Comment on acp-2020-1262
Anonymous Referee #1

Referee comment on "Assessing Urban Methane Emissions using Column Observing Portable FTIR Spectrometers and a Novel Bayesian Inversion Framework - Revision 1" by Taylor S. Jones et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1262-RC1, 2021

Jones et al. present an inverse modeling study on methane emissions from the city of Indianapolis. While a number of studies have been published to address the same problem, the study is unique and novel by using data from five portable solar-tracking Fourier transform infrared (FTIR) spectrometers and an inversion method devised to comprehensively account for uncertainties, especially those uncertainties that are not often adequately addressed in the literature, namely, uncertainties of the background methane concentration, and uncertainties in the spatial pattern of the prior inventory. As such, this study is positioned to complement previous efforts in painting a fuller picture on the issue and to help point to future directions of research. The paper is well written, including key information regarding the inversion method and a concise presentation of the key results in figures and tables. I recommend publication of the paper, and provide the following questions and comments for the authors to consider. I realize addressing the comments below might require some additional model experiments. I encourage the authors to do what they deem as appropriate with available resources and time, as I believe a better understanding and discussion of these issues would further strengthen the paper.

I think the section on the results (Section 3) should be expanded. In its current form, with text of less than 30 lines, this section does not go much further from summarizing the numbers from the figures and tables and comparison with previous estimates. I suggest the authors to include more discussions, highlighting the unique findings from the study with previous studies as the context, and offer some experiences and suggestions for future work based on the results. Here are some comments and questions for the authors to consider:

The authors offer an explicit approach to calculating the background concentration with STILT (Eq. 5), which to me is necessary and a preferred approach in future studies. I suggest the authors further explore and demonstrate the benefit of using this approach, e.g., by doing another inversion that does not treat the background with a background influence matrix. How would the emission estimates change?
One intriguing finding from the study is that two prior spatial patterns have led to drastically different results. This finding should be further explored and discussed. The study uses a small state vector with just a few terms as scaling factors for the emissions from different sectors; this approach implies considerable confidence/weight on the prior spatial distribution in fitting to the observations. An alternative approach that has been used previously would be to solve for the spatial pattern with a bigger state vector that leaves more degrees of freedom on top of the prior spatial distributions, i.e., scaling factors of emissions from different parts of the domain, for example, by dividing the domain into a number of squares a few kilometers in dimension. This approach would mean the data can have more freedom in informing the spatial pattern of the posterior. What are the considerations being such a choice of state vector? Is it because of the lack of data points from FTIR, or is it because the results make more sense this way? Have you tried, and if not, is it feasible to try another inversion with a different configuration of the state vector?

The results presented in the figures and tables need some discussions and explanations. For instance, Figure 15 shows large day-to-day variations of diffuse methane emissions of about a factor of 5 (if taking the minimum of 50 mol s\(^{-1}\) on 5/13 and maximum of 250 mol s\(^{-1}\) on 5/22). Can you explain the reasons behind this change? Figure 15 apparently shows that using roads as the spatial pattern for the prior, an inversion using all data gives a higher emission estimate than estimates on all individual days. Why is this?

The data used by the study, i.e., FTIR column data, is somewhat unique by its own right. It would be very beneficial for future studies if the authors can, based on their experiences, summarize and discuss a few key considerations future studies should bear in mind when using this type of data. For example, what are the favorable situations to use these data, what supplemental data are needed, and what would be the minimum number of sensors needed.

Technical errors:

Line 54: “We focus of” should be “we focus on”

Line 55: “have proven” should be “have been proven”