Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-321-AC1, 2020

Atmospheric Chemistry and Physics Discussions

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.

Sören Johansson et al.

soeren.johansson@kit.edu

Received and published: 22 September 2020

We thank Michelle Santee for her thorough and very valuable comments and suggestions. We changed all minor language and wording corrections according to her 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.
Our answers are given below. The original referee comment is repeated in **bold**, changes in the manuscript text are printed in *italics*.

P1, L16-17: It is not clear to me what the assertion that the models reproduce the large-scale structures of the pollutant distributions "if the convective influence on the measured air masses is captured by the meteorological fields used by these simulations" is based on, since this study does nothing to demonstrate that the models capture convective influence well, and in fact numerous prior studies have shown that they do not. Perhaps the large-scale trace gas distributions are controlled mainly by the large-scale circulation, which global models do simulate reasonably well.
We agree with the referee that we have not unambiguously demonstrated the influence of convective events to be responsible for the disagreement between models and GLORIA observations. Given the changes in the main part of the manuscript, we changed this part of the abstract to: *It is shown that these simulation results are able to reproduce large scale structures of the pollution trace gas distributions for one part of the flight, while the other part of the flight reveals large discrepancies between models and measurement. These discrepancies possibly result from convective events that are not resolved or parameterized in the models, uncertainties in the emissions of source gases, and uncertainties in the rate constants of chemical reactions.*

P2, L4: The manuscript by Basha et al. (2019) has been rejected and should not be cited.
We thank the referee for pointing that out! We missed to check this reference before the submission of the manuscript.

P2, L7-8: Some references for the sentence about airborne in situ measurements inside the AMA would be appropriate.

We added the Bourtsoukidis et al., 2017 (10.5194/amt-10-5089-2017) and Gottschaldt et al., 2018 (10.5194/acp-18-5655-2018) references. Because both references (and other references we know) only describe measurements of air masses of the AMA edge outflow, we rephrased to: *Airborne in-situ observations of air masses belonging to the AMA are extremely sparse and often sample only filaments, border areas, or outflow of the AMA (e.g., Bourtsoukidis et al., 2017, Gottschaldt et al., 2018.)*

P2, L10-23: This paragraph as a whole is rather disjoint, with multiple independent thoughts assembled together with no thread connecting them. The last two sentences in particular seem out of place and do not follow from previous lines, and it's not clear why the last one begins with "However". I suggest rewriting to improve the cohesion and flow.
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) for the ASM UTLS.

P3, L30: This is a very abrupt transition to tropospheric ozone; it would be better to say something about background values of tropospheric ozone, and possibly its sources as well, before talking about the magnitude of enhancements.
We added typical O_3 VMRs in the ASM and restructured the paragraph, so that the sources are discussed before the magnitude of enhancements.

P4, L4-5: Numerous papers (some of which are referenced elsewhere in this manuscript) have discussed the low abundances of ozone inside the AMA, so it is not appropriate to cite only a single paper for this point; at the very least an "e.g." is needed here.

We added an "e.g.", and Santee et al., 2017 and Brunamonti et al., 2018 as additional references.

P4, L10-11: This sentence is somewhat inaccurate. Ozone is typically low inside the AMA; the Park et al. papers cited here use low ozone abundances (along with enhanced CO) as a marker of tropospheric air trapped inside the AMA. Park et al. (and others) have used larger abundances of ozone as an indicator of the presence stratospheric air, but not "polluted air" as stated here. If the authors are referring to the findings of Gottschaldt et al. (2017), then that paper should be cited here. In addition: measurements of O3 . . . is –> O3 is.
We rephrased this sentence. In addition, we added the suggested reference: *Similarly to HNO_3, enhanced O_3, within the generally O_3 poor AMA upper tropospheric air, is either interpreted as indicator of stratospheric air (e.g., Park et al.,2007,2008) or connected to uplift of O_3 precursor species of polluted air (Gottschaldt et al., 2017).*

P4, section 2.1.3: Typical background abundances of PAN should be stated here, as they are in the respective subsections for HNO3 and O3. This information is given in Section 3, but for completeness it should appear here as well.
According to the referee's suggestion we added: *Typical background abundances of PAN in the upper troposphere are below 100 pptv (Glatthor et al., 2007).*

P4, L17-18: It is stated that photolysis plays a minor role, but according to Fadnavis et al. (ACP, 2015), photolysis is the dominant loss process for PAN in the UTLS. In addition, 250 K is not a higher altitude than 298 K.
Our original statement was meant for the whole troposphere. We added Fadnavis et al. (ACP,2014) as reference for this sentence and formulated more precisely: *[...] and photolysis play a minor role for lower tropospheric altitudes (e.g., Fadnavis et al., 2014). In the upper troposphere instead, photolysis is the dominant loss process for PAN (e.g., Fadnavis et al., 2015).*

In addition, we clarified, that the numbers given are temperatures, not potential temperatures.

P4, section 2.1.4: It is even more critical to help readers by providing some idea of typical background values for acetylene since that information is not given in Section 3.
We added: *Typical background values for C_2H_2 are below 75 pptv (e.g., Xiao et al., 2007; Wiegele et al., 2012).*

P5, L13-14: Why is only a single research flight singled out for analysis in this study? Unless some explanation is given about why the data available from the other three flights are not considered, readers may draw their own inferences about their quality or consistency.
We added these sentences for explanation: *This research flight was selected for this work due to high flight altitudes and low cloud top altitudes within the AMA, which are both optimal measurement conditions for the infrared limb instrument GLORIA. This research flight was by far the best, due to the flight length allowing different air masses to be sampled and the low cloud top altitude.*

P7, L10-15: These sentences are poorly written and unclear. Was the extension of the MECCA model performed by the authors as part of this work, or by the EMAC team? Are the values quoted for the number of reactions, etc., for the "standard" MECCA submodel or the "extended" one?
Sorry for the confusion, we selected a more comprehensive chemistry set-up as usual in our simulations. The MECCA submodel was not extended. We removed the word "standard", because we do not explain it in the text and changed the sentence accordingly: *The chemical setup of the chemistry submodel MECCA (Sander et al., 2011) was selected with focus on the simulation of PAN and tropospheric chemistry.*

P7, section 2.3.1: How are emissions prescribed in the EMAC runs done for this study? This information seems just as critical to me as the details of the chemical submodel. In particular, if emissions were prescribed using RCP scenarios, which do not include specific events, such as major fires in any given year, then even specified-dynamics EMAC simulations cannot be expected to replicate the observations closely.
The referee is right, the emissions do not include the specific events of the year 2017. We use an emission scenario, which is quite common in the climate modeling community and currently, we do not have more recent emission data for the year 2017. We will express this more clearly in the paper. Nevertheless, we are convinced that the EMAC results should remain in the paper, because we think that simulation results based on these commonly used emission scenarios should be compared to measurements.

P7, section 2.3.2: Similarly, information about the emissions in CAMS also needs to be given.
We added: *Anthropogenic emissions are prescribed by MACCity (MACC/CityZEN; Granier et al., 2011), biogenic emissions by MEGAN2.1 (Model of Emissions of Gases and Aerosols from Nature; Guenther et al., 2012), and biomass burning emissions by GFAS v1.2 (Global Fire Assimilation System; Kaiser et al., 2012).*

P7, L23-24: This sentence mentions a study evaluating the CAMS chemical reanalysis using aircraft measurements but provides no information about the results of those comparisons. Did Wang et al. (2020) find that CAMS fields match the measured species well or not? What are the implications for this work? In addition, the paper by Wang et al. has now been published, so the reference needs to be updated.
Thanks for reminding us of the updated Wang et al., 2020 paper. We added to the section: *Profiles of O_3, HNO_3, and PAN above Hawaii showed an agreement within the*

uncertainties of measurement and model. These agreements encourage the model evaluation of this study at altitudes of the upper troposphere in the ASM.

P8, L9-10: Is this 3-h ERA5 product different from the one mentioned on P7, L28 with 1-h temporal resolution?

Both trajectory models (TRACZILLA and ATLAS) use the same ERA5 product, but ATLAS used a 3 h temporal resolution and a different spatial grid. We changed the manuscript to: *Trajectories from the ATLAS model (Wohltmann et al., 2009) are driven by the same ECMWF ERA5 meteorological fields as TRACZILLA, but with a temporal resolution of 3 h and a horizontal resolution of 1.125° × 1.125°.*

P8, L22-26: The investigation described in these sentences is interesting, but the results reported here are vague and their implications for this study are unclear (and the last sentence in this paragraph could also be better composed). What exactly is meant by "major differences" and "minor influences"? This discussion should be more quantitative. Do the findings from these ATLAS and TRACZILLA tests have any implications for the results from EMAC, since those runs were driven with ERA-I?

We added an additional figure to the supplement (Suppl. Fig. 19) to exemplarily show the influence of the reanalyses, trajectory type, and diffusion on the trajectory paths and location of convective events. We now refer to this supplementary figure and rephrased these sentences. In addition, we now reference to this investigation in the discussion of possible improvements of the EMAC simulations: *In an analysis of the ATLAS trajectories, the influence of the usage of ERA5 or ERA-Interim as meteorological fields, the influence of applied vertical diffusion, and the influence of the usage of kinematic or diabatic trajectories was investigated (shown in the supplementary information). This analysis (and also similar analyses by Legras and Bucci (2019)) revealed that major differences occur between ATLAS trajectories that use ERA5 or ERA-Interim meteorological fields. These major differences are exemplarily visible*

in Supplementary Fig. 19, where trajectory paths and locations of convective events are considerably different between ERA-Interim and ERA5. Compared to these large discrepancies, differences in trajectory paths and locations of convective events due to the usage of kinematic or diabatic trajectories, or due to the application of vertical diffusion are small.

P8, section 2.3.5: It is necessary to provide information on the quality and resolution of the OMI tropospheric column NO2 data, as well as a suitable reference for this specific product (beyond the general OMI instrument paper and the Krotkov (2013) citation, which is just for the L3 files and which is also incomplete).

We added: *The version 3 standard retrieval of tropospheric column NO_2 comes with a spatial resolution of 1.0° × 1.25° (latitude × longitude), and showed an overall agreement with other satellite and ground based measurements of NO_2 (Krotkov et al., 2017).*

The Krotov (2013) citation was meant as a documentation of the data file we used for this work, which is encouraged to be used by ACP. Due to technical issues, the DOIs were not displayed in the bibliography, which is now fixed.

P9, L6-7: This wording is unclear. By "local enhancements up to 0.5 ppbv", do the authors mean that the measured mixing ratios approach 0.5 ppbv, or that they are 0.5 ppbv larger than the regional background values (it looks like the latter to me). Some of these enhancements appear to be located at altitudes higher than 16 km. In fact, the particular structure noted at 4:00 UTC is at more like 16.5 km.

We tried to formulate more precisely: *In the first part of the flight (until 4:45 UTC), also a local maximum of VMRs up to 1.0 ppbv is visible below the tropopause at altitudes between 15.5 km and 17 km (close to the red box in Fig. 2b).*

P9, L8: Why is the magenta box drawn so as to exclude the peak in this enhancement at 4:00 UTC, and also the higher values right at 16 km just before 4:15 UTC? If this enhanced structure is of interest for further analysis, I would think that it would be desirable to encompass the region of its strongest signature.
We added the (slightly adjusted) red box to the HNO_3 cross section plot and clarified: *This maximum is continued by enhancements noted at 16 km at 4:00 UTC moving down to 15 km at 4:15-4:50 UTC with VMRs up to 0.75 ppbv (marked with a magenta box). The shape and position of the red and magenta boxes are optimized for the pollution trace gases PAN and* C_2H_2 *discussed later in this section to have a local maximum in the red and a local minimum in the magenta box. Thus, these boxes do not exactly match the structure in* HNO_3*. In addition, Höpfner et al. (2019) reported enhanced ammonium nitrate abundances in the red air masses, and a local minimum of ammonium nitrate in the magenta box. Given these different pollution trace gas and aerosol concentrations in the red and magenta boxes, it is assumed that these air masses have different origin, even though the structure in* HNO_3 *appears to be connected.*
The adjustment of the red box induced changes in Sec. 4.

P9, L14-15: It would be helpful if the colored boxes on Figure 2 were also overlaid on Supplementary Figures 2, 4, 6, 8, and 10.
We updated these Supplementary Figures according to the referee's suggestion. In addition, we refined the statement about the O_3 error within the purple box.

P11, L5-9: I'm wondering why the authors have chosen not to highlight the region with the minimum in HCOOH where PAN and C2H2 are present in its own colored box. Considerable discussion is devoted to this part of the flight, possibly more than for some of the regions that are enclosed within boxes.
We added a green box to highlight this minimum in HCOOH. We know, that the green

color might be difficult to see on top of the cross section, but with the white border all colored boxes have, it should be possible. We decided for this color because it is also easy to separate from the other colors in the written discussion later in the manuscript.

Figure 2: It would be extremely helpful to the reader to: (1) enlarge the major tick marks on both x and y-axes, (2) add minor tick marks, and (3) include tick marks on the right-hand y-axis and the top x-axis. Without them, it is very difficult to judge the values quoted in the text
We changed the figure (and similar figures later in the manuscript) according to the suggestions.

The colored boxes on both the maps and the curtain plots are a little hard to see, as is the green line marking the tropopause. Perhaps it would help to make these lines a bit thicker.
We increased line thicknesses according to the suggestions. In line with suggestions from the second referee, we also changed the color of the 380 K tropopause line to dark gray.

P12, L11: Although the overlaid boxes in Figure 3 facilitate comparison with Figure 2, the authors should consider adding an altitude scale on the right-hand y-axis of the panels as well. It would also be helpful to state the approximate pressure level corresponding to 15 km in this line.
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.

P12, L16-19: My apologies, but I am missing something here. I don't quite understand how the densities of convective events discussed in this paragraph relate to the convection probabilities shown in Figure 3 and discussed in the

previous paragraph (which are an order of magnitude larger). Please clarify the relationship between these two quantities.
We have substantially rephrased the complete paragraph to make more clear what is shown in Figures 3 and 4 (also in response to your comment on P12, L20). The text was confusing and did not contain sufficient information for the reader to understand the method and the figures. In addition, there was a factual error in the text which increased the confusion: "the smallest bordered regions include at least 0.1% of convective events" should have been "the outermost contours include at least 0.1% (per square degree) of convective events" (i.e. just the opposite of what was written). We have also made more clear now that the unit of the fractions shown in Figure 4 is "percent per square degree", i.e. the quantity shown is a fraction per area and not just a fraction.

P12, L20: I'm confused here too – why would it necessarily be the case that "larger regions contain accordingly a larger fraction"? A large region encompassed by a single colored contour but no inner contours would still have convective densities between 0.1% and 1.0%, no matter its size. Unless an inner contour is present, the fraction does not reach 1.0%. In addition, I have looked closely at Figure 4, and I am not convinced that any of the outlined regions contain the innermost contour representing 10%, except for one orange region in the TRACZILLA panel. Perhaps the rarity of that occurrence should be pointed out.
The statement was incorrect and we have rephrased the paragraph (see also reply to P12, L16-19).

P14, L8-9: The flow in this paragraph needs to be improved. The sentence about the small fraction of trajectories experiencing convection in the 5 days leading up to the measurement is ambiguous; it immediately follows a sentence on the magenta region and thus appears to be about that area, but in fact I think

it is referring to the red region. This should be clarified.
We removed the reference to the magenta region, which is not needed in this paragraph. We apologize for the confusion.

P14, L10-12: The writing in these lines is very unclear. Assuming that I have interpreted them correctly, I suggest instead: "For most regions marked red, only the 0.1% contours are present; thus convective influence along the trajectories was weak. However, most regions marked red in northeastern China lie close to areas with enhanced NO2, so these regions may possibly have contributed to the measured enhanced pollution trace gases."
We changed the manuscript in line with the referee's suggestion. In addition, we changed the order of words in the first sentence to make cause and effect more clear: Because convective influence was weak, only the 0.1 percent per square degree contour is present.

P14, L13-14: Again, I am confused about how the 30% value quoted here for the red regions can be reconciled with the 1% contour outlining those regions in Figure 4. The sentence in these lines is quite unclear. I'm also confused about exactly what is being shown in Supplementary Figure 12. As I understand it, the trajectories are launched from the GLORIA measurement locations, which in many/most cases are not characterized by ongoing convection. However, although the caption to Figure S12 is unclear, particularly the description of panel (c), it seems to suggest that a convective event was occurring at the time the trajectories were launched, and that 30% of those back trajectories had experienced convection leading up to that point. Please clarify.
We have substantially rephrased the text and the caption in the supplement. In particular, we did not want to suggest that a convective event was occurring at the time the trajectories were launched, which is not the case. We changed the text in the manuscript to: *For the ATLAS model, it is shown in Supplementary Fig. 12 that for the*

red region, less than 30% of all started trajectories experienced a convective event within 10 days before the measurement, showing the weak convective influence.
In addition, we changed the caption of Fig. S12 to: *In b) and c), dots mark the location of all convective events experienced by backward trajectories starting in the red region (with the convection scheme switched on). b) is color-coded with the time difference between the convective event and the time of measurement, and c) is color-coded with the percentage of the other backward trajectories that already had experienced convection when the trajectory represented by the dot went into convection.*

P14, L15-16: The magenta box is not shown on Fig. 2i, j, nor was a minimum in HCOOH in this region discussed (P11, L1-17). If anything, HCOOH looks slightly high in that area. I assume that "close to the red maximum" is referring to the pollutant enhancements in the red box?
We thank the referee for pointing that out! HCOOH appears in that list by mistake. We removed it from this paragraph. We changed the formulation "close to the red maximum" to *close to the maximum of the pollutant species marked with the red box*.

P14, L19-20: This sentence is badly written and hard to read. I suggest instead: "However, in this case, it is likely that convection in the regions above the South China and Philippine Seas brought up clean maritime air." But perhaps I have not understood this sentence. I can see that convective transport of clean maritime air could produce a local minimum in the pollutants, but how could it have led to enhanced HNO3 in this region?
We changed the sentence according to the referee's suggestion. In addition, for the explanation of the enhanced HNO_3, we add: *Enhanced HNO_3 concentrations within these air masses possibly result from reaction of lightning NO_x with OH to HNO_3 (see e.g., Schuhmann et al., 2007).*
We compared typical lifetimes of NO_x in the upper troposphere (4-7 days according to Schumann et al., 2007; 10.5194/acp-7-3823-2007) with the time since the convective

event above the South China and Philippine Seas for the magenta air masses (3-5 days; see Suppl. Tab. 1). Together with observations of several ppbv of lightning NO_x (Schuhmann et al., 2007), and HNO_3 as main sink of lightning NO_x, we consider this to be the most likely origin of the enhanced HNO_3 concentrations. In addition, we added in response to the referee's comment on "P9, L8" a comment on the structure of HNO_3 in the first part of the flight.

P14, L28-29: This sentence is unclear. More plausible than what? More likely than what?
We rephrased this sentence to: *This corresponds to the orange region in India with enhanced NO_2 columns.*
Other information in the original sentence was redundant to preceding sentences.

P15, L3-4: Why would bringing up relatively pristine marine boundary layer air lead to a local enhancement in ozone?
We added an interpretation of this result from the trajectory analysis: *These areas marked by the trajectories show low OMI NO_2 and indicate relatively clean boundary layer air, which cannot explain the measured local enhancement of O_3. This suggests that the measured local maximum of O_3 is of other than convective origin; possibly, the measured maximum is a pollution remainder transported for more than 10 days, or an intrusion of stratospheric air.*

P15, L6-7: I do not follow the logic here. The relevant sentence in Section 3 "suggests that these air masses are older than a few days (lifetime of HCOOH), but younger than 2 weeks (lifetime of C2H2)". How does that lead to the statement here that "convection 10 days before the measurement only had a minor influence" – that is, where does the value of 10 days come from? Perhaps the authors mean "convection any time in the last two weeks"?
This sentence was confusing and we changed it to: *In Sec. 3, it is suggested that*

these air masses are transported for more than a few days, but for less than two weeks. For this reason, it is not expected to see strong convective influence in the trajectories a few days prior to the measurement.

P15, L15: I'm not sure what the take-away message for the reader is. Does the fact that both models seem to identify source regions that are less "plausible" call into question the entire source attribution analysis? Are these regions really less plausible as source regions because they are characterized by low OMI tropospheric column NO2? As mentioned in connection with Section 2.3.5, some discussion of the reliability and sensitivity of these OMI data is needed. Moreover, can it necessarily be assumed that tropospheric column NO2 is a robust proxy that reflects *all* possible sources for these NMVOCs? In particular, according to Section 2.1.5, formic acid arises in part from biogenic emissions. Would those be captured in the NO2 measurements? Some further discussion is warranted here.
We rephrased and extended the last paragraph of this section, after a summary of air mass origins (as asked for by referee 2): *The comparison of ATLAS and TRACZILLA calculations of convective origin of the measured pollution species shows that there are few differences between these model results. Both models give results for the source regions and convective age of air that are broadly consistent with the measurements. Due to the numerous uncertainties, there are, however, also some results which seem to be less plausible. However, OMI NO_2, which is shown as proxy for boundary layer pollution, does not account for biogenic sources and is shown as average over 14 days prior to the measurement (see Sec. 2.4). Due to these limitations, additional pollution sources may have been overlooked in this analysis. Still, similar origins of highly polluted air masses, indicated by two independent backward trajectory models, agree with enhanced surface pollution, measured by OMI. This agreement within the anticipated accuracy of the two backward trajectory models suggests that both models use reliable schemes for convection detection.*

P15, L25-26: The EMAC HNO3 mixing ratios at the tropopause look quite a bit smaller than 0.75 ppbv to me. In addition, the writing in this sentence is very awkward; I suggest rewriting as: "... flight; they decrease to values of 0.75 ppbv at the tropopause. Simulated maximum stratospheric values are not always as high as those measured, but they agree to within ... ."
This is correct! We checked again in the data and at the tropopause in the second part of the flight, HNO_3 actually goes down to 0.5 ppbv. We changed the formulation according to the referee's suggestion.

P15, L31-P16, L2: The authors posit that the diagonal feature in the HNO3 field simulated by EMAC may originate from reactions with NO2, and the tone of the discussion seems to suggest that this may be a model artifact, especially in the latter portion of the flight. But they have made no attempt, here or in the previous section, to account for the similar feature seen in the GLORIA measurements in the first half of the flight. What is the explanation for the observed structure in HNO3?
We added a short discussion about this diagonal feature in HNO_3: *The difference in this diagonal structure between GLORIA and EMAC in the second part of the flight may result from a spatial displacement of the whole structure in the model, which is, however, unlikely due to the agreement of this HNO_3 structure in the first part of the flight, and due to the agreement of structures in pollution trace gases in the second part of the flight (see below). It is more likely that EMAC overestimates the production of or underestimates the loss of HNO_3 here at altitudes below 14 km.*

P16, L5: It would be appropriate to include a reference for the CAMS assimilation of O3.
We added the Inness et al., 2015 reference (10.5194/acp-15-5275-2015). It describes the assimilation scheme for the MACC data product, a precursor of CAMS.

P17, L15: In addition to pointing out that the EMAC C2H2 enhancement is in the same geolocation as the measured enhancement, it would be good to note that the simulated enhancement is much weaker and less extensive than the measured enhancement; it would also be helpful to add "(cyan box)" here.
We changed the sentence to: *In the second part of the flight, again a very small enhancement at the tropopause at 6:00 UTC (cyan box) is visible in EMAC, which is at the same geolocation as the enhancement in the measurements, but much weaker and less extensive.*

P17, L12-15: A point that is missing from the C2H2 discussion is the fact that EMAC completely fails to simulate the maxima in the red and orange boxes and the minimum in the magenta box, even in a relative sense.
We added the sentence: *Measured maxima of C_2H_2 are not reproduced by the EMAC model.*

P17, L25-26: The authors state that their results indicate that the meteorological fields used to prescribe transport in the simulations do not include processes relevant for the observed situation. I presume that they are referring to deep convection, which is not resolved by the reanalyses, but that should be clarified. I am wondering, however, why this would be a factor only for the first part of the flight (which the sentence in question is about). According to Figure 3, as well as much of the discussion over the preceding pages of the manuscript, the second half of the flight was influenced by convection up to 150 hPa to a similar degree.
We agree that the paragraph this sentence originates from was badly formulated. We restructured the whole paragraph and tried to be more precisely.

P18, L57: The writing in these sentences is clumsy. Moreover, I'm afraid

that I don't follow the logic of the arguments. First, as mentioned in an earlier comment, both portions of the flight are characterized by high convection probabilities up to 150 hPa, so for that reason alone it doesn't make sense to focus only on the second half. Second, the authors appear to be saying that *because* the first part of the flight is strongly influenced by convection, the simulated results would not be affected by increased emissions. But that seems backwards to me – in the absence of convection, the strength of the surface emissions would be of little consequence. This discussion needs to be clarified.
Also based on the feedback from referee 2, we decided to move the sensitivity test that is discussed in Sec. 6 to the Supplementary Materials. We only provide a short summary of the quite lengthy discussion of Sec. 6 at the end of Sec. 5. For this reason, the sentences that are addressed by this comment are no longer part of the revised manuscript.

P19, L6-8: I'm not sure that it is true that GLORIA did not observe the slight enhancement in HCOOH at 6:00 UTC and 16 km. There may be a faint hint of this structure in the data. Perhaps this feature should have been introduced earlier in the discussion, e.g., P17, L16-22.
We added to Sec. 5: *In the averaged GLORIA cross sections, a small local maximum of 60 pptv is visible after 6:00 UTC, which coincides with the small enhancement in EMAC.*
However, the sentences that are addressed by this comment are no longer part of the revised manuscript.

P19, L10-14: Of course, although the increased emissions led to larger maximum values of PAN that matched the observed peak abundances better, they did nothing to improve the structure of the simulated field. I do not think that this is an unanticipated result. I would have expected background abundances of these tropospheric tracers to rise along with peak abundances in

the increased-emissions scenario. So I am slightly puzzled by the discussion in these lines, which focuses on the impact of vertical resolution on the modeled fields. Its placement in this paragraph seems to imply that the smoothing effect of the coarser resolution of EMAC, which blunts peak abundances and blurs or erases fine-scale features, is somehow responsible for the background values of PAN being too high in this sensitivity test. In fact, I think that the resolution issue is just as relevant for the baseline model run, in which background abundances were also overestimated, and it would be more appropriate to move the discussion about it to Section 5.
We added the discussion of overestimated background VMRs and the resolution issue to Sec. 5. However, the sentences that are addressed by this comment are no longer part of the revised manuscript.

P19, L16: The possibility that model/measurement discrepancies may be partly attributable to emission sources not represented in the inventory used in these EMAC runs is mentioned. As I noted in connection with Section 2.3.1, which emission inventories were used in these simulations is a critical piece of information that has been omitted from the manuscript.
We added the missing information to Section 2.3.1. However, the sentences that are addressed by this comment are no longer part of the revised manuscript.

P19, L18: That the meteorological reanalyses do not resolve local deep convection is a wellknown issue that is presented here as a finding of this study. In addition, another aspect (besides the reanalyses) that does not appear to have been considered by the authors is the convective parameterization being used for these EMAC simulations. The choice of which convective parameterization is used has been shown to have a substantial impact on modeled trace gas distributions.
For convection, we use the parameterization introduced by Tiedtke (1989) with

modifications by Nordeng (1994) as described in Tost et al. (2006). So far, this has been the best choice for our EMAC simulations. We will add this information to the EMAC description.
In addition, in the discussion of Sec. 5, we now refer to the large uncertainties of convection parameterizations used by EMAC, as reported by Tost et al. (2006).

P19, L23: In my opinion, the statement that this study discusses "the first measurements of HNO3, O3, PAN, C2H2, and HCOOH in the center of the AMA UTLS" is too broad. While that may be true for some species of the species listed, it is not true for all of them. This statement should be qualified in some way, e.g.: first airborne measurements, or first measurements by GLORIA.
We formulated more precisely: *This study discusses the first simultaneous airborne measurements of HNO_3, O_3, PAN, C_2H_2, and HCOOH in high spatial resolution in the center of the AMA UTLS.*

P19, L29-30: This study is not the first to show that PAN is efficiently transported to the UTLS by deep convection, as is implied by the wording in these lines.
We changed the sentence to: *These measurements and their analysis confirm that PAN, a precursor of O_3, is efficiently transported upwards by convection, and transported for a longer time in the tropopause region, as shown earlier by Glatthor et al. (2007), Fadnavis et al. (2015), and Ungermann et al. (2016).*

P20, L6-8: Some of the discussion here is appearing for the first time in this manuscript. I do not think that it is appropriate to introduce new concepts in a section entitled "Conclusions".
We moved this sentence to Sec. 3 and referenced this thought only briefly here.

P20, L23-25: As noted earlier, the fact that EMAC overestimates tropospheric background mixing ratios is not unique to the increased-emissions scenario – it

was also the case for the baseline run, and increased emissions are expected to affect background as well as peak abundances. The same comment regarding vertical resolution applies here as well.
We added the aspects of overestimated background VMRs and resolution to the discussion of the baseline run, while the original sentence has been omitted in the revision of the manuscript.

P20, L31-34: These sentences are poorly written. "enhancements" are not transported upward – pollution is transported upward, leading to enhancements in the UTLS. Likewise, a "region" is not transported "around the tropopause" – the measured air masses in that region are transported. And I'm not sure what is meant by "around the tropopause".
We changed the sentence to: *Some pollutants have been transported into the upper troposphere by convection within days before the measurements, while one part of the observed air masses remained at UTLS altitudes for a longer time.*

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-321, 2020.