GENERAL COMMENTS

In this study, Mengze et al. present an analysis of observed (airborne) vs modeled ethane, propane and methane trends for the period 2006-2016. They estimate a global emission of ethane of 19.28 Tg/yr. Their results show trends for upper tropospheric and stratospheric ethane, propane and methane.

The paper is generally well written and contributes to the scientific understanding of ethane, propane and methane trends globally. I recommend this paper for publication after major revisions.

My biggest concern is that authors jump into conclusions too fast in some sections. There is also a lack of information regarding the model simulation setup. There are multiple places where it is hard to distinguish if the authors are referring to modeled results or observations, making the reading a bit confusing. Also, almost all figure captions (in the manuscript and the supplemental material) should be improved by adding information regarding the legends, and the type of information shown (see specific comments below). Lastly, even though this study shows results for propane and methane, these compounds are barely discussed in the text and their results are not even mentioned in the abstract. I wonder if somehow the title should be changed to clarify the readers that ethane is the main compound discussed in the paper and just a few propane and methane results will be shown.

We thank the reviewer for the helpful comments, suggestions and discussion. We have addressed all points raised in the revised manuscript. The original comments from the reviewer are marked as bold in black, and our replies are in blue.

More information regarding model setup has been now added, and the wording of the figure captions improved. Details are given below.

Indeed the paper is focused primarily on ethane rather than methane and propane but the latter two molecules are used as less and more reactive comparators respectively. To make this focus clearer we have added the following sentence in the Abstract to clarify this point: "The model simulations, and methane and propane observations provide additional information for understanding northern hemispheric ethane trends and emissions, which is the primary focus of

this study." We have also changed the title to "Northern hemispheric atmospheric ethane trends (2006-2016) with reference to methane and propane"

The revised manuscript will be uploaded soon.

SPECIFIC COMMENTS

Section 3.2

1. **There is no comment on the most significant feature of Figure 1, which is the much stronger and different seasonality of ethane for EUR compared to the rest of the regions. Why are concentrations so high for this region and why the peak shows up 1-2 months prior compared to NAM and whole NH?**

The reviewer has raised many interesting scientific questions and discussions. According to the journal's author guidelines "Any interpretation of data is outside the scope of regular articles." We therefore limited our interpretation. Nevertheless, in answer to the point raised we now add more information and discussion.

Figure 1 shows the ethane mole fraction in the upper troposphere which can be influenced by both surface air and stratospheric air in the vertical, and by adjacent regions through advective transport. The observed upper tropospheric ethane concentration therefore doesn't necessarily reflect perfectly the surface emissions. One possible explanation for higher ethane over EUR is that at similar flight altitudes (~10km), upper tropospheric air over EUR is influenced less by the in-mixing of stratospheric air than over NAM or ASI (e.g. Asian monsoon, etc.). Support for this explanation can be seen in the stratospheric ethane concentration, ASI has higher ethane concentrations than EUR, probably due to more stratosphere-troposphere exchange. As our dataset, which is limited in time and space, doesn't provide enough information to confirm this, we have not speculated further on this point.

To make mention of this point we now add the following text:

"Ethane mole fractions show a stronger and different seasonality in EUR compared to the other regions. One possible explanation for this is relatively less influence by the in-mixing of stratospheric air over EUR."

2. **How does stratospheric ethane lack of general seasonality from this study compares to others?**

To the best of our knowledge, there is no study on stratospheric ethane seasonality based on in-situ aircraft measurements with which we can compare. Normally aircraft campaigns sampling in the stratosphere are only for short time periods and therefore do not cover a whole year or multiple years. Other observations of stratospheric ethane are based on FTIR techniques which report the total ethane column. Such studies have reported a lack of seasonality in that stratospheric ethane (column). For instance, Helmig et al., 2016 (cited in our manuscript) showed UTLS ethane column (8-21km) above Jungfraujoch exhibited little evidence of seasonality (see their Fig 1c).

We now add the following text to the manuscript to capture this point.

"There is little seasonality evident in the ethane mole fractions in the stratosphere. Since stratospheric aircraft measurement campaigns are generally of short duration (several weeks), a direct comparison to previous data is not possible, however, vertical column data obtained by ground based FTIR for 8-21km reported by Helmig et al., 2016 also showed no clear seasonal variation."

Reference:

Helmig, D., Rossabi, S., Hueber, J., Tans, P., Montzka, S. A., Masarie, K., Thoning, K., Plass-Duelmer, C., Claude, A., Carpenter, L. J., Lewis, A. C., Punjabi, S., Reimann, S., Vollmer, M. K., Steinbrecher, R., Hannigan, J. W., Emmons, L. K., Mahieu, E., Franco, B., Smale, D., and Pozzer, A.: Reversal of global atmospheric ethane and propane trends largely due to US oil and natural gas production, Nature Geoscience, 9, 490-495, 2016.

3. **Why does NAM show such seasonality? Any insights of the reasons behind it?**

Given that the data shown in Figure 1 are the average of 10 years thus the trends can be assumed to be robust seasonal patterns. In the case of NAM upper troposphere, the high springtime values in the Northern Hemisphere closely match that of the whole Northern Hemisphere so we may

interpret this as the hemispheric optimum between high emissions in winter and highest removal in summer. For the stratospheric NAM there is a peak in the summer months. This may be caused by the injection of ethane into the stratosphere from intense biomass burning episodes that occur periodically in summer generating pyrocumulus clouds that can inject emissions into the relatively low-lying stratosphere.

Lines 219-221.

1. **Figure S3 does not indicate the region that the uncertainties correspond to (e.g. NAM, EUR). If it is for the whole globe, it needs to be stated. Again, captions and descriptions in the text should be improved.**

We now add the corresponding regions in the descriptions of Figure S3 and improved the captions and descriptions of other figures.

2. **".. and model optimizations (section 2.2)". I couldn't find the description of the model optimizations in section 2.2. Check or add this information clearly. Use the same terms/words, so that is clear and easy for the reader to find this information. This is extremely important to better understand Figure 2.**

We now add more description of model optimizations in section 2.2, the text reads as follows:

"We further optimized modeled ethane mole fractions for each emission sector (indicated as "opt" in the later figures and "optimized" in the later texts). The model optimization is done by increasing the emissions of each input emission sector by 45%. We found that the root mean squared error (RMSE) between the modeled and observational ethane mole fractions for the whole dataset was the minimum by 45% increase in the input emissions."

Line 222-224. Map with location and time frames of sample collection is needed.

We now include the spatial segregation plot as a map in the manuscript showing the delineation of NAM, EUR, ASI and tropospheric, stratospheric air samples (see below). While it is not possible to display the time variable in this plot we make the data file including altitude, latitude,

longitude and time available at https://doi.org/10.5281/zenodo.6301729 and refer to it in the text. We also add the caveat that the region designated must not correspond to the source region, only the geographical location of the data points.

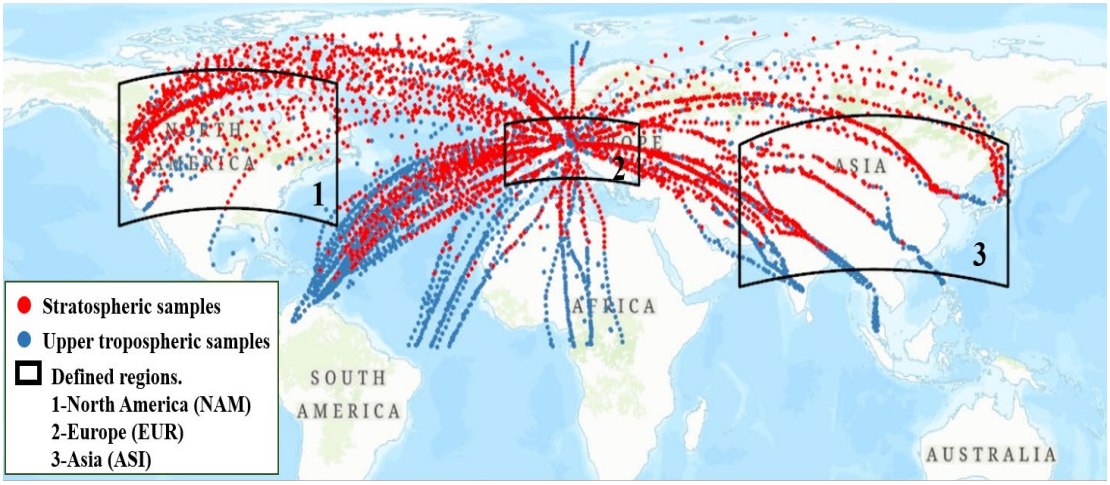

Figure 1. Geographical locations of aircraft samples (distinguished as upper tropospheric samples and stratospheric samples) and spatial segregation.

Lines 224-225. How were these observations selected? Which are those observations selected?

This sentence is indeed confusing. We meant that observations were selected or filtered by measurement location and regions of interest (NAM, EUR, ASI, ROW). We used all the available observations, none was removed any prior to analysis. To avoid confusion, we now delete this sentence.

Figure 2 – Panel a) I am surprised by seen that the FEF opt contribution is almost similar to RES opt and how little BIB opt contributes from 2006-2014. Also, more information of model setup is needed. Specifically a table with emission inventories used, global and regional (NAM, EUR, etc) totals per sector will be helpful.

We used CAMS-GLOB-ANT v4.2 emission inventory data for each model sector, and then applied the same optimization factor (1.45) to each sector in order to match the observation. Fugitive emission (FEF) has been shown in previous studies to be one of the major sources of global ethane (e.g. Helmig et al., 2016). Parts of oil and gas emission were included in FEF in

the emission inventory that we used in this study, and other parts were included in ENE. In this study, we use model simulations to validate the observational trends, and assume that the order of contribution to each sector from this inventory is correct or close to reality and only upscaling is required. Note that the trends from this study are determined from aircraft observations at ~10km height, where air samples do not only reflect surface emissions, therefore, the sectoral contribution from this study may not match exactly with the studies using ground measurements. We now add the following text in section 3.3.1:

"Interestingly the FEF opt contribution is comparable to RES opt, which highlights the importance of fugitive emissions to the global ethane budget as has been previously noted by (Helmig et al., 2016)."

Table 1 shows the estimated ethane emissions (Tg/yr) from this study for each sector and geographical region. We now add a column showing ethane emissions from the inventories used.

Line 267. Add the word "modeled" in "… the modeled NH upper tropospheric ethane…".

Done.

Line 269. State that SWD and TRO are modeled results.

We have stated in the beginning of that paragraph with "…top 5 contributing model sectors … are … TRO (road transportation), SWD (solid waste and waste water)…".

Nonetheless we now add the sentence "We note that TRO, SWD, and other sectors listed in Table 1 are modeled results" for greater clarity.

Lines 269. How can contributions of SWD and TRO be so high in NAM? Can these contributions be explained? Do these modeled results make sense? Also, how can SWD and TRO have such high sectoral contributions, but low contributions when considering trends from the optimized models (Figure 2).

SWD and TRO were the sectors with large contribution in CAMS-GLOB-ANT v4.2 emission inventory (a prior assumption), we input these emissions in our model, and thus high

contributions of SWD and TRO are expected. We note this in the following paragraph with "The contribution of TRO from this study is more than that of ~10% estimated by Peischl et al. (2013); Warneke et al. (2012); Wunch et al. (2016)." No independent estimation of SWD contribution was found in the literature.

Figure 2 shows the optimized sectoral contributions of SWD and TRO, but not their contributions before optimization. We only apply a correction factor (1.45) for optimizing each sector.

Line 271. Add the word "modeled" in "Figure 3 shows the modeledsectoral…"

Done.

Figure 3. Even though this figure looks really nice, it does not provide clear information on the contribution of each sector and region. The size of Whole NH, NAM, EUR, and ASI on the right side is the same for each region. On the left side, the sectors have different sizes, but there is no axis or value assigned to each contribution and also, for each sector the slice corresponding to each region is always the same. My suggestion is to change this figure for another one that shows the contributions by region as stated in lines 271-280. The way it is right now, those contributions are not clear.

To improve the clarity of this figure we now move it to the Supplement and furnish more explanatory text. The right side of the figure shows the fractional contributions of all sectors for each region, assuming 100% for each region. This is why the sizes are the same. With this figure we are trying to show the relative contributions among sectors for each region. Unfortunately, we cannot derive emissions of each sector for troposphere and stratosphere separately from our model.

Lines 283-284:

1. **"Five geographical sectors" ?? Do you mean geographical regions? Explain what do you mean by geographical sectors because mixing regions with emission sources is confusing.**

We now add a map that shows the geographical regions and the spatial distributions of aircraft samples. We use the term "geographical sectors" to differentiate "geographical regions". We

refer “geographical regions” as the locations where the aircraft samples were collected, “geographical sectors” as the regions where the emissions came from. An example: for a sample collected at NAM (geographical region), 50% of its emissions may come from ASI (geographical sector), 30% from NAM (geographical sector), 20% from EUR (geographical sector). We now add more explanation in section 3.3.2 and it reads:
“Four geographical sectors, i.e. ASI, NAM, EUR, and ROWwere included to investigate the origin of the ethane emissions (Figure 4, Figure S5). Geographical sectors refer to the regions where the emissions came from, whereas “geographical regions” (Table S1) refer to the locations where the aircraft samples were collected.”

Why can’t BIB and BIO emissions be separated by regions? Are the emissions not gridded in the model? Clarification is needed.

In principle we could separate them. However, by separating BIB and BIO more uncertainty will be added. In addition biogenic emissions are very small (almost negligible). For biomass burning a different regional division is needed, based on bio-types rather than continent/region, and therefore would have made the analysis even more difficult and uncertain. Furthermore biomass burning is only ~20% or less of the total emissions for ethane and therefore we can consider them less important compared to the anthropogenic emissions. It was not accurate in the manuscript that “AIR+BIB+BIO (as they cannot be separated into regions)”, we now change it to “AIR+BIB+BIO (combined as one sector to reduce uncertainty)”

We add the following text in section 2.2:

“It is noted that AIR, BIB and BIO were combined as one sector to reduce the uncertainty.”

Figure 4: Is Model Opt the sum of all the optimized geographical regions? The authors need to clarify this.

We add the following in Figure 2 legend where “Model_opt” first appeared in the manuscript:

“Model_opt is the sum of the optimized model contributions listed in the corresponding figure.”

Lines 285-286.

1. **"… 30%~55%, 35%~50%, 50%~65%, and 30%~40%...", the symbol "~" must be changed for "-".**

Done.

2. **Are these percentages average values from 2006-2016? Specify.**

Done.

3. **"Ethane emissions from ASI dominates the trends…" What are the model estimated ethane emissions (in Tg/yr) for ASI compared to the rest of the regions? A table with regional emissions used in the model is needed.**

In Table 1 (b) we have listed the estimated ethane emissions from different regions from this study (ASI: 7.48 Tg/yr, EUR: 2.32 Tg/yr, NAM: 1.46 Tg/yr).

Lines 288-289. Clearly state to which atmospheric layer correspond these trends. For example: "…contribution to ethane trends in the upper troposphere and/or stratosphere".

Done.

Lines 295-301. How does these results compare with other studies?

We have listed the comparison with other studies for ethane in Table 2. We have also extended the comparison with previous studies.

Section 3.3.3 has "methane" in its title, but it is barely discussed in the text.

As noted in the previous responses, the main focus of this paper is the northern hemispheric budget of ethane, which is in part elucidated by reference to methane and propane data collected in parallel. The title has been changed to reflect this more clearly. Nonetheless we now provide more details in the Method section about collection and measurement of methane. In this section 3.3.3 as well as section 3.4.2, we presented methane trends from four regions (whole northern hemisphere, NAM, ASI, and EUR) in the upper troposphere and stratosphere using high-quality dataset. Another peer-reviewed article by Zimmermann, et al., 2020 has used the same aircraft methane dataset and discussed methane more in detail in combination with atmospheric model.

Thus we want to avoid publishing overlap with this article. We now add some literature comparison for methane in the text.

Reference:

Zimmermann, P. H., Brenninkmeijer, C. A. M., Pozzer, A., Jöckel, P., Winterstein, F., Zahn, A., Houweling, S., and Lelieveld, J.: Model simulations of atmospheric methane (1997–2016) and their evaluation using NOAA and AGAGE surface and IAGOS-CARIBIC aircraft observations, Atmos. Chem. Phys., 20, 5787–5809, https://doi.org/10.5194/acp-20-5787-2020, 2020.

Section 3.3.4

1. **This section should be named differently because it discusses the comparison of the model to ONLY two observations. It's current name gives the false idea that there was a model simulation using ground station data. I suggest calling it: "Model results compared to two ground stations".**

Done. We have now removed this section and moved the figures into Supplementary.

2. **The conclusion from the last sentence should be erased. It cannot be concluded just by comparing 2 stations that the model provides realistic values for ethane surface level. A thorough analysis of multiple surface stations must be done before jumping to that conclusion.**

Done. We have deleted this sentence.

3. **I suggest completely getting rid of this section and mention that the use of this model results for surface-level ethane should be studied in the future.**

We have combined this section with section 3.3.5 and noted in the manuscript that further model results for surface ethane is needed in the future.

Section 3.3.5

1. **Erase the term "budget" in the title and text because this implies sources and sinks of a certain compound and here, only global emission totals are presented.**

Done.

2. **Thus, modify the title of this section.**

Done.

Lines 334-335. How does the authors conclude that the stratosphere has a minor contribution for observed trends? Authors need to explain clearly why location biased trends can be discounted, even if it is for the same reasons that were discounted for the upper troposphere.

We didn't discuss or conclude that the stratosphere has a minor contribution for observed trends in lines 334-335. We concluded that the location bias has a minor contribution to the observed trends. This point is now addressed more clearly in the following text:

"The stratospheric climatology (Figure 6 (a)) indicates the contribution from sampling location for the observed stratospheric ethane trend. It varies more than the tropospheric one, but it is again a minor contribution, so that location biased trends can be discounted."

Lines 335-336. Add "over the Whole NH" before "shows a general…" to specify which panel the authors are referring to.

Done.

Lines 336-339. Suggest the following change: "The peak in 2010 is not seen at regional levels (NAM, ASI, EUR, Figure 6 (b)(c)(d)), which suggestglobal upward transport of the upper tropospheric ethane emissions (peaking in 2010-2011) into the stratosphere." No analysis was made to clearly indicate that there was an upward transport from the upper troposphere, therefore, it cannot be stated as such.

Done.

Line 339. Suggest the following change: "The second peak in 2013 can be due to the regional…". Same reason as explained in the previous comment.

Done.

Section 3.4.2 Add (if any) references and comparisons to other studies.

Done.

Lines 396-397. Specify which "current inventory" the authors are referring to.

Done.

TECHNICAL CORRECTIONS

Figures 1, 2, 3, 4, 6, and 7: Explanation of legends and terms used in the figures are missing in the caption.

We have added explanation of legends and terms in the figures.

There are two figures with number 5 (Lines 640 and 645).

We have changed the figure numbering.

Caption Figure 3. Clearly state sectoral contributions are model results.

Done.

Caption of Figure 4 should be revised because not all results correspond to "Optimized geographical sector contribution" as stated in the first sentence (Observations are not included in the caption). Also, clearly state that the optimized geographical contributions are model results.

Done.

Suggest the use of a different color scheme for the EUR, NAM, etc regions in Figures 4 and 7 to avoid confusion model optimizations in figure 2 and figure 6.

We tried to make the same category (e.g. model sector, observation) with same color, for both upper tropospheric and stratospheric ethane. We have used 8 colors in figures for the upper troposphere, finding different colors for the stratosphere is therefore difficult.

Sections 3.3.3 and 3.4.2 have exactly the same name. Indications to "troposphere" or "stratosphere" should be added to avoid confusion. The same goes for their corresponding figures in the supplemental material.

Done. We have changed the titles for sections 3.3.3 and 3.4.2.