We would like to thank Anonymous Referee #2 for insightful comments and suggestions to improve the manuscript. Please see specific responses below.

Reviewer 2

Specific comments:

P. 14 L. 444: It is true that changes in the background may lead to uncorrelated Ethane/Methane changes. But it is hard to tell what causes the changes in the ratio unless one permanently measures zero air. In practice a change in Ethane/Methane ratio could also point to a different Methane source. The uncertainty estimation later in the manuscript is very good in this regard as it uses frequent airborne zero air measurements to identify and quantify instrument drift The reviewer is correct. Only looking the methane measurements is not enough judge the validity of the measurements. The paragraph is re-worded as follows " ... Obviously, non-linear drifts or jumps in the true background will cause data errors. Our subsequent data analysis using our ethane data flags such time periods, especially where there are large background changes and/or the ethane data shows such an artificial time dependence. The flagged time period are then manually examined for validity..."

P. 17 L. 529: I would write out the formula

Added an equation showing the relationship TMU and subcomponents. The section now reads as below.

The TMU at the 1- σ level is comprised of 5-terms:

$$TMU = \sqrt{A^2 + B^2 + C^2 + D^2 + E^2}$$
(3)

These terms are: A) the background precisions prior to each ambient acquisition period; B) temporal changes in the background differences over the course of each ambient acquisition, as discussed in the previous section; C) the uncertainty in the methane interference correction (0.342-ppbv/2000 ppbv [CH₄ \pm 0.006]), as determined in the laboratory; D) the PICARRO methane measurement error (\pm 1-ppbv x 0.342/2000 = \pm 0.0002 ppbv, https://doi.org/10.3334/ORNLDAAC/1556); and E) the uncertainty in the fitting correction factor employing the input calibration standards."

P. 19 L. 587 and Fig. 9: Figure 9 shows 5 data points between 0..5 ppm Ethane that are clearly below the regression line, but they have a very similar slope. Are these from the same flight? Perhaps a systematic offset? Also in Figure 9 you excluded the 10 ppm Ethane data point. Please explain why. The error bars are just slightly larger than another point at 5 ppm, and the bias from the fit is very similar to the other 5 points mentioned above.

The five points below the correlation line was caused by an intermittent timing problem during the transition between background and ambient measurements that only affected the linear interpolation routines when drift was large during the background acquisition. All data have been re-processed to remove these artifacts. Instead of using the "notch" concentration value as a starting point for the linear background interpolation we instead set the concentration in the beginning of the interpolation to zero regardless of the notch value, which is only used as a diagnostic to flag regions that needs a closer look. The corresponding sections (2.8 and 3.3) have been updated as well as Fig. 9a and Table 2 (see below). Over the 4 mission phases of this study, 95% of the re-processed data resulted in an absolute difference less than 225 pptv and 90% of the data resulted in a change less than 133 pptv. The 10 ppm point is removed by the arbitrary 0.6 - ppbv standard deviation filter designed to catch points where the fill profile of the flask package could compromise the comparison due to rapid changes in the concentration. Please see new fig 9b to better illustrate this. This filter cutoff is higher than our previous, but captures the most egregious point.

Part of Section 2.8

"...Although the background profiles, and hence the quality of the ambient ethane data, were significantly improved during the 4th field deployment phase, as shown in Fig. 7a, we still observed moderate background shifts even after system temperature stabilization. Figure 7d, which was acquired on the same day as Fig. 7a, provides one such example. The background data during Period A reveals essentially the same excellent performance as Fig. 7a. However, the background data in Period B reveals a residual system sensitivity to what we believe are caused by rapid changes in aircraft pitch as the aircraft was preparing for landing, but have been observed during other occasions even after improved optics stabilization. Although the precisions are still excellent, here the background jumps from an average value of -0.002 ppbv to 0.188-ppbv, and the 3-second Snapshot period B. To account for such additional time

background changes we applied an additional correction to the final ambient ethane data. Referring to Fig. 7d, we linearly interpolate the background data between a zero concentration at the end of Period A (which represents the new background that is applied to the subsequent ambient data) and the average background data at the start of Period B. This linear background temporal interpolation, which is subtracted from the ambient data between the two background periods, accounts for linear background drifts. Obviously, non-linear drifts or jumps in the true background will cause data errors. Our subsequent data analysis using our ethane data flags such time periods, especially where there are large background changes and/or the ethane data shows such an artificial time dependence. Flagged time period are manually examined for validity. Using this same logic for the next ambient period, we interpolate between 0 ppb (Period B) to the mean background at the start of the next background period (0.038 ± 0.032 ppb). We estimate the component of uncertainty due to such background changes over each ambient time period by 1/2 of the mean value at the beginning of the next background period. Section 3.1 further discusses the various components to our estimated total measurement uncertainties..."

Section 3.3

"Figure 9a shows a linear regression fit (orthogonal distance regressions, ODR) of the fast CAMS ethane data averaged over the PFP time base (Y-axis) vs the PFP measurements (X-axis), and the results are shown in the ODR inset and in Table 2. During each field deployment (II-IV), we carried out comparisons of our continuous 1-second ethane measurements (not corrected by the calibration standards comparisons) with the NOAA PFP results by averaging our results over the flask fill start and stop times of the PFP system. This procedure is accurate during constant ethane mixing ratios when rapid ethane changes in plumes are not sampled. When sampling plumes, by contrast, one would need to know the exact temporal filling profile of the PFP system in order to modify the CAMS averaging kernel. This is further discussed by Baier et al. (2019). In plumes without taking this into account, one can thus obtain fast averages that are both too high, too low, and in agreement with the PFP measurements, depending upon the slope of the changes. Thus, to reduce such effects, we exclude CAMS data whose standard deviation over the PFP sampling period is greater than 0.6-ppbv, and the highest point of the regression plot of Fig. 9a (blue point with blue circle) was eliminated for this purpose. As shown Fig. 9b, the atmospheric ethane (blue lines) and methane (red lines) concentrations were rapidly changing, resulting in PFP underestimations in both cases. The PFP results are highlighted in the shaded region by crosses in both cases. Our 0.6-ppbv ambient ethane standard deviation cut-off filter in this case flagged this point. Here the ambient standard deviation for the CAMS data averaged over the PFP time base was 1.006 ppb.

The average slope value for the 3 field deployment phases is 1.030 ± 0.005 , which falls within the -1.2% and +4.8% range for the calibration standards comparisons. At present, we do not have an explanation for the small but persistent negative intercepts that average to a value of -0.097 ± 0.021 ppbv. This could imply that either the PFP measurements could have a small positive interference or the CAMS direct absorption measurements could have a small negative interference from the tails of nearby absorptions."



Figure 9a: Spring 2018 IV field deployment phase final comparisons of CAMS average on PFP time base vs PFP.



Figure 9b: Temporal profile of ethane from the CAMS (blue lines) and PFP (blue crosses) measurements and PICARRO methane (red line) and PFP (red crosses) measurements. The gray shaded region shows the highly variable ambient results for the point eliminated in Fig. 9a. lines.

Deployment Phase	Slope	Intercept (ppb)	R ²	Ν
Winter 2017, II	1.029 ± 0.0047	-0.119 ± 0.021	0.998	80
Fall 2017, III	1.031 ± 0.0039	-0.093 ± 0.014	0.998	101
Spring 2018, IV	1.031 ± 0.0063	-0.078 ± 0.017	0.994	173
Average	1.030 ± 0.005	-0.097 ± 0.021	0.997	

Table 2: Orthogonal Linear Regressions of the fast CAMS data averaged over the PFP time base vs the PFP data for 3 of the field deployment phases.

Technical corrections

P. 7 L. 210: I would rephrase to: "A sample flow rate of ... yields a cell response time of Re-worded the sentence "A sample flow "to "For a typical sample flow rate of 4 slm we achieve a cell resonance time of \sim 1 s (1/e)"

P. 14 L.427: should be take off Changed "...take..." to "...takeoff..."

Fig. 11: The small labels and wind speed are not readable. Neither the legend for EPA emission rates. The legend has been made bigger as well as wind arrows.

Fig. 12a: Increase fonts of inset to same size as major axes Increased the font size of inset