



Comment on **essd-2021-246**

Anonymous Referee #3

Referee comment on "Northern hemispheric atmospheric ethane trends in the upper troposphere and lower stratosphere (2006-2016) with reference to methane and propane" by Mengze Li et al., Earth Syst. Sci. Data Discuss., <https://doi.org/10.5194/essd-2021-246-RC3>, 2022

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.

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.

L10 “global”: Same as the comment in the title.

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

L84 “global”: Same as the comment in the title.

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.

L124: CI 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).

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.

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)?

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

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.

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

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

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.

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?

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).

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.

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.

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

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

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

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?

L316-317: Which figure are you referring to?

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.

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.

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.

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?

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

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?

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".

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

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.

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.

Technical corrections

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

L56: Tg/yr² ?

L394: Same as L19.

Figure 5: There are two Figure 5!

References

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