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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.

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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 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

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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.

âĂć We agree with this assessment and plan to address flux and global implications in a future paper. Thus, we have deleted the discussion as recommended by the reviewer. We have clarified our argument that the approach is better and under what conditions, based on the philosophy that reducing extrapolation improves outcomes. We now devote the first paragraph of section 4.2 to this important issue. Additionally, we have re-arranged section 4.2 to remove some duplication. Also, we have toned down the title.

âĂć WRT to the approach being better – our contention is simple – more information or more complete characterization of the PBL means less interpolation and extrapolation which cannot help but reduce uncertainty for any method. As to whether the improvement is significant or trivial, that of course depends on the situation. In any case, the text no longer makes the claim.

âĂć With respect to leveraging terrain, I fully accept it would be better if I had an airplane and could fly and collect data. But most scientists on this planet very seldom have access to an airplane, particularly on a regular basis, and as a result, by leveraging terrain, scientists like myself, can collect useful vertical atmospheric profile data. In the San Joaquin Valley, agreement between the airplane data and car data was very good over a wide range of altitudes indicating that at a minimum, the method has significant promise. We have added a paragraph at the beginning of section 4.3 that now recommends further research and highlights that it may not be appropriate elsewhere.

âĂć Based on the comments from the reviewer, we decided to add a paragraph to the methodology that explains why we are using an anomaly approach rather than an upwind/downwind mass balance approach. See the paragraph, which is presented below. Since this is key, a short summary now also appears in the study motivation section.

âĂć “Unfortunately, the upwind data showed a lateral gradient, which coupled with uncertainty in precisely where the downwind air originated (given the topography, which features a gentle incline towards the northeast, this gradient is unsurprising, in retrospect). Thus a very small shift in the winds between the upwind and downwind curtains results in a significant shift in CB, with a very large effect on Q. As a result, the more traditional upwind/downwind mass balance approach was abandoned for an anomaly approach.”

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).

âĂć As this is background material, we have added to the current text the Saunois reference to highlight that there is uncertainty in the CH_4 budget.

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 disagreement 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].

âĂć We agree and delete the sentence as ancillary to the main focus of the study.

L62 - Peischl, White and Karion all use essentially 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?

âĂć Yes, the difference is whether transport (i.e., transport) are measured in the study domain, or need to be modeled correctly. The sentence has been re-written to clarify what is meant by direct assessment.

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.

âĂć Agreed. We missed Smith et al 2015 due to the long time frame between writing the paper and submission – the intro material was written in 2015! We have also added a citation to the more recent Peischl et al., 2016, too. In the same vein, we have added a citation to a recent paper by Schwietzke et al. 2016 to bring the introduction more up to date.

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

âĂć Replaced by a higher resolution figure. North is now indicated on panel b.

L163-165 repetitive? awkward.

âĂć Thanks, rewritten less repetitive

L167 CGE should be GCE?

âĂć Thanks, yes. Fixed.

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.

âĂć Stranded plumes are plumes that due to wind shifts are no longer contiguous to their source (at least in the transit type of data we collect). Stranded plumes have been observed frequently in the SJV by AMOG.

âĂć That said, whether they are stranded or connected to their source, is not relevant to whether they can disrupt the experiment, so the term stranded is dropped.

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?

âĂć Added detailed criteria. Winds speeds typically less than $\sim$2 m s-1, and variability less than 30°. Flush the nocturnal (i.e., no CH_4 cloud at or nearby upwind of the site, which means that winds could not have been light as recently as several hours prior; however, winds are not measured several hours prior),

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

âĂć True, and we do calibrations daily. Text reflected to note that the calibration gas does not need to be onboard the platform – it can be at the lab / hotel/ base site, etc.

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

âĂć Sample is drawn from between 2 m above the roof and 3 m above the roof, depending on speed. This has been clarified by rewriting the paragraph.

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)?

âĂć Pressure: We have compared with the Bakersfield airport, and agreement is within our uncertainty on the altitude of the height of the sensor, and pressure changes over an hour (the airport reporting time).

âĂć Winds are much more challenging they are always changing and always spatially varying. Our best efforts have been to compare short data sets collected at a range of speeds on an open road, early in the morning, to compare wind measurements for driving the road in both directions (with the wind at an angle). Worst performance is at around 45 mph for winds of less than 1 m/s where errors are on the order of 20% in speed and direction. At higher wind speeds and/or lower driving speeds, error decreased rapidly. The GPS correction to real speed error is much smaller as it corrects itself after a few readings, which we distribute across the data by spatial filtering that limits accelerations in the along travel direction to physical limits and to near zero in the direction transverse to the direction of travel. Discussion with Vaisala, indicate that there is no need to send their wind sensor in for annual calibration (barring it being hit by a large branch). We also optimized the wind sensor positioning to minimize uncertainty for winds within $\sim 30^{\circ}$ from the front of the vehicle. Since the relative wind always has a very strong along travel direction component, this criteria is almost always met for driving at all but the slowest speeds and/or the strongest cross winds. We have not spent effort at looking at accuracy for very high cross wind data (have measured to 17 m/s) because we do not analyze such collected data in our studies to date. Additionally, such strong winds in California tend to be very strongly modified by topography – making them particularly challenging to validate.

âĂć This above discussion has been added to the supplementary material.

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?

âĂć This paper is in a trade journal (it was peer reviewed) and will be attached.

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).

âĂć Done. We have also added our linear cell pressure calibration to the supplemental material.

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.

âĂć Thanks. Typo corrected.

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).

âĂć Equation added, units added, and it is noted that there is a conversion factor between ppm and moles.

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.

âĂć It is split into a right and a left half due to gradients across the field. Since CBL and CBR are average values, they do not have an x dependency

– 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.

âĂć An equation has been added, and text now notes that C is in ppm and that there is a conversion factor to moles per volume.

âĂć When there is a gradient as there almost always is in nature, it is unclear as to which part of the background is transported to the measurement plane, introducing uncertainty. We address this by derive background from the downwind dataplane.

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?

âĂć Equations have been added and the section has been clarified. Phis is a distribution for each left or right transect at each altitude (hence the z dependency). Interpolation, prior to integration, is linear, now stated

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

âĂć Equation 2 was rewritten to not use x1 and x2

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

âĂć Data key added. The purpose of this figure is to show the typical surface obstacles to surface winds along the profile, elevation is not particularly relevant.

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?

âĂć Since 2200 m is background, there is no need to separate the background concentration from a plume and it is used without smoothing or analysis in the linear interpolation.

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?

âĂć There was a mistake in figure S6 caption – the probability distributions are all for CH_4, half for the right side of the data field and half for the left side of the data field. This has been corrected.

Additionally, the methodology of filling in the background data plane is now also described in the supplemental material.

Is x in Fig S6 going from west to east?

âĂć The caption for Fig S6 has been improved to include the definition of x and z, as in the main text.

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?)

âĂć The is a good suggestion. Changed to representative and to spatial and temporal variability. Also changed to 1800 masl.

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

âĂć It was a typo – corrected to 2058 m here and elsewhere. This was above the airplane profile. Data were collected in the open to compare with the direction and speed of winds near the top of the overlapping profiles where AMOG was surrounded by tall trees, and showed good agreement This clarification now is added.

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

âĂć Done.

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).

âĂć Moved to section 2.2 where calibration also is mentioned for the GHG analyzer.

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?

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(from reading on, we see this is the "upwind" profile, so this should be mentioned here or somewhere nearby).

âĂć The upwind profile is to characterize the atmospheric structure, not to provide input to the mass balance, and air from the profile will pass to the east of the oil fields. That this is the upwind profile is now mentioned.

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

âĂć This would require doing so for all greek letters in all captions or the entire text, which seems excessive. We will discuss with the copy editor.

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?

âĂć The upwind profile is at alpha-alpha', Supp. Fig. S6 shows the background curtain (not upwind) curtain, correctly labeled in the caption and section heading, and is at delta-delta' and is derived from are data outside the plume. The methodology is described in Section 2.4, now corrected in the supplemental to refer to the right section.

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

âĂć AMOG is on the surface, now labeled.

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

âĂć As noted above, the profile is not for directly estimating emissions, but to characterize the atmospheric profile structure, primarily the location of the PBL. The figure was very busy already, so only the north component (ascent and descent) are shown. The purpose of showing the winds is to note they are not (in this case, useful for identifying the PBL, and that they show a change between the earlier and later profiles. The east component conveys similar (lack of useful) information.

âĂć Text now notes “Winds were not useful for deriving the location of the PBL.”

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

âĂć Yes – text clarified.

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?

âĂć For clarity, text changed to “$\sim$100 m growth to $\sim$1675 m,”

L368 Westerlies?

âĂć Changed.

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 background 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 âĹij1100 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.

âĂć Agreed. Results are now reported in Mol s-1 (with the Gg yr-1 reported in parentheses for comparison to inventory). We have also expanded the Monte Carlo approach section

L431: Could you look at a slope of the CH_4 to CO_2 tracer plot in the plume to show this consistency with the reported ratio?

âĂć Its really the anomaly, and we are comparing the emissions, which accounts for this in a way that a scatterplot would not.

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

âĂć Calibration tanks for the two platforms were different, but from the same vendor. Agreement was 99.7% for CH_4 and 99.9% for CO_2. Comparison of the median winds showed is 38% for Ueast is 27%. A figure of the altitude variations is shown as Supp. Fig. S7.

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?

âĂć As noted in the text, this disagreement above the PBL is due to AJAX data being influenced by being collected in the lee of a mountain peak. This paragraph is onluy for winds above the PBL, The next paragraph is for winds in the PBL, where there was good agreement.

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.

âĂć The upwind profile and similarly the downwind profile are not for the purpose of characterizing the upwind or background concentrations, but solely for the purpose of identifying the PBL. We do compare extensively a range of parameters between AJAX and AMOG to show that surface profiles using topography can be used to characterize atmospheric structure. This is a second very important purpose of the manuscript, because not everyone has an airplane or access to an airplane, particularly in the developing world. We fully agree it is nice to have an airplane and I wish I had one of my own.

Additionally, the upwind and downwind profiles are "inconveniently" located with respect to the study area (which is approximately 50 km to the south of the upwind profile. In the case of the flux calculation, stranded layers are not evident in the airborne profile data. This could be because they are more likely to occur in the mountains used for the surface profile, which is speculative, and thus is not mentioned in the manuscript. Given the typical air flow patterns, such features will typically not return to the SJV center when formed on the east mountains, but would for formation on the west mountains.

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 incidentally)? Seems to me the emissions estimate actually overlaps with the inventory quite well given the uncertainties that are reported.

âĂć Text adjusted to note that the derived flux lies within the inventory uncertainty, but is higher, and then continues to note this is consistent with the metastudy of Brandt et al., that inventories likely are too low.

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?

âĂć Agreed, rewritten – see response above.

Conclusion

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

âĂć Removed

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!

âĂć Rewritten.

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.

âĂć Good points, and an additional sentence has been added to highlight.

Supplement: L26 cfm should be given in metric

âĂć Yes, done.

What are some estimated uncertainties on the FGGA CH_4 measurements based on the calibration standard - how often is it sampled, is 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?

âĂć Added

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 measurements - which number applies here? Where do the authors get the accuracies reported for the other analyzers (ozone, etc)? Manufacturer?

âĂć This has been clarified. The accuracies are from the manufacturer and include 24 hour drift.

If the main paper is not about these auxiliary gases, this information should not really be mentioned and could be removed.

âĂć We prefer to describe the system completely, and to include (with better explanation) the improvement in accuracy of the 450i by hourly zero measurement, as this could be of interest to other researchers

Interestingly, no accuracy or uncertainty is reported for CO_2 or CH_4, the main gases of interest in this work (for the AMOG measurements).

âĂć Added

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.

âĂć The MMS is a NASA developed system that has not been published. We provide a link to the homepage, and report its accuracies. Additionally, information on AMOG winds is now included, and explained as it depends on velocity of AMOG and the velocity of the winds.

Please also note the supplement to this comment:
https://www.atmos-meas-tech-discuss.net/amt-2017-133/amt-2017-133-AC1-supplement.pdf

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