We would like to thank the reviewers for their useful and detailed comments, and we are happy with the positive feedback from both reviewers. In the revised paper, we have addressed the important comments regarding the lack of laboratory test information, poor figure resolution, the apparent trend in the difference between active AirCore and tower  $CO_2$  measurements seen in figure 7 (e), and the different spatial resolution due to diffusion of  $CO_2$  and  $CH_4$ . We feel these have especially improved the scientific quality and technical details of the paper. In addition, we have addressed the many minor remarks that was pointed out by the reviewers. A point-by-point answer to all the remarks from reviewer #2 can be seen below, given in red text.

## **Reviewer #2 comments**

## General comments

The paper reports on the development and on a field test of an UAV-based active AirCore system for measurements of greenhouse gases. The subject of the paper fits AMT perfectly; it describes a method that bears a lot of potential for future research on GHG fluxes on scales of 100's to 1000's of meters. It is an important contribution to this field of research and should thus be accepted for publication – but only if the comments below are adequately and fully addressed.

The presented tests, validation and field deployment are in my opinion sharp at the necessary minimum maturity for the paper to be accepted. The paper shows an interesting way forward, but fails to present a robust method/application for the time being. Only after additional work it will become clear for what applications and to what extent an active AirCore can be used best. It is critical that this is clearly communicated in the Discussion and Conclusion parts of the paper

- The usefulness of a UAV platform to quantify instantaneous CH4 fluxes from a landfill has been demonstrated by Allen et al., 2018. Following this manuscript, we have used our UAV active AirCore system to quantify CH4 emissions from a coal mining shaft in Poland, and from a dairy farm in the Netherlands. The results of these studies will be published in separate papers, and are beyond the scope of this paper. We added a sentence in the conclusions and discussion "The usefulness of a UAV platform to quantify instantaneous CH4 fluxes from a landfill has been demonstrated by Allen et al., 2018."

I fully agree with Anonymous Referee #1 regarding the comment concerning Page 1/Introduction. Furthermore, writing style improvements are possible throughout the text; avoid using statements devoid of a clear meaning where they are not adding any information or the reader expects clear, often quantitative information. The resolution of the figures must be improved. Also, every figure must be readable also if printed on an A4 sheet of paper.

- We have deleted four paragraphs, P2L21 to P3L22, regarding satellite, FTS and aircraft measurements, cutting out a big portion of the introduction to reduce the lengthiness.
- All figures are now high-resolution PNGs, PDFs or EPS.

Specific comments and technical corrections

Note on Technical corrections: in some cases, I have marked a word or formatting only once, but make sure to apply the corrections throughout the text where relevant.

Page 1, Line 1 (1/1): the word "accurate" from the title is not backed up by the paper's content (see e.g. 1/24!) -

- acceptable title is:" A UAV-based active AirCore system for measurements of greenhouse gases"
  Changed the title to the suggested one. We would like to point out that the comparison results (P1/L24) are also affected by the real atmospheric variability, which adds noise/mean differences for short-term comparisons in general, and the accuracy of our active AirCore measurements is likely better than that.
- 1/10: In some places, you are describing too many detailed information for an abstract (e.g. tubing dimensions).
   Removed the tube dimension like the, i.e. Swagelok type, wall thickness etc.
- 1/14: Replace "...sample atmospheric air in both vertical and horizontal planes."...spatially sample atmospheric air."
   Replaced this sentence with "spatially sample atmospheric air".
- 1/16: Delete "small" in "a small KNF micropump" - Deleted throughout the paper
- 1/18: What is "...shortly after landing..."? for example use at least "not more than xx min after".
  Replaced "shortly after landing" with "not more than 7 minutes after landing"
- 1/18: H<sub>2</sub>O should not be stated here; you are not calibrating for it, your sampled air is dried, it is not discussed in the text.
  - Deleted H<sub>2</sub>O. H<sub>2</sub>O is still measured, and used to correct for the dilution and the pressure-broadening effects to derive dry mole fractions of CO<sub>2</sub>, CH<sub>4</sub> and CO, as stated in section 2.3 and 2.6.
- 1/28: AirCore is not a platform there are several platforms that would have access to locations you measure, but

the question is what data and at what resolution it could collect (and at what operational costs) – rephrase. Changed 'platforms' to 'techniques'.

- 2/24: Be clearer on the "vertical distribution" I presume you are referring to the total column (as opposed to tall towers providing profiles, but only up to some 100's of meters).
  - This whole section of the Introduction has been removed in an attempt to shorten the introduction.
- 4/1: Here some relevant references are missing: *Khan et al. 2012, doi:10.3390/rs4051355 ; Kunz et al. 2017, doi: 10.5194/amt-2017-207; Watai et al. 2006, doi:10.1175/JTECH1866.1.* 
  - We added the above mentioned references to the introduction.
    P3L2-L3: "... investigation of temporal and spatial variations of atmospheric CO<sub>2</sub> using a unique CO<sub>2</sub> measurement device attached to a small UAV (Kite plane) (Watai et al., 2006), ..."
    P3L6-L7: "..., a small atmospheric sensor measuring CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>O attached to a robotic helicopter (Khan et al., 2012), ..."
    P3L8-L10: "..., and a dedicated CO<sub>2</sub> analyzer, COmpact Carbon dioxide analyzer for Airborne Platforms (COCAP), capable of being flown onboard small UAVs (Kunz et al., 2017). ..."
- 4/23: Please define "lightweight".
  - Added a parenthesis after "...lightweight UAV..." to specify: "... (total weight below 4 kg) ..."
- 4/29: Be more explicit on the analyzer you have used.
  Added a parenthesis specifying the analyzer: "... (CRDS, Picarro, Inc., CA, model G2401) ...".
- 5/Table 1: is +- 1 g a meaningful information? Are the numbers after the comma for the vertical and horizontal resolution representative/funded in your calculations?
  - No, you are correct. The small uncertainty numbers were there to add all the information we had. We have removed these numbers.
- 5/4: "inner diameter (ID)", not "ID (inner diameter)", same for 5/5 - Switched the order
- 5/6: Rather use "glue for ceramics".
  - Changed "ceramic glue" to "glue for ceramics".
- 5/11: Unclear sentence please rewrite. "in the laboratory." implies that somewhere else, at another altitude this is different clarify.
  - Removed "in the laboratory". The vacuum the pump could provide was determined in laboratory, but you are correct in that the pressure is always monitored in the system, and does not change much from this value.
- 5/13: Since you start describing the setup here, you should already here give details on the used hardware (e.g. pressure sensor type/model).
  - Added a parenthesis with the model number of the pressure sensor.
- 6/2: Delete "product".
  - Deleted.

6/16: fox -> box - Changed; "fox" is now "box".

- 6/Fig 1: You can drop a and an before schematic and image. The photo should be cropped nearer to the box. - Done. The "a" and "an" are now gone, and the photo has been cropped closed to the box.
- 6/21: Not necessarily true for H<sub>2</sub>O delete this sentence; the relevant information on the analyzer's precision is included in line 7/2
  - Deleted
- 7/2: Please clarify where you got the numbers for precision from.
  - They are based on cylinder measurements. We have added a sentence to express this: "..., based on cylinder measurements before and after analysis of the AirCore. ...".
- 7/4 and 7/6: How do you measure/monitor the flow rates?
  - The flow rates were measured with an Alicat flowmeter located at the exhaust of the pump. The flowrate was noted at the beginning of the analysis and assumed constant throughout the analysis. It was monitored, but not logged. We have added a sentence on P6L13 L15 stating this:

"The flowrate was monitored using an Alicat MB-100SCCM-D/5M flowmeter located at the exhaust of the pump, and was noted down at the beginning of the analysis and assumed constant throughout the analysis of the AirCore."

- 7/8: There must be more details / figures related to laboratory tests in the paper as they are a corner stone for the validation of any method/measurement setup. You are testing a setup that later flies on an UAV, but have not done any tests where the pressure on the inlet side varies. How many experiments were there (several?)? How large was the variation of the measured trace gases? Also, the parameters measured during tests and flights (e.g. pressure) is nowhere shown but should be.
  - Three laboratory experiments were conducted to verify the consistency of the results. We have added this number to the text (P6L28). During the experiment, the datalogger tracked the inside pressure, the ambient pressure, and the temperature of the AirCore; all these parameters are essential for the processing of the data. The range of mole fractions during the experiments were between 394 to 417 ppm for CO<sub>2</sub>, 2009 to 2120 ppb for CH<sub>4</sub> and 118 to 1657 ppb for CO. We have included the information regarding the range of mole fractions and which parameters were measured from P6L32 P7L2:

"Figure 3 shows the time series of one of the experiments, The mole fractions during the three tests ranged from 394 to 417 ppm for  $CO_2$ , 2009 to 2120 ppb for  $CH_4$  and 118 to 1657 ppb for CO. During the roof air tests, the datalogger tracked the inside pressure, outside pressure, and the temperature of the AirCore, which are the essential parameters that goes into the processing. From figure 3 (a) and (c), a small time lag between the AirCore measurement and the direct measurement can be seen. This is believed to be due to water vapor effects, as the air was not fully dried. "

We have also added a figure describing the laboratory setup (figure 2), and we have also added a figure showing the results of one of the three laboratory tests ( $CO_2$ ,  $CH_4$ , and CO - Figure 3). a figure showing the results of one of the three laboratory tests.

- 7/27: quad copter -> quadcopter
  - Changed "quad copter" to "quadcopter"
- 7/31: How exactly was it attached/what was the position of the inlet? The position of the inlet is important as the sampled air is influenced by the rotors (also depending on the direction and speed of the movement particularly at relatively low speeds as are 1.5-2.5 m/s that you discuss in connection with the resolution).
  - The inlet was facing downwards, sticking out from the bottom of the AirCore box. Added a sentence "..., so that the inlet was facing downwards towards the ground, ...", indicating the inlets orientation.
- 8/1: Specify the hardware used (i.e., quick connects, rotary valve, solenoid valve,:::).
   Added manufacturer and model name to the individual parts.
- 8/3: What do you mean by "contamination of room air"?
  - Changed from "... contamination of room air ..." to "... reduce the potential contamination of the sample from non-sampled air...".
- 8/17: "reference gas"? In Fig. 2 you have Cal and Fill gases, only please clarify.
  True, this is indeed confusing. We have changed the figure-text to "Reference gas", and changed the text accordingly.
- 8/18: Replace "chase" (with "push" or " force")
  - Changed to "push".
- 8/21: This leads to a well-defined sample between the two reference gas mole fraction values.
  Added: "... between the two cylinder gas mole fraction values...". We did not want to state "...two reference..", as this may be seen as confusing with respect to the previously mentioned "reference" and "fill" gas terms.
- 9/14: This cannot be correct see GAW Report No. 229 (2016; on p. 6: "The current scales are (as of June 2016): WMO CO2 X2007, WMO CH4 X2004A, WMO CO X2014A,: : :") and edit accordingly.
  - We have edited accordingly. The calibration scales used are X2007 for CO, and X2004A for CH4 and CO.
- 9/Table 2: The WMO CO calibration range is currently 30-500 ppb; see also 9/14
  - Yes, it is. The fill gas CO content is not meant as an accurate number, it is there to be a clear indication as to when the sample ends. The calibrated CO value of the fill gas has a large uncertainty.
- 9/8: Please explain what you mean with "::: to correct for the small nonlinearity if there is any,...".
  Changed the sentence from: "... to correct for the small nonlinearity if there is any, ... " to "... to correct for drift in the linear calibration curve, ...".
- 9/12: "3/4 ways" why? (and it is a rise, not a dip). In general, explain the starting and ending point choices.
  Changed from a "... dip ... " to " ... increase ... ", and added a sentence stating that the points were empirically determined from the fifth flight.

P10L15: "... These points were empirically determined from the fifth flight, where the maximum correlation between the active AirCore and the 60m continuous measurements was found. These points were consistently selected for all the flights. ...".

10/Fig.3: "H<sub>2</sub>O [%%]"?

- A typo; fixed by removing one "%".
- 11/9: A map shoving the station and enough surroundings to meaningfully support the description of the station/the field experiment described later in the text is missing here.
  - We have added a google maps image showing the atmospheric station and its surroundings.
- 11/12 "Situated directly behind" is unclear how far is "directly"? (see previous comment)
  Changed "... directly behind ... " with "... roughly 50 m behind ... "
- 11/13 Unclear/wrong sentence rephrase sentence.
  Agreed, it was a bit unclear. Decided to remove the sentence.
- 12/3: "The observatory itself is surrounded by insignificant shrubs and grass." what exactly are "insignificant shrubs"?
  - Changed to "... small shrubs..."
- 12/8: I guess you mean 60 m a.g.l.? Any references describing the Lutjewad station measurement setup that you could cite here?
  - Yes, been changed to "60 m a.g.l.". We have added a reference that describes the station measurements setup: van der Laan et al., 2009.

van der Laan, S., Neubert, R. E. M., Meijer, H. A. J., and Simpson, W. R.: A single gas chromatograph for accurate atmospheric mixing ratio measurements of  $CO_2$ ,  $CH_4$ ,  $N_2O$ ,  $SF_6$  and CO, Atmospheric Measurement Techniques, 2, 549-559, doi: 10.5194/amt-2-549-2009, 2009.

- 12/18: Replace "happened" with "took place" - Changed "happened" with "took place".
- 12/19: Instead of "right before sunrise", better state the exact time of sunrise on that day.
  - Time of sunrise has now been stated.
    - "... The sunrise occurred at 06:05 UTC. ... "
- 13/Table 3: Mean speeds are misleading as there was also some hovering involved in some flights (see also 7/31). - Mean speeds have been removed from Table 3.
- 14/Fig.5: Zooming into area / time of interest (all graphs) and adding measurement points from AirCore would largely increase the usefulness of these plots.
  - Now zoomed in to focus on the times around the flights instead of the full day. Sample data has also been included in the figure, along with a caption description of the altitude range for each flight: "... The altitude covered during the flights were 485m, 301m, 478m, 23m, and hovering at 60m for flights #1, #2, #3, #4, and #5 respectively. ... ".
- 15/Fig.6: Same as for comment above .
  - Now zoomed in to focus on the times around the flights instead of the full day
- 15/4: Is the time lag due to the long inlet lines at the tower taken into account in your calculations?
  Yes, the time lag has been accounted for.
- 16/Fig.7: The titles above the graphs are not necessary. What are the fine dots in 7.c? As the five flights were so few and different it is difficult to say anything concrete on the quality or interpretation of the flights (particularly on flight #1). I therefore strongly suggest avoiding highly speculative interpretation attempts (as in 18/6-9). Some retro trajectories might help (even if a trajectory does not explicitly give information of the fluxes), but that might be already beyond the scope of this paper. 7 a and b are so much zoomed out that we can only poorly evaluate the performance of AirCore vs.tower Table 4 is more helpful some discussion is needed on why flight #3 seems to be giving the "best fit" profile, judging from Table 4 (even if there was no data recorded on the SD card).
  - Titles have been removed. The fine dots in Figure 7 (c) represent the CO data points with a time resolution of 3. The lines are drawn with a 5 data point average, as stated in the figure description. We've added a sentence in the figure text stating that each dot represents a data point:
    - "..., with each dot representing a data point with a time resolution of 3 seconds. ...". Added a possible explanation as to why the fit seems to be better for the third flight: "... From table 4, it is seen that the best fit

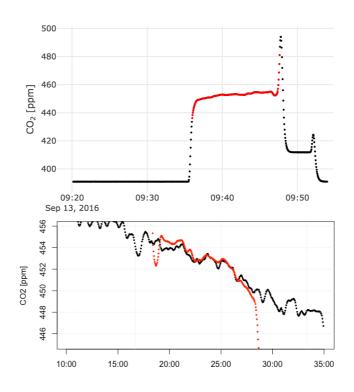
between data and atmospheric tower data occurred during the third flight. A possible explanation for this could be the smaller variability of mole fractions within the boundary layer. ...".

We decide to keep the sentence that  $CO_2$  may have been originating from Eemshaven (east of Lutjewad), because this is the most likely interpretation we have based on available information. However, we have added more information to the matter, by including an additional sentence: "... Hysplit backward trajectories show that the winds emanated from the south-east during the time of the campaign. ...".

- 18/10: "Both the descending and ascending mole fraction profiles during all the flights compare well with the continuous measurements of CO<sub>2</sub>, CH<sub>4</sub>, and CO at 60 m and 7 m, indicating that the features seen during the first flight's CO<sub>2</sub> profile is indeed real.": I cannot agree with this statement the unexplained features of flight #1 are above the 60 m level. The fact that the measurements agree somewhat (not well) at 60 and 7 m does not imply that one knows what happened above 60 m please rephrase.
  - We have removed the sentence " ... , indicating that the features seen during the first flight's CO<sub>2</sub> profile is indeed real. ... ".
- 18/19: From where did you obtain the information on the wind speed and direction (as the instrument on the tower was not recording this information)? For which altitude are the 2.5 3.0 m/s?
  - The wind speed was recorded at the tower, just not at 60m. As figure 6 shows, the wind speed was recorded at both 40m and 7m, however, the tower did not provide wind direction. The wind direction, and also speed, was obtained from a monitoring station of the Royal Netherlands Meteorological Institute (KNMI) that is located in Lauwersoog (Latitude, Longitude, Altitude: [6.200E, 53.413N, 2.9m]). Link to KNMI data: http://projects.knmi.nl/klimatologie/uurgegevens/selectie.cgi
- 18/21: With what std.dev. for the mean mole fractions? And, you should state at first mention of "mole fraction" in the text that you are referring to "dry air mole fractions" (c.f. GAW Report No. 229, p.2)
  Added the standard deviation as well.
- 18/30-31: Mentioned already in 18/19-20
  - Deleted the sentences 30-32.
- 19/Fig.8: Using the rainbow scale is not recommended (see e.g. <u>https://www.climate-lab-book.ac.uk/2016/why-rainbow-colour-scales-can-bemisleading/</u><u>https://www.poynter.org/news/why-rainbow-colors-arent-best-optiondata-visualizations</u>,etc.)
  Changed from the color scale "jet" (Rainbow) to "hot" (Black-Red-Yellow-White).
- 19/4: Chapter 3.5 is actually an attempt of validating the active AirCore measurements and could thus be part of Chapter 3.3.1. It is important to note that this "verification" is informative, but that it has only limited informative value for active flights, where the position of the UAV is changing (rapidly).
  - Moved section 3.5 into chapter 3.3.1.

20/Fig.9e: There seems to be a clear trend – any ideas how to explain it? Could there be a systematic bias introduced during data processing?

- We believe the bias is due to a contamination from unwanted air at the beginning of the sample, likely a contamination of human breath that introduced a big spike of CO<sub>2</sub> to the end of the sample, and hence contaminates that end of the AirCore sample with a higher concentration of CO<sub>2</sub>. On the other end of the sample, the reference gas carried a lower concentration of CO<sub>2</sub> than the sampled air, and has likely contaminated the other end of the sample by lowering the overall concentration. This leads to the trend that is seen in the figure 11 (e). We've attached two figures to illustrate what we mean; The first figure is the AirCore analysis, with the black curve being the raw data and the red area the cut AirCore sample. The lower figure shows the comparison of this cut AirCore sample (still the red curve) along with a longer time series of the 60m Picarro measurements.
- We have added a sentence to explain this: "... Figure 11 (e) also shows a slight downward trend in the difference. This can be explained by contamination of the AirCore sample at both ends, where the end has been contaminated by a high mole-fraction CO<sub>2</sub> spike in one end, likely due to human breath while disconnecting the AirCore and preparing for flight, and the other side by the reference gas, which held a lower concentration of CO<sub>2</sub> than the sampled air. ..."



- 20/7: Do you mean transport delay and time constant?
  - No, The analyzer smearing effect is an effect due to the mixing of the air samples in the cavity of the CRDS analyzer.
- 21/20: How different are the uncertainties for  $CO_2$  and  $CH_4$  (having in mind their different molecular diffusivity)?
  - The diffusiveness is not the dominant uncertainty in the calculation, the analyzer smearing effect is. However, it is true that you mention that CO<sub>2</sub> and CH<sub>4</sub> has different molecular diffusivity, which will influence the final uncertainty. The diffusivity is larger for CH<sub>4</sub>, which will lead to a larger uncertainty for CH<sub>4</sub> than for CO<sub>2</sub>, and so we've added a sentence stating the difference between the two:
    "... Due to the difference in molecular diffusion for CO<sub>2</sub> and CH<sub>4</sub>, the spatial resolution differs between the GHGs. When the UAV is flying with an average speed of 1.5 m/s, the uncertainties range from 7.6 m to 15.2 m for CO<sub>2</sub> depending on the storage time, while for CH<sub>4</sub> the uncertainty ranges from 9.1 m to 18.2 m depending on the storage time ranges from 10 to 40 minutes. ..."
    We have changed the numbers in the paper abstract and conclusion to state the ones for CH<sub>4</sub>, seeing how they carry the lowest spatial resolution.
- 22/29: "This study shows the active AirCore's ability to capture both vertical and horizontal trace gas profiles with high precision and accuracy." I strongly disagree with this statement. Unless you find a definition of high precision and accuracy that fits your results, this sentence should be deleted/rephrased.
  - Deleted "... with high precision and accuracy...".
- 23/25: Only cite what is published at time of writing.
  - According to the AMT guidelines: Works "submitted to", "in preparation", "in review", or only available as preprint should also be included in the reference list.