We would like to thank Anonymous Referee #1 for insightful comments and suggestions to improve the manuscript. Please see specific responses below.

Reviewer 1

Major comment:
The overall procedures for determining the background variability are robust and well described, and I especially appreciate the histogram shown in figure 8a. I am concerned, however, that the authors equate precision with the limit of detection (LOD).While the detection limit is determined by the noise of the measurements (and thus directly connected to the precision), presumably it is also affected by the uncertainty introduced by the need to subtract out the contribution of methane's absorption using data from the PICARRO instrument. At least one equation which defines the LOD (not the same as precision) at a given signal-to-noise ratio should be included. Note that for many other absorption-based measurements the LOD is defined as twice the 1-sigmaprecision (i.e., signal-to-noise ratio of 2).

We agree the reviewer's comments regarding limits of detection and we have replaced the term "LOD" with "precision" throughout the article. The term "TMU" Total Measurement Uncertainty (Lines 530-550) includes the PICCARO measurement uncertainty among other factors (please see modified paragraph and new Equation 3 below). The TMU is defined as 1-sigma.

"As previously stated, the measurement precisions only reveal part of the performance story as changes in background structure acquired during zeroing between ambient acquisitions dictates the overall total measurement uncertainty (TMU). The TMU at the 1-σ level is comprised of 5-terms:

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

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 [CH_4 ± 0.006]), as determined in the laboratory; D) the PICARRO methane measurement error (± 1-ppbv x 0.342/2000 = ± 0.0002 ppbv, https://doi.org/10.3334/ORNLDAAC/1556); and E) the uncertainty in the fitting correction factor employing the input calibration standards."

Minor Comments:
line 34 onward – sentence starting with "There are a..." has multiple grammatical errors: 1. remove colon 2. in the parentheses, do not write "i.e., x, y, z, etc.". Just write "e.g., x, y, and z" 3. write "...coal mines, wildfires, ruminants and associated manure, landfills water treatment plants, wetlands, and stagnant water ponds" – leave off "as well as biogenic emissions from" and "to name a few".
Corrected multiple awkward sentences as well as comments regarding lines 43-44 (see below for specific response).

The paragraph now reads
"The Atmospheric Carbon and Transport-America (ACT-America) campaign was a four year study composed of five different aircraft campaigns over the continental U.S. to quantify sources, sinks and transport of carbon dioxide (CO_2) and methane (CH_4), two of the major greenhouse gases. There are a multitude of sources of methane emitting into the atmosphere, such as oil & natural gas exploration and production (e.g., emissions from drilling, on-site processing, storage, flaring and transmission), coal mines, wildfires, ruminants and associated manure, landfills, water treatment plants, wetlands, and stagnant water ponds. In order to evaluate their respective contribution of total emissions, it is important to distinguish and quantify these various sources. One method that has successfully been employed is to utilize fast simultaneous measurements of CH_4 with ethane (C_2H_6). Both gases are co-emitted from oil & natural gas production in varying amounts depending upon the particular shale formation and specific production activity. By contrast, biogenic methane sources are usually not also ethane sources. In addition to its role in characterizing methane sources, ethane is long lived and one of the most abundant non-methane hydrocarbons. Since its reaction rate with OH is ~ 40 times higher than the methane reaction rate with OH at 298-K, large enhancements in ethane relative to methane can dramatically affect local OH levels, and hence ethane acts as an indirect greenhouse gas (Kort et al., 2016). This

paper discusses the development and deployment of a precise, accurate, and fast instrument that can reliably measure ethane on small low-flying aircraft and provide invaluable information related to greenhouse emissions."

line 43: "ethane is the longest-lived and most abundant non-methane hydrocarbon" –longest lived, really? In some environments, other non-methane hydrocarbons could be more abundant (e.g., isoprene).
Typically ethane has much higher concentration than other non-methane hydrocarbons, but as pointed out by the reviewer, isoprene concentrations can under certain conditions get higher. The sentence is modified from "ethane is the longest…" to "ethane is long lived…" to account for this.

line 44: awkward: "higher than methane-OH"
Re- worded "…with OH is ~ 40 times higher than methane reaction rate with OH" and "…dramatically affect OH levels.." to "…dramatically affect local OH levels.."

line 44: I do not follow the logic regarding ethane acting as an indirect greenhouse gas. This needs to be better explained, and with a reference. "Greenhouse" need not be capitalized.
At the local level changes to the OH concentration by ethane will affect the lifetime of other gases (some of which are greenhouse gases). A reference (Kort et al, 2015) has been added.

line 53: should be "DFG-based
"DFG based " changed to "DFG-based"

lines 59 and 619: metric units please!
Units changed from lbs to kg

line 66: should be "Aerodyne Research, Inc"
Changed to "Aerodyne Research, Inc"

lines 70-71 are awkward: "...systems e.g. Pal et al., (2020), quantification of regional,season fluxes of CO2 (Feng et al., 2019; and Zhou et al., 2020) and CH4 Barkley et al,(2019a,b), and evaluation of the Orbiting Carbon Observatory-2 (OCO-2) satellite Bellet al., (in press)
Awkward sentences re-worded (see below)

line 72: Replace "on average fell in the 80 pptv range" with "were approximately 80pptv"
Replaced "on average fell .." with "were approximately 80 pptv …"

This section now reads
Yacovitch et al. (2014), Smith et al. (2015), and most recently Kostinek et al. (2019) reported the use of a smaller and lighter weight high performance IR laser system from Aerodyne Research, Inc. and successfully recorded high quality and fast ethane measurements. The latter paper describes improvements to such systems for high performance measurements of CH_4, CO_2, CO, N_2O in addition to C_2H_6 on the NASA C-130 aircraft during ACT-America. Both the C-130 and B200 were deployed with similar payloads and coordinated flight paths to study the transport of greenhouse gases, primely CO_2 and CH_4, by mid latitude weather systems. Papers describing these activities are Pal et al., (2020), Feng et al., (2019), Zhou et al., (2020), Barkley et al, (2019a,b), and Bell et al., (in press). Typical airborne ethane measurement precisions reported by Yacovitch et al. (2014) and Smith et al. (2015) were approximately 80 pptv, which is about a factor of 4 higher than when the aircraft was on the ground.

line 85: "The cabin pressure effect is endemic to all such spectrometers without optical compartment pressure control" The authors should clarify that "optical compartment pressure" refers not just to the optical cell itself but the entire optical set-up.
Added clarification of pressure controlled compartment by "The cabin pressure effect is endemic to all such spectrometers without pressure control of the entire optical set-up"

line 87: remove the colon!!! Later in the sentence, replace the semicolon with a comma
Removed and replaced comma and semicolon

line 102: remove "as will be discussed"

Removed "..as will be discussed"

Figure 1 - nice photo, but please clarify what's inside the black cylinder. That's the optical compartment I assume?! It's only labeled as "TEC Temperature Control" and "Vibration/Shock Isolated and Pressurized Enclosure
Figure label re-worded to clarify "Vibration/Shock isolated and pressurized optical compartment"

Line 119: remove comma
Overall I would have liked to have seen much better usage of commas and colons. I find it surprising that none of the co-authors objected to their frequent incorrect usage.
Comma removed

line 141: replace "...except for a couple of beam dumps" with "...except for two beam dumps"
Replaced with "…except for two beam dumps"

line 151: "two orthogonally placed spherical mirrors" insert hyphen between "orthogonally" and "placed". Moreover, I am confused by what that means. The two Herriott cell mirrors are at right angles to each other? That can't be right! Also, how many passes are used for this multi-pass cell and what is the effective path length? The path length (48 m) is only noted in figure 2 but not the text.
The mirrors are parallel and the re-worded section now reads

"…Similar to the patented multi-pass cell design employed in CAMS-1 (Richter et al., 2015) the present MP offers long path length (47.6 m, 49 roundtrips) and smaller sampling volume (~ 1 liter) than traditional Herriott cells. This is accomplished employing a sealed hollow core tube in addition to an outer cylindrical tube that provides a vacuum-tight optical sampling cell. The inner tube is mounted centered to the cell's longitudinal optical axis, reducing the sampling volume between the two spherical mirrors of a traditional Herriott cell. Its diameter is limited to a radius that provides sufficient clearance of the recirculating beams between the two spherical mirrors…"

lines 205 – 209 – What kind of Teflon - PTFE? PFA? Note that not all fluoropolymers areactually "Teflon" brand (from Dupont/Chemours). Easier to just not use the commonly-used word "teflon" and describe what it actually is!
Replaced brand name with "…teflon (PTFE)..".

line 210: remove comma
Re-worded awkward sentence to "For a typical sample flow rate of 4 slm we achieve a cell resonance time of ~1 s (1/e)"

line 215 – this paragraph seems out of place. I recommend placing it after the sampling train and calibrations are described
Paragraph moved to end of section 2.4

line 244 "Calibration standards are measured before and after each flight" The wording is a bit confusing – it could be interpreted to mean that the calibration standard cylinder was measured with something else before and after flights. Perhaps "Calibrations were performed before and after each flight" instead.
The instrument is not "calibrated" as it's a first principle/absolute measurement technique. The calibration standards are used to verify that the instrument absolute accuracy. The section is re-worded as below to clarify.

"…During the Calibration Standard mode, a known mixing ratio C_2H_6/CH_4 (20/ 2000 ppbv) is fed into the zero air stream by a flow controller, which is then added to the inlet. This was performed before and after each flight to verify instrument accuracy. During Ambient and Background modes, a small suck-back flow (0.3-slm) is engaged to draw away any residual standard trapped in the addition line…"

line 322: not "times the" but "multiplied by..."
Replaced with "…multiplied by"

lines 330 onward: I highly recommend using single-letter variables, e.g. P for pathlength rather than PL.
Replaced double letter variables with single letter

line 360: “we introduce known C2H6/CH4 calibration standards in compressed gas cylinders from Scott Marrin into the inlet before and after each flight. CAMS direct ab-sorption measurements retrieved ethane mixing ratios that were too low by 6% and all ACT-America data have been subsequently raised by this number” awkward sentences. Raised by what number? Were all the measurements lower than the standards by 6%?Or just some? Please clarify.
We agree with the reviewer that the calibration section was confusing and we re-worded the entire section (see below) to clarify and reflect the composite of all calibrations. Please note that an error was discovered by Reviewer 2 and the absolute numbers (not the conclusions) have changed slightly after re-processing all data affecting figure 9 as well as table 2.

“As indicated, known calibration mixtures of ethane/methane diluted in zero air from a set of working Scott Marrin standards were introduced into the inlet (20 and 2000 ppb) before and after each flight to further validate the direct absorption retrieved values and the fitting approach implemented. Typically, the retrieved ethane values for the working standards were lower than the expected input values based on the manufacturer assigned values times the measured dilution ratio. All reported ambient ethane data were thus based on direct absorption values corrected by the daily working standard correction factors (Assigned cylinder mixing ratio/retrieved values during pre-and post-flight calibrations). Since this procedure relied upon the accuracy of the Scott Marrin working standards, we also verified these standards in the laboratory based on multiple direct absorption measurements employing the CAMS-1 and CAMS-2 instruments. In addition, prior to the 5th field deployment we measured the mixing ratios of various additional ethane standards by direct absorption. These standards included: 1) a gravimetrically prepared ethane/air standard (nominal 5 ppm) from the Apel-Reimer Corporation, which in turn was evaluated by Reimer against NIST Standard Reference Material (SRM) gases; and 2) two additional ethane standards in the 0.3 and 3 ppb range employing Niwot Ridge air prepared and analyzed by the NOAA/ESRL Global Monitoring Division and subsequently analyzed by Detlev Helmig’s Atmospheric Research Laboratory at the University of Colorado Institute of Arctic and Alpine Research using standards tied to the Global Greenhouse Gas Reference Network (see for example Helmig et al., 2016). The latter two standards were measured in our laboratory by direct absorption without dilution. Collectively, all the ethane standards comparisons resulted in agreement between our direct absorption values and the assigned cylinder values in the range between – 1.2% and + 4.8%. It is important to note that the NOAA standards were used by Baier et al. (2019) in their programmable flask package (PFP) ethane measurements. Figure 9a shows an Orthogonal Distance Regression (ODR) linear regression plot of our direct absorption results (with the daily corrections applied) integrated over the PFP time base as a function of the PFP ethane results for the entire spring 2018 4th deployment. Additional ambient ethane ODR comparisons for the 2nd through the 4th field campaigns are provided in Table 2, and these results show agreement between CAMS and the PFP to within 3%. Collectively, all ethane comparisons (ambient and cylinder standard measurements) show agreement within the ± 6% (1σ) Harrison (2010) cross section uncertainty value.”

line 374: “are accurate on average to within ±6% range” please clarify if you are referring to 1 or 2 sigma accuracy
The accuracy is 1 sigma and it is clarified in the re-written section (see above)

Figure 6, caption: “The ethane fits out to 4.23±0.025-ppbv,” awkward language (“fitsout”). Perhaps “The fit indicates an ethane mixing ratio of 4.23...”?
Caption sentence is re-worded “…The spectra are in channel numbers. The fit indicates an ethane mixing ratio of 4.23 ± 0.025-ppbv, while the methane corresponds to 1591 ± 30-ppbv….”

line 393 and Figure 7a: “In the case of Period A, we show the residual fit of Bkgn acquired during this period minus Bkgn-1, acquired 7 minutes prior (not shown)” This is confusing. In the figure I do not see any “fits” – just concentrations. Should “fit” be replaced with “derived mixing ratio” or something similar? Ethane-methane slopes: personally I’d prefer to not see them expressed as percentages. ie, just 0.184, not18.4%
To clarify that the concentration is based on the residual fit of two consecutive backgrounds the section is re-written as below ” … In the case of Period A, we show the derived mixing ratio results from the residual fit of Bkg_n acquired during this period minus Bkg_{n-1}, acquired 7 minutes prior (not shown). As illustrated, the fit of the resulting background difference (Bkg_n - Bkg_{n-1}) yields a stable background difference (0.020 ± 0.018 ppb) close to zero…”

Figure 7a
The sentence in caption text is re-worded as “…The dark blue circles represent the derived mixing ratios of the background difference (Present - Previous)…”