 1995, MEASUREMENTS OF STRATOSPHERIC CARBON-DIOXIDE AND WATER-VAPOR AT NORTHERN MIDLATITUDES - IMPLICATIONS FOR TROPOSPHERE-TO-STRATOSPHERE TRANSPORT, JGR, DOI: 10.1029/95GL02337,*
- *Boering et al., 1996, Stratospheric mean ages and transport rates from observations of carbon dioxide and nitrous oxide, Science, 10.1126/science.274.5291.1340*

- *Park, S et al., 2007, The CO2 tracer clock for the Tropical Tropopause Layer, ACP, DOI10.5194/acp-7-3989-2007*
- *Andrews A et al., 2001, Empirical age spectra for the midlatitude lower stratosphere from in situ observations of CO2: Quantitative evidence for a subtropical "barrier" to horizontal transport, JGR, 10.1029/2000JD900703*
- *Andrews et al., 2001, Mean ages of stratospheric air derived from in situ observations of CO2, CH4, and N2O, JGR, 10.1029/2001JD000465*
- *Ray et al., 2017, Quantification of the SF6 lifetime based on mesospheric loss measured in the stratospheric polar vortex, JGR, doi:10.1002/2016JD026198*

We are grateful to Referee #3's proposed references and added a selection of them into the revised version of the manuscript. We have to consider that the number of references in Communications Earth & Environment is restricted to 70.

Specific Comments:

1. *Suggestion to remove or greatly reduce verbiage about N2O from abstract and intro, since there is actually very little analysis of the N2O profiles in the current version of the paper.*

   We prefer to keep the discussion about N_2O in the abstract and introduction as in the submitted version. We added a Figure showing the reconstruction of N_2O (Fig. 7b of the revised version of the manuscript) to enhance the role of N_2O within the paper (details see below).

2. *CLaMS driven by ERA-5 seems to be ideally formulated for reproducing mid/upper troposphere profiles in the NH subtropics. Is it expected to work as well elsewhere?*

   ERA5 is the newest reanalysis provided by ECMWF. There are already a few case studies using CLaMS driven by ERA-5 regarding different atmospheric regions and using different versions of ERA5 (e.g. Hoffmann et al., 2019; Li et al., 2020; Ploeger et al., 2021; Konopka et al., 2022). ERA5 has a better representation of convective updrafts, gravity waves, tropical cyclones, and other meso- to synoptic-scale features of the atmosphere attributed to its better spatial and temporal resolution compared to ERA-Interim reanalysis. However, the vertical transport in the upper troposphere and stratosphere is somewhat slower compared to ERA-Interim. Further studies are necessary to demonstrate the strength and weakness of CLaMS simulations driven by ERA5 reanalysis in different atmopsheric regions.

3. *The abstract and intro include a lot of verbiage about N2O emissions, but the paper is really about CO2 as reflected in the title. I suggest removing mention N2O from the abstract and limiting discussion in the intro, since it's really just functioning as a tracer of stratospheric air. Although reframing as suggested above might lead to other changes in these sections such that N2O has a larger role in interpreting the stratospheric overworld data.*

   We added a Figure showing the N_2O Reconstruction (Fig. 7b of the revised version of the manuscript) to enhance the role of N_2O within the paper (details see below).

4. *Line 640: I agree that the rapid ventilation up to 14 km is consistent with significant influence from India and Tibet, but I think stating that the influence of asia and india can be discerned up to 22km is an overstatement. At those highest altitudes, the signature of surface emissions is tho*

   We added a sentence in the revised version of the manuscript for clarification:

   Our study shows that spatio-temporal patterns of CO_2 in India and Tibet driven by regional flux variations are readily ventilated to at least 14 km during the Asian Monsoon and can be discerned up to 20 km as they ascend further into the stratosphere. However in the stratosphere, the fraction of air originating in India and Tibet is low compared to contributions from the tropics and of aged air from the stratosphere.

5. *Figure 1 shows that BKT has lower values than MLO, WLG and other stations, suggesting that BKT is impacted by local CO2 uptake by vegetation. Some explanation about why CO2 at BKT is so much lower than the other sites is needed. In contrast, MLO, WLG, and SMO are regionally representative records. Due to it's high elevation (nearly 4km above sea level), MLO is representative of the free troposphere rather than the marine boundary layer. I recommend evaluating the extent to which BKT is regionally representative by considering the NIES CONTRAIL dataset of measurement from commercial aircraft flights. There are many vertical profile data in the vicinity of Indonesia. However, the contribution of the Warm Pool region is rather small and likely does not contribute significantly to the reconstructed*

profile. I would venture to guess that the BKT timeseries could be replaced with either SMO or MLO or the average of the two with minimal impact on the reconstructed CO2 profile. I'm not sure the success of the method is dependent on treating the Warm Pool as a separate region. If so, then that would be an interesting result worth highlighting a bit more.

Many thanks for this important comment. We did the same CO_2 reconstruction as presented in the manuscript (Fig. 2 (left) of this reply), but without the Warm Pool region (Wpool). We divided the Warm Pool region into the northern and southern hemispheric part and used ground-based measurements from SMO, MLO and WLG as indicated in Fig. 2 (right) instead of using ground-based data from Bukit Kototabang (BKT) (Fig. 2 (left)). The CO_2 reconstruction is shown for trajectories until the monsoon season 2016 (1 June 2016) to show the entire simulation period. The decrease in measured CO_2 between 400 K and 420 K is better represented in the CO_2 reconstruction considering the Warm Pool region by using low CO_2 ground-based measurements from Bukit Kototabang (BKT). Because of this impact of the Warm Pool region on the CO_2 reconstruction, we keep the regional mask as in our submitted version of the manuscript. However, at altitudes above 420 K reconstructed CO_2 is somewhat closer to the HAGAR measurements without considering the Warm Pool region (Fig. 2 (right). However at this altitudes also the uncertainty of reconstructed CO_2 is increasing cause by uncertainties of the stratospheric background inferred from GOSAT-L4B data.

To to hightlight these results a bit more, we added the following sentence to the revised version of our manuscript:

Air from the boundary layer above 400 K originates mainly in the southern and northern ITCZ. Extreme low CO_2 values from ground-based measurements in the Warm Pool region have to be taken into account to reconstructed CO_2 in this altitude range.

6. *Regarding the use of surface CO2 observations from the World Data Center for Greenhouse Gases. I think they include a fair use statement and metadata requesting that the actual data providers be contacted about how they should be acknowledged in publications. The WDCGG functions as a clearinghouse for datasets, they do not directly provide observations. Please look at the metadata and consult the actual data providers about how they would like to be acknowledged. If the data are from NOAA's*

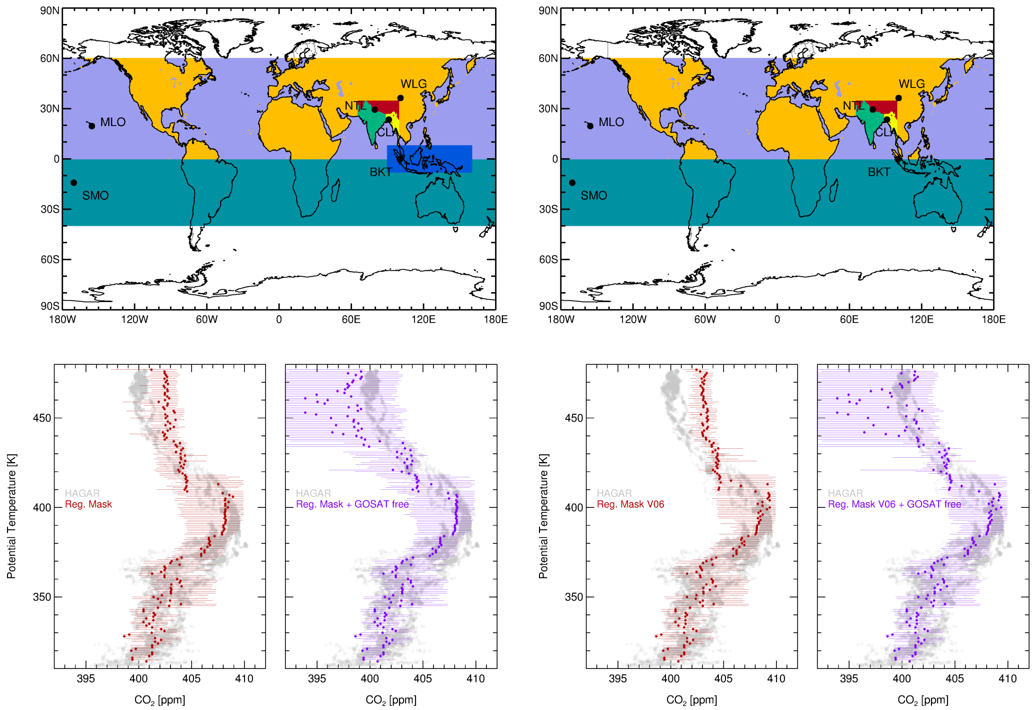

Figure 2: **Regional masks and** CO_2 **reconstruction.** CO_2 reconstruction as shown in the main paper (left) and without the Warm Pool region (Right; Regional Mask Version V06). The CO_2 reconstruction is shown for trajectories until the monsoon season 2016 (1 June 2016).

Global Greenhouse Gas Reference Network as I suspect, then please contact Xin.Lan@noaa.gov. She may also have some advice about the represtativeness of BKT data.

Many thanks for this hint. We checked already the data policy of WDCGG and considered that in the acknowledgement and cited references.

7. *Figure 3:There are a lot of black points at low altitudes that I would assume are the youngest particles. This is confusing since the caption says that aged parcels > 1 yr are shown in black. Are both the oldest and the youngest particles shown in black? Maybe the figure should be reworked so that the oldest parcels are shown in grey/black and the youngest are shown in color. The trajectories corresponding to the oldest air are likely the least reliable,*

so maybe not useful to show different colors for e.g. air > 365 days. This would allow color scale to be used to full effect in the portion of the profiles between 350-410K where the CLaMS reconstruction is most successful.

Many thanks for this comment. We agree, that Fig. 3 needs some modifications and revised the figures as shown in Fig. 3 of this reply.

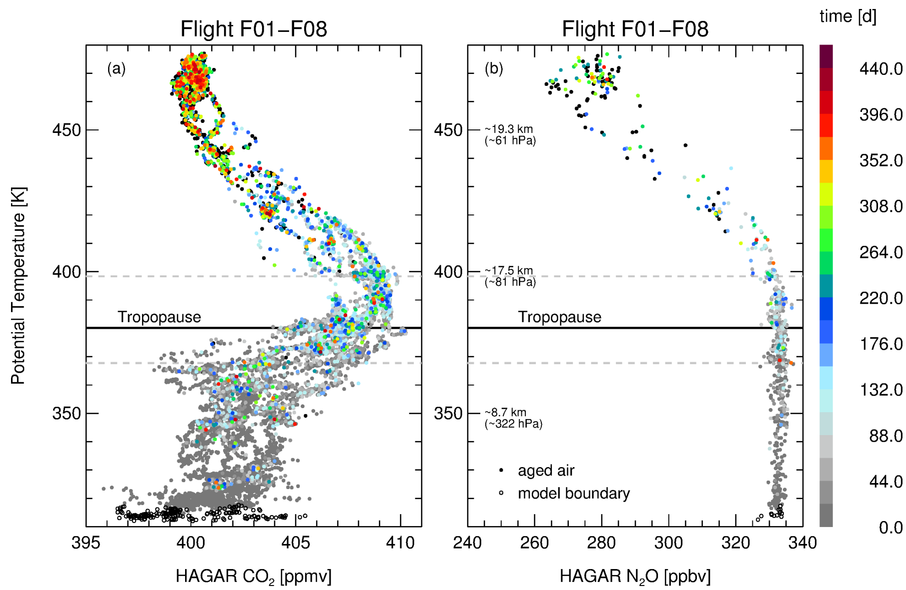

Figure 3: **Airborne** CO_2 **and** N_2O **measurements from the StratoClim campaign in Kathmandu (Nepal) during July and August 2017.** Each air parcel is coloured by the transport time from the model boundary layer (BL) to the time of measurements inferred by Lagrangian back-trajectory calculations. Air parcels located in the model BL as well as aged air (air located in the free atmosphere on 1 June 2016) are marked. The number of air parcels is determined by the different temporal resolution of the CO_2 **(a)** and N_2O **(b)** measurements (details see Methods). In addition, the mean WMO tropopause (Hoffmann and Spang, 2022) as well as the lowest and highest tropopause (grey dashed lines) over Kathmandu during the flight days are shown.

8. *Line 227: N2O emissions are found... What is the source of this information about the geographic distribution of N2O emissions? Inventory estimates?*

   We rephrased the sentence 'Higher N_2O emissions are found in the eastern Indo-Gangetic Plain, including Comilla, compared to the western Indo-Gangetic Plain influencing Nainital (Nomura et al., 2021).' as follows:

Higher N_2O values are found in Comilla located in the eastern Indo-Gangetic Plain compared to Nainital located in the western Indo-Gangetic Plain (Fig. 1c).

9. *Line 257: "CO2 is chemically intet in the troposphere and stratosphere." CO2 has a small source from CH4 oxidation in the stratosphere/mesosphere that is not completely negligible in the lower stratosphere. The contribution of CO2 from this source to the observed profile can easily be estimated from coincident measurements of CH4 (i.e. it is equivalent to the observed CH4 minus CH4 at 400 K).*

   Thank you for this comment. However, we have only CH_4 measurements for flight F05 until F08 with a much lower temporal resolution than the CO_2 measurements (see Fig. 4). Assuming 400 ppm CO_2 at 470 K and 0.35 ppm CO_2 from CH_4 oxidation ($\sim$ 1850 ppb-1500 ppb), we can estimate an contribution CO_2 from CH_4 oxidation of 0.09% at 470 K. This error is much lower, than the variability of reconstructed CO_2 by the uncertainties of the GOSAT-L4B data (see Fig. 3 of the revised supplement). Therefore, CO_2 from CH_4 oxidation plays a minor role for the CO_2 reconstruction approach we are using.

   We added the following sentence to the reviser version of the manuscript.

   The contribution of CH_4 oxidation in the stratosphere is estimated to be much lower (0.09% at 470 K) than the variability of reconstructed CO_2 in this altitude region, therefore, CO_2 from CH_4 oxidation is not considered in our approach.

10. *Line 297/298: I'm not sure what is meant by "most of the trajectories were released at the model BL much earlier". Perhaps it could be reworded something like: "Air parcel trajectories were calculated backward in time until June 2016 by which time approximately X% of the air parcels reached the model BL"*

    We rephrased the paragraph 'The trajectories are calculated back to 1 June 2016, although most air parcels were released at the model BL much earlier, and are analysed within different time periods. Further these back-trajectories are used to identify the source regions at the model BL depending on season (see Tab. 1). ' as follows:

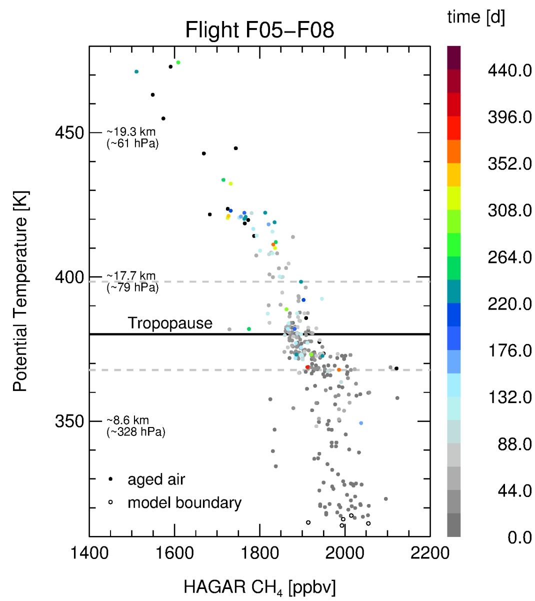

Figure 4: Same as Fig. 3, but for CH_4. However CH_4 measurements are only available for research flights F05-F08.

The trajectories are calculated back to 1 June 2016 and are analysed within different time periods to identify the source regions at the model BL depending on season (see Tab. 1). However, most air parcels were released at the model BL much earlier than 1 June 2016 (e.g. 64% of all air parcels are from the monsoon season 2017).

11. *Line 313: summery − > summary*

    done

12. *I would omit most of the N2O paragraph starting line 472 unless you add the analog of Figure 8a. It makes sense that the N2O profile above 400K cannot be reconstructed due to chemical loss, but what about the tropospheric profile? I understand that the profile is nearly vertical, but do you get the right tropospheric N2O value based on these sites? Or are all the tropospheric values essentially the same?*

    We aggree that it would be interesting to show also the reconstruction of N_2O (Fig. 5) and add it as Fig. 7b to the revised version of the paper.

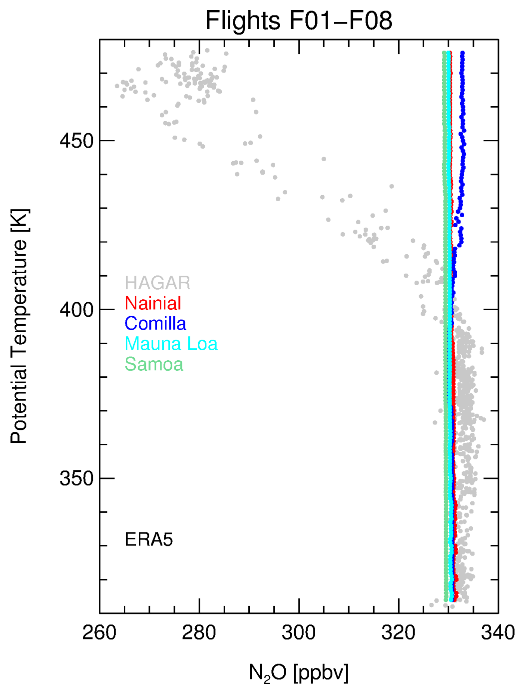

Figure 5: N_2O airborne-measurements and reconstructed N_2O (case S1). Similar as Fig. 7 from the submitted version of the paper, but for N_2O.

13. *Figure 6 warrants more discussion, in particular, the progression back through the seasons across the panels is interesting in that for panels d and e show the surface influences above 410 K are about equally distributed between tSH and mNH,with a large fraction of older air. The region above 400K is the stratospheric overworld where isentropes are contained wholly within the stratosphere. The difference between the overworld transport regime and the subtropical UT/lowermost stratosphere corresponding to 400 K.*

    See above 'Overarching comments/concerns' (2nd point).

14. *The distribution of influences is consistent with papers using NASA ER-2 CO2 measurements in the 1990's through the early 2000's showing that the stratospheric overworld boundary condition can be well-represented by the average of MLO and SMO data lagged by 2 months (references). It may also be possible to compare the relative contributions of surface air and aged stratospheric air in the lower stratospheric overworld region (i.e.*

above 380/400K) with the empirical age spectra derived in Andrews et al. I think the label "free atmos" and corresponding references in the text should simply be changed to "aged stratospheric air". When I think of free atmospheric air, I usually think of free troposphere above the boundary layer. Irene Xeuref has a relevant paper on CO2 as a clock tracer in the UT/LS that seems highly relevant. Kristie Boering had a paper on using CO2:H2O relationships to estimate transit times from the tropical tropopause to the lower mid-latitude stratosphere that is also relevant.

Many thanks for this comment. We decided to use the label "free atmos" in Fig. 6 of the submitted version of the manuscript, because the label "aged air" is already used for air older than 1 June 2016 (see Table 1, submitted version of the paper). In Fig. 6, the fraction of the "free atmos" depends on the used trajectory lengths (Fig. 6a until 1 June 2017, Fig. 6b until 1 March 2017, Fig. 6c until 1 December 2016, ...). For the shortest back-trajectory calculation (Fig. 6a), indeed the fraction of the "free atmos" includes air masses from free troposphere above the boundary layer, thus your thinking is correct here. In Fig. 6e the fraction of the "free atmos" corresponds to the fraction of "aged air".

We added the following explanation to the caption of Fig. 6 in the revised version of the paper.

In Fig. 6e, the fraction of air referred to as the free atmosphere corresponds to the fraction of 'aged air' defined in Tab. 1.

15. *I don't think the inclusion of the GOSAT-L4B product to improve the profile reconstruction at the highest altitudes adds much to the paper. The error bars are very large, and a mismatch of several ppm in the stratosphere cannot be considered a good fit to the observations. Why not also include CAMS and CarbonTracker in the main paper along with GOSAT L4B and make a stronger point about the poor performance of the available model products? Figure S2B is underwhelming. How does the GOSAT-L4B compare directly with the HAGAR data? My understanding is that the GOSAT-L4B trajectories are sampled at their endpoints and that extrapolation is necessary above 10hPa. This doesn't seem at all compelling. I'm just not sure what we are learning from this part of the analysis. Maybe I am missing something?*

    Many thanks for this comment. We included a comparison of HAGAR CO_2 profiles with both CarbonTracker and GOSAT-L4B data in the revised version of the supplement (see Supplementary Fig. S3).

The comparison with aircraft CO_2 profiles demonstrates, that in the troposphere GOSAT-L4B agree much better with measured CO_2 profiles as CarbonTracker (which is in general too high), reflecting that GOSAT-L4B data are based on column-averaged satellite measurements compared to CarbonTracker that does not include ground-based measurements from the Indian subcontinent after 2013 (detail see main paper). Further, Fig. S3 shows that within the UTLS the vertical resolution of both CarbonTracker and GOSAT-L4B is too low to reproduce the vertical variability of CO_2 visible in the airborne measurements and caused by the seasonal variability of CO_2 at the ground (details see main paper). Despite GOSAT-L4B and CarbonTracker fail to reproduce HAGAR CO_2 in the UTLS, CO_2 values of the stratospheric background (above 450 K / 70 hPa) from GOSAT-L4B and CarbonTracker show a reasonable agreement with HAGAR.

Because GOSAT-L4B data have in in general a better agreement with HAGAR CO_2, we use GOSAT-L4B date for reconstruct the stratospheric background in our approach.

16. *Line 621: "Thus not surprisingly, the CO2 distribution at the ground nor the vertical distribution of CO2 over South Asia during summer 2017 is not well represented in CarbonTracker (35)." I think maybe this is supposed to be Ref #34 (Jacobson et al) rather than Ref #35 (Konopka et al, submitted).*

    Konopka et al. (2022) use the CarbonTracker CO_2 distribution at the ground as a lower boundary condition to simulate CO_2 in the Asian monsoon region 2017 as well as in the northern extra-tropics. We rephrased the sentence as follows:

    Thus not surprisingly, the CO_2 distribution at the ground over South Asia during summer 2017 is not well represented in CarbonTracker (see Supplementary Fig. S3 for further discussion on this issue). A consequence when using CarbonTracker CO_2 as the lower boundary condition the vertical distribution of CO_2 over South Asia during summer 2017 can not be well represented in 3-dimensional model simulations in Konopka et al. (2022).

17. *In the discussion of Figure 7 the agreement between the observed profile and the CO2 reconstruction based on Nainital >410 K is described as remarkable. I would instead describe it as coincidental. It's not really that surprising given the range of observed surface CO2 across the cases that*

one of them falls on top of the obs. I don't think there is any physical significance to this result.

We agree, we have dropped any discussion of the quality of the CO_2 reconstruction above 410 K. Further, we changed in Fig. 7 the trajectory length for the CO_2 reconstruction (from Dec 2016 to 1 June 2016).

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5th Dec 22

Dear Dr Vogel,

Your manuscript titled "Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region" has now been seen by our reviewers, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. We strongly encourage you to carefully reconsider the framing of your manuscript in light of the reviewer recommendations on improving the impact of your paper. Please state clearly and explicitly what your study contributes to our understanding, and what the applications and implications of your reconstructions are. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

I am satisfied with the authors' response.

Reviewer #2 (Remarks to the Author):

Dear authores,

2nd review of “Reconstructing high-resolution in-situ vertical carbon dioxide proles in the sparsely monitored Asian monsoon region” submitted to Communications Earth & Environment by Bärbel Vogel et al.

The revised manuscript answered the reviewer’s questions and comments sincerely. This reviewer almost satisfied the answers from authors, however, before accepting the publication and more useful to be the scientific manuscript, this reviewer recommends to add the discussion or explanation as follow.

The following points would be added to the section of Discussion/conclusion.
What kind of research can be applied by using the characteristics of this data? It would be better for users if there is a concrete example. For example, a quantitative discussion of STE process studies on the intraseasonal variability associated with the deep convection and anticyclonic of Asian monsoon. Further the possibility of applying this method to other aircraft observation cases.

Reviewer #3 (Remarks to the Author):

Recommendation: The revision falls short of the major reframing that I was hoping to see. In my opinion, the paper is still lacking as described below. The current analysis is fine, in the sense that the method does not have major errors, and the model trajectory analysis provides interesting insight about regional influences on air aloft, especially for the tropospheric portion of the profile and through the TTL. Of course I understand that it may not be practical for the authors to undertake further revision, in which case I defer to the editor about whether the current version of the manuscript is suitable for publication in Communications Earth & Environment. The suggestions below are offered in the spirit of increasing the impact of the paper. These ideas could be pursued in subsequent papers once the data are publicly available.

Overarching comments:

Regarding Age(N2O) & Age(CO2).
I am disappointed that the authors did not take this to completion. I understand that computing CO2(Age) is not trivial (i.e. there is not a simple equation, it requires numerical convolution of reasonable age spectra from a model with a CO2 stratospheric boundary condition). Nevertheless, I am confident that this avenue is well worth pursuing based on my own quick analysis of the CO2 and N2O profiles presented in the manuscript (from manually picking points off of Fig 3).

A similar approach to what I suggest has been demonstrated to improve the a priori trace gas profiles used for the TCCON spectrometers (https://doi.org/10.5194/amt-2022-267) and these new priors will soon or are already be used in an updated retrieval for the OCO-2 satellite sensor. Josh Laughner, the lead author for the TCCON paper noted above, would easily be able to provide expected CO2:N2O:Age stratospheric overworld relationships for the dates corresponding to the HAGAR flights. This could be directly compared to the CO2:N2O relationship observed during HAGAR for air with potential temperature > ~380-400K. And CO2 profiles versus potential temperature could be estimated from the observed N2O profile. Such an analysis would allow the authors to directly relate their findings to prior mean age of air work by Engel, Andrews, Eric Ray and others and to contribute to the ongoing discussion in the literature about whether there are observable trends in the stratospheric Age of Air inferred from observations of long-lived tracers.

The authors made a reasonable attempt at addressing my suggestions, and Figure 1 of their response to my earlier review is very interesting. The figure shows a comparison of Age(N2O) from the Engel 2002 and the Andrews 2001 equations along with age from the CLaMs trajectories. The age(n2o) equations agree within +/- 2 months (better than 10% for air with N2O 260-280 ppm and age ~ 2.5-2.9 years) for the lowest values observed during HAGAR. Note that differences are unsurprising due to different assumptions about the stratospheric boundary condition for CO2 and possible calibration-scale differences for CO2 and N2O for the data used to derive the relationships). Meanwhile, the age inferred from CLaMS trajectories is only sensitive to the youngest air, since backward trajectories were truncated on 1 June 2016, approx 14 months prior to the HAGAR flights. The difference between the CLaMS age and age(n2o) corresponds to the portion of old air descended from aloft, and a frequency distribution of CLaMS age for a particular potential temperature level (e.g. 450+/- 5 ppb) would provide insight into the shape of the age spectrum according to the model.

Regarding direct evaluation of CarbonTracker and GOSAT-L4B.
The direct comparison of HAGAR CO2 profiles with CarbonTracker and the GOSAT-L4B is very nice and showcases the good performance of GOSAT-L4B and the poor performance of CarbonTracker for these flights. If possible, it would be great to see this incorporated into the body of the paper and expanded to include a larger suite of models (e.g. the OCO-2 Model Intercomparison suite presented by Piero et al., 2022 https://doi.org/10.5194/acp-22-1097-2022), and to consider whether general conclusions could be made about the impact of assimilating satellite observations for improving simulated CO2 in this region. Specifically, I wonder if the OCO2 MIP model simulations that assimilate the Land Nadir Land Glint measurements from OCO-2 show better agreement than those that only assimilate in situ measurements. I expect that the OCO-2 MIP team would be very interested in such a comparison and willing to assist with obtaining the simulated profiles.

Specific comments:

Revised Manuscript 101-112: The additional information is helpful, but I still think that it would be useful to state explicitly that altitudes corresponding to theta > ~360K (380K?) represent an entirely different transport regime because the isentropes are wholly contained within the stratosphere. The point of my earlier comment is that within the stratospheric overworld CO2 variability is explained comprehensively by the simple conceptual models that were developed following the Holton paper e.g. the leaky pipe model developed by Jessica Neu with Alan Plumb and more recently in a study led by Marianna Linz, (10.5194/egusphere-egu2020-12704). It would be great to explor the extent to which the new stratospheric overworld data from HAGAR can also be explained using the simple conceptual models that have been used to explain the previously observed consistency of relationships among long-lived trace gases in the lower stratosphere.

Revised Manuscript 306-306: It would be helpful to add something like…"which occurs primarily in the tropical mid-/upper stratosphere (24-40 km, 30S – 30N; Prather et al., 2015, doi: 10.1002/2015JD023267).

349-350: "Most air parcels were released at the model BL much later than 1 June 2016." I think there is some confusion between backward/forward time in this discussion. My understanding is that the particles were released along the flight tracks and run backward in time until they enter the model boundary layer at which point the trajectory is truncated. The use of the verb "released" is confusing because it seems to imply forward trajectory runs where the particles started in the B: and were run forward in time. Maybe it would be more clear if rephrased something like: Most air parcels encounter the model BL within a few months of backward transport at which point the trajectories are truncated (e.g. 64% of all trajectories intersect with the model boundary layer during the monsoon season 2017).

Line 352: Something like "As expected, simulated transport times increase with altitude (Fig. 3)."

Line 357: Something like "…it is essential to determine the location where the back trajectories intersect the model BL so that they can be tagged with the closest ground-based measurement."

Fig 3: Suggest using "ppm" and "ppb" instead of ppmv/ppbv, since the former is the correct unit for the dry air mole fraction preferred by the CO2 community (i.e. "by volume" is an approximation that is not strictly correct).

Fig 8 and S1, S2: I agree that the reconstruction of the upper portion of the profiles using the GOSAT-L4B product is reasonably successful, but the error bars are very large, and the present discussion does not provide much physical insight.

Some notes should be included about traceability of the HAGAR CO2 and N2O profiles to the WMO scales that are maintained by NOAA. If the data are not reported on the current WMO X2019 scale for CO2 and the NOAA-2006A scale for N2O, then equations to convert the observations to the current scales should be included.

Author Comment to Referee #1 - #3

Communications Earth & Environment (COMMSNV-22-0506-T), 'Reconstructing high-resolution in-situ vertical carbon dioxide profiles in the sparsely monitored Asian monsoon region' by B. Vogel et al.

We thank Referees #1-3 for their positive reviews and for further guidance on how to revise our manuscript. Our reply to the reviewers' comments is listed in detail below. Questions and comments of the referees are shown in italics. Passages from the revised version of the manuscript are shown in blue.

Reviewer #1 (Remarks to the Author)

I am satisfied with the authors' response.

We are pleased that we could respond satisfactorily to all of the reviewer's comments.

Reviewer #2 (Remarks to the Author)

Dear authores, 2nd review of "Reconstructing high-resolution in-situ vertical carbon dioxide proles in the sparsely monitored Asian monsoon region" submitted to Communications Earth & Environment by Baerbel Vogel et al.

The revised manuscript answered the reviewer's questions and comments sincerely. This reviewer almost satisfied the answers from authors, however, before accepting the publication and more useful to be the scientific manuscript, this reviewer recommends to add the discussion or explanation as follow.

The following points would be added to the section of Discussion/conclusion. What kind of research can be applied by using the characteristics of this data? It would be better for users if there is a concrete example. For example, a quantitative discussion of STE process studies on the intraseasonal variability associated with the deep convection and anticyclonic of Asian monsoon. Further the possibility of applying this method to other aircraft observation cases.

Many thanks for the helpful comment. We included the comment in the revised version as follows:

Our findings show that the Lagrangian transport in CLaMS using diabatic vertical velocities and driven by the European Centre for Medium-Range Weather Forecasts' new high-resolution reanalysis ERA5 is very well suited for CO_2 reconstruction (see Supplementary Fig. S2 for further discussion on this issue) and could be applied to other CO_2 aircraft observations.

We suggest that the propagation of these signals from the surface to the stratosphere constitutes a stringent test for atmospheric transport simulations and thus the data presented here provide unprecedented opportunity for CO_2 inversion systems to critically evaluate model transport and assess the derived CO_2 fluxes in South Asia. High-resolution CO_2 profiles can further be used to study stratosphere-troposphere-exchange processes as well as the intra-seasonal variability during the Asian monsoon season.

Reviewer #3 (Remarks to the Author)

Recommendation: The revision falls short of the major reframing that I was hoping to see. In my opinion, the paper is still lacking as described below. The current analysis is fine, in the sense that the method does not have major errors, and the model trajectory analysis provides interesting insight about regional influences on air aloft, especially for the tropospheric portion of the profile and through the TTL. Of course I understand that it may not be practical for the authors to undertake further revision, in which case I defer to the editor about whether the current version of the manuscript is suitable for publication in Communications Earth & Environment. The suggestions below are offered in the spirit of increasing the impact of the paper. These ideas could be pursued in subsequent papers once the data are publicly available.

We are pleased that Reviewer #3 did not find any major errors in our analysis and that he/she sees new interesting insights in our study. We are impressed of the number of good proposals for further analysis using the presented unique measurements in the region of the Asian monsoon. We do not want to give the impression to be lazy, but a publication in Communications Earth & Environment has restrictions in terms of number of words and number of figures. In the revised version of our paper, there are a series of interesting figures in an electronic supplement because we already exceeded the number of tables/figures permitted in the main body of the paper. Therefore we would prefer to do further analysis as proposed by Reviewer #3 in subsequent papers. Maybe Reviewer #3 would like to join this activities. We would definitely appreciate to be contacted.

Overarching comments:

1. *Regarding Age(N2O) & Age(CO2). I am disappointed that the authors did not take this to completion. I understand that computing CO2(Age) is not trivial (i.e. there is not a simple equation, it requires numerical convolution of reasonable age spectra from a model with a CO2 stratospheric boundary condition). Nevertheless, I am confident that this avenue is well worth pursuing based on my own quick analysis of the CO2 and N2O profiles presented in the manuscript (from manually picking points off of Fig 3).*

   That would be an interesting follow-up study using multi-annual 3-dimensional CLaMS simulations driven by ERA5 high-resolution data. Such 3-dimensional CLaMS simulations allows age-spectra to be reconstructed. However at the moment, we are restricted in driving multi-annual CLaMS simulations with high-resolution ERA5 reanalysis because of required computer resources. However, we are working on that issue.

2. *A similar approach to what I suggest has been demonstrated to improve the a priori trace gas profiles used for the TCCON spectrometers (https://doi.org/10.5194/amt-2022-267) and these new priors will soon or are already be used in an updated retrieval for the OCO-2 satellite sensor. Josh Laughner, the lead author for the TCCON paper noted above, would easily be able to provide expected CO2:N2O:Age stratospheric overworld relationships for the dates corresponding to the HAGAR flights. This could be directly compared to the CO2:N2O relationship observed during HAGAR for air with potential temperature $>\sim$380-400K. And CO2 profiles versus potential temperature could be estimated from the observed N2O profile. Such an analysis would allow the authors to directly relate their findings to prior mean age of air work by Engel, Andrews, Eric Ray and others and to contribute to the ongoing discussion in the literature about whether there are observable trends in the stratospheric Age of Air inferred from observations of long-lived tracers.*

Yes, the authors agree that this proposal would be an interesting follow up study. Unfortunately, there are not many prior measurements in the region of the Asian monsoon and it would be difficult to discuss trends in the stratospheric age of air in limited space. A large amount of analysis would be required to harmonize data and interpret the results. A detailed discussion on age of air analysis is outside the scope of this study.

3. *The authors made a reasonable attempt at addressing my suggestions, and Figure 1 of their response to my earlier review is very interesting. The figure shows a comparison of Age(N2O) from the Engel 2002 and the Andrews 2001 equations along with age from the CLaMs trajectories. The age(n2o) equations agree within +/- 2 months (better than 10% for air with N2O 260-280 ppm and age $\sim$ 2.5-2.9 years) for the lowest values observed during HAGAR. Note that differences are unsurprising due to different assumptions about the stratospheric boundary condition for CO2 and possible calibration-scale differences for CO2 and N2O for the data used to derive the relationships). Meanwhile, the age inferred from CLaMS trajectories is only sensitive to the youngest air, since backward trajectories were truncated on 1 June 2016, approx 14 months prior to the HAGAR flights. The difference between the CLaMS age and age(n2o) corresponds to the portion of old air descended from aloft, and a frequency distribution of CLaMS age for a particular potential temperature level (e.g. 450+/- 5 ppb) would provide insight into the shape of the age spectrum according to the model.*

   It is good to see that Reviewer #3 acknowledges our efforts to reasonably answer Reviewer #3's previous comments. Again a full 3-dimensional model simulation yielding age spectra must be deferred to future work. To be focused, we have restricted our analysis to CLaMS simulations driven by ERA5 only.

4. *Regarding direct evaluation of CarbonTracker and GOSAT-L4B. The direct comparison of HAGAR CO2 profiles with CarbonTracker and the GOSAT-L4B is very nice and showcases the good performance of GOSAT-L4B and the poor performance of CarbonTracker for these flights. If possible, it would be great to see this incorporated into the body of the paper and expanded to include a larger suite of models (e.g. the OCO-2 Model Intercom-*

parison suite presented by Piero et al., 2022 https://doi.org/10.5194/acp-22-1097-2022), and to consider whether general conclusions could be made about the impact of assimilating satellite observations for improving simulated CO2 in this region. Specifically, I wonder if the OCO2 MIP model simulations that assimilate the Land Nadir Land Glint measurements from OCO-2 show better agreement than those that only assimilate in situ measurements. I expect that the OCO-2 MIP team would be very interested in such a comparison and willing to assist with obtaining the simulated profiles.

We are grateful to Reviewer #3's comment to include a direct evaluation of CarbonTracker and GOSAT-L4B. This comparison is a significant added value for our manuscript. We agree that this comparison would be of worth to show it in the main body of the paper. However as mentioned above, the number of figures/tables in a publication in Communications Earth & Environment is restricted to the number of 10 in the main body of the paper. So unless the editor does not decide otherwise, we cannot change the situation. The measurements will be made available for the public along with this publication. Thus, the measurements can be used for the evaluation of further model simulations.

Specific comments:

- *Revised Manuscript 101-112: The additional information is helpful, but I still think that it would be useful to state explicitly that altitudes corresponding to theta $> \sim$360K (380K?) represent an entirely different transport regime because the isentropes are wholly contained within the stratosphere. The point of my earlier comment is that within the stratospheric overworld CO2 variability is explained comprehensively by the simple conceptual models that were developed following the Holton paper e.g. the leaky pipe model developed by Jessica Neu with Alan Plumb and more recently in a study led by Marianna Linz, (10.5194/egusphere-egu2020-12704). It would be great to explor the extent to which the new stratospheric overworld data from HAGAR can also be explained using the simple conceptual models that have been used to explain the previously observed consistency of relationships among long-lived trace gases in the lower strato-*

sphere.

We agree in principle with the suggestion in the review regarding the cited simple conceptual models. However, it is not straightforward to apply these ideas (that are describing a two-dimensional, zonally averaged world) to the Asian monsoon anticyclone. The vertical structure in the Asian monsoon region is different compared to the residual tropical tropopause layer (TTL). The thermal tropopause as well as isentropes (in log-pressure altitude coordinates) are enhanced in the region of the Asian monsoon anticyclone compared to the residual TTL (e.g. Vogel et al., 2016). Even when an isentrope crosses the anticyclonic monsoon circulation, air masses cannot freely move along this isentrope. The edge of the anticyclone is characterized by a PV (transport) barrier located at levels of potential temperature between $\sim$370 K and $\sim$390 K (see (e.g. Ploeger et al., 2015)). And transported air masses also need to (approximately) preserve PV. Therefore, the application of conceptual models to the monsoon circulation (and the CO_2 data described in our study) constitutes a special effort that cannot be incorporated in the present paper.

In the revised version of the paper we add the following sentence:

Air parcels are uplifted quickly by convection followed by slow diabatic uplift in the UTLS superimposed by the anticyclonic flow, while in other regions within the tropical transition layer (TTL) the heating rates are in general smaller during boreal summer (Vogel et al., 2019). Further, the thermal tropopause as well as isentropes (in log-pressure altitude coordinates) are enhanced in the region of the Asian monsoon anticyclone compared to the residual TTL (e.g. Vogel et al., 2016).

- *Revised Manuscript 306-306: It would be helpful to add something like ..."which occurs primarily in the tropical mid-/upper stratosphere (24-40 km, 30S – 30N; Prather et al., 2015, doi: 10.1002/2015JD023267).*

The reduction of N_2O in the lower stratosphere occurs via photolysis and reaction with excited atomic oxygen ($O(^1D)$).

We revised this sentences as follows:

The critical region for N_2O loss is the tropical middle stratosphere (24-40 km) (Prather et al., 2015) where destruction of N_2O occurs via photolysis and reaction with excited atomic oxygen ($O(^1D)$).

- *349-350: "Most air parcels were released at the model BL much later than 1 June 2016." I think there is some confusion between backward/forward time in this discussion. My understanding is that the particles were released along the flight tracks and run backward in time until they enter the model boundary layer at which point the trajectory is truncated. The use of the verb "released" is confusing because it seems to imply forward trajectory runs where the particles started in the B: and were run forward in time. Maybe it would be more clear if rephrased something like: Most air parcels encounter the model BL within a few months of backward transport at which point the trajectories are truncated (e.g. 64% of all trajectories intersect with the model boundary layer during the monsoon season 2017).*

  However, most air parcels were released at the model BL much later than 1 June 2016 (e.g. 64% of all air parcels are from the monsoon season 2017).

  We revised this sentences as follows:

  However, most air parcels encounter the model BL within a few months of backward transport (e.g. 64% of all trajectories reach the model BL during the monsoon season 2017).

- *Line 352: Something like "As expected, simulated transport times increase with altitude (Fig. 3)."*

  The higher the sampled air parcels are located the longer are their simulated transport times (see Fig. 3) which is to be expected.

  We revised this sentences as follows:

  As expected, simulated transport times increase with the altitude of sampled air parcels (Fig. 3).

- *Line 357: Something like "...it is essential to determine the location where the back trajectories intersect the model BL so that they can be tagged with the closest ground-based measurement."*

  For the CO_2 reconstruction it is essential to determine where the air parcels were released at the model BL to use the closest ground-based measurement.

  We revised this sentences as follows:

  For the CO_2 reconstruction it is essential to determine the location where the back trajectories intersect the model BL so that they can be tagged with the closest ground-based measurement.

- *Fig 3: Suggest using "ppm" and "ppb" instead of ppmv/ppbv, since the former is the correct unit for the dry air mole fraction preferred by the CO2 community (i.e. "by volume" is an approximation that is not strictly correct).*

  done

- *Fig 8 and S1, S2: I agree that the reconstruction of the upper portion of the profiles using the GOSAT-L4B product is reasonably successful, but the error bars are very large, and the present discussion does not provide much physical insight.*

  We agree that the error bars are very large, reflecting the uncertainties using GOSAT-L4B product for the reconstruction of stratospheric background.

- *Some notes should be included about traceability of the HAGAR CO2 and N2O profiles to the WMO scales that are maintained by NOAA. If the data are not reported on the current WMO X2019 scale for CO2 and the NOAA-2006A scale for N2O, then equations to convert the observations to the current scales should be included.*

  The ground-based measurements in Nainital and Camilla were calibrated with NIES secondary standard gas series (CO_2-NIES09 scale and N_2O-NIES01 scale) with differences of -0.04 to -0.09 ppm for CO_2 relative to the WMO X2007 scale and -0.61 to -0.69 ppb for N_2O relative to the NOAA-2006A scale (Nomura et al., 2021).

HAGAR data were referenced to standards provided by NOAA and are based on the CO_2 WMO X2007 scale and the N_2O NOAA-2006 scale. The data can be converted to the current CO_2 WMO X2019 and N_2O NOAA-2006a scales using the following equations, which are based on reassigned standard values on the current scales:

$$X2019 = 1.00033 * X2007 + 0.467$$

$$2006a = 0.99841 * X2006 + 0.587$$

These conversions amount to small positive shifts of about 0.18 ppm for CO_2 and 0.05 to 0.17 ppm for N_2O.

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