Response to Reviewer #1 of "Application of Gauss's Theorem to quantify localized surface emissions from airborne measurements of wind and trace gases" by Conley et al. submitted to Atmospheric Measurement Techniques 18-Apr-2017

General Comments

The paper by Conley et al. describes the use of aircraft-borne in-situ measurements for the quantification of localized greenhouse gas sources in a heterogenous field of potential sources. Overall, the paper is well written and the well described theoretical method may be a powerful tool to improve quantification of greenhouse gas emissions, especially in a complex area. My main point to criticize is that there is a missing link between the performed LES simulations and the presented aircraft measurements. Therefore it is not totally clear to me whether you use the LES simulations to show whether the suggested flight pattern is suitable in general, or if you use the simulations to actually design the flight pattern (e.g. the loop diameter) for each single mission. Independent from this, if d suggest to focus on one specific flight/flight series throughout the paper. This would simply help to evaluate how good the LES simulations agree with the observations and e.g. how useful the simulations are to optimize the circling radius etc.

Also, a summarizing paragraph ("Summary/Conclusions/Outlook") is missing (by accident?) and essentially needs to be added.

We apologize for the unintentional obfuscation of the link between the LES computational results and the observational ones. The LES work was performed for just three different conditions as outlined in Table 1, and was not meant to specifically represent any of the individual observational surveys. Our intention is to very generally illustrate the dispersion of a plume in a convective boundary layer and to use those results to help guide the observational strategy development, such as optimal distance downwind and flux divergence profile extrapolation. We have added some text in the introduction (new line #103) to help clarify the approach:

"Because the wind fields of turbulent flows cannot be predicted in detail, we do not attempt to compare specific features of our observations with specific LES results, but rather we use the numerical experiments to guide the development of the observational methodology. For example, by investigating the LES flux divergence profiles in the layer below the lowest flight altitude, we are able to estimate the contribution of this unmeasured component to the overall source strength."

As recommended, we have added a Conclusions Section (Section 5) to the end of the manuscript to help summarize and point to further method development directions as recommended:

This technique was developed out of the necessity to identify and quantify individual well pads in an extensive oil and gas production field. Consequently the frequent tracking of the upwind and downwind side of the source provides a very accurate determination of the location and magnitude of a given emission site. The main uncertainty arises from the effluent below the lowest flight altitude, but this is minimized by targeting a downwind distance determined by LES studies to provide very little change in the plume flux divergence from the lowest loop to the ground. In addition to the controlled release experiments, hundreds of sites have been measured using this technique with varying levels of success. Ideal conditions include flat terrain, ample sunlight to promote vertical mixing, consistent winds, and no nearby competing sources. Under optimal conditions we have demonstrated that measurement uncertainties are quite low, often better than 10%. As the conditions deteriorate from the ideal to situations involving complex terrain, variable winds or nearby upwind sources, measured uncertainties can increase to be as large or larger than the emission estimates themselves. In the worst case of stably stratified conditions (winter or night time), for instance, the lack of vertical mixing may preclude the trace gases emitted at the surface from reaching the minimum safe flight altitude. Complex terrain provides a challenge to the method because the aircraft is unable to maintain a constant altitude above the ground. A possible future refinement of this technique to be applied in complex terrain would be to fit the measurements of both wind and mixing ratio to a uniform 3-dimensional surface surrounding the source, where the grid passes through the terrain and then integrate the flux normal to this irregular virtual flight path. This would not assume level loop flight legs and would, in principle, account for individual loops being flown at differing altitudes and thus more closely track mass continuity near the terrain elevation.

Main comments:

<u>Line 141:</u> According to table 1, you release the emissions in a box of 50x50mx8m (table 1). What means the question mark after 8 in the column dz?

Those were typos and have been removed.

Especially the (center of the) release height is a very critical parameter. Have you done sensitivity studies by varying the release height to e.g. account for buoyancy?

We have not experimented with elevated releases or lofting due to initial buoyancy of plumes. Because any elevated or buoyant release would only make the plume easier to detect from the aircraft, we considered a non-buoyant surface source as the limiting condition of detection. We feel that this complication is beyond the scope of this paper, but merits further investigation in the future.

I assume the release rate is constant after the start of the release? Why is the release rate so small ($^{\sim}$ 3kg/h), especially compared to the stated detection limits of 5 kg/h?

Because LES is not subject to instrument noise or variability in the background - both of which determine the detection limit for actual measurements - the size of the release does not need

to be as large as in the actual atmosphere. The magnitude was chosen long before we had a very solid idea of what the actual measurement detection limit would eventually turn out to be.

Figure 1/3: See my main point: Is there any possibility to combine/compare both figures in order to see how good the plume shape is represented in the model, compared to the (lower resolved) measurements? At least, you should be able to virtually fly along the flight path, extract the concentration levels and plot it together with the measurements along the timeseries of the flight (although I know that this kind of graphic representation may be misleading if the plume is slightly shifted). Line 299ff/Figure 6 and 7: See my main point: Why don't you compare the LES results with the Aerodyne real test case? I'd suggest using the same flight example for both simulations and measurements (you may also discuss the variability based on your set of simulations).

As we have attempted to elaborate in the revised introduction (above), we do not feel that it is very informative to compare specific LES results with specific realizations observed in the real atmosphere. Because of the inherently random nature of turbulence these are bound to differ in their specific details (e.g., position or even structure of the effluent plume at any given instance.) While Figure 1 shows a sampled "snapshot" of the plume encountered during the ~30 minutes on station, the simulation results presented in Figure 3 are heavily averaged crosswind integrated concentrations that are meant to illustrate the average structure of the plume in the downwind direction. Any single aircraft crossing of the plume is going to deviate from this picture, and therefore we feel that a direct comparison would not convey anything new or instructive.

In the case of Figure 6 and 7, again, we do not feel that a specific comparison between the details of the LES and observations is going to show anything informative. However, comparison of the Figures does show that the average behavior of the asymptotic approach to the actual emission rate appears similar.

Minor comments:

Line 111: I assume that the flow rate is controlled in a way that the lag time of both instruments is independent from the ambient pressure?

We have added the following text in Section 2.1 to clarify this point, "Both lag times are slightly dependent on pressure, i.e., with a typical altitude change of ~1 km, the change in lag time is less than 10%, and is inconsequential when applying this method within a few hundred meters from the surface."

Figure 1: Please provide more details such as date/time/duration of flight, (derived) source strength, loop diameter.

We have added specific flight information to the caption as requested. The revised caption of Figure 1 reads:

Figure 1 - Map of the airplane flight pattern sampling a methane plume emanating from an underground storage facility. Wind direction is indicated by the white arrow and the methane mixing ratio is given by the color bar to the right. This flight was conducted on June 28, 2016 and took place between 12:46PM and 1:52PM LT at altitudes ranging from 91 m to 560 m with a loop diameter of approximately 3 km. The measured methane emission rate was 763 \pm 127 kg hr⁻¹.

Line 300: What means similar? The number of passes?

We have deleted the word 'similar' to avoid confusion.

Line 374: Please give the uncertainty of the release rate.

An estimate of the uncertainty in the release rate has been added.

Technical Comments:

Please check the number of the equation in section 3.8

The equation in Section 3.8 has been eliminated in response to a recommendation by another reviewer.

Figure 10: What is the unit of the x-axis? Please correct "geographic distribution of methane" to "ethane" in the caption. Please increase the dot size in the right figure.

Figure 10 has become Figure 9 and the caption and axis labels have been expanded to clarify.