

***Interactive comment on* “Quantifying methane emissions from natural gas production in northeastern Pennsylvania” by Zachary R. Barkley et al.**

Anonymous Referee #2

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The manuscript "Quantifying methane emissions from natural gas production in north-eastern Pennsylvania" by Barkley and co-authors presents CH₄ emission rate estimates for a natural gas extraction area in the north-eastern United States. The authors use observations from 10 individual flights, high-resolution CH₄ transport simulations in combination with an optimisation approach (inverse modelling) and mass balance calculations for their estimates. Although, there is large interest in such independent emission rate estimates throughout the US, the presented results somewhat fail to convince that the applied methods are suitable for a robust estimation. Especially the presented uncertainty assessment remains largely arbitrary and needs further improvement. In addition, the manuscript should be shortened and needs a number of

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small and technical corrections.

Major comments:

I have three major areas of concern that are briefly mentioned here, but more details follow below along the course of the manuscript.

Model optimisation technique (inverse modelling): The applied optimisation technique uses a very simple cost function to find an optimal factor between 'prior' and 'posterior' emission rates. Why did you not apply a Bayesian approach that could take uncertainties in the prior values and observations/simulations into account? Much of the following uncertainty assessment could then be used in the optimisation step and also more reliably give posterior uncertainties. Also, why do you not use sum of squares in the cost function as is commonly done? Some explanation is given later in the text, but without further discussion of Figure 16 this remains useless. Please discuss why your method should provide more reliable results as other similar studies that have used a Bayesian framework.

Uncertainty assessment of obtained emission rates: The uncertainty assessment of the obtained emission rates for both the 'model optimisation' as well as the mass balance method contains a lot of arbitrary assumptions and does not seem to be statistically sound. There seems to be an overall assumption of Gaussian uncertainties (although not explicitly mentioned), which does not seem to be justified for several of the sources of uncertainty. Also the way the uncertainties are propagated through the individual methods remains very vague and could be improved by using a Monte Carlo type uncertainty assessment. These concerns are repeated in detail below.

Length: With 21 figures, 8 tables and a total of 50 draft pages the manuscript is quite lengthy and it would profit from shortening and restructuring. Below I suggest a number of figures that could easily be omitted or moved into a supplement without loss of information. Furthermore, I strongly encourage to incorporate the description of the uncertainty assessment method, which is now given within the results section, into

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the methods section along with or following the description of emission rate estimation itself. In this was many repetitions could be avoided and the paper could really focus onto the results in section 3.

Minor comments:

L51: Shouldn't you rather say that for emission rates larger than 3 %? Otherwise the sentence does not define a threshold.

L232-238: This paragraph should be directly in front of the one describing the data assimilation strategy finally used in the study (L246-253).

L261: The important time scale for the mass balance approach is not alone the flight duration but also the transport time from the upwind to the downwind interface of the box. Also the time from emission to sample should be discussed, as this is the crucial one for the inverse modelling.

L312 and elsewhere: I don't like the terminology here. "observed CH₄ enhancement associated with upstream natural gas". The described quantity contains information from both model and observation and, thus, is not purely 'observed'. Maybe the term 'observation-based' or something else that identifies the real character of the data could be used instead.

L321: How frequent and how large are negative values of X_{GasO}?

L352: Were individual atmospheric densities for each flight neglected? Why?

L416: If this is a good correlation coefficient, what is the range obtained for the other flights? Could this information be added to Table 4 or 5. It could also be used as part of the uncertainty assessment. See comment below.

L418f: Be a bit more specific concerning the near zero differences between model and observation. For the PBL height this statement may be true, however, it is also clear that the model is not able to capture the very sharp increase in potential temperature. I

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also wonder why the two profiles are shown separately, would be nice to have them on top of each other. Concerning the wind there is a directional shift of about +10 degree in the model. You may call this small, but for the kind of plumes simulated here it might be of importance and should be mentioned.

L424ff: Please add a number for the average and maximum enhancement due to up-stream natural gas sources at the northern transect to this discussion. From Figure 9 I would think these numbers are around 30 and 80 ppb. It puts the other contributions into perspective.

L473ff: This discussion on the uncertainties due to shifts in the simulated wind direction is very hard to follow. Figure 16 does not speak for itself. You will need to explain in more detail what was done to derive the figure. This discussion should actually given when the cost function is introduced.

L486: Should it be wind speed instead of wind direction?

L498ff: The way the wind speed error is assessed biases could be introduced. The average over all wind observations by the aircraft does not necessarily reflect the average wind speed within the boundary layer, since the sampling is not uniform (especially with height). It seems calculating an average vertical profile of wind speed from all the observations and comparing that to an average model profile generated from the same sampling locations should be more robust. Was this considered and decided that the aircraft sampling was sufficiently uniform?

L506ff: There are more advanced techniques for estimating boundary layer heights from model and observational data than just 'looking' at the potential temperature. For example Bulk Richardson methods are quite useful in situations when potential temperature alone is not providing explicit results. The example shown here is an easy case but I wonder about the other situations mentioned in the text and especially how large the uncertainty of the estimates becomes in such situations.

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L542f: How was this done exactly? Did you simply run 3 cases with 1) reference background, 2) background -5 ppb, 3) background +5 ppb? Or did you do a Monte Carlo approach where the +/- 5 ppb could be understood as the standard deviation of a normal distribution? Please clarify and justify the approach also in the light of possible non-linear effects that would not be covered by simply running 3 cases.

L548f: It is mentioned that the +/- 5 ppb assumption for the background uncertainty can have different impacts depending on the magnitude of the observed plume and this is related to wind speeds and PBL heights. How can you justify the use of a constant value for the background uncertainty? Shouldn't this also change with meteorological conditions? For example it is mentioned earlier that it will be large when there is more entrainment, which is usually the case while the PBL is still developing, and when plumes from other sources are advected. Wouldn't it make sense to take this variability in the background uncertainty into account?

L557f: Again the question: is this done by running by simply running 3 cases or in a Monte Carlo fashion. Did you scale all categories with the same factor at the same time or test combinations as well? How do you obtain symmetric uncertainties in the final emission rate when you use asymmetric uncertainties in the non-upstream natural gas emissions?

L505, L562: Table 5 should already be referenced here and not only at the very end of the section.

L571: The estimator for the model performance uncertainty is a normalised root mean square error (NRMS) and it should be called like this to reduce the confusion. However, I am not convinced that this is the best estimator for what you want to achieve. One problem is the use of the optimised simulation, which means you have already used the information contained in the observations and thus the uncertainty estimator is not independent anymore. Furthermore, NRMS is not a good measure of what you call "similarity of pattern". The Pearson correlation coefficient should be better suited for

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this purpose. Please justify your choice. If you stay with a RMS estimation it should be applied to the prior simulations and the bias should be removed before the RMS is estimated.

L579f: You also assume Gaussian uncertainty distributions, which seems oversimplified.

L645: Figure 18 can go into a supplement without any loss of information. Instead give the rate of change of the background with time as a number in the text.

L652: Does this mean the final uncertainty estimate is the mean over the uncertainties for each flight?

L663ff and Fig. 19: The variability in emission rate estimates for individual days as shown in the figure are not discussed in any detail. Of special interest is the question of agreement between the two methods as it could lend some additional trust in the applied methods. However, the figure seems to raise more questions in the reliability. For example if 29 May is the golden day (as described earlier in the text), why are estimates from both methods so different? Also there seems to be no agreement in the variability for the 4 common days. How could you explain the large variability from the point of the emission processes? Is there any dependency on the total production on these days (following the line of thought used later for the discussion of different basins)? Please speculate on this otherwise the reader is left with the impression that your given uncertainty estimate is rather optimistic.

L666f: Repeat the range of emission rates from other studies here.

L696ff: The flight described in this section and shown in Fig. 21 is not part of the main analysis of the paper and should be omitted as it does not provide any additional insights and aspects that have not or could not be shown on the other flight examples.

Technical comments:

L26: Abbreviation (WRF-Chem) not introduced before. Do so on line 25.

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L48: "transforms" instead of "transform".

L59: Add "United States" in front of "Environmental Protection Agency".

L348: Why are the terms U, D, and L given in braces? Not necessary.

L496: "as long as" instead of "so long as".

Fig. 7: Do not show wind arrows in center panel. Not visible anyway and one gets the general flow direction from the shape of the plumes. Show center panel with same size as the individual tracers. Also use exactly the same color scale for all sub-panels. It looks like the center plot does have fewer color levels than the others. Each panel (also the center panel) requires a label.

Fig. 11: Show observed and simulated profile on top of each other (see also comment above).

Fig. 12: There is no mentioning of the wind arrows in the figure legend. Are these near surface or mean PBL winds? There is also no reference vector that would allow inferring the wind speeds from the arrow length. Choose a different color for the wind vectors. They are not well visible in the righthand figure.

Fig. 13: There is no mentioning of the wind arrows in the figure legend. Are these near surface or mean PBL winds? There is also no reference vector that would allow inferring the wind speeds from the arrow length.

Fig. 14: There is no mentioning of the wind arrows in the figure legend. Are these near surface or mean PBL winds? There is also no reference vector that would allow inferring the wind speeds from the arrow length. Choose a different color for the wind vectors.

Fig. 15: There is no mentioning of the wind arrows in the figure legend. Are these near surface or mean PBL winds? There is also no reference vector that would allow inferring the wind speeds from the arrow length.

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Fig. 19: Mention in the caption what error bars represent.

Fig. 20: This information could also be merged into Fig. 1 and save the additional figure here.

Fig. 21: There is no mentioning of the wind arrows in the figure legend. Are these near surface or mean PBL winds? There is also no reference vector that would allow inferring the wind speeds from the arrow length.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-200>, 2017.

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