General comments

Li et al. present long-term atmospheric ethane, propane, and methane data obtained from airborne observation in the UTLS region in the Northern Hemisphere through IAGOC-CARIBIC project for the period 2006–2016. The authors also present simulation outputs from the EMAC model to interpret the contributions of different source regions or different source sectors to the observed ethane trend.

The manuscript is generally well written. The airborne observational ethane, propane, and methane data in the UTLS region is unique and useful because there is only a limited platform that can measure with high precision such trace gases regularly in this region - perhaps only IAGOC-CARIBIC (this project) and CONTRAIL project (e.g., Machida et al. 2008; Sawa et al. 2015). The observational data will certainly be reused by many researchers in this field, e.g., validate the emission inventories (as the authors did), interpret the atmospheric transport and chemical processes in the UTLS region, through the comparison with model simulations.

However, the current manuscript is lacking the information on methods (e.g., model simulation setup, trend analysis, measurement uncertainty), making the readers a bit confused and difficult to interpret the presented results (and dataset). For example, I couldn't find how the emission inventories are optimized from prior emission inventories to match the observation data. Under the lack of such important information, it is difficult to judge these model outputs are worth publication or not, despite the uniqueness and potential usefulness of the observational data itself. Also as for the measurements, the authors just cited the previous (~10 years ago) publications for ethane and propane measurements (Baker et al. 2010) and for methane measurements (Schuck et al. 2009), but I would recommend the author to describe information on the data quality control over the whole analysis period (2006–2016). For example, how the long-term stability of the standard gases has been maintained? Such information should be necessary for the readers and data users when using the data and interpreting the long-term trend of these trace gases over the decades, especially for ethane and

propane, whose measurement uncertainties are comparable with the discussed observed trends.

Considering the above, I would recommend this paper for publication after major revisions.

We thank the reviewer for the helpful comments, suggestions and discussion. We appreciate that the reviewer finds our data unique, precise and useful for future studies. 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.

We have added more information on model setup and optimization in section 2.2, measurement details (data collection, quality control, instrumentations, measurement procedures, uncertainties, etc.) in section 2.1, and trend analysis in section 2.3. We have also included a section to address the limitations and implications of our study.

The revised manuscript will be uploaded soon.

Specific comments

Title: I would recommend that the authors reconsider the title because this study present almost only the ethane trends and the data is obtained in the Upper Troposphere – Lower Stratosphere (UTLS) in the northern hemisphere, not in the global.

We now change the title to "Northern hemispheric atmospheric ethane trends (2006-2016) with reference to methane and propane". We also add the following texts 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."

L10 "global": Same as the comment in the title.

Done.

L66: Please clarify "the global ethane trend" and "long-term global ethane datasets". Emissions or atmospheric mole fractions? Also, what is "local influences"?

We now revise the text as follows:

"Therefore, to determine the global ethane trends in terms of mole fractions and emissions with greater certainty, long-term global ethane datasets from observations and model simulations with minimal influences from local sources (e.g. observations at higher altitudes) are required"

L84 "global": Same as the comment in the title.

Done.

L105-108: Please describe the calibration scales used in this study. Just "please see the references" is inconvenient for the readers who expect to use the dataset for this study. Also please mention the long-term stability of calibration gasses, as well as long-term reproducibility of the sample measurements.

We now add more information in section 2.1 (see text below) regarding data collection, analytical protocols, measurement and calibration procedures, and quality control, etc.

"Methane, ethane, and propane were measured with a HP 6890 GC coupled with a flame ionization detector (FID) with a polymer Porapak Q 3/4" column (10 ft, 100/120 mesh) installed in a single oven. Nitrogen (N_2, purity 99.999%) was used as carrier gas at a constant flow rate of 50ml/min. The FID was operated at oven temperature of 220°C with flow rates of synthetic air of 250ml/min and hydrogen of 80ml/min. Water vapor in samples was removed by passing through a drying tube at the start of the analysis. The calibration standards were ordered from NOAA (for methane), and the National Physical Laboratory (for ethane and propane) which are certified against World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) program scale, and they are regularly renewed within every three years which warrants the stability of calibration gases. Three injections of calibration standards were made in between samples of each flight sequence in order to maintain the quality of measurements and reduce uncertainty. "

L124: Cl is important for the modeling of ethane, methane, and propane. The authors need to discuss the issue more in the section 3 (e.g., L345-350 and L374-375).

Chlorine chemistry is more important for the stratosphere than for the troposphere. We add the following text to address it:

"The loss of ethane in the stratosphere by reaction with Cl radicals is about 40 times more than that by OH radicals (reaction rate of ethane with Cl is about 400 times faster than with OH at 250K (Atkinson et al., 2001), and stratospheric OH is about ten times more abundant than stratospheric Cl (Li et al., 2018)), whereas the ethane loss in the troposphere by Cl is negligible compared with by OH due to the small amounts of tropospheric Cl (OH:Cl around 10,000) (Gromov et al., 2018; Lelieveld et al., 1999)."

L129-135: I think this part is very important for readers to know what the model optimization setup is. However, I don't think it is well written at this stage.

We now add more information on model optimization in section 2.2. Now the section 2.2 reads:

"The ECHAM/MESSy Atmospheric Chemistry (EMAC) model is a numerical chemistry and climate simulation system that includes sub-models describing tropospheric and middle atmosphere processes and their interaction with oceans, land, and human influences (Jöckel et al., 2010). It uses the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes. The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al. (2006)). For the present study, we applied EMAC (ECHAM5 version 5.3.02, MESSy version 2.55.0) in the T63L47MA-resolution, i.e. with a spherical truncation of T63 (corresponding to a quadratic Gaussian grid of approx. 1.8 by 1.8 degrees in latitude and longitude) with 47 vertical hybrid pressure levels up to 0.01 hPa (~80 km). The model has been weakly nudged towards the ERA5 reanalysis data of the ECMWF (Hersbach et al., 2020). The chemical mechanism comprises methane, alkanes, and alkenes up to C4, ozone, odd nitrogen, some selected non-methane hydrocarbons (NMHCs), heterogeneous reactions, etc. In total, 310 reactions of 155 species are included in the model. The photolysis rates are calculated following Sander et al. (2014). No chlorine chemistry is included in the model. To account for realistic emissions, the CAMS-GLOB-ANT v4.2 emission inventory data is used for model simulations (Granier et al., 2019; Guevara et al., 2020). In this study, we have included 13 emission sectors (shown in Table 1) which are BIO (biogenic emission), BIB (biomass burning), AWB (agricultural waste burning), ENE (power generation), FEF (fugitives), IND (industrial processes), RES (residential energy use), SHP (ships), SLV (solvents), SWD (solid waste and waste water), TNR (off-road transportation), TRO (road transportation), and AIR (aviation). It is noted that AIR, BIB and BIO were combined as one

sector to reduce the uncertainty. AIR is not shown in Table 1 as its contribution is negligible. It has been shown by multiple studies that the ethane emissions due to fossil fuel combustion are strongly underestimated in the emissions database (Guevara et al., 2021; Pozzer et al., 2020; Helmig et al., 2016). In this work, we therefore increased the anthropogenic emissions of ethane of a factor of 2.47 to match (for the year 2010) the total amount suggested by Pozzer et al. (2020) although the value used in this study (~11.8 Tg/yr) slightly underestimates the measured mole fraction as shown in Pozzer et al. (2020) (13.2 Tg/yr). We further optimized modeled ethane mole fractions for each emission sector (referred to 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 at a minimum after a 45% increase in the input emissions. The input ethane emissions from natural and anthropogenic sources are presented in Table 1, together with description for each sector and optimized sectoral emissions (will be discussed in the Results and Discussion section).

In this study, two types of ethane trends were presented with the model simulation: (1) constant meteorology and constant emission (hereafter called climatology), sampled at the IAGOS-CARIBIC sampling location with S4D algorithm (sampling in 4 dimensions) described in Jöckel et al. (2010). Any trends (or changes) detected in this simulation would be caused by differences in sample location and timing. (2) real meteorological conditions from ECMWF and the adjusted emissions described above, sampled at the IAGOS-CARIBIC sampling location with S4D algorithm (Jöckel et al., 2010). "

L129-132: Did you increase not the fossil fuel combustion only, but the total anthropogenic emissions? If so, what is the justification of this? Then did you increase the total anthropogenic emissions to match which Tg/yr in Pozzer et al. (2020) (13.2 or 15.3 Tg/yr, as in Section 1)?

We increased all the emission sectors including anthropogenic, biogenic and biomass burning emissions by 45%. As our model and emission input have been examined many times before, we only did this simple optimization. And the aim of our study is not to improve model prediction, but to use model to explain the changes in observed trends. We now add more explanation in section 2 to clarify this.

L132-135: How to estimate the "45%"? Without the information of the optimization method, it is difficult to interpret the model results hereafter.

We found that the root mean square error (RMSE) is minimum between model and observations when the model results are increased by 45%. We use this as the method for model optimization. We now add the information in section 2.2.

L144 "Prophet": As long as I see the main figures (e.g. Fig. 2, 4, 5), I wonder how this algorithm can capture the discussed long-term trend over the decade. The results show noisy inter-annual variations, not averaged smoothed trend. Maybe the author also need to show the raw data obtained in this study, as done in Fig. S1.

Our aircraft sampling is different to surface measurement in terms of temporal and spatial distributions of samples. Normally to estimate a trend of surface measurements, one needs to make sure the time series data are continuous (e.g. weekly, monthly), when not, missing values need to be filled by various approaches (e.g. using the same value as the previous value). This is possible and the uncertainty is small for surface observations, as they are often regular (e.g. weekly for NOAA VOCs) and with little missing values. Whereas it is more challenging for aircraft measurements, because (1) the time gap between two flights was not always exactly the same, (2) the sampling locations are rarely or impossibly the same (latitude, longitude, altitude). From what we have seen in the previous studies, trend analysis was often done by applying a simple linear fit. Such linear fit approach has shortcomings, e.g. (1) the estimated trend is influenced by the time period of choice (for a same dataset, the trend of one season may look much different as the trend of one year), (2) it doesn't capture inter-annual variability, (3) it can be influenced by outliers, etc. To improve these shortcomings, we used the "Prophet" algorithm which is an open source software and suitable for non-continuous time series data. It is robust to missing data and the influence from outliers is minimized. It is a non-linear model with seasonal effects included. Thus we think this algorithm will work better with our data than a linear fit. Due to the flight frequency of our aircraft measurements which is often one or two intensive days of measurements in a month, the temporal distribution of our data is ca. monthly, instead of being evenly distributed through every day. The "Prophet" algorithm outputs a value for trend for each measurement, and no outputs if no measurement exists on a day. Also, in the case of our data, the sampling locations may be different even if samples are collected above the same regions (EUR, ASI, etc), which results in different sample trajectories and abrupt changes in

trend may occur. That is why our trend results do not look smooth. We are aware that our trend analysis still has shortcomings, but we believe it is a better and more suitable approach for our data in comparison with linear fit. We now add more information in the manuscript to clarify the advantages and limitations of our trend analysis method (section 3.5). We also add ethane mole fractions from aircraft observations in figures in Supplementary Information (SI) together with the trends from Prophet. These figures clearly show that Prophet captures the trends well.

L150-162 "changepoint_prior_scale": Please explain the definition of the parameter; otherwise, the readers cannot understand what the uncertainty interval represents.

We now clarify it in the section 2.3.

L155-158: If so, please show the comparison of fitting between NOAA algorithm and Prophet algorithm in the supplement Fig. S1?

For Iceland surface ethane observation data from NOAA, both algorithms estimated the trend of ~900 ppt for Jan 2006, NOAA algorithm estimated the trend for Jan 2007 of ~1400 ppt, and Prophet algorithm estimated it to be 1350~1500 ppt. For the years 2008-2015, both algorithms estimated the trends to be around 1400 ppt. As we don't have the results from NOAA algorithm from Helmig et al., 2016 and including the figure from Helmig et al., 2016 may potentially bring copyright issues, we only describe the agreement between two algorithms in the manuscript, and provide the reference of Helmig et al., 2016 for readers who are interested in looking into more details.

L165-205: I wonder why the estimated ethane trend of this study is so different from those of previous studies (same as in L237-245, L364-371). Calculating the trend for another periods would be useful to directly compare the results (e.g., 2009-2014), but I also wonder how the "Prophet" can robustly estimate the trend, due to the large inter-annual variations (as commented in L144). Also, the authors need to explain how they separate the air mass origin between troposphere and stratosphere before presenting the result (e.g., in Method). If the criteria of troposphere and stratosphere is set to be "PV = 2", as in L204, please describe it earlier.

Previous studies provided ethane trends based on surface observations which are largely influenced by local ethane sources and sinks. Our aircraft sampled air at high altitude (~10km) on a global scale. This dataset is extremely useful to understand trends in regions where

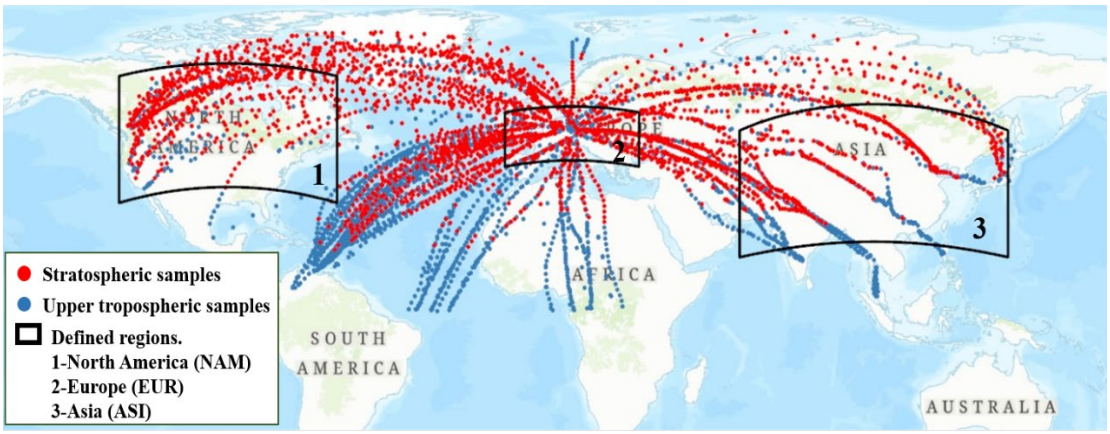
hydrocarbon emission inventories are poorly known. It is noted that the observed trends from aircraft measurements can be influenced by surface emissions such as convected plumes, or changes in stratosphere-troposphere exchange. Thus it is expected to see different ethane trends compared with other surface observations.

We now calculate the ethane trend for another period (2007-2014) to directly compare with other studies, and the values are added in Table 2.

We now add more explanation on the criteria of the tropospheric and stratospheric air samples in section 2.1.

L207: According to Fig. S2, the sample number obtained in Rest of the world (ROW) is dominant more than half of all data in the stratosphere. Then I wonder where the air samples are collected in the ROW and wonder if the current air mass classification is valid for interpreting the trend?

Our aircraft flight paths are from Germany in Europe to the other continents/regions defined in our study (ASI, NAM, ROW). ROW samples are mostly collected above the oceans (see newly added Figure 1 in the revised manuscript and below) and have broad spatial distributions. These samples have diverse trajectories and thus cannot be easily classified. Indeed ROW is dominant for stratospheric samples and also for upper tropospheric samples in the year 2010 and 2011 when we observed a spike in the whole NH upper tropospheric ethane trend. We now add figures for ethane, methane and propane trends at ROW (troposphere and stratosphere) in SI. We don't observe a spike at ROW for 2010-2011. We suspect that the spike for the whole NH was due to the uneven temporal distributions of the origin of samples. There was no ASI samples in 2009, thus the 2009 trend was influenced by ROW, NAM and EUR. In 2010 and 2011, there were more ASI samples and that was the time period when ASI trend reached peak, thus the whole NH trend shows a spike at that time. This reveals the limitations of our trend analysis, we add a new section to discuss the limitations of our study.



L213-215: Is the seasonality in ASI not statistically significant? It is interestingly showing the same peak timing with UT in June. Is it potentially indicating the intrusion of tropospheric air masses into stratosphere due to Asia summer monsoon (e.g., Park et al. 2007; Xiong et al. 2009).

This is a good point. We modify the text in section 3.2 and it reads now:

"In contrast, the stratospheric ethane mole fractions do not show strong seasonality, except that NAM has a seasonal trend with 3-month later shift compared to the upper tropospheric NAM trend, and stratospheric ASI ethane shows the same timing peak in June with upper tropospheric ASI ethane which potentially indicates the intrusion of tropospheric air masses into the stratosphere due to Asian summer monsoon (Xiong et al., 2009; Park et al., 2007)."

L228-234: It seems saying about the discrepancy of the inter-annual variations between observation and model climatology, not that of the whole trend for 2006–2016. Also, the observational trends estimated by Prophet show very large inter-annual variations and thus it seems difficult to compare the long-term trends by this analysis.

The climatology simulation was performed to test the importance of the sampling location on the trend estimation. The climatology, by definition in its construction, does not present trends nor interannual variability. Therefore, any trend observable by the sampling of the climatological data along the flight tracks is purely due to the aircraft position and not from changes in atmospheric composition. Therefore, the fact that no noticeable trends are present confirms that the frequency and location of the sampling did not influence the estimation of the trends, while those are due to changes in the atmospheric composition (by e.g. changes in emissions, in

meteorology and hence transport, or in chemical transformation). We now add the climatology trends for each region in the figures.

Regarding Prophet, please see our replies to a previous comment (L144).

L264-270: I would suggest the authors describe the source sub-category information in the method section. And then explain which sub-category emissions are optimized.

We have added descriptions of each emission sector in the method section and explained how the optimization was done.

L272-274: Same as the general comment and specific comment (L129-135 and L264-270). I wonder how the author optimized the emission inventories.

We now explain more about the model optimization in the section 2.2.

L283-284 "AIR+BIB+BIO": It is said they cannot be separated into regions. Then how are they included into the model simulation?

These sectors were kept global, i.e. they represent the global emissions. In principle, we could separate them. However, more uncertainty will be added by separating BIB and BIO. In addition, biogenic emissions are very small (almost negligible). For biomass burning (BIB), a more detailed analysis could be performed, although the regions should be based on fire type/burned biomes (e.g. savanna, boreal forest, and others), rather than geographical area, and therefore we preferred to avoid and focus on the geographical area. For AIR, it is difficult to assign singularly to a region. 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 now add more clarification in section 2.2.

L295-301 & L364-371: How the authors calculate the long-term trend and uncertainties? Please describe it somewhere.

We now describe it in section 2.3 as follow:

"In this study, we also calculated simple linear trends (hereafter as linear trend to distinguish with the trends derived from "Prophet" algorithm) within a time period as follows:

$$\text{Linear trend} = (c_{End}-c_{Start})/(t_{End}-t_{Start}) \quad (1)$$

where t_{End} and t_{Start} represent the end and start date and time of the target time period, c_{End} or c_{Start} is the mole fraction of trace gases (ethane, methane or propane) at the end or start date and time. ”

L304-307: But as presented in L264-270 and Fig. 2b-d and, the pronounced peak in 2010-2011 for NAM, ASI, and EUR are mainly explained by SWD (solid waste and waste water) and TRO (road transportation). Are these results consistent with the sentence here? As seen in Fig.2a, the peaks in FEF and RES are actually seen in 2010-2011. Are these happened in ROW then?

The sectoral emissions (e.g. FEF, SWD, TRO, etc) were optimized based on the input emission inventory. We see that the optimized modeled emissions (sum of all sectoral emissions) captured the similar ethane trends as from observations, whereas not for the peak in 2010-2011. We now include trends at ROW (in SI) to better understand this. We see a peak in 2010-2011 for ROW trend as well. Because about half of the samples were collected above ROW, the whole NH trend may be largely influenced by ROW, thus its trend and sectoral emission signature might be different from these at regional scale (NAM, EUR, ASI). We point this out in section 3.5 of limitations.

L316-317: Which figure are you referring to?

It refers to figure 5 which shows the observations and model simulations for ethane at surface stations. Now this figure has been moved to the Supplementary.

L321-L333: Again how to estimate the optimised emissions? Just by comparing with the observation with model output with “try and error” method? Needs more explanation. Also I wonder how meaningful to present the emission amount under 2 decimal places. As long as I understand, the presented optimization method is very rough and seems not to be able to estimate within such small amount of emissions.

We now explain how the model optimization was done in the manuscript, and reduce the decimal of emission amount.

L329-330: But, please be noted that the atmospheric ethane, methane, and propane in the UTLS region can be largely influenced by atmospheric transport and chemical destruction processes.

We now add this in section 3.5 to discuss limitations of our analysis. Section 3.5 reads:

"Despite the usefulness, uniqueness and high quality of our datasets, several limitations of our study should be noted. (a) representativeness of the presented trends. Although our flight sampling is frequent and covers a large area of the NH, the spatial and temporal distributions of our samples are not even. This may cause the trends being influenced by a specific regions where more samples were collected. (b) chlorine chemistry is missing in the EMAC model. Chlorine radicals are much more abundant in the stratosphere than the surface, thus the change in chlorine plays a great role in the observed trends. (c) our samples were collected in the UTLS region and can be influenced by atmospheric transport (e.g. troposphere-stratosphere exchange), surface sources, and chemical destruction processes. Therefore, the trends represent the net effects of these factors making the interpretation on a single factor difficult. (d) PV choice of identifying upper tropospheric and stratospheric samples. In this study, we used PV=2 to define the tropopause, whereas other approaches exist. It is shown that on large space and time scales in the extratropics, the WMO tropopause corresponds rather well to a surface of constant potential vorticity (PV), although there exist systematic differences on smaller scales (Stohl et al., 2003; Wirth, 2000). (e) trend analysis tool "Prophet". One needs some experience with the algorithm to choose and tune some parameters to get the best results for individual datasets, i.e. settings for our dataset may not be suitable for other datasets. (f) model optimization. Our EMAC model and input values for sectoral emissions have been examined and optimized in many previous studies, therefore, in this study we simply increased each emission sector by 45% to match the observations. The aim of model simulations is to better understand the contributions from each emission sector, rather than improving the performance of model and emission inventories. (g) interpretation of results. This article is designed as a data description article to provide high quality and useful dataset for scientific use. There are many interesting features in the presented trends to be explored, however, it is beyond the aim of this study.

Implications. (a) observations of ethane, methane and propane were often restricted at regional scale or short-duration. We presented a long-term (10 years) airborne observations of ethane, methane and propane in the UTLS region at northern hemispheric scale. This dataset is unique and can be used to examine long-term troposphere-stratosphere exchange, chemical and dynamical changes in the UTLS region, and improve model performance. To our best knowledge, such long-term aircraft observations are only available from IAGOS-CARIBIC project (our study) and CONTRAIL project (Machida et al., 2008; Sawa et al., 2015).(b) The

"Prophet" algorithm is an open source software, and suitable for non-continuous time-series datasets. Unlike the commonly used linear fit approach for trend analysis in other studies, the "Prophet" algorithm is robust to missing data and the influence from outliers is minimized. It better captures the inter-annual variability and is not influenced by the time period of choice. (c) other analysis approaches such as machine learning techniques can be used on our dataset to enlarge the spatial and temporal distributions. Combining our dataset with space-borne observations will provide a better view of global distributions and trends of trace gases."

L336-337: It is not clear why the peaks in 2010 not seen at regional levels even though it was caused by global upward transport of the upper tropospheric ethane emissions. Maybe the contribution of ROW is important? Also see the comment in L207.

We now add ROW trends in SI. We do see a peak in 2010 at ROW, which may suggest that the contribution of ROW is important. We now add this point in the manuscript.

L345-350: Any possible explanation that the Cl in the stratosphere has increased after 2013? As mentioned here, the discrepancy between model and observation since 2013 is mainly seen only in ASI. Does it mean the Cl destruction process is only influential in the stratosphere in ASI?

The reviewer has raised 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. The discrepancy between model and observations since 2013 is seen in ASI but not NAM or EUR, we suspect that it is caused by regional signature in ASI. It has been shown that the global and Asian emissions of some chlorinated compounds (e.g. CFC-11, CHCl3) were increasing in 2012-2016 (Fang et al., 2019; Rigby et al., 2019), and strong chlorine chemistry associated with Asian outflow in the UTLS region in 2013 (Baker et al., 2016). Thus the Cl destruction of ethane in ASI may explain the discrepancy between model and observation. We now add more texts in the manuscript to clarify these points:

"Previous studies have shown that the global and Asian emissions of some chlorinated trace gases (e.g. CFC-11, CHCl3) were increasing during 2012-2016 (Rigby et al., 2019; Fang et al., 2019; Montzka et al., 2021), and a strong chlorine chemistry was associated with Asian outflow

in the UTLS region in 2013 (Baker et al., 2016).This could be an explanation for the larger discrepancy between model and observation since 2013 in ASI."

References:

Baker, A. K., Sauvage, C., Thorenz, U. R., van Velthoven, P., Oram, D. E., Zahn, A., Brenninkmeijer, C. A., and Williams, J.: Evidence for strong, widespread chlorine radical chemistry associated with pollution outflow from continental Asia, Scientific reports, 6, 1-9, 2016.

Fang, X., Park, S., Saito, T., Tunnicliffe, R., Ganesan, A. L., Rigby, M., Li, S., Yokouchi, Y., Fraser, P. J., and Harth, C. M.: Rapid increase in ozone-depleting chloroform emissions from China, Nature Geoscience, 12, 89-93, 2019.

Rigby, M., Park, S., Saito, T., Western, L., Redington, A., Fang, X., Henne, S., Manning, A., Prinn, R., and Dutton, G.: Increase in CFC-11 emissions from eastern China based on atmospheric observations, Nature, 569, 546-550, 2019.

Montzka et al. (2021). A decline in global CFC-11 emissions during 2018− 2019. Nature, 590(7846), 428-432. https://doi.org/10.1038/s41586-021-03260-5

L373-374: But why it is not seen in methane? Needs more explanation.

Different from ethane and propane, methane has more complex emission sources. These sources can originate from different regions, which makes the interpretation more difficult. As replied to previous comments, the whole NH trends can be largely influenced by ROW, thus we suspect that maybe some unique sources of methane at ROW were changing differently as at other regions.

L374-375: Any possibility of ethane destruction by Cl increased after 2013? For example, the emissions CFCs from eastern mainland China has increased after around 2013 (Rigby et al. 2019; Montzke et al. 2021), which may have affected the increase of Cl production in the stratosphere and may have caused the larger destruction of ethane in ASI?

As replied to a previous comment, we add the following text in the manuscript:

"Previous studies have shown that the global and Asian emissions of some chlorinated trace gases (e.g. CFC-11, CHCl3) were increasing during 2012-2016 (Rigby et al., 2019; Fang et al., 2019; Montzka et al., 2021), and a strong chlorine chemistry was associated with Asian outflow in the UTLS region in 2013 (Baker et al., 2016).This could be an explanation for the larger discrepancy between model and observation since 2013 in ASI."

L379 Data availability: Please include the PV values, as well as other observed meteorological information, into the dataset for each air sample. These are important for readers to know how the air masses are classified into troposphere or stratosphere. Also please add the measurement information in more detail (e.g., calibration scales, precision, measurement number, standard deviation (if exists), and reference lists of the measurements). For the simplicity and usability, I would even suggest that the tropospheric and stratospheric data sheet can be combined into one sheet with timeseries, just by adding a flag column like "trop" or "strat".

We now change the dataset as the reviewer suggested.

The new link for the dataset: https://zenodo.org/record/6301729

Figure 2: Gray lines are not plotted in Fig 2b–d

We now add gray lines for all the subplots.

Figure 3: I'm not sure how this figure could be useful for readers. It is said the vertical scale of each source sector represents the source contributions, but the contributions to each geographical region shouldn't be the same.

We realize that this figure can cause confusion, therefore, we moved it to the Supplement and add more explanatory text. The right side of the figure shows the contributions of all sectors for each region, assuming 100% for each region which is why the sizes of them are the same. We were 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.

Figure 7: First I wondered why the authors need to show the "AIR + BIB + BIO_opt" here, with geographical sector contributions. After reading the sentence L283-284, I found it is said "AIR+BIB+BIO" cannot be separated into regions. But, then I wonder how they are included into the model simulation.

We now remove the “AIR + BIB + BIO_opt” from those figures.

Technical corrections

L19: As already commented, I don’t think the two decimal place is meaningful under the current rough model optimization method.

Done.

L56: Tg/yr2 ?

We present the ethane emission growth rate which in unit of Tg/yr.

L394: Same as L19.

Done.

Figure 5: There are two Figure 5!

Done.

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