

Earth Syst. Sci. Data Discuss., referee comment RC2
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Comment on **essd-2021-246**

Anonymous Referee #2

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-RC2>, 2021

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; Tzompa-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.

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.

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.

Specific comments:

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

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)?

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

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.

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

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

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

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.

139: Replace 'of' with 'by'.

156: Insert 'the' before NOAA.

174: ...related to emissions from the

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

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

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

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.

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

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.

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.

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.

262: Insert 'the' after although.

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

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?

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.

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

321: Unreasonable number of significant figures.

334: ...is again a minor

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

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

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

396: Please name the inventory.

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

411: ..revised ...

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

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?

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

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.

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?

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.

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