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:

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.

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.

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.

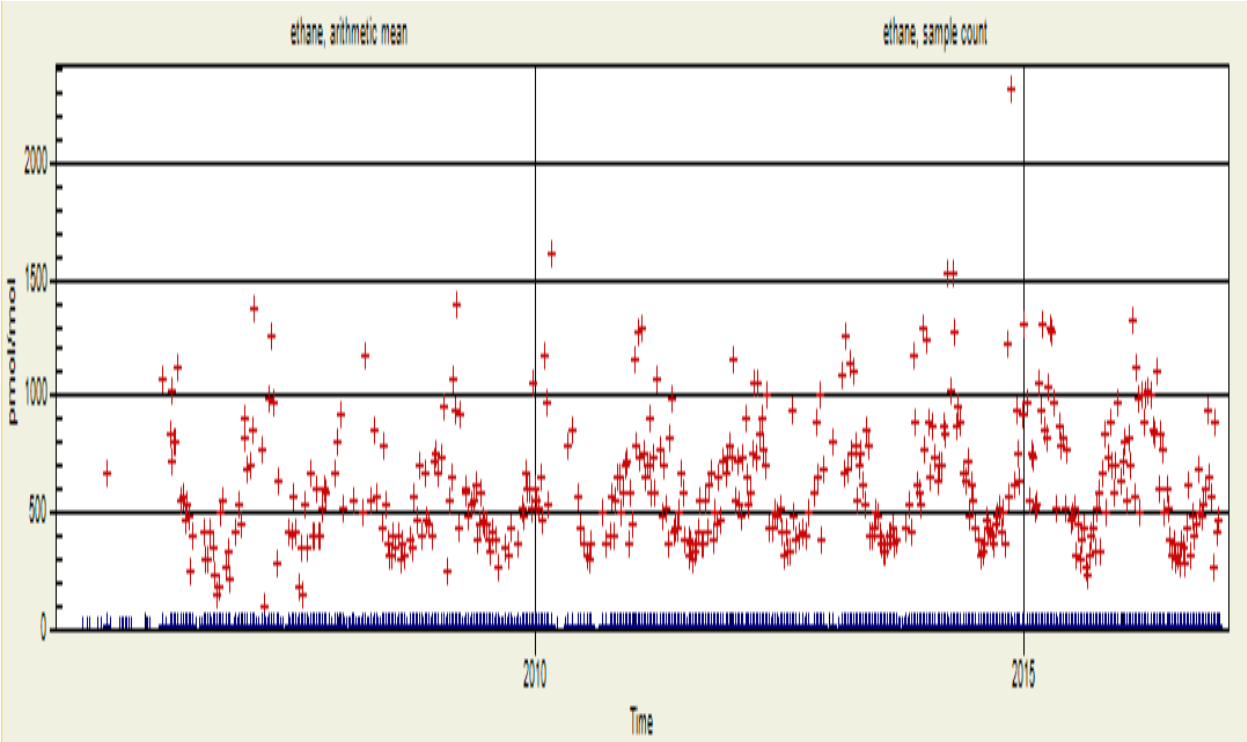

Figure 1: Ethane atmospheric mole fractions (red crosses) at Mauna Loa Observatory (3397 m asl, data and graph retrieved from https://www.nilu.no/).

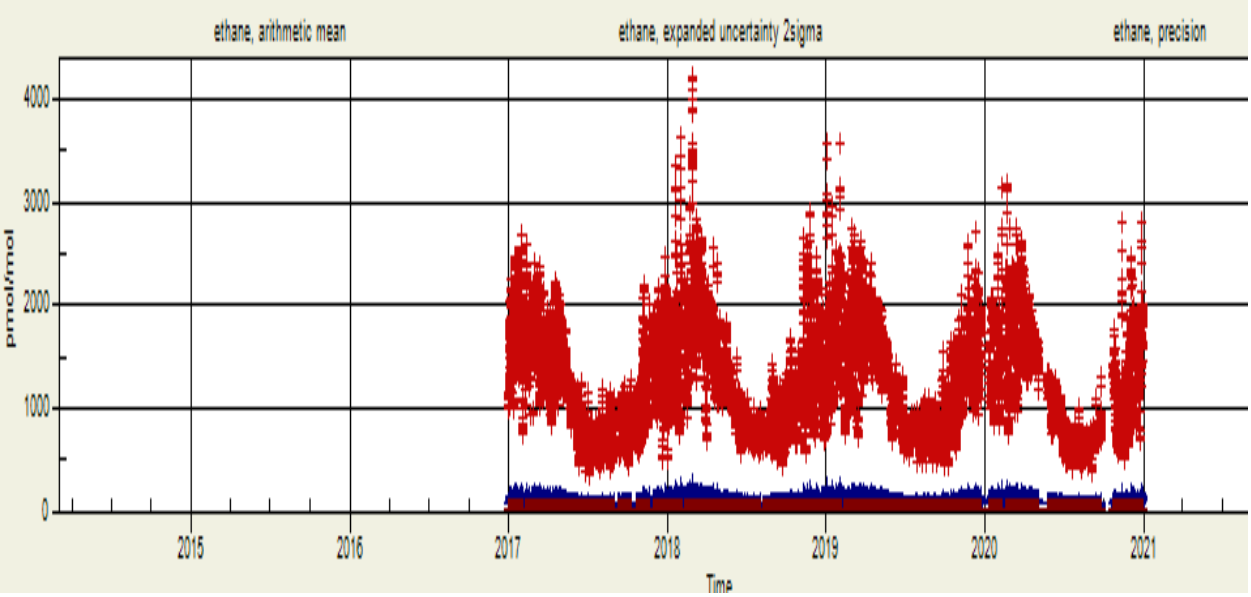

Figure 2: Ethane atmospheric mole fractions (red crosses) at Jungfraujoch (3463 m asl, data and graph retrieved from https://www.nilu.no/).

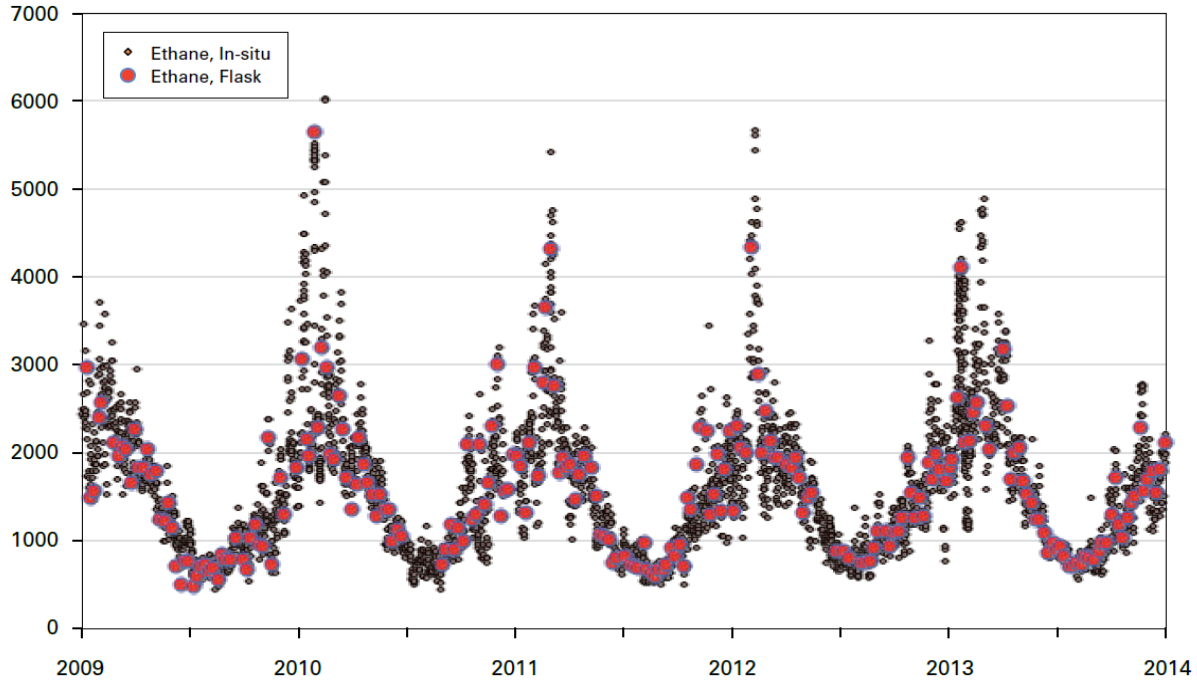

Figure 3: Ethane atmospheric mole fractions from flask sampling (red dots) and in-situ monitoring (black dots) at Summit, Greenland (3200 m asl, graph reproduced from WMO Reactive Gases Bulletin (https://library.wmo.int/index.php?lvl=notice_display&id=19877#.Yr9JIXbMKUk).

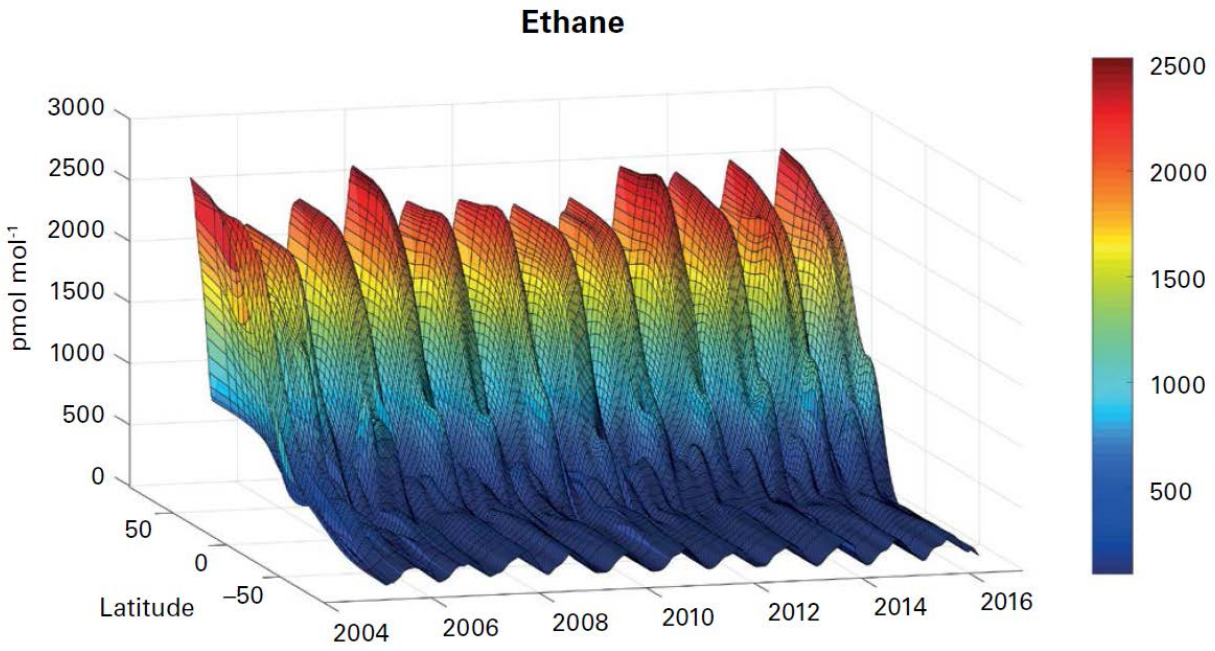

Figure 4: Latitudinal gradient and seasonal cycle of atmospheric ethane (graph reproduced from WMO Reactive Gases Bulletin (https://library.wmo.int/index.php?lvl=notice_display&id=19877#.Yr9JIXbMKUk)).