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Comment on acp-2020-1262

Anonymous Referee #2

Referee comment on "Assessing urban methane emissions using column-observing portable Fourier transform infrared (FTIR) spectrometers and a novel Bayesian inversion framework" by Taylor S. Jones et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1262-RC2, 2021

Summary

The paper presents quantification of methane emissions using a network of five portable solar-tracking Fourier transform infrared (FTIR) spectrometers during a field campaign at the city of Indianapolis, USA in May of 2016. Methane emissions are estimated using a combination of Lagrangian transport model with a Bayesian inversion framework. The approach estimated both, surface emissions and background methane concentrations flowing into the city. Diffuse emissions, presumably leaks from the natural gas infrastructure, were found to be 73 ± 22 mol s^{-1}, 68% higher than estimated from bottom-up methods.

General Comments

The paper performs a valuable experiment by trying to use only a several days of total column methane measurements from EM27/SUN solar-tracking total column Fourier transform infrared (FTIR) spectrometers to estimate methane emissions at Indianapolis. In comparison to the complexities of aircraft flights or of setting up long-term tower measurements, EM27/SUN instruments are relatively easy to move around and in principle could be used at multiple urban centers over the span of several months, which could significantly help with the goal of quick greenhouse gas emission estimation required by policymakers to efficiently address the issue of climate change. A novel Bayesian inversion framework is advocated in synergy with EM27/SUN instruments to perform the mentioned methane emission estimation. In this regard, the paper is appropriate for the journal and helps to advance the field of carbon cycle. However, despite these positive attributes, there are a number of critical omissions in the description of the work's methodology that need to be fixed before this paper can be published. Additionally, the article lacks interpretation and proper discussion of the results.

Specific Comments

Lines 174-175: If there are no particles released above 2500 m, is there an assumption that layers above are homogenous or somehow not important to the total column methane measurements? What about potential transport of significant methane plumes in the mid-troposphere?

Line 186: Variable *s* and indices *i* and *j* are not clearly defined. Please define.

Lines 205-215: the units of vector **b** and matrix **B** are not properly defined. Text indicates that units of vector **b** are ppb, while units of **B** are hr^{-1} (according to Figure 5). Given such units, equation 5 does not work (since all the sums must maintain units of ppb). It is possible that authors treat **B** as unitless, a fraction of particles affected by the defined edges. But this needs to be clearly stated. Units and variables must always be defined in a proper mathematical notation.

Line 213: Please explain how the error term, ε_b is calculated?

Line 221: The definition implies that you are solving for matrix **x**, that is for an emission scaling factor $\mathbf{a_s}$ (unitless) and background time series **b** (ppb), is that correct?

Line 224: Background error was not described previously (please see Line 213 comment).

Line 273: Part of a sentence is missing, please fix.

Equation 10: if a priori scaling factors are all 1 and a prior background mixing ratio is 1.84 ppm, what is the point of the following conditions: $1 \leq i \leq n_{sec}$ and otherwise? Define i.

Equation 11: the conditions (such as $i, j \leq n_{sec}$ and $i = j$) are not explained.

Line 284: It is not clear what is meant by observations i and j. Are these the same as in section 2.4?

Lines 301-302: On these lines there is a following statement, "…the diagonals of which

describe the model framework's ability to reduce the uncertainties of the priors…" Please explain what do you mean by an ability to reduce the uncertainties of the priors? How an uncertainty could be reduced given that it is determined subjectively (as described in the methodology section)?

Lines 315-316: Although the diffuse emission value makes sense with all the days combined, individually the emissions look unrealistic as they are shown in Table 4. How can emissions jump from 40.4 mol/s to 146.3 mol/s in 2 days? And what is the meaning of negative emissions? It is not clear why the uncertainty is only 22 mol/s given a sample of only 5 days (each with significantly different values). Physically this is highly improbable. As an experimental product this result may be of an interest, but it should not be interpreted as an actual value of diffuse methane emissions for Indianapolis. As a cross check, it may be a good idea to take data from INFLUX towers and perform an inversion for the same days to see if the emission results will be comparable. Also, aircraft can be used to perform mass balance when available. Now, it is understandable that these comparisons may be outside of the scope of this work, but then the presented results must note that they are experimental in nature and are not to be interpreted as actual natural gas emissions from Indianapolis. Please address this in the results/discussion section.

Figure 11: What part of edge do dashed optimized background values represent? Is that combined optimization of background from all the instruments?

Figure 13: This figure seems cluttered, is it possible to present these data in the time series format? Also, what is the significance of the selected bins?

Figure 14: It is not exactly clear how the model decides whether to adjust diffuse emissions or background mixing ratios (the choice seems subjective as it should depend on the prescribed a priori error). It would be great to have a sensitivity analysis where it could be clearly shown what happens when the errors of both, diffuse emissions and background mixing ratios, are allowed to vary to see if there is any stability in the shown solution.

Lines 326-331: Varying the prior of the spatial distribution of emissions significantly changes the result of the total diffuse emission estimation. That in itself shows that each posterior result should be approached with caution.

Figure 16: Please define on one of the maps what is precisely meant by "Marion County".