REVISION MENGZE ET AL.

GENERAL COMMENTS

After carefully revising all the responses to the comments made to reviewers, I see that the authors have done a thorough revision of the previously submitted manuscript. Information regarding the model setup and important descriptions in the methodology were added. Also, numerous changes to the discussion section were made. The revised manuscript was greatly improved by the new information, clarifications, and changes to the title.

That being said, I have significant concerns regarding the model setup as described in the revised manuscript and concluded that their modeling approach has serious flaws, which are then reflected in the results. The authors used to run their model an ethane emission inventory (Table 1) that does not reflect the current scientific knowledge of 1) the sectoral distribution (percent-wise), 2) the magnitude of the emissions by sector and region, and 3) the geographical distribution of emissions for the represented period (2006-2016). I will further explain these:

1 and 2) Some emission sectors and their associated emission estimates are unrealistic (e.i. SWD-solid waste and wastewater, TNR, TRO). Just as the author responded to line 269 in response to reviewer comments, "No independent estimation of SWD contribution was found in the literature". And even so, authors included that sector without justification with emissions equal to other well-known sources such as biomass burning-BIB.

2 and 3) The geographical distribution of anthropogenic emissions presented in Table 1 is not based on any study, and no justification is offered for such values. Asian emissions are too high compared to North America and Europe (Xiao et al., 2008, Tzompa-Sosa et al., 2017). Furthermore, emissions from the ROW-Rest-of-the-World (excluding Asia, Europe, and North America) are estimated to contribute about ~17% to global emissions, whereas Mengze et al. report a 30% contribution, again without providing any justification.

The lack of attention to these characteristics in the model input ethane emissions ultimately diminishes the quality of the results and conclusions therein. Currently, the model simulations constitute a fundamental part of the manuscript since they were used to determine the upper-tropospheric and low stratospheric trends. It is no surprise that the model did a poor job determining regional trends (Lines 327-328 from the red-line manuscript essd-2021-246-ATC2.pdf). It is important to note that the global ethane emission in the optimized model simulation (19.3 Tg/yr) is similar to other studies (Franco et al. 2016), and this could explain why the model did a better job at determining the NH upper tropospheric trend (lines 326-327).

Overall, I can see a significant scientific value in the measurement dataset they present and the linear trends derived from the observations only. I suggest either removing all the sections and conclusions related to the model results or re-run the model with science-based emissions of ethane, methane, and propane. Additionally, it would be interesting to run the model using the trend derived from observations and see how similar or different the model is compared to observations. Major changes need to be made to this manuscript before acceptance.

Line 324. "The model incorporates all known emissions via emission inventories so any…"

Suggest changing the wording to "The model incorporates emissions from various emission inventories described in table XX". In that table, provide a list of all the names (with references) of the emission inventories used for each relevant compound (i.e., ethane, methane, propane). Also, include emission totals by region and sector for methane and propane since both compounds are mentioned in the paper (similar to Table 1).

Authors need to avoid subjective adjectives such as "well" (line 327), "good" (line 409) and instead provide values for every comparison made.

Suggest adding "in the upper-troposphere and lower-stratosphere" in the title.

Line 160 & line 490. Does the model include chemistry for other halogens, such as Br? Also, suggest adding a reference to Sherwen et al. [2016] that concludes that Cl may be an important C2H6 sink that can decrease the simulated global burden of C2H6 by about ~20%. Also, add other studies that estimated the impact of the lack of other halogens like Br.

Line 175. It seems that there is confusion between the mole fraction term and emissions. The emissions were optimized to match the observed mole fractions. Thus, "We further optimized ethane mole fractions for each emission section" is technically incorrect.

Line 179. What is the value of this minimum RMSE?

Lines 209-210. If the Prophet algorithm is designed for non-continuous datasets, why do results match with the NOAA algorithm even though it is designed for continuous datasets? Is this an expected result, or what does having similar values mean?

The entire Section 3.1 should be moved to the introduction. Also, the first paragraph of section 3.2 should be moved to the introduction. The time of collection and location of the sampling should not be in the discussion but in the introduction and methodology section.

Lines 277-278. Are there any studies that talk about a weaker mixing of stratospheric air over EUR? If so, add references.

Lines 304-307. Can authors provide values for trends due to sampling location vs. trends derived from observations to support this conclusion?

Lines 338-371. As suggested in the general comments, model sectors and model geographical sectors (section 3.3.2) contribution analyses should be deleted because sources, totals, geographic distributions, and trends do not reflect current scientific knowledge.

Lines 390-392. Having peaks in all regions does suggest regional and global increases in emissions, but not an increase in fossil fuel emissions. Thus, remove "fossil fuel" from this sentence.

Lines 412-414. This result should show the estimated emission in 2006 and the subsequent increase due to the increasing trend derived from observations. Otherwise, state the exact year at which those estimated emissions by sector correspond.

Line 541. Remove the word "budget". Also, see the previous comment on how to present this trend value. Possibly write it as "averaged" emission for the same period.

Line 545. The underestimation is only ~30% based on the optimized model emissions (13.3 Tg vs. 19.3 Tg). When re-writing this sentence, make it clear which emissions this comparison refers to.

Lines 551-552. "…was caused by fossil fuel-related emissions, likely from oil associated and natural gas sources". The conclusion of the 2010-2011 peak coming from oil and gas emissions is stated in the text (line 390) only as a possible explanation and is not based on any of the main results (see Fig 3) and discussions made in the text (lines 330, 390, 430, 478). Even figure 3 (lower panel with model optimized sectoral emissions) does not reflect this statement, as other sectors such as SWD, RES, and TRO show the same increase in 2010-2011. Therefore, this conclusion should be removed from here and from the abstract since the authors have no strong results to reach such a conclusion in the manuscript.

Lines 552-553. "The global ethane trends cannot be well simulated by advanced atmospheric chemistry modeling ". This is a strong claim that is not supported in the text. If there are clear examples of studies that have had struggles modeling ethane trends, they should be cited here and in the text so that this conclusion is consistent with the main document.

TECHNICAL CORRECTIONS

Line 174- Move "(13.2 Tg/yr)" to the end of line 172.

Line 187- Add upper case in "Real..."

Line 321. Model estimation or optimized model results?

Add the word "modeled" every time the text refers to regional trends. For example, in line 475, it should say: "The modeled regional trends..."