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Atmospheric Measurement Techniques Discussions

Interactive comment on "Improved Atmospheric Characterization through Fused Mobile Airborne Surface *In Situ* Surveys: Methane Emissions Quantification from a Producing Oil Field" *by* Ira Leifer et al.

Anonymous Referee #2

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"Improved Atmospheric Characterization through Fused Mobile Airborne & Surface In Situ Surveys: Methane Emissions Quantification from a Producing Oil Field" Leifer et al.

Reviever Comments:

This manuscript describes a campaign conducted using a surface mobile platform along with an airborne platform, to estimate CH4 and CO2 emissions from California's Kern Oil Field using one day of measurements. In principle, this is a valid attempt

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Discussion paper

to use two different platforms and merge the data sets to create a better picture of emissions, which would be worthwhile and could help improve current methodologies. But in practice, in my opinion, this manuscript does not accomplish that. For a submission to AMT, there is very little precision in the method description or the description of the measurements themselves. There is no quantification of how well the concentration measurements from the two platforms compare, nor is there any description of the interpolation method for the sparse measurements in space and time. The description of the flux calculation is confusing and the description of the uncertainty analysis is too short, especially given the focus of this journal on measurement techniques and methods. In the conclusion several statements are made that are, in my opinion, overly broad and have not been shown in the work, including the interpretation of the final emissions estimate in relation to a bottom-up inventory estimate and in relation to global trends. In addition, it is not clear to me even that this method works in such complex terrain and conditions (the authors do spend a large portion of the paper describing the terrain, which they use to their advantage) which the authors do note. It is certainly not shown that this method might be better than any other method in terms of accuracy - this may not be required for publication, but it is claimed by the authors. The manuscript would be improved by focusing on the methodology itself and justifying the various methods used to perform the estimate, and remove the focus on the estimated emissions result and the global methane budget.

Specific comments:

L 54-55 "most" should be "largest" L55 Does the EPA inventory discuss the global budget? What about agriculture? (The latest Global Methane Budget Saunois et al may be a better citation here, and they place agriculture and waste to be significantly larger contributor than fossil fuel, so this statement does not seem to be supported by the literature).

L56-60 - Turner and Bruhwiler disagree as to whether a methane trend is detectable over the US; the Bruhwiler paper specifically refutes the results in Turner. So the

phrasing here is not really correct, but could be phrased to simply emphasize that there is diagreement in the literature over whether a methane emissions trend in the US exists. More recent literature on the global methane is also available suggesting that the OH sink is the cause of the global increase (as mentioned in lines 41-46). References to a more recent Turner et al and Rigby et al, both in PNAS, 2017 should be made if this is to be discussed. [However, as noted above, perhaps devoting a large portion of the paper to the global methane budget is not really in the scope of this work].

L62 - Peischl, White and Karion all use esssentially the same method (mass balance) - not sure what is meant by "direct assessment". Perhaps because of the aircraft-based winds used in the first two while Karion relied on model or ground observations?

L69 - This discussion should include a citation to Smith et al, 2015, ES&T where the ethane/methane ratio was not assumed a priori but determined from the airborne data, but still used to apportion emissions in the Barnett.

Fig 1: a) Panel in figure is not clear - much too small to read. (b) North should be indicated.

L163-165 repetitive? awkward.

L167 CGE should be GCE?

Stranded plumes: This should be more clearly described as plumes that are coming into the domain from upwind or outside the domain. At least this seems like what is being described here, that there is a criterion of a "clean" or at least relatively uniform upwind condition.

L170: What is the specific criterion for "too light or variable" on the wind speeds? In my opinion, this is very subjective in the description, including "flush nocturnal accumulations before the overpass" - so this is a restriction on wind speed in terms of transit time?

L179: These studies or most of them used compressed gases as standards either on-board or prior and after the campaigns - calibration is still required with CEAS systems.

L193 At what height is the air sample drawn relative to the roof and the anemometer?

L184-205 Have the environmental variables been compared with local weather stations or other sensors for validation (i.e. of wind measurements, which as is noted in the text, can be difficult because of the need to account for vehicular motion)?

I was looking for this reference, which is cited for the instrumentation: "Leifer, I., Melton, C., Manish, G., and Leen, B.: Mobile monitoring of methane leakage, Gases and Instrumentation, July/August 2014, 20-24, 2014.", not clear what journal or no way to find this?

Information should be given in a similar fashion as they are in 2.3 for AJAX on calibration methods. (see later comments on the supplement).

When merging the two data sets for a single analysis it becomes even more important to show that measurements of methane are on the same scale relative to the same standards, etc. and have been intercompared. The vertical profile indicates that they compare "well" at high altitudes, but no quantity is given.

L230-231 U_n and U_N are both representing perpendicular winds.

Section 2.4: This is not clear enough. There should be an equation here for Q as a function of U and C - initially it is reported that Q is simply the product but later that C is converted to a mass (density?) to derive an emission rate. What are the units of Q (flux of what? grams or moles?). If the emission rate units are in moles, then this is not required (as in Cambaliza et al). The derivation of the background is also not very clear here - why it must be split into a right and left half? Might be more clear to describe x as the coordinate from the beginning of a transect to the end, and xmax/2 is the midpoint for each transect upwind? Is there no x-dependence of CBL and CBR? (L235 indicates they are only functions of z?). An example would be nice here.

– Just looking now at the list of definitions (thank you, this clarifies things!), and it becomes clear that when the authors refer to concentration they are actually referring to a mole fraction, i.e. micromoles of CH4 per mole of dry air (this should be defined), or ppm. Concentration is usually (if molar concentration) in units of moles per meter cubed (in SI units - the authors use mol per cm cubed), which could make it a "molar mass anomaly" for the authors (N'). These should be re-defined correctly in the future draft for section 2.4 - call C a mole fraction and N a concentration.

This section is unclear with equation (1) not clear to me why the integral of a gaussian distribution would be zero. Not clear how C' is related to Phi_P. A reader has to work way too hard to make sense of this method. Also, from Figure S6, it seems that Phi is a distribution for each transect, but in the equation C is a function of z. How is the vertical interpolation done, and how is C' defined?

Equation 2 does make sense, although x1 and x2 should be defined.

Figure S5: What is this figure telling us? What are the colors in the tracks (yellow/green?). Could an elevation map suffice here?

Figure S6: Is there a transect upwind at 2200 m (as in (c)?), but there is no dashed line in (a) corresponding to this one? The data should be shown as well as the interpolated curtain, to show if there is spatial structure in x and z of this background that is being smoothed?

Supplement S5: L168 should read section 2.4 in text. L167. not clear. So the peak of the distribution (is this the mode?, i.e. the value most commonly seen in this upwind transect?), is used as the value for the entire transect, and then the background was interpolated vertically - how? Is x in Fig 6 going from west to east?

L253, I agree, but wouldn't call this "appropriateness" - more specific. Maybe appropriateness of the measurements for the assumptions made for the analysis? What assumptions are being made that need to be satisfied? I would call it representativity

or just say that spatial and temporal variability are the dominant sources of uncertainty.

L276: to 1800 masl (from what base elevation?)

L277: at 2258 is this part of the profile? Isn't this higher then?

Fig 4 (indicate masl rather than just meters for clarity)

L314, this is nice to note, but should also be included in the supplementary section on measurements, as well as to what precision they agreed (within X ppb agreement on average, or something quantitative).

L325, don't remnant structures from the prior day make the mass balance or emissions estimate not correct, according to earlier text about flushing out prior days' emissions? (from reading on, we see this is the "upwind" profile, so this should be mentioned here or somewhere nearby).

L329 (alpha - alpha' should be used here for clarity for the reader).

Fig 4: For the upwind profile, alpha-alpha', the CH4 is lower in the PBL than above. However, in Fig S6, the upwind "curtain" or plane, is showing higher CH4 at lower altitudes. How are these two figures consistent?

In Figure S6, which of the transects are AJAX, and which AMOG?

Fig 5 why only is the north wind shown? These are very low wind speeds indeed, esp. for doing an emissions estimate.

L345 yes at 4 m/s - is that the wind speed? It's not shown. Was that the wind speed for 5 hours?

L347 how is growing from 100 to 1675m a stable PBL? Also, is stable referring to the atmospheric stability class (i.e turbulence) or the fact that the PBL depth is not changing much in time?

L368 Westerlies?

Figure 8: what is the time difference between when these transects were measured, as well as when the transects for the background (showin in Figure S6) sampled? Was the bacgkround plane subtracted point by point, i.e. in x, z space so that a higher background was subtracted on the east side, (L386)? Still don't understand where the distributions Phi come in to this picture.

Fig. 8 how was the vertical interpolation done, and the extrapolation above the highest flight transect at ∼1100 masl? It seems like a different method was used for U_n and CH4, noting where they drop off in the vertical. Figure 4 indicates that AMOG was driving the surface transect much earlier than the AJAX transects (or perhaps I misinterpreted this), so how can we combine them when we know the PBL is growing?

L400: Extrapolating these emissions to an annual average is a stretch and not at all defensible. This is one of the reasons that recent similar studies that are performed over a short time frame report their emission rates in moles per second or kg per hour or such. The section on the uncertainty estimate is short and not thorough - the distributions that go into the Monte Carlo would need to be explained better.

L431: Could you look at a slope of the CH4 to CO2 tracer plot in the plume to show this consistency with the reported ratio?

L496 I would say that these complexities also challenge this method because of the variability that you are not measuring - and the model you are using assuming some constancy in wind.

Overall, this method does not fully account or try to discount the possibility of unsteadiness in the winds between upwind and downwind transects that could lead to accumulation of emissions during slower wind speed periods (night time but also could just be earlier in the day). Perhaps this is dealt with in the uncertainty calculation but that is not clear in the text as written here.

L499-501, Please indicate some quantification of the differences here. This is a meth-

ods paper - how well did the concentrations (mole fractions) agree (above the PBL), in ppb? Were any calibration tanks measured on both systems?

L502-505, yes it is true that we could not expect the winds to agree, but what does this indicate for the interpolation of wind in the vertical from the different platforms? Is that variability captured at all?

L523+ What about plumes of CH4 that are following these complex winds and topography? the simple interpolation and treatment of the surface data is troublesome under these conditions.

A mass balance equation is a conservation of mass and the equations (although not written out here) assume some steady uniform wind condition. This is clearly violated here. Perhaps the uncertainty calculation deals with this problem but it is not clear.

L541: Are these factors not accounted for in the inventory? What about temporal variability? Also, what about the uncertainties on both numbers, assuming they are 1-sigma (which should also be noted incidentaly)? Seems to me the emissions estimate actually overlaps with the inventory quite well given the uncertainties that are reported.

L552. In my opinion, this should be toned down - this one measurement supports the conclusion is that the global loss rate of CH4 to OH (or soils) is underestimated? What percentage of the global methane budget does 25 or 32 Gg/year actually represent?

Conclusion

L559. This statement implies that the uncertainty has been reduced from other methods, which is not the case, and has not been shown.

L562 But this method relies on the aircraft measurements as well as the surface, so could not be applied in the absence of those resources!

L564 - The flux quantification is "direct", meaning measured winds and concentrations were used, but that is the flux through a point in space and time (Q(x,z)) - the rest is

a simple model: you must integrate that flux based on an interpolation (in space and time), and must subtract a background that has its own model and interpolation, and the attribute that flux to a surface emission which requires some Eulerian conditions - steady flow through a control volume. All are a "model" - just a simple one.

However this point can be made differently - that one should measure before adding one more assumption to the model, which is that of a vertically well-mixed plume. Other studies have moved away from this assumption of vertical well-mixedness as well: Cambaliza et al., Heimburger et al. (Elementa 2017), Lavoie et al. (ES&T 2015 and 2017), Conley et al (both 2016 as well as 2017: http://www.atmos-meas-tech-discuss.net/amt-2017-55/), and numerous others, especially when sampling in the near field. I agree that this is a valid point to make using these observations.

Supplement:

L26 cfm should be given in metric

What are some estimated uncertainties on the FGGA CH4 measurements based on the calibration standard - how often is it sampled, si there noise/drift, etc? A sentence or two on this is warranted beyond just the statement that a calibration was performed. Was there a water correction, or were the dry values reported by the FGGA used?

The additional accuracy of the 450C sentence should go where it is first discussed, before the sentence about the FGGA. Earlier it says it achieves 1ppb accuracy, but now it says that it can achieve 50ppt if calibrated with hourly zero gas mesaurements - which number applies here? Where do the authors get the accuracies reported for the other analyzers (ozone, etc)? Manufacturer? If the main paper is not about these auxiliary gases, this information should not really be mentioned and could be removed. Interestingly, no accuracy or uncertainty is reported for CO2 or CH4, the main gases of interest in this work (for the AMOG measurements).

S2.2: Is there a reference for the MMS wind system? There is no information given

here, and this is a key measurement for flux studies. Uncertainty on winds should be reported for both platforms.

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