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# Reply on RC1

Hélène Angot et al.

Author 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-AC2, 2021

RC: Reviewer Comment

AC: Authors Comment

RC: 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.

AC: Thank you for the overall positive feedback. Our responses to the specific comments are provided below.

**Transport (section 3.3)**

RC: 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?

AC: Thank you for this suggestion. The following sentence has been added to section 3.3:

"*We investigated the potential influence of the NAO using monthly mean values from the NOAA Climate Prediction Center. We found a somewhat weak but significant positive correlation between the NAO and monthly-averaged mixing ratios over the 2008-2019 period ($R^2$ = 0.4, p-value < 0.01 for both ethane and propane), in line with enhanced transport of pollution to the Arctic during positive phases of the NAO*".

Please note that we found a weaker correlation between the Arctic Oscillation and monthly-averaged mixing ratios ($R^2$ < 0.2, p-value = 0.1 for both ethane and propane).

RC: 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.

AC: The message has been clarified in the revised manuscript. First of all, we no longer state that changes in transport do not play a role here, and the title of section 3.3 has been revised accordingly (now: "Changes in transport from source regions"). The key message of this section is that changes in transport must be associated with changes in emissions to explain the observed trends (see lines 377-385 of the revised manuscript). Changes in emissions are then discussed in section 3.4. We also tried to better link results from the back-trajectory analysis to the background provided in the first paragraph (and correlation to NAO is discussed above). For instance, we now mention that "*years with enhanced transport from North America (e.g., 2012, 2019) coincided with a negative NAO index*", which is in line with the background provided in the first paragraph: "*Negative phases of the NAO are associated with decreased transport from Europe and Siberia and increased transport from North America*". Regarding the duration of the back-trajectory: we believe that using 5-day backward trajectories is appropriate to get an idea of the origin of air masses (e.g., North America vs. Europe or Siberia). Indeed, the results we show here (GEOSummit mostly influenced by transport from North America and Europe) are in agreement with the isobaric 10-day back-trajectory study by Kahl et al. (1997) and the 20-day backward FLEXPART simulations by Hirdman et al. (2010). This has been clarified in the revised manuscript.

**Biomass burning (section 3.4.1)**

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

AC: We agree that biomass burning can impact the interannual variability of observed ambient air ethane and propane mixing ratios, and this is actually why we investigate the correlation between observed mixing ratios and biomass burning emissions in section 3.4.1. This is done using the Fire INventory from NCAR (FINNv2.2) emission estimates driven by *daily* MODIS fire detections (Wiedinmyer et al., 2011). As such, any ENSO driven variability in fire counts (and thus, in biomass burning emissions) should already be taken into account in this analysis.

RC: 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 density 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 (~9x more than boreal) (Nicewonger et al., JGR, 2020). So, if there are correlated changes in boreal and non-boreal fires that are similar strengths in a relative sense (e.g., 50% of 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 have over the discussion at the very end of the paper relating propane trends over Greenland to propane production trends shown in Fig. 8?

AC: Thank you for raising this point. In light of the Nicewonger et al. (2020) paper, we agree that only considering boreal fires is a shortcoming. However, we did not find any correlation between observed mixing ratios and Northern Hemisphere (NH) biomass burning emission estimates. We have modified the following paragraph in the revised manuscript and added NH emission estimates to Fig. 6b (see attachment for the Figure):

"*For ethane, the sensitivity to biomass burning emissions from boreal fires is almost entirely balanced by the larger magnitude of emissions from non-boreal fires (Nicewonger et al., 2020). Propane being shorter-lived, the fire component over Greenland should be dominated by emissions from boreal fires. We thus investigate the interannual variability of biomass burning emissions from both all open biomass burning north of 45°N (boreal fires) and north of the equator (all NH fires). (…) NH ethane and propane emissions slightly decreased in 2017 and 2018 but remained fairly stable over the 2009-2016 time period. We did not find any significant correlation between annual biomass burning emissions and annually-averaged mixing ratios (true using either 2009-2018 or 2015-2018 data, and true using either all open burning north of 45°N or north of the equator)*".

**Specific comments**

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

AC: This sentence has been removed in the revised manuscript.

RC: Line 36: No need for “however”.

AC: Done.

RC: Also, asking for better emission inventories is good, but isn’t one of the purposes of long-term measurements 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?

AC: We agree and this is actually mentioned at the end of section 3.4.2: *“A number of top-down studies, focusing on specific regions or time-periods (e.g., 2010-2014), have shown that current inventories underestimate ethane emissions (e.g., Tzompa□Sosa et al., 2017; Pétron et al., 2014). The modeling study led by Dalsøren et al. (2018) focusing on year 2011 showed that fossil fuel emissions of ethane are likely biased-low by a factor of 2-3. In this highly dynamic context, where ethane production and volume rejected continuously vary and where leak rates change over time (Schwietzke et al., 2014), there is a need for further hemispheric- or global-scale top-down studies focusing on the interannual variability of ethane emissions*”.

RC: Line 82-86: Rephrase or break up the sentence to clarify.

AC: Done.

RC: Line 117: Replace “i.e.,” with which.

AC: Done.

RC: Line 203: Grouped instead of “filtered out”?

AC: “Filtered out” has been replaced by “removed” in the revised manuscript.

RC: Line 248-250: 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?

AC: On second thought, this Figure does not bring anything and has been removed from the revised manuscript.

RC: Line 301-302: Changes in instead of "a change in".

AC: Done.

RC: Line 335-338: How far back do the back trajectory go?

AC: As mentioned in the Methods section, we used 5-day air-mass back trajectories.

RC: Line 368: Possibility of instead of "assumption of".

AC: Done.

RC: Line 370: Is there fire activity in or very near Greenland?

AC: Fires can occur in Greenland but are not frequent (https://earthobservatory.nasa.gov/images/145302/another-fire-in-greenland).

RC: Line 375 – Table 1: Are the trends in this table determined from single year averages for end-point years or do they reflect linear fits to de-seasonalized time series data? Showing the data would be preferable, perhaps in the supplement.

AC: The trend analysis was done as described in section 2.4, i.e., using de-seasonalized time-series. The ethane and propane time-series at the different Northern Hemisphere sites have been included in the revised supplement.

RC: Line 395: 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.

AC: We assume you actually refer to Fig. S4. As mentioned in the caption, we only used data from the biomass burning plumes. The caption has been revised and now includes the following sentence: "*This figure was made using data from July 14-23, 2019 and from August 15-23, 2019 for the July and August biomass burning plumes, respectively*". For you reference, please find attached the plots for the entire July-Aug 2019 dataset. Emission ratios derived from these two methods (plume vs. entire dataset) are similar – that was a good sanity check though, thank you for asking.

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Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2021-285/acp-2021-285-AC2-supplement.pdf