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Atmospheric Chemistry and Physics Discussions
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Interactive comment on "Pollution trace gas distributions and their transport in the Asian monsoon upper troposphere and lowermost stratosphere during the StratoClim campaign 2017" *by* Sören Johansson et al.

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We thank the referee for valuable comments and suggestions. We changed all minor language and wording corrections according to the suggestions without listing all of the changes in this answer. Instead, a latexdiff document that tracks all changes made in the revised manuscript is provided in the author's response file.
To our knowledge, Copernicus will have the manuscript copy-edited by a professional writer in case of acceptance and before publication in ACP. In addition, translation services at our institution (KIT) checked the language of the revised manuscript.

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Discussion paper

Our answers are given below. The original referee comment is repeated in **bold**, changes in the manuscript text are printed in *italics*.

P2, L16-17, what's the name of the aircraft campaign?
We added the name of the Earth System Model Validation campaign to the manuscript. For the next sentence, we also mention now the Oxidation Mechanism Observations campaign.

P2, L22-24. The logical connections of these two sentences and the previous section seems to be amiss. What's the relationship between radiative heating rates and transport in reanalyses with trace gases? Observations are sparse, so what? How do observations help? The last sentence in the next paragraph (atmospheric chemistry models ...) is out of place. It fits much better in this paragraph, instead.
According to the comments of both referees, we reformulated and restructured this paragraph into two paragraphs: The second paragraph of the introduction now mentions studies about the transport in the ASM, focusing on open issues of vertical transport. The third paragraph of the introduction now summarizes studies of pollution trace gas measurements (and their implications) in the ASM.
In addition, we removed the "atmospheric chemistry models" sentence from the next paragraph.

P2, L24-28. These three sentences seem to repeat themselves in various ways. It can be easily condensed into a single sentence but capture all essential elements. Please revise.
We agree that the third sentence did not provide much new information. It would be possible to also merge the first two sentences, but in our opinion, this would not improve readability. We propose to change the first four sentences to these two sentences: *The first observations of the upper tropospheric chemical composition*

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in the region of the ASM in high vertical resolution have been obtained during the high-altitude airborne StratoClim (Stratospheric and upper tropospheric processes for better climate predictions) campaign. This study presents a unique data set of pollution trace gases, in particular non-methane volatile organic compounds (NMVOCs) obtained with the Gimballed Limb Observer for Radiance Imaging of the Atmosphere (GLORIA) during this StratoClim campaign based in Kathmandu, Nepal, 2017.

P3, L1. Which two models? EMAC and CAMS? Please specify. And I am sure this is not the "first evaluation" of these two atmospheric chemistry models.
We changed the sentence to: *Second, a first evaluation in high spatial resolution in the ASM UTLS of the atmospheric chemistry models EMAC and CAMS is provided [...]*

P3, L30. WMO, 2019 $->$ This one is actually "WMO, 2018"
You are right that the WMO report referenced here is named "Scientific assessment of ozone depletion: 2018", but it was published in January 2019 (see https://www.esrl.noaa.gov/csl/assessments/ozone/2018/downloads/2018OzoneAssessment.pdf, page ii), which makes it a publication of the year 2019. However, in case of publication in ACP, Copernicus Publications will adjust the references according to their standards.

P4, L1. Catalytic reactions with nitrogen oxides is a sink of ozone in the stratosphere and to some extent in the upper troposphere. Primary sources of ozone is the troposphere is in situ production of NOx +HOx, and NOx + peroxide radicals from VOC oxidation, hence indicator of polluted air masses.
We refined our explanation of tropospheric ozone sources according to your comment and restructured the section according to the comment in Michelle Santee's review. In addition, we added reactions with NO_x to the loss processes (Bozem et al., 2007; 10.5194/acp-17-10565-2017) and mentioned lightning as a source of NO_x.

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P4, L27. Maximum tropospheric mixing ratios of a few ppt for C2H2? Are you sure it is not a few ppb?? Check Xiao et al., (2007).
Yes, you are right, that was a typo.

P7, L14. Can you describe what are the NMVOC emission sources used in EMAC? When you say 50% and 100% additional emissions, do you mean from all emission sources, e.g. biomass burning, biofuel, fossil fuel, etc., and globally or just over Asia?
There are anthropogenic emissions sources from biomass burning, agricultural waste burning, fossil fuels, ship, road and aircraft emissions, as well as biogenic emissions. We included this in the paper. The emissions are from MACCity, ACCMIP and RCP6.0. In our sensitivity studies, we added 50% or 100%, respectively to all NMVOC emission sources globally. We explain this now also in the description of the EMAC model simulations.

P8, L3-4. I find this sentence very awkward, with no clear description of what was actually done.
We changed this sentence to: *A trajectory is considered to be influenced by convection if it encounters a convective cloud during its advection, with a pressure higher than the cloud top pressure (similarly done by Tissier et al.,2016). The location of cloud encounter is then identified as a convective source.*

P7-8. In sections 2.3, could you also provide the details on which year, time period of the model simulations that were conducted?
For EMAC, it is already mentioned at the end of Sec. 2.3.1 that all model runs were initialized on 1 May 2017. The CAMS reanalysis is an operational data product from ECMWF and similarly to other reanalyses continuously updated. We added to the manuscript: *CAMS reanalysis data is available for the time between 2003 and 2018.*
Both backward trajectory models simulate the time before the measurement. For

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TRACZILLA, it is already stated in Sec. 2.3.3 that they are simulated for one month before the measurement. For ATLAS, we added: *Trajectories are calculated for 30 days prior to the measurement.*

P8. Section 2.3.5 is out of place. This is observations and it should be listed in Section 2.1 or Section 2.2, not in the modeling subsection.
Thanks for pointing that out! We modified the structure so that the OMI part is now in its own subsection.

P9, L3 and Figure 2. I think it is more accurate to say these are colored boxes are "air masses" of interest, rather than "regions" of interest. Also in figure 2 caption, add "shows" after "the green line". I find the green line very hard to see. A thick solid dark gray line would be much better. It also distinguishes its functionality from the color boxes.
We agree that the formulation "air masses of interest" is more accurate and changed this term throughout the manuscript. We also followed your suggestion to change the color of the 380 K tropopause to dark gray and thickened the line.

P10, Figure 2. I find all panels very noisy, which is not surprising due to the large errors in GLORIA measurements as listed in Table 1. I would suggest average the measurement samples to larger temporal and vertical bins. This way you can average down the noise and illustrate the discussed features much better. In the present form, these features are barely distinctive from the surrounding background air masses. This is particularly problematic from C2H2 and HCOOH.
One goal of this manuscript is to publish the GLORIA data set for this StratoClim science flight, characterized by estimated error and vertical resolution. For this reason, we prefer to present the data in the full spatial resolution. Later in the manuscript, for comparisons with the EMAC and CAMS models, we horizontally average GLORIA

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profiles for a better comparison of the major structures. This averaging also reduces the noise error, which is a large contribution but not the total estimated error (see Suppl. Figs. 1, 3, 5, 7, 9). The other major contribution to the total error is the pointing error, which is not a statistical error and is thus not reduced by averaging of profiles. Vertical averaging, as suggested by the referee, would make the vertical resolution, which is an important characteristic of the retrieval, very difficult to interpret by the reader. As a compromise to better illustrate the discussed features, we changed the colorbars to discrete values instead of a continuous color spectrum, which also reduces noisy structures that are in the order of magnitude of the total estimated errors.

P12, Figure 3 and the corresponding discussion. (a) In the text, the relevant discussion uses km as a unit while the y-axis only shows pressure. Please add the corresponding km on y-axis. (b) The cyan box in TRACZILLA show likely convective influences while ATLAS shows none. Why the two models are showing such different results? And how can the GLORIA measurements help in assessing which back trajectory model is more accurate. Also, overall, I can see TRACZILLA shows more convective influences that ATLAS. How can you assess which one is more accurate?

(a) We followed the suggestion of the referee and added an additional y-axis with an approximation of altitude to the plots. In addition, we also mentioned the corresponding pressures in the text.

(b) “The cyan box in TRACZILLA show likely convective influences while ATLAS shows none. Why the two models are showing such different results?”

After the review, we noticed that there was a missing line in the analysis code. The enhancement in the TRACZILLA cyan box was corresponding to trajectories that were leaving the meteorological domain at higher altitudes, and they were not actually associated with convection. After applying the correct analysis, the structures of convective influence are very similar between the two models.

“Also, overall, I can see TRACZILLA shows more convective influences that ATLAS.

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Discussion paper

How can you assess which one is more accurate?"
The differences in the intensity of the convective influences between ATLAS and TRACZILLA are expected and are related to the different approaches used in the two methods, both relying on a different set of assumptions.
In ATLAS, the convective influence is estimated from the modeled detrainment rates of ECMWF ERA5 with a stochastic approach.
In TRACZILLA, the convective influence is estimated from the satellite-observed thick and high clouds. This has the advantage to give an observation-based information on the convective events based on a high temporal and spatial resolution (15 and 20 minutes and 3 and 2 km for MSG1 and Himawari data, respectively), reducing the spatial and temporal uncertainties of their identification with respect to the model-based approach. However, there are uncertainties in the determination of cloud altitudes using only passive sensors, and we have no information on the amount of mixing with ambient air at the point where the backward trajectories hits the cloud.
To conclude, ATLAS and TRACZILLA use different methods relying on different data sets (cloud satellite measurements and ERA5 detrainment rates), which have different temporal and spatial resolutions. It is expected to have different results from these different models. However, it is out of the scope of the paper to compare the two models and, in addition, it would be difficult to assess the performances of the two models from this specific case study. This issue is instead more extensively treated in the following papers: Bucci et al. (2020; 10.5194/acp-2019-1053), Legras and Bucci (2019; 10.5194/acp-2019-1075), and Wohltmann et al. (2019; 10.5194/gmd-12-4387-2019). The intent of this manuscript is to exploit the complementary information from the two models to interpret the data.
We added to the manuscript: *The different absolute percentages for convection probability for ATLAS and TRACZILLA are likely the result of the different underlying data sets and different methods for detection of convection along the backward trajectories by the models.*

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P14-P15, the discussion on various air mass signatures. For clarity and easy-to-follow purposes, I highly recommend you assemble all this information into a table. In the table, please list the type of targeting air masses, altitude at which they are sampled, surface regions where they were originated from, average measured HNO3, O3, PAN, C2H2, HCOOH concentrations within these colored boxed, transport time since they left the surface, etc. Second, please add a summary discussion on the different chemical signature of airmasses from different regions, e.g. the purple/blue box air from the marine background vs. the orange/red box air from China, etc.
(1) We followed the suggestion of the referee and added such a table to the manuscript.
(2) We expanded the discussion at the end of Sec. 4.

Sections 5 & 6. I found the observation-model comparison and evaluation a major weakness of this study. Neither CAMS nor EMAC produces well the observed features and gradients of all five species.This is particularly the problem for C2H2 and HCOOH. I also have problems with the brutal way of increasing NMVOC emissions by 50% and 100%. I don't see any improvements in model performance with such approach. By matching with observations better in a few patchy spots, you are also creating huge biases in other places (Figure 6) for all three species. PAN, C2H2 and HCOOH can be emitted and/or formed from various sources, i.e. anthropogenic emissions and biomass burning emissions being the highly relevant sources. The differences in the regional distributions of these sources can have a dominant impact on tropospheric distribution of these gases after they are being lofted and formed during transport. A proper way to address this model bias is to adjust the emission strength of these individual sources in separate runs and assess how do the resulted distribution change. This way, one can potentially assess the sources of these biases. You are only presenting analysis of one single flight. Therefore, such model sensitivity simulations can be easily conducted within a few days. The new

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model results and the corresponding discussion should be included in the revised manuscript before the paper is moving forward for publication.
We agree that the sensitivity test performed with EMAC is a very simple analysis, based on the findings by Monks et al. (2018), and not a full sensitivity study. Unfortunately, we do not share the referee's optimism to perform a full sensitivity study "easily [...] within a few days", given our workforce and computational resources. Even though we only compare and discuss one flight, every sensitivity simulation needs to include the time of at least several weeks before the measurement, in order to account for the transport of pollution from the boundary layer to the upper troposphere, where the measurements are performed.
For that reason, we decided to remove Sec. 6 from our manuscript, show results of the simple sensitivity test with +50% increased NMVOC emissions in the supplementary materials, and only briefly summarize the results of this test in Sec. 5. We think that the comparisons of GLORIA measurements to CAMS and EMAC without a detailed sensitivity study are of value on their own, as these comparisons point out considerable weaknesses of well-known atmospheric models in the upper troposphere of the ASM. In our opinion, it is also important to document negative outcomes of model evaluation studies, in order to motivate further sensitivity studies and model improvements.

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