This manuscript reports on measurement data for methane, ethane, and propane from the IAGOS-CARIBIC commercial aircraft sampling program. Data from the upper troposphere/lower stratosphere are compared with outputs from the EMAC model. The conclusion from the disagreement in this comparison is that there are discrepancies between inventory and actual emissions of ethane.

This paper follows up on a series of other recent studies that have investigated the burden and atmospheric ethane, for instance [Aydin et al., 2011; Simpson et al., 2012; Franco et al., 2015; Franco et al., 2016; Hausmann et al., 2016; Helmig et al., 2016; Dalsoren et al., 2017; Tzompa-Sosa et al., 2017; Dalsoren et al., 2018; Monks et al., 2018; Tzompaâ□□Sosa et al., 2019; Angot et al., 2021].

I see a plethora of issues with this manuscript that dampen my enthusiasm for this work.

Overall, the manuscript does not seem all that carefully proofread, having an unusual high number of punctuation and formal errors. The concept of deriving a linear trend determination for the 2006-2016 window seems ill directed, given that it has been demonstrated that there was a decline in atmospheric ethane to approximately 2009, and then an uptick of ethane after that. Ignoring this trend reversal and then averaging trends over these two contrasting periods of atmospheric ethane changes does not seem to make much sense.

I am skeptical of the quality of the observational data and that the authors did not address any of the very obvious discrepancy of their findings with the prior published literature (see listing above). Year-to-year variability and trends in the ethane mole fraction are ways higher than reports in any of these prior publications. This points towards technical inconsistencies in the sampling program, possible analyte losses in the analytical system, or calibration issues.

We thank the reviewer for the forthright and critical comments which are all addressed in the text below. The original comments from the reviewer are marked as bold in black, and our replies are marked in blue.

We agree that there are differences with other published articles and we have clearly indicated it in the manuscript, for example in section 3.1 we have tried to summarize how the observed ethane trend from our unique long-term airborne dataset compares to other studies. Some

differences are clear and the comparison is straightforward. This makes the work of interest, particularly so when the data are compared to our best-modeled version of comparable data. Our observational trends have been evaluated with the atmospheric chemistry model (EMAC), the agreement between observation and model simulations supports the consistency of our observational data, and there is no reason to question the quality. One significant difference between our study and previously published works is the sampling location. Our aircraft sampled air at the tropopause region, whereas most other studies used ground-level measurement. This makes our reported trends and yearly variability inherently less dependent on sources directly upwind as is the case for a surface sampling site. 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 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 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 and have large year-to-year variability. 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 analysis method (section 3.5).

There are many possible reasons to explain the differences with other studies (will be discussed in detail in the later replies). We respectfully disagree with the reviewer's statement that "This points towards technical inconsistencies in the sampling program, possible analyte losses in the analytical system, or calibration issues." Our analytical procedures have been well documented in publications and have been consistently applied using the same measurement system for many years (e.g. Schuck et al., 2009 for methane measurement, Baker et al., 2010 for ethane and propane measurements). These papers are also referenced in the manuscript. Operational lab notes are maintained and any measurements with potential interference from analytical issues were excluded. Our data has been examined by various journals, scientists, and no concerns about the quality of our data was reported. Data from the same instruments and projects are published, see e.g. Thorenz et al., 2017, Li et al., 2018, and Baker et al., 2016. The modeled data based on an updated emission inventory for ethane shows similar trends which lends further support for the dataset quality.

While we accept it is a good scientific practice to question any analytical method when the results obtained differ from previously held expectations, however, in this case we are confident that the analytical method and quality controls applied therein make this dataset robust. To strengthen this point we have now added more details in the manuscript about instrumentation, analytical and operational procedures in section 2, and limitations of our study in section 3.5.

Nevertheless, we understand the point of the referee, and in this manuscript, we have described the data and have an initial interpretation with comparison with model results, although we would be grateful to see further interpretation/studies based on our data, we have raised this point in the limitations and implications section.

The revised manuscript will be uploaded soon.

References:

Schuck, T. J., Brenninkmeijer, C. A. M., Slemr, F., Xueref-Remy, I., and Zahn, A.: Greenhouse gas analysis of air samples collected onboard the CARIBIC passenger aircraft, Atmos. Meas. Tech., 2009.

Baker, A. K., Slemr, F., and Brenninkmeijer, C. A. M.: Analysis of non-methane hydrocarbons in air samples collected aboard the CARIBIC passenger aircraft, Atmos. Meas. Tech., 2010.

Thorenz, U. R.; Baker, A. K.; Leedham Elvidge, E. C.; Sauvage, C.; Riede, H.; Velthoven, P. F. J. van; Hermann, M.; Weigelt, A.; Oram, D. E.; Brenninkmeijer, C. A. M.; Zahn, A.; Williams, J., Investigating African trace gas sources, vertical transport, and oxidation using IAGOS-CARIBIC measurements between Germany and South Africa between 2009 and 2011, Atmospheric Environment, 2017.

Li, M., Karu, E., Brenninkmeijer, C. et al. Tropospheric OH and stratospheric OH and Cl concentrations determined from CH4, CH3Cl, and SF6 measurements, Nature Clim Atmos Sci, 2018.

Baker, A. K., C. Sauvage, U. R. Thorenz, P. van Velthoven, D. E. Oram, A. Zahn, C. A. M. Brenninkmeijer & J. Williams, Evidence for strong, widespread chlorine radical chemistry associated with pollution outflow from continental Asia, Scientific Reports, 2016.

Unfortunately, this work does not provide sufficient insight into the analytical protocols and quality controls to evaluate this question. Has the sampling apparatus and analytical lab ever been audited by the World Calibration Centre for VOCs [WCC-VOC, 2021], which most labs with global monitoring programs have been subjected to during the past ten years? If so, then the results from this audit should be presented and critically evaluated.

Our measurement apparatus and the papers describing it predate the establishment of the "World Calibration Centre for VOCs" and as yet we have not been offered the opportunity to undergo an analytical audit. If this airborne measurement program restarts, as is hoped, then we would gladly participate. In lieu we have now added more information in the manuscript about the analytical protocols and quality controls applied. The procedures described in the previous published papers (Schuck et al., 2009, Baker et al., 2010) have been applied throughout this dataset so that long term data consistency and quality are assured. Certainly, having a world

calibration center to ensure comparability between different measurement methods and sites is important to address atmospheric budgets on a global scale. We now make this point in the text to advertise this point as follows:

"When comparing surface and airborne datasets from multiple locations to assess global atmospheric changes it will become increasingly important to ensure comparability of data quality, a process that has begun through the grounding of a World Calibration Center for VOCs, although this dataset predates this initiative."

The reason for the difference in surface versus upper tropospheric trends needs more discussion/justification. Obviously, this defies the findings from column FTIR observations, e.g. [Helmig et al., 2016] who showed that, while lower in magnitude, lower and upper tropospheric trends were approximately in sink.

Helmig et al., 2016 showed the ethane total column from FTIR observations at two stations: Jungfraujoch (for column 8-21km, and 3.6-8km) and Lauder (total column). Some of their trends are different from the upper tropospheric trends from our study. There are several possible explanations for this:

(1) Trends at the two stations at Jungfraujoch and Lauder do not represent the trends of the whole atmosphere (neither surface nor upper troposphere nor stratosphere). This has been shown in Figure 3(a) of Helmig et al., 2016 that large difference in ethane growth rates at different ground stations is observed (from -50 to 50 pmol $mol^{-1}y^{-1}$).

(2) FTIR observes the total (or partial) column of ethane. Considering the short lifetime of ethane, the observed trend from FTIR will likely be weighted more with the trend at near surface regions, whereas the observational data in our study do better represent the upper troposphere and stratosphere.

(3) Our aircraft sampled air at high altitudes 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, or

changes in chemical destruction processes, and these can be reasons for the large year-to-year variability and trends observed from our study.

We now add more discussion in the manuscript to address this issue, though we must limit ourselves to the stated scope of the journal which is "Any interpretation of data is outside the scope of regular articles".

Specific comments:

Abstract: The source/program of the measurement data should be mentioned. The model that was used should be mentioned.

Done.

Line 14-16: I am surprised that the oil and gas sector is not clearly listed as an emissions category given its significant contribution to ethane emissions (see the above listed literature that overwhelmingly focused on the oil and gas emissions sector)?

We used anthropogenic emission sectors from CAMS-GLOB-ANT v4.2 ethane emission inventory for model simulation. Oil and gas are included in the FEF (fugitives) and ENE (power generation) sectors, however, the emission inventory does not combine the oil and gas as one emission category. To our knowledge, the oil and gas emission studies from above-listed literature were not retrieved from model and emission inventories, but from the ratio of methane and ethane from observations, e.g. Helmig et al., 2016, Aydin et al., 2011.

19: Given the overall uncertainty in emissions, four significant figures for the global ethane emissions do not seem justified.

We categorized our samples into four regions, based on the number of samples we have in order to make sense in statistics. We agree that zooming into smaller regions would help better to understand global emissions and reduce uncertainty.

23: Instead of using the term 'discrepancy' it would be more meaningful to use a term that clearly identifies if inventory emissions are over- or under-estimated.

Done.

30, 32, 41, etc: There are many, many cases throughout the manuscript where a comma should be placed within a sentence.

Commas have been added to sentences on lines 30, 32, 41, etc to ease reading. We have carefully rechecked and improved the spelling. The professional editorial team from the journal will further assist with proofreading before this manuscript is published.

56: Please explain what the number in parenthesis stands for.

Done.

103: Are methane and CO2 indeed measured with an ECD?

Methane and CO2 are measured with FID, now we have revised it in the text.

105-109: Details on the calibration scale, source of calibration standard should be provided, and how and when the scale was verified against the World Meteorological Organization Global Atmospheric Watch program that is used by the international community for global observations of ethane.

We have provided more information on the calibration scale, calibration standard and other quality controls and protocols in the manuscript. The text reads:

" 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 (N2, 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."

139: Replace ‘of’ with ‘by’.

We don’t find any “of” in line 139 or anywhere near it.

156: Insert ‘the’ before NOAA.

Done.

174: …related to emissions from the ….

Done.

188: What is the reference year for the % trend determination?

We used Feb 2006 as the reference, now added in the text.

193/194: No need to put parentheses around the time intervals.

Done.

204: Please provide more explanation on the criteria that were used for differentiating upper tropospheric air from stratospheric air and associated uncertainties.

The criteria was using PV values to differentiate samples. In the meteorological community, PV is preferred over the thermal definition of the tropopause (the lowest level at which the temperature lapse rate decreases to 2 K km^{-1} or less). PV is conserved under adiabatic and frictionless conditions, and it can capture tropospheric folds. Other tropopause definitions such as one based on ozone (O3) or an O3 to CO ratio also show a sharp transition at the tropopause. All these definitions give broadly consistent delineation of the tropopause although the chemically defined tropopause tends to be somewhat lower than the thermal one. PV was preferred to ensure that tropopause data was always available even if a particularly chemical measurement was not available.

As noted in the review of Stohl et al., 2003, on large space and timescales, in the extratropics, the WMO tropopause corresponds rather well to a surface of constant potential vorticity (PV) (e.g., Reed and Danielsen, 1958; Danielsen, 1968; Hoerling et al., 1991), although there exist systematic differences on smaller scales (Wirth, 2000). We now refer the reader to this review and provide more details about how the differentiating was done in the text.

References:

Stohl, A., et al. (2003), Stratosphere-troposphere exchange: A review, and what we have learned from STACCATO, J. Geophys. Res., 108, 8516, doi:10.1029/2002JD002490, D12.

Reed, R. J., & Danielsen, E. F. (1958). Fronts in the vicinity of the tropopause. Archiv für Meteorologie, Geophysik und Bioklimatologie, Ser. A, Meteorologie und Geophysik, 11(1), 1-17.

Danielsen, E. F. (1968). Stratospheric-tropospheric exchange based on radioactivity, ozone and potential vorticity. Journal of Atmospheric Sciences, 25(3), 502-518.

Hoerling, M. P., Schaack, T. K., & Lenzen, A. J. (1991). Global objective tropopause analysis. Monthly Weather Review, 119(8), 1816-1831.

Wirth, V. (2000), Thermal versus dynamical tropopause in upper-tropospheric balanced flow anomalies. Q.J.R. Meteorol. Soc., 126: 299-317. https://doi.org/10.1002/qj.49712656215

208, Figure 1 caption: Ethane is presented in mole fraction units (ppb) and not as a concentration. Use of the term 'concentration' is therefore technically not correct.

We have revised it.

219: Wording is not correct. Uncertainties are presented in Figure S3.

We have revised the wording.

246 - 262: A more convincing approach would be to first evaluate and optimize the modeled trends using available and much more abundant (and likely better quality) surface data (possibly high elevation mountaintop data, e.g. Jungfraujoch, Mauna Loa, Summit) or FTIR column records, and then move on towards evaluating the upper troposphere trends.

We agree that this is a valid approach to evaluate model and emission inventories. However, we did not proceed in this way as our EMAC model has been evaluated for ethane using surface data, e.g. Pozzer et al., 2021. The ethane emission inventory used in our study has been also optimized with ground observations from our previous study (Pozzer et al.,2020).

References:

Pozzer, A., Reifenberg, S., Kumar, V., Franco, B., Taraborrelli, D., Gromov, S., Ehrhart, S., Jöckel, P., Sander, R., Fall, V., Rosanka, S., Karydis, V., Akritidis, D., Emmerichs, T., Crippa, M., Guizzardi, D., Kaiser, J. W., Clarisse, L., Kiendler-Scharr, A., Tost, H., and Tsimpidi, A.: Simulation of organics in the atmosphere: evaluation of EMACv2.54 with the Mainz Organic Mechanism (MOM) coupled to the ORACLE (v1.0) submodel, Geosci. Model Dev. Discuss., 2021.

Pozzer, A., Schultz, M. G., and Helmig, D.: Impact of U.S. Oil and Natural Gas Emission Increases on Surface Ozone Is Most Pronounced in the Central United States, Environmental Science & Technology, 54, 12423-12433, 2020.

249: It shouldn't really be a surprise that the model performs better for methane, given the much lower variability of methane in time and geographically, and given its 40 times longer lifetime? Please avoid subjective terms such as 'better', and instead give a concrete quantitative evaluation.

We have revised it. Now it reads:

"The initial model results underestimate ethane mole fraction by about 45%, whereas the model estimation is closer to observation for methane with the same model and observation dataset (Zimmermann et al., 2020)."

259: The oil spillage doesn't really matter for ethane. It would make more sense to provide emissions/leakage/spill estimates for natural gas, including ethane.

We have deleted this statement.

262: Insert 'the' after although.

Done.

283: Please explain/spell out all abbreviations when they are used for the first time.

The abbreviations for regions (ASI, EUR, etc.) are explained in the first paragraph of section 3.2 and Table S1. All abbreviations for emission sectors are now added in section 2.2 and explained in Table 1.

285: Doesn't this result contradict the findings from other prior literature, e.g.[Franco et al., 2015; Franco et al., 2016; Hausmann et al., 2016; Helmig et al., 2016] who claim that North America is the main contributing region for the rise in global ethane post 2009?

We don't think our findings contradict previous studies. Some previous studies have indeed shown that surface ethane emissions from North America have been increasing since 2009, contributing to the rise in global surface ethane. This reported increase was observed from surface measurements, whereas the contributions of ethane from each region reported in our study (e.g. Figure 4) were simulated by the atmospheric model and emission inventory which may not include such ethane rise in North America from bottom-up approach. Our findings stated in line 285 indicate the relative contribution of air emitted from each region to the observed upper tropospheric ethane trend. We find that about 50% of upper tropospheric ethane of the whole Northern Hemisphere was transported from Asia, which makes sense considering the transport of pollutants from Asia into the stratosphere via Asian Monsoon. An example to better understand this concept: for Figure 4(b), the black line indicates the upper tropospheric ethane trends derived from samples collected above NAM, the red line (from model simulations) indicates the ethane emissions originated from ASI which influenced the black line. In this case, we see that a large fraction of ethane from samples collected above NAM was influenced by ASI emissions, which indicates that our samples and observed trends are influenced by global transport, and do not represent local emissions, and thus should not be interpreted as local ethane trends or be compared with surface observations. We now add these points in the manuscript.

295-301: As mentioned previously, I question that trend results over this time window are meaningful, given that this spans over a point in time when there was a reversal of the ethane trend.

We have presented the trends over two time spans when there was a turning point in 2010-2011 in the fourth paragraph of section 3.3.1, i.e. trends between 2006-2010, and 2010-2016. In section 3.3.3, we provided an overall trend for 2006-2016, calculated as the difference of ethane mole fraction in 2016 and 2006, divided by 10 years: $(c_{(2016)} - c_{(2006)})/10yr$. We now add this in section 2.3 for better clarification. This approach minimizes the influence of any short-term change in ethane sources or sinks over that time period. As we cannot confirm what are the drivers of trend reversal (location bias with emission plume, or change in stratosphere-

troposphere exchange, etc.), we preferred to present the observed trends in both ways (one as an overall trend, the other one to separate the reversal trend).

314-318: Again, 'well' is a subjective qualifier. Please provide a concrete quantitative evaluation.

We have now revised it and moved this section to Supplementary as suggested by the other reviewer.

321: Unreasonable number of significant figures.

Done.

334: …is again a minor ….

Done.

338: What do you mean by 'upper tropospheric ethane emissions'? I am not aware of ethane emission sources in the upper troposphere?

We have removed the word "emissions".

353: Figure 6 shows time series data, not trends.

Figure 6 shows the stratospheric ethane trends which were derived from observational data by a statistical model "Prophet" in Python (see section 2.3). The trends shown in Figure 6 are not the observed ethane mole fractions from aircraft sampling. Our trends are equivalent to the orange line in Fig.1(b) of Helmig et al., 2016. We now also add the time series of observed mole fractions in the figures in SI.

378: Shouldn't the NOAA data source be included here as well?

Now we add the NOAA data source.

396: Please name the inventory.

Done.

402: 'Trend' is not a suitable term here.

Done.

411: ..revised …

Done

605: The listed ethane sources do not add up to the total that is listed at the bottom?

The sum of BIO, BIB and Table 1(a) is equal to the sum of BIO, BIB and Table 1(b), which is 19.28 Tg/yr. Note that Table 1(a) and (b) are only for anthropogenic emissions.

Figure 2: To the very best of my knowledge, the ethane spike that is reported in 2010 has not been seen in any other data products, including FTIR column observations and surface measurements, including those taken at mountain sites. This makes me quite skeptical of these results. In my opinion, this requires a much more thorough evaluation/verification, including through careful consideration of available other data sources. Can this be explained by interannual differences in the fraction of stratosphere-troposphere exchange that the aircrafts encountered during their sampling? That would then raise the question or representative each year's aircraft data are?

As replied to the previous comments, there are differences between our aircraft measurements and ground observations. Indeed aircraft observations at UTLS region can be influenced by the change in stratosphere-troposphere exchange and sampling locations. Ground-based trends are easier to interpret as they are dominated by the upwind activities. High altitude trends are more difficult due to wider source footprint, stochastic convection events, and stratospheric mixing. This is why here we have applied a global model for comparison. One may indeed question the representativeness of aircraft data as it is dependent on sampling location. To address this, we have examined the sampling location bias in the manuscript (method described in section 2.2) by simulating ethane mole fraction for each aircraft sampling location with constant meteorology and constant ethane emission as model input (so called "climatology").We find this bias has little influence (discussed in section 3.3.1 and 3.4.1). Compared to surface observations, aircraft observations provide unique information to express a more homogenized global trend which is inevitably a variable composite of multiple global sources. We have deposited our data including sampling date and time, location (longitude, latitude, altitude), pressure, together with observed and modeled trace gas mole fractions on a publicly accessible repository. This guarantees the

reuse of our data in future studies to examine/be aware of the representative of aircraft data. We also add a new section (3.5) to discuss the limitations of our study including the representativeness of aircraft data.

Figure 3: The size of the four colored emission regions does not seem to agree with the data in Table 1.

We realize that this figure can cause confusion, therefore, we moved it to the Supplement and added 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 5: Graph shows time series data, not trend results. The 2011 propane spike is very significant, representing a 50% increase from prior years. This dramatic change is much larger than year-to-year variability seen in other data products. This seems unreasonable and makes me question the quality of the underlying data. This calls for much more documentation on the quality control procedures and a convincing demonstration of the long-term stability of the sampling and analysis program.

Figure 5 shows stratospheric ethane trends which were derived from observational data by a statistical model "Prophet" in Python (see section 2.3). We have interpreted the spike in 2011 as a plume of oil and gas emission. We also see a peak in 2010-2011 for ROW trend as well (figure added in SI). Because about half of the samples were collected above ROW (SI), the whole NH trend may be largely influenced by ROW. If we exclude the spike from the trend, we see that the change between 2006 and 2016 is small and comparable with other studies. Observing a spike by aircraft does not mean that the aircraft observation is wrong or is incompatible with a global trend since the observed ethane trends match reasonably well with trends from atmospheric chemistry model simulations. As requested, we have documented our measurement procedure further. These have been consistently applied so that we are confident of the quality of our data.

Fig 6/7: I would like to see information on how the data were treated, averaged, binned, and how uncertainty ranges were determined. These figures are the pivotal example for my

skepticism of the data quality and the knowledge/understanding of the authors of the atmospheric behavior of these NMHC. For the ASI panel, there is a more than 50% drop of ethane in the observations from 2013 to 2015. So, that's a reduction of ethane to less than half in two years. No other data set that is referred to in the above-cited literature shows year-to-year changes of that magnitude. A concentration drop like this is highly unreasonable, most likely impossible. The best explanation that I can think of is that there were sampling or measurement problems that the authors did not become aware of and investigate. Or that there were strong differences in the fraction of stratospheric versus tropospheric air that were sampled each year and that the differentiation wasn't accomplished properly?

Regarding the drop of stratospheric ethane above ASI shown in Figure 6/7, it should be noted that there is a very large uncertainty associated. Such large uncertainty is seen only for ASI, very likely due to the smaller amount of sampling numbers. As replied to previous comments, whole NH trends can be influenced by the regions with more samples collected (we include this point in the discussion of limitations). Trend variability within only two years may be even more influenced by that. This is also the reason why we only split our data into four regions for the presentation and not into more. Secondly, Asia is a region with strong stratosphere-troposphere exchange, and as well as surface emissions. Thirdly, if we compare the model and observation of stratospheric ethane above ASI, we see a relatively good match before 2011, and for other regions like NAM and EUR, the modeled trends match well with observation thorough the entire time period, this indicates that our measurement is not wrong, so we exclude the possibility of measurement problems. The discrepancy between model and observation for ASI after 2014 may indicate changes in stratosphere-troposphere exchange, or changes in ethane sources or sinks, which are not captured by model or emission inventory.

Figure 8: Similar reservations as already explained for Figure 6/7: I can not believe how propane would drop in the stratosphere by a factor of more than four from 2006 to 2011. This defies every other data set on atmospheric NMHCs that I have seen in the published literature.

To our best of knowledge, there is no study reporting stratospheric propane trends which covers the time period as our study, therefore, we kindly ask the reviewer to provide the literature which shows different stratospheric propane trends. As replied to the previous comments, stratospheric trends are not only being influenced by surface emissions, but also atmospheric transport

(stratosphere-troposphere exchange), and chemical destructions. For example, chlorine radicals are abundant in the stratosphere and are important oxidants for stratospheric propane, ethane and methane, whereas chlorine concentration on the surface is much smaller (~100 times lower than in the stratosphere) (Gromov et al., 2018). We now add this point in the manuscript, and we think comparing surface trends with stratospheric trends needs to be done in a careful manner.

As for the surface propane and ethane, Warneke et al., 2012 summarized a series of studies and showed that surface propane mole fractions have been decreasing at the rate of -4.6%/year in the LA basin (1960-2010), and at -3%/year in London (1998-2008), whereas ethane had even higher decreasing rate over the same time period and locations. Although surface trends of ethane or propane should not be directly compared with stratospheric trends, the fast drop of surface ethane and propane in some regions may play a role in the observed stratospheric trends.

Reference:

Gromov, S., Brenninkmeijer, C. A. M., and Jöckel, P.: A very limited role of tropospheric chlorine as a sink of the greenhouse gas methane, Atmos. Chem. Phys., 18, 9831–9843, https://doi.org/10.5194/acp-18-9831-2018, 2018.

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