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Interactive comment on "Improving Mobile Platform Gaussian-Derived Emission Estimates Using Hierarchical Sampling and Large Eddy Simulation" *by* Dana R. Caulton et al.

Anonymous Referee #1

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This paper presents an ambitious and valuable attempt to quantify uncertainty in estimate of methane emissions from natural gas wells using downwind, automobile-based measurements of methane concentrations. I believe that the strengths of the manuscript include the assessment of sampling error derived from idealized LES of plumes, and the attempt to provide an overall uncertainty assessment for this methodology. The methodology is not especially unique, but the breadth of the effort is important and worthy of publication.

The manuscript at present, however, contains shortcomings that make it unsuitable for publication in its current form. Major revisions are necessary. I first present my

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overarching concerns, followed by a line-by-line assessment following the order of the manuscript.

Major issues.

1. Basic issues concerning the methods are not explained, thus many of the most important results cannot be clearly interpreted. I have included specific comments for many figures, tables, and text in my detailed comments. I mention what I consider to be the most important overall points here.

1.a) Table 5 is arguably the most important product of the manuscript, but the basic elements that are used to build this table are never clearly articulated. The manuscript should be reorganized to clearly explain the basis for the uncertainties being imposed in the inputs to the Gaussian plume emissions estimates, and the uncertainties in these inputs should be clearly stated. This is true with a limited number of inputs in this table (instrument precision, stability class), though even for one of these inputs, the source of the uncertainty in the inputs to the model is not clear (e.g. why is one class the assumed bound of uncertainty?, what time resolution is associated with 5 ppb precision in the LiCor 7700?). Atmospheric variability, the most important issue according to the table, is never defined. The main finding of the manuscript must be clearly defined to be useful and interpretable.

1.b) The method for determining single vs. multiple sources, or the method for evaluating the number and spatial distribution of multiple sources, is never explained.

1.c) The method used to remove background concentrations from the field measurements is critical, and is never discussed. This is a major source of uncertainty in plume dispersion estimates, and is not included in the results. This must be addressed.

2. The averaging applied to the downwind transects is arguably inappropriate and suggests a fundamental misuse of the Gaussian plume model. The Gaussian plume model describes the ensemble average of atmospheric concentrations downwind of a

point source. This includes the fact that the instantaneous plume will meander over the course of time due to atmospheric turbulence. The model is based on the assumption of a separation of scales between atmospheric turbulence, and mesoscale or synoptic-scale atmospheric flow variations. The authors have chosen, however, to align each measurement downwind of their point sources by the peak concentration measured on each transect. The wind direction, therefore, changes with every transect. The mean wind in the Gaussian plume model should be the average wind across the time span of all of the downwind transects (which are conducted within an hour, over which the atmospheric dispersion conditions are assumed to be steady-state), and should not vary every few minutes with each transect. This approach to averaging would create a significantly broader observed plume. It is not clear to me how this might alter the estimated emissions, but this is a misapplication of the plume dispersion model that should be corrected. If the dispersion conditions within the time span of the downwind transects are not steady, this must be explicitly identified and treated in the analyses.

3. Given the limited spatial domain, the LES used in this study has no large-scale turbulence, thus its value for comparison to plume observations is uncertain. Its use for an observational system simulation experiment is acceptable, given the caveats that it only simulated neutral conditions and has no larger-scale turbulence. The authors do explain the limitations of neutral stability, but do not acknowledge that the simulation will by necessity truncate the spectrum of atmospheric turbulence that will contribute to issues like meandering of the downwind plume.

4. A major element of the uncertainty assessment relies on the controlled release experiments. This requires characterization of the uncertainty in the rate of release. No assessment of the uncertainty in the release rates are given. This must be addressed.

5. The measurement protocol chosen focused on morning and evening conditions to search for neutral atmospheric stability. Sounding on calm mornings, however, are far neutral, and morning and evening conditions are characterized in general by rapidly changing atmospheric stability. Plume dispersion modeling assumes steady atmo-

spheric stability and mixing conditions. What is done to screen the observations to avoid rapid transitions in stability that clearly violate the conditions for the Gaussian plume model?

Detailed comments.

1. Page 1, Line 14-15. " ...a hierarchical sampling with increasing complexity was implemented" is, I believe, what you mean to say. Though I'm not sure what "with increasing complexity" means. It is hierarchical, thus complex.

2. Page 1, transects, lines 15-20. How are multiple transects separated in time? Across multiple days? Or all on the same day / within the same hour? This is an important methodological distinction.

3. Page 1, Line 20. "in most cases with average differences" Be more precise. Is 25% the average difference or not?

4. Page 1, Line 24-25. "Approximately 10 repeat transects spaced at least 1 min apart are required to produce statistics similar to the observed variability over the entire LES simulation period of 30 min." This is not very informative. What is relationship between number of transects and precision of determination of the emission rate?

5. Page 1, Line 25-26. "In addition, other sources of uncertainty including source location, wind speed and stability were analyzed." This is too vague to be informative.

6. Page 1, Line 26. "atmospheric variability" is too vague. Do you mean to say sampling error caused by atmospheric turbulence? If so, isn't that a function of the sampling strategy? Is atmospheric sampling still the dominant source of uncertainty with 20 downwind transects?

7. Page 1, Lines 28-30. What is "this condition?" What is "this metric?"

8. Page 3, line 1. "lag behind the standard." In what respect?

9. Page 3, equation 1. Please incorporate the equation into the English, and please

define C and the origin.

10. Page 3, line 15. “Gifford’s”

11. Page 3, line 16-17. It is not necessary to describe the stability classes. The model does not predict a PDF of the concentration. It predicts the average concentration. These are very different.

12. The uncertainties in Table 1 are not helpful as currently presented. This Table must be revised prior to publication. I understand that they are all reported by the individual authors, but this manuscript does present them for the purpose of comparison. Many of the techniques and papers have radically different spatial and temporal domains and sampling density, and the uncertainties quoted are thus not at all comparable. How, for example, can aircraft estimates of the mass balance for an entire basin be compared directly to the uncertainty of a single chamber sample, or a single Gaussian dispersion estimate, without an understanding of what this uncertainty bound is trying to represent? This table could be enhanced to include some information about the domain and sampling density associated with these studies. This would be challenging, but much more interpretable.

13. Page 4, lines 3-12. Are these uncertainties for single emissions estimates from a site using an individual “transect” of data? Please clarify. See my concerns above. Without specifying more about the temporal and spatial domains, these uncertainties are difficult to compare or interpret.

14. Page 4, line 19. What is “an averaging plume for unconstrained instantaneous measurements”? Please clarify. I study boundary layer meteorology but do not understand what you are trying to say.

15. Page 4, line 25. I don’t understand, “this method.” Are the authors proposed a new standardized methodology for quantifying emissions? Or is this a study of the uncertainties in plume measurement / source quantification methods?

16. Page 5, line 5. The methane span gas concentration is quite low for the measurement of plumes at close range. Are there any tests of the linearity of the instrument at higher concentrations that I assume are encountered driving downwind of strong methane sources?

17. Page 5, lines 19-20. “site were screened”: Does this mean that sites with trees were eliminated from the sample? The text later states that it is hard to measure a plume at a distance greater than 300m if trees are present. Can you please explain the site selection algorithm more carefully?

18. Page 6, line 7-9. Does the NOAA web site provide observations, or numerical model reanalyses? Please clarify. How does this NOAA web site account for the impact of local roughness on atmospheric stability? The hilly, forested, heterogeneous landscape of Pennsylvania will have a significant impact on local dispersion.

19, Page 6, equation 2. As with equation 1, please make equation 2 part of the sentence.

20. Page 6, line 20. This description is insufficient. The Gaussian plume model describes only the concentrations caused by the local source. The measurements do not. A concentration background must be defined and subtracted from the observed time series. This is a critical and non-trivial step. How was this done? I am also concerned that this method does not take into account the correlation between the modeled and observed concentration enhancements. Systematic errors in the dispersion would not be identified by simply matching the integrated downwind enhancement.

21. Figure 2. This figure is concerning. First, the figure shows concentrations that differ by a factor of 1000, and no explanation is offered. Second, the background that must have been removed from the observations is not discussed. Third, there appears to be a systematic difference in the width of the observed vs. modeled plume. This implies a systematic error in the dispersion coefficient, which will lead to a systematic error in the emissions estimate. How is this addressed? Finally, given that instrument

characteristics are expressed in units of ppb, it would be useful to also present downwind concentrations in these units. It would be easy to include both mass density and mole fraction as two different y axis labels.

22. Page 7, line 11. How is steady-state evaluated?

23. Page 7, line 11. What does it mean to “rescale” the LES output to the measured momentum flux? Isn’t the LES set up to simulate these sites? Why is rescaling necessary?

24. Section 3.1 The LESs are truncated in the vertical and thus cannot include large-scale turbulence that will influence the sampling statistics. This will be particularly important for unstable conditions. How is this dealt with in designing the sampling strategy?

25. Section 3.1. The LESs say nothing about sensible heat fluxes. Turbulence statistics change dramatically as a function of stability conditions. What stability conditions are simulated?

26. Page 7, line 25. Short intervals will sample the same structures in the turbulence. This is well known. Some of the existing literature on sampling statistics in atmospheric turbulence should be cited here.

27. Page 7, line 26. What does “1-N” mean? What is N? I read the description, but I do not understand what this means. What defines the number of samples available in a numerical simulation?

28. Page 8, lines 8-9. What is the difference between turbulence and plume meandering? Isn’t plume meandering caused by atmospheric turbulence?

29. Page 8, lines 10-12. What does it mean to “reflect the actual variability in the atmosphere”? This is not meaningful. Please define a quantitative tradeoff between transects and flux retrieval.

30. Figure 3 is not very helpful. Please include vegetation and topography.

31. Figure 4. Images of instantaneous vs. averaged plume structures do not need to be published. This is well known. The manuscript has no references to several decades of simulations of plume dispersion. This is a serious hole in the scholarship of this manuscript. Look up, for example, the publications of Jeffrey Weil, circa 1990.

32. Figure 5. Issues. 1. There is a lot of blank space. 2. The percent difference figures show results from a distribution. What are the elements of the distribution? Is each “pseudo-transect” an element of the distribution? Please define this clearly in the caption. 3. A standard deviation is a description of a population. Are there multiple populations whose statistics are being compared? Please clarify. 4. What does “random seconds” mean? Why is this useful? 5. There is little additional information given in the 30/60/120 second sample repeat figures, and the differences are difficult to see. It would be more effective to show one of these, then have one additional figure showing the differences caused by changing the sampling interval.

33. Page 8, lines 18-19. The original plan is not relevant at this point. Present the number of samples obtained.

34. Page 8, line 24. What is the “average maximum percent difference”? Maximum among what? Average across what? And why is this relevant? Please also explain the populations used to define the standard deviations.

35. Page 8, last sentence. What is the population being discussed here? Why are these numbers important?

36. Figure 7 does not explain how the number of sources will be determined. I also disagree with the far right-hand bar. A more complex simulation does not ensure less uncertainty, and “model uncertainty” is not defined. If there is a precise definition of uncertainty that can be shown to be reduced with the LES, please explain and define this.

37. Page 9, line 17. What is a "warm up time"? (I see here that the determination of steady state is explained.)

38. Page 9, line 21-24, and figure 8. This discussion and figure displays a fundamental problem with the LES setup, and the authors' interpretation of these data. The "real world" meandering of the wind that is being described is turbulence in the atmospheric boundary layer. A Gaussian plume model describes the average state downwind of a point source, which would be properly represented by the average of all of the transects, without "aligning" them to match the peak concentration on each transect. Aligning these is the equivalent of performing some sort of high-pass filter.

39. Similarly, I am now concerned about the processing of all of the transect data. In Figure 2a, for example, have all of the transects been arranged so that the x direction changes from transect to transect? If so, the same odd filtering of turbulence has been applied. This is not appropriate for comparison to a Gaussian plume model. I expect this could explain the difference in plume widths that is evident in Figure 2 (note the modeled plume is wider that the observed plumes that may have been filtered to remove large-scale turbulence)/.

40. Please explain the theoretical basis for equation (3)?

41. Section 4.1. How do you distinguish among single vs. multiple sources? This is not explained.

42. Page 10, line 2. The two Gaussian retrievals "compare"? What does this mean?

43. Figure 9. Once again, please explain the populations of data that go into these box and whisker plots. Is each point a transect? If so, how many transects make up each population?

44. Page 10, line 13-14. I do not agree that the results show no apparent bias. Figure 9a has winds that are known to be too high, according to the text. Figure 9b shows a systematic underestimate of emissions. Figure 9c shows varied results.

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45. Figure 10. What is the level of uncertainty in the release rate? If this release is being used to evaluate the methodology, then the release rate must be carefully calibrated. How has this been done?

46. Page 10, line 22-23. Only one outlier with zero emission is shown in Figure 10.

47. Table 2. Please report the observe and simulated winds, and the observed and simulated stability conditions. Please include more descriptive information in the table caption. In particular, please note how many transects were collected at each site, the time span of the transects, and the source of the uncertainty values given in the table. If these are standard deviations based on emissions derived from individual transects, and the purpose is to compare mean values, the authors should report the standard deviation of the mean, not the standard deviation.

48. Table 3. Please include more descriptive information in the table caption. In particular, please note how many transects were collected at each site, the time span of the transects, and the source of the uncertainty values given in the table. If these are standard deviations based on emissions derived from individual transects, and the purpose is to compare mean values, the authors should report the standard deviation of the mean, not the standard deviation. Also, please report winds and stability. And were the winds observed, or taken from NOAA reanalyses?

49. Page 10, lines 27-28. See my earlier concerns about centering of the plumes on each transect. I believe this is an erroneous interpretation of plume dispersion.

50. Page 11, lines 7-8. The overall differences are small? The flux estimates in Table 3 differ by as much as a factor of two. This seems large to me.

51. Figure 11. This figure points to many questions and problems. 1) What are the equations for the vertical and horizontal flux profiles in the Gaussian plume model? 2) What is the position in the domain where the profiles (figure d-g) are computed? 3) What is the distance downwind for everything shown in this figure? 4) How are the

multiple sources chosen? As best I can tell this is never described anywhere in the manuscript. 5) Does the LES flux include subgrid and resolved fluxes? Please explain, and delineate these two sources so that it is clear what fluxes are explicitly resolved. 6) What does z=3 mean? 3 what? The same comments and consternation hold for the similar figures in the supplementary materials.

52. Page 11, lines 12-14, sentence starting with “regardless.” I do not understand what you are trying to say.

53. Table 4. What is the distance from source of the measurements? Are “emissions” those estimated from transect measurements? How many transects? What are the meteorological conditions for these measurements?

54. The text in the first paragraph of section 5.1 does not appear to match the results in Table 4. The text notes that unless the change in source location in the x direction is similar to the distance of the measurement downwind, that the impact of the estimated emission is small. Table 4 shows two examples, one of which has a 150% change in source strength estimate, and another a 7.5% change in source strength estimate, and as best I can tell (Table S1?) the measurements are both from about 150m downwind of the source, with the site with a larger % change having measurements farther downwind. Based on the results presented by the authors, as best I can interpret them, I disagree with their conclusions.

55. Figure S10 (b) is uninterpretable. What is the “Ratio between the sum in y and distance x of the scenarios”? Is this truly a ratio of distances that is being plotted? Please clarify.

56. Figure S11 (b). See comment above regarding Figure S10.

57. Page 11. “NOAA wind speeds differed from the tower data on average by 50%.” This is uninterpretable. Please rewrite this to be meaningful.

58. Figure S12. Please define this ratio, as with Figures S10 and S11.

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59. Page 12, lines 7-8, “The magnitude of the difference between consecutive stability classes is relatively consistent, averaging 40%.” Figure S12 does not suggest simply defined average, as the signs and magnitudes of the differences change as a function of distance downwind. Please explain what this 40% value means.

60. Page 12, lines 12. Please define “absolute terrain slope.”

61. Page 12, lines 24-28. The methods for quantifying the uncertainties should be explained carefully in the methods. This rapid-fire, qualitative overview of the methods behind Table 5, arguably the most important result of the manuscript, is insufficient.

62. Page 12, lines 30-34. See previous concerns about the lack of larger scale turbulent motions in the LES used for this study. I agree in general that the LES used here should provide a lower estimate of turbulent sampling error, but it also will not contain the full spectrum of atmospheric turbulence and true turbulent sampling error. This should be explained, and references to the rich literature of the full spectrum of atmospheric turbulence and the limitations of LES that is limited to the atmospheric surface layer should be included in the manuscript.

63. Page 13, line 6. The ranges of the input values used in the Monte Carlo simulation and their distributions must be described, or these results are meaningless.

64. Table 5. See prior concerns about the lack of documentation of the methods for assessing these uncertainties. In addition, how is the “total”uncertainty assessed? Is this the result of the Monte Carlo simulation described in the text? If so, see prior concerns about documentation of this experiment. If not, please explain how this total is computed.

65. Figure 12 results vary by a factor of 1000 with no explanation.

66. Section 5.3. As best I understand this work, the order of averaging that is varied is certain to cause no significant change since the relationship between concentration and source strength is linear. Thus this comparison is not informative or significant. If

anything is nonlinear that could cause a difference, please explain.

67. Page 14, lines 14-15. I do not see how your results justify this statement.

68. Page 14, lines 22-23. I do not yet agree with the assessment that no bias is observable. This cannot be assessed until the uncertainty bounds in Table 2 are clarified. If the mean values differ significantly, then there is observable bias.

69. Page 14, line 23,"LES is therefore not required for studies where source strength calculation is the main goal." The authors have clearly avoided complex terrain where LES is most likely to be needed. This broad and general statement is not justified by the research presented in this manuscript.

70. Page 14, lines 24-25. "From this we use Monte Carlo analysis to extrapolate that the 95% confidence interval for sites with standard sampling (n=2) ranges from 0.05x–6.0x where x is the emission rate." This is a potentially important result, but this is offered as a passing comment. The methods behind this calculation are not presented. This is not acceptable for publication. This is an important result whose methods must be clearly explained and defended.

71. Page 15, lines 3-4. Rella et al, (2015) did not use a Gaussian plume dispersion approach, and attempted to measure the entire vertical extent of the plume. This manuscript notes that vertical dispersion is a major source of uncertainty in their results. It seems very likely that Rella et al's approach should, therefore, yield a significantly smaller uncertainty estimate than a method that relies on a Gaussian plume dispersion model to quantify vertical dispersion.

72. Page 15, lines 5-6. "As described in Sect. 3.2, the observed atmospheric variability can range from 10-200% meaning that in-situ observations of variability and post screening out conditions with unacceptably high variability may be a viable way to reduce uncertainty." What is the quantity "atmospheric variability," that can range from 10-200%, and 10-200% of what? What criteria of "unacceptably high variability" can

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be used to screen out data and thus reduce uncertainty? This reasoning is either explained poorly or imprecise, and the conclusions stated are thus either uninterpretable or inaccurate. This statement is not suitable for publication.

73. Page 15, recommendations. Point 1 cannot be satisfied in many cases. What happens when measurements are needed for a location that does not satisfy these criteria? I don't understand point 5.

74. Point 6 is not a significant recommendation resulting from this research.

75. I do not understand "the strategy" proposed in Point 8. It is not clear how this collection of measurements is proposed as an integrated sampling strategy.

76. Section 8. Data should be publicly available, and not restricted to access only via correspondence with the authors.

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