referee-report-1

REVISION LI ET AL.

GENERAL COMMENTS

After the second revision, the authors followed the suggestions made by reviewers and eliminated the model simulation previously included. The new manuscript focuses on the observational dataset and the trends derived from it.

This new manuscript is well written and the methods and results are presented in a clear manner. I recommend this paper for publication after minor revisions.

We appreciate a lot that the reviewer provided lots of helpful comments and suggestions through all rounds of revisions, and the quality of our manuscript has been improved a lot and it now fits the journal much better. We have now revised the manuscript according to the reviewer's comments.

SPECIFIC COMMENTS

Line 90. Suggest changing the second part of this statement to: "…, including regions without ground measurements."

Done.

Line 146-150. If the term "whole NH" will be used, it is better to change the name of ROW to RNH "Rest of Northern Hemisphere" and make sure data from the Southern Hemisphere is not included. The way it is written right now gives the impression that some data in ROW corresponds to the Southern Hemisphere, and thus, combining the four regions into one does not make sense.

Done. We have replaced ROW with RNH in text and figures.

Line 154. Change "must" for "does".

Done.

Lines 194-201. These lines should be moved to the methods section (2.1).

Done.

Line 238. Specify what you mean by “low” (maybe add a cut-off number).

Done.

Section 3.3. Add information regarding other halogens like Bromine.

Done. We have added more information and the text reads:

“In the stratosphere, the OH radical concentration on average decreases by a factor of 10 compared with tropospheric OH levels, whereas halogen (e.g. chlorine (Cl) and bromine (Br)) are more abundant and react faster with ethane, methane and propane, therefore they play a greater relative role in ethane, methane and propane oxidation (Li et al., 2018).”

Line 285. Define “large”. Provide a value for this difference.

Done.

Line 388. Since some values are negative, “growth rates” should be changed for “annual rates of change in atmospheric abundances of”.

Done.

referee-report-2

Manuscript Review for ESSD

Li et al.,: Northern hemispheric atmospheric ethane trends in the upper troposphere and lower stratosphere (2006-2016) with reference to methane and propane

This manuscript is a revision/resubmission of a previously evaluated submission to the same journal. The authors made several changes from the prior submission. Despite their efforts, in my opinion, some important shortcomings remain. Concerns by Reviewer #3 about the classification and filtering of air samples have not been fully resolved. I have also reviewed comments and revisions from the very first submission. Several of the previously articulated problems are still valid:

We thank the reviewer for the comments. We have addressed them and our replies are shown below.

As pointed out in the prior reviews and comments, there appears to have been a minimum in the atmospheric mole fraction of ethane around the year 2009-2010, with declining levels prior, and increasing levels afterwards. These trends were seen consistently in monitoring records from the Northern Hemisphere. With this having been well established, it does not seem appropriate to conduct linear trend analysis across this minimum. Obviously, the trend results will then all depend on which starting and end time one chooses before and after the minimum. Despite this having been emphasized before, the authors adhere to this analysis and dedicate a full section to this method and discussion of the results.

We understand the reviewer's point on separating the linear trend into different time periods, instead of across all time. This is seen in some literature, e.g. in Helmig et al. (2016) and Franco et al. (2015). As the reviewer commented, the selection of starting and end time will influence the trend results. This is true for either applying one linear trend across all time (the way we did in our study), or multiple time periods (as the reviewer suggested). Both Helmig et al. (2016) and Franco et al. (2015) have estimated ethane trends over Jungfraujoch, but the authors chose different starting and end time and their trends are different. Helmig et al. (2016) used May 2014 as the ethane turnover time, whereas Franco et al. (2015) used Jan 2014. The increasing

ethane trends after turnover in 2014 was estimated as 4.2%/yr by Helmig et al. (2016) and 4.9%/yr by Franco et al. (2015), showing a ~17% difference. The choice of the turnover time of ethane trends is often somewhat subjective, lacking objective criteria. Due to these reasons, we don't think separating trends into different time periods will significantly improve the robustness of linear trend analysis. There are also some studies using only one linear fit across all time, e.g. Simpson et al. (2012) (published in *Nature*) applied one linear fit for ethane trend over 1985-2010. Although their study didn't cover the ethane turnover time in 2014, we can still see by eye from Fig.4 of their paper that the ethane trends during 1985-2000 vs 2000-2010 are very different. As the scope of this journal is to present an overview of the datasets, instead of data interpretation, we therefore applied linear trend analysis across all time to provide a general overview of our datasets. The trend analysis results in Figs.3 and 6 show non-linear trends with higher temporal variability and are less influenced by the choice of starting and end time. Our linear and non-linear trend analysis helps readers to understand our dataset from different aspects.

The second problem I see, and this is a big one, is that to the best of my understanding and evaluation, it seems that the data that are used for attempting these trend analyses are ways to variable in its source and representation of a particular altitude (troposphere versus stratosphere) as well as latitude for justifying this analysis. The authors conducted filtering of their data based on polar vorticity (PV) and continental areas. I don't think this worked. The data that are displayed in Figures 2 and the derived trend results in Figure 3 and 5 defy the atmospheric behavior of methane, ethane, and propane that has been well established in other literature. Ethane is a relatively long-lived gas in the atmosphere, with its lifetime being on the scale of or longer than the mixing time scale in the Northern Hemisphere troposphere.

Therefore, it does not seem plausible that upper troposphere ethane would behave so completely different than ethane in mid to lower tropospheric records. I have pulled out some data examples in the figures below, i.e. from Mauna Loa Observatory, HI, USA, Jungfraujoch, Switzerland, and Summit, Greenland. There are many more that can be easily reproduced from records in the World Data Center for Reactive Gases (EBAS/Nilu). I have intentionally chosen examples from high altitude monitoring stations as those are deemed to best describe the behavior in the lower free troposphere. The important take home messages are that seasonal cycles of ethane are very regular. Year-to-year changes are relatively small, just a few percent, if at all. These are all deemed high quality observations, reflecting the seasonal and year-to-year changes at a fixed site. However, there is a very substantial latitudinal gradient, with ethane dropping steeply from mid-latitudes to the tropics (see Figure 4).

The data presented in this paper look very different, showing a much higher variability and much higher year-to-year changes, which the authors interpret as atmospheric concentration trends and attempt to relate to surface emissions changes. I don't think this is appropriate and correct. I think it is much more likely that the variability seen in these data to a large extend reflects differences in the origin of the air masses that were sampled, both in altitude (tropospheric versus stratospheric contribution) and with latitude. What the authors interpret as trends likely are year-to-year changes in the fraction of collected samples having one sort of these influences versus another.

We would like to emphasize again the difference between surface and lower stratosphere. Many surface observation sites are remote from any known anthropogenic or natural sources to represent the background conditions. At these sites, the ethane, methane or propane trends are largely influenced by the variation of their sink, i.e. OH radicals. As OH radicals are high in summer and low in winter, the trends of ethane, methane and propane observed at ground are low in summer and high in winter. This explains why surface seasonal cycles of ethane are regular and their yearly variation are small. Whereas it is very different for the upper troposphere/lower stratosphere (UT/LS). The air at UT/LS region has a broad range of age (often 0-3 years) (Bönisch et al., 2009), which means the air represents a mixture of air parcels emitted 0-3 years ago. The transport time from surface to UT/LS is about 3 months which is similar to ethane's atmospheric lifetime. Orbe et al. (2015) has shown that compounds within the lower stratosphere of the northern hemisphere are predominately influenced by the Brewer-Dobson circulation, not the northern mid-latitude emissions (~ 10%). As we have stated clearly in the manuscript, region designated does not correspond to the source region, only the geographical location of the data points. Due to these reasons, we don't think surface trends are comparable with UT/LS trends.

As for the reviewer's comment on the use of PV filter, it is a commonly used method in the community and has been examined many times in the literature, e.g. Sprenger et al. (2007); Sprenger et al. (2003). Air samples with PV larger than 2 are commonly defined as stratospheric samples. Creating a new filter for stratospheric samples is out of our scope and will make our results difficult to be compared with other studies.

I don't think the paper is publishable as it is. Since this journal focuses on presentation of data, a way to proceed may be to just publish the data by themselves without the trends analyses.

Another, totally different approach might be to focus instead on the analysis of ratios

of the hydrocarbon over methane. One could also utilize the propane/ethane ratio as an indicator of photochemical age and possibly derive a fractional contribution of stratospheric air on the samples and then compare that with the PV rating. Or maybe one could attempt filtering the data based on these ratios and attempt a trend analysis on the ratio-filtered data. But all of that may be well beyond the scope of an ESSD publication and be better suited for a regular Atmospheric Science journal.

We think trend analysis provides important information about our data, and is within the scope of the journal (e.g. https://essd.copernicus.org/articles/14/1917/2022/). We present the ethane, methane and propane trends from different analysis approaches (seasonal, linear, and non-linear trends) which provide the readers a broad spectrum of understanding our dataset and therefore the possibility of using our data for their work in the future. The discussion about differences with trends and variability derived from surface data is very interesting and indispensable, but will need extensive interpretation supported by atmospheric modelling.

We thank the reviewer for providing ideas on data analysis and interpretation. These are important points that we will consider for our future work. Indeed, as the reviewer wrote, they are beyond the scope of this journal.

References:

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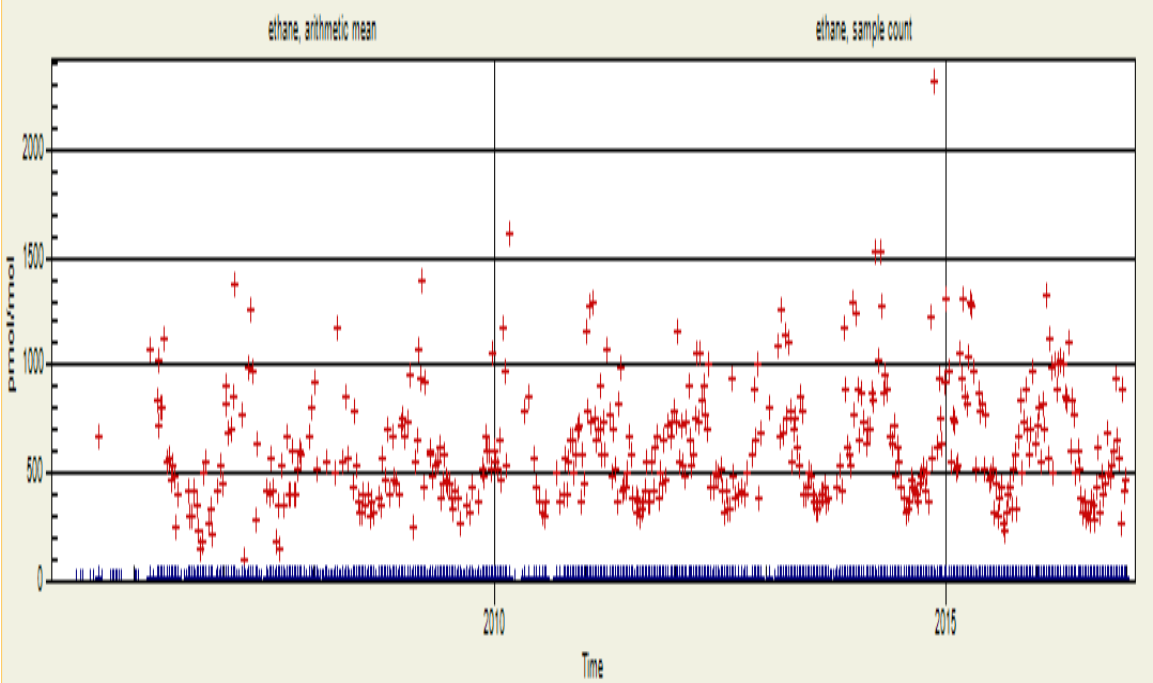

Figure 1: Ethane atmospheric mole fractions (red crosses) at Mauna Loa Observatory (3397 m asl, data and graph retrieved from https://www.nilu.no/).

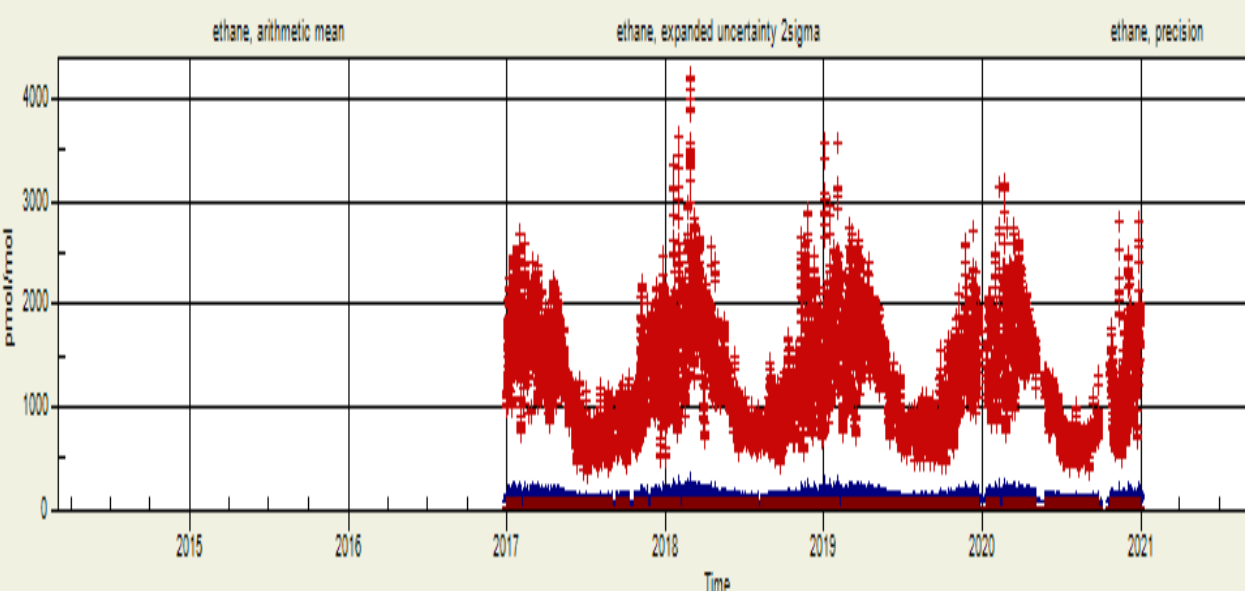

Figure 2: Ethane atmospheric mole fractions (red crosses) at Jungfraujoch (3463 m asl, data and graph retrieved from https://www.nilu.no/).

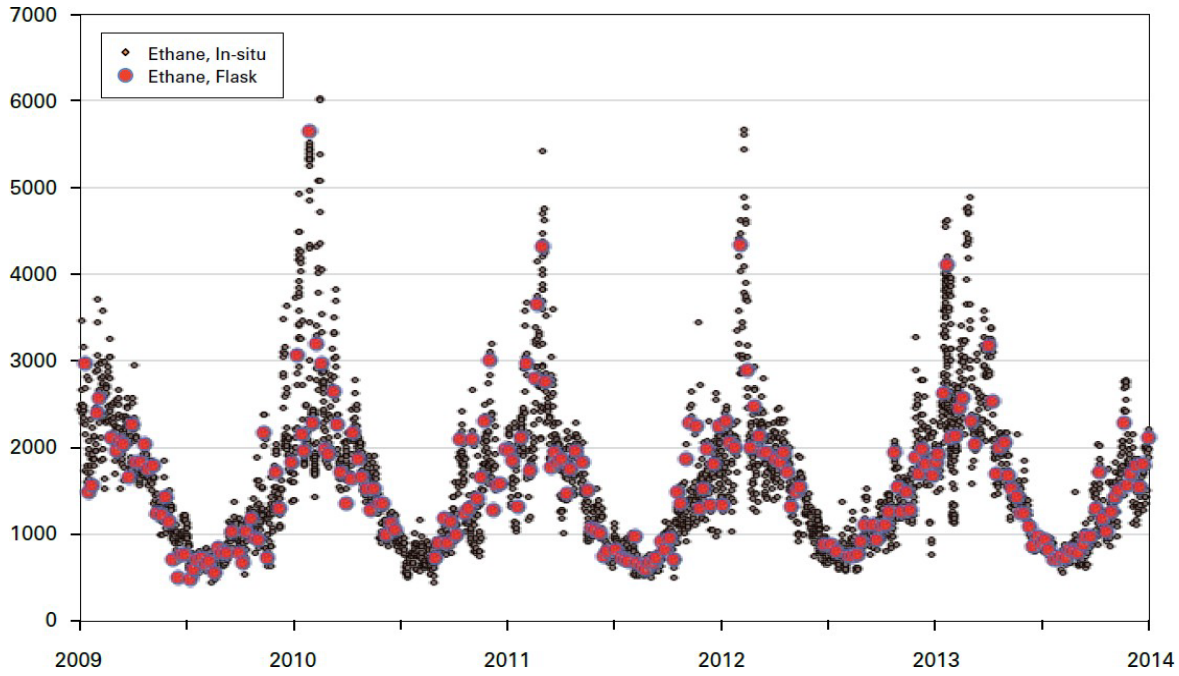

Figure 3: Ethane atmospheric mole fractions from flask sampling (red dots) and in-situ monitoring (black dots) at Summit, Greenland (3200 m asl, graph reproduced from WMO Reactive Gases Bulletin (https://library.wmo.int/index.php?lvl=notice_display&id=19877#.Yr9JIXbMKUk).

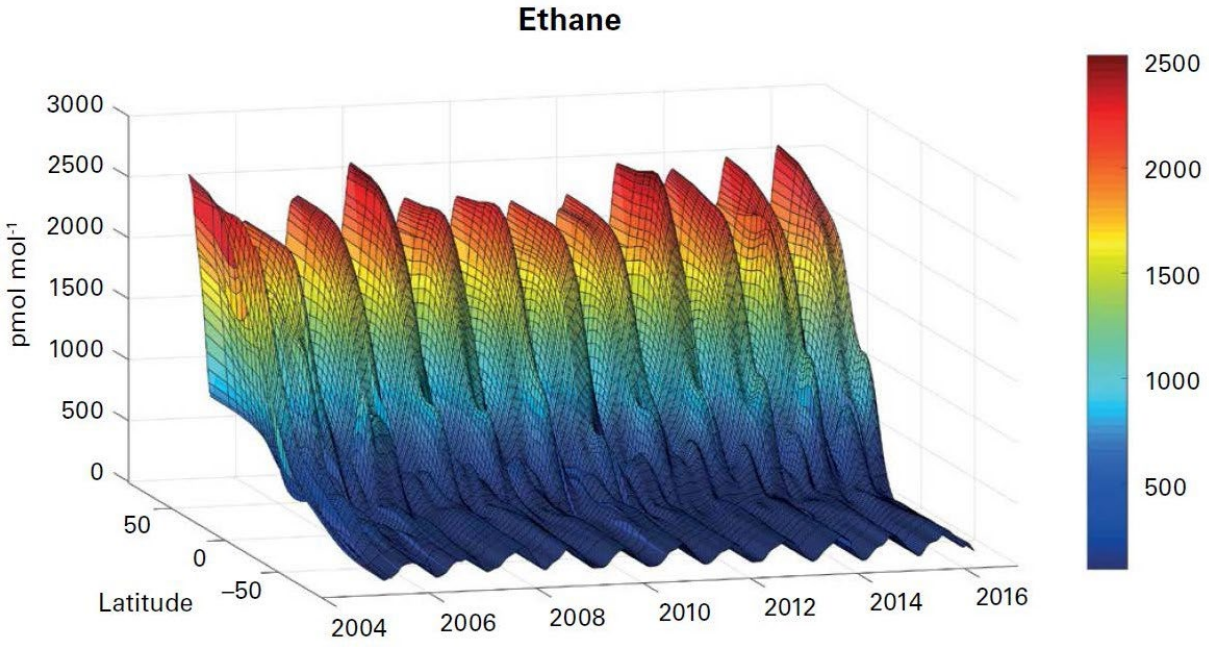

Figure 4: Latitudinal gradient and seasonal cycle of atmospheric ethane (graph reproduced from WMO Reactive Gases Bulletin (https://library.wmo.int/index.php?lvl=notice_display&id=19877#.Yr9JIXbMKUk)).