

Atmos. Chem. Phys. Discuss., referee comment RC1 https://doi.org/10.5194/acp-2021-285-RC1, 2021 © Author(s) 2021. This work is distributed under the Creative Commons Attribution 4.0 License.

Comment on acp-2021-285

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Referee comment on "Temporary pause in the growth of atmospheric ethane and propane in 2015–2018" by Hélène Angot et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-285-RC1, 2021

The paper by Angot et al. analyzes data from the GEOSummit station since 2008. They present data for C2-C7 NMHCs but the analysis primarily focuses understanding the causes of interannual trends in ethane and propane measurements. The paper concludes that the trends are driven primarily by emissions from O&NG industry in North America. The paper is well written, easy to understand, and presentation quality is good. The measurements are based on established methods and traceable calibrations. The analysis is also quite detailed; the authors put in considerable effort to address the different complexities that go into interpretation of short-lived gas measurements from a remote site. The paper will be a valuable contribution to ACP after revisions.

My primary concerns are with regards to how possible contributions from transport and biomass burning to the observed interannual trends is addressed (see below). I also listed specific line-by-line comments in the order that they appear in the manuscript.

Transport (section 3.3)

Section 3.3 starts out with a brief description of pressure systems that control atmospheric transport and the NAO. NAO is commonly recognized as a decadal oscillation, although the index can go through more rapid phase changes. I'm assuming the observed interannual variability patterns do not correlate with NAO phases? How about Northern Annual Mode, which tends to vary more on interannual time scales?

The section transitions into the back trajectory analysis in the second paragraph and I

struggled to draw a connection between the background provided in the first paragraph onto the second paragraph. I'm not sure how to interpret a back trajectory analysis for investigating the transport variability question for ethane and propane. How far back do the back trajectories go? Mean annual lifetime of ethane is 2 months. In the winter, even the shorter-lived propane can be transported from several weeks away. I find it difficult to dismiss transport changes playing a role in observed interannual trends over Greenland without analysis of data from other regions in the NH. This is done in the following section 3.4 with results from other stations summarized in Table 1. Instead of conclusively rejecting transport contributions in section 3.3, this should be done in conjunction with a more NH wide analysis. Within this context, it would strengthen the paper to show the data that underlie the results shown in Table 1.

Biomass burning (section 3.4.1)

The discussions addressing the biomass burning contribution question are purely qualitative and leaves some question marks. I think more caveat is required to better convey the full scope of the complexity of this issue.

It is established that fossil-fuel sources are larger than biomass burning emissions in the present day budgets of NMHCs, but biomass burning can still impact variability, especially on interannual time scales. For example, Simpson et al. (GRL, 2006) suggested that ENSO driven variability in biomass burning emissions accounted for most of the observed interannual changes in NH ethane levels during 1996-2004. Did you check any possible correlation with ENSO?

Correlation analysis will reveal whether a particular source is the primary driver of observed variability, and the lack of correlation between boreal fires and observed gas mixing ratios makes a strong case that there were large changes in ONG emissions during the study period. However, this does not preclude additional significant impacts from biomass burning. Fig. 6b shows max year-to-year changes on the order of 60-70% (0.3-0.5 Tg/y) of total boreal fire emissions. This is equivalent to 50-100 ppt change for ethane over Greenland based on published sensitivity estimates (Nicewonger et al., 2020). The paper also only considers boreal fires. It is true that levels of short-lived gases at Summit are much more sensitive to boreal emissions than from low latitude fires, but emission magnitudes also matter. For ethane, the sensitivity to emissions from boreal fires (roughly 10x the sensitivity from non-boreal emissions) is almost entirely balanced by the larger magnitude of emissions from non-boreal fires (\sim 9x more than boreal) (Nicewonger et al., JGR, 2020). So, if there are correlated changes in boreal and nonboreal fires that are similar strengths in a relative sense (e.g. 50% in each), the impact in ppts could easily reach 100-200 ppt/y level for ethane. Propane is shorter lived so the fire component over Greenland should be dominated by emissions from boreal fires. Emissions from non-boreal fires is another mechanism - in addition to differences in the nature of ONG sources - that can cause Greenland records of ethane and propane to trend differently.

The paper should need some justification as to why only boreal fire emissions are considered and why no attempt is made to quantify what the expected contributions are from interannual fire emission variability. What impact does this has over the discussion at the very end of the paper relating propane trends over Greenland to propane production trends shown in Fig. 8?

Specific comments

Line 31: What is meant by regional, Greenland or the Arctic?

L36: No need for "however." Also, asking for better emission inventories is good, but isn't one of the purposes of long-term measurement networks to provide top-down estimates of emissions? Is this possible for ONG emissions from North America and Europe and what needs to be done to get there? The paper can offer some future direction perhaps?

L82-86: Rephrase or break up the sentence to clarify.

L117: Replace "i.e." with which.

L203: Grouped instead of "filtered out"?

L248-250: Is there a significant correlation without ethane in Fig. S1? I'm not sure what inference to draw from this figure; some very short-lived gases have significant local sources during summer and not the others, or measurement noise (blanks?) is significant for some gases when levels are too low?

L301-302: Changes in instead of "a change in".

L335-338: How far back do the back trajectories go?

L368: Possibility of instead of "assumption of".

L370: Is there fire activity in or very near Greenland?

L375 – Table 1: Are the trends in this table determined from single year averages for endpoint years or do they reflect linear fits to de-seasonalized time series data? Showing the data would be preferable, perhaps in the supplement.

L393: Is Fig. S5 all the data visible in Fig. 7, or just the plume? If just the plume, indicate how you define the plume, and it would be interesting to see how the property-property plots for the entire data set from July-Aug 2019 look like.