

Review of “Optimization of the Gaussian Dispersion Model Inversion for Estimating Facility Scale Methane Emissions in Canada”

By Yamanouchi et al.

Anonymous Reviewer

Summary:

Yamanouchi et al present a method to estimate source emission rates from mobile atmospheric sampling platforms. Here, they use a Bayesian approach to solve a Gaussian Plume model. The novelty of the approach, like many efforts before, is to parameterize the atmospheric dispersion coefficients. In sampling a transect, this parameter constrains the width of the Gaussian shape that is fit to the transect data. The goal is to establish a reliable and quick method to find and quantify unwanted emissions. Yamanouchi test their method against controlled releases and around several facilities (an oil refinery, a recycling center, and a recycling plant).

Overall, this work provides another application of Gaussian Plume Models in mobile systems, which is valuable, but the methodology has several limitations that need to be addressed. First, the methodology is not well described, omitting crucial details of the experiments (see General Comment 1). Next, in applying a Gaussian Plume Model there are important caveats that need to be considered that are not addressed here (see General Comment 3). It is possible that these caveats are addressed and just not described in this manuscript.

This manuscript will require major edits prior to publication.

General Comments:

1. Description of Experiments and Data Collection: More detail is needed to describe the data, including how the experiments were set up, what sensors were used, and how the sampling system was set up. Below is a list of parameters that absolutely need to be included. I suggest editing Table 1 to add some of these parameters.
 - Release rate of controlled experiments
 - What sensors (gps, ...)
 - Sensor flow rates
 - Distance downwind of the source which the transects were collected
 - # of transects
 - Speed of mobile station
 - Time sampling

- Height of release
2. Validity of HRRR Data: More description is needed that can help the reader follow and recreate what the authors perform. This work relies on outputs of this model to determine the dispersion parameters. In comparison to the time and spatial scales of the measurements, the resolution of HRRR is very coarse.
 - a. Are the authors using output from HRRR (operational) or HRRR-x (experimental)?
 - b. HRRR provides hourly output. Is that high enough resolution to meaningfully compare to in situ (1-Hz) measurements or transects that lasted for an unknown amount of time?
 - c. Can HRRR provide valid wind data for a sampling height of 2.5 m agl? The lowest atmospheric layer in HRRR is typically ~6-12 m agl meaning that the lowest elevation wind parameter (e.g. U1000, V1000) is typically well above the sampling height presented.

Broadly, it would be expected that wind speed at 2.5 m agl is less than that at 10 m implying that using HRRR will overestimate the emission rate. However, the impact is probably more complex than that and probably depends on the height of emissions and vertical velocity components, which are not currently discussed in this manuscript.

- d. Please provide the specific parameters used from the HRRR output.
 - e. General description of the parameters that the authors used from HRRR is needed. For example, Surface roughness in HRRR is assumed from the MYNN Surface Layer Scheme. In MYNN, that is largely dependent on land use, greenness fraction, and snow depth. This allows the reader/reviewer to assess whether these are reasonable parameterizations. Please include citations for HRRR (<https://journals.ametsoc.org/view/journals/mwre/144/4/mwr-d-15-0242.1.xml>) and MYNN (<https://repository.library.noaa.gov/view/noaa/30605>)
3. Gaussian Model and Fitting location: There are a number of assumptions and limitations within a Gaussian Plume model that are not addressed. Please see Seinfeld & Pandis “Atmospheric Chemistry and Physics” (chapter 18, see references therein). A very simplistic version of the Gaussian Plume equation that assumes total reflection at z=0 can be written as (S&P eq 18.88):

$$\Delta C(x, y, z) = \frac{Q}{U} \frac{1}{2\pi \sigma_y(x) \sigma_z(x)} e^{\frac{-y^2}{2\sigma_y(x)^2}} \left[e^{\frac{-(z-Z(x))^2}{2\sigma_z(x)^2}} + e^{\frac{-(z+Z(x))^2}{2\sigma_z(x)^2}} \right]$$

Where ΔC is the methane mixing ratio above background at location x, y, z . Q is the flux rate. U is the windspeed along flow of plume. $\sigma_y(x)$ and $\sigma_z(x)$ describe the Gaussian width horizontally across the plume and vertically at distance x from the source. $Z(x)$ is the height of the plume at distance x from the source.

In this equation there are essentially 9 parameters: 4 that should be known (3 for the sample locations, 1 for the release height), 3 derived from HRRR (windspeed, $\sigma_y(x)$, $\sigma_z(x)$), 1 measured value ($\Delta C(x,y,z)$), and essentially 1 parameter (Q) that is solved for with the Bayesian approach. A similar equation must be used and all of the parameters need to be discussed in terms of what they are, how they influence the interpretation of Q, and how they are measured/modeled/parameterized.

- a. What version of Gaussian Plume equation is used? Please provide the equation. Are you assuming 100% absorption by the ground or ground reflection? Reflection of the plume off the PBL? Please provide the Gaussian Plume equation
- b. How is vertical velocity accounted for? The authors mention "Briggs plume-rise scheme" (L144), but do not provide a citation. How is a reviewer or reader supposed to reproduce this?
- c. Specify, maybe in a table, the values of parameters that are modeled/derived/parameterized, including $\sigma_y(x)$, $\sigma_z(x)$, plume height at the transect ($Z(x)$) for each transect measurement set? What are their uncertainties? How do these uncertainties affect the interpretation of Q?
- d. Why initiate the model for the selected parameters, e.g. 1 g (CH₄)/sec? How would changing to a different initialization state affect the results? My guess is that because the Bayesian approach solves for only 1 parameter, it is insensitive to the initialization state.
- e. Practically, it was unlikely that the transects were perfectly orthogonal to the direction of flow (wind direction). How is the non-orthogonal transects accounted for? If plumes were perfectly Gaussian, then a non-orthogonal transect would introduce skewness to the measured concentration profile.
- f. How was weighting b/w stability classes performed?
 - i. Which set of stability classes are used? I can infer from relation to surface roughness and Obukhov length, but there are other classifications that are in use, including; US EPA 1970 ("Workbook of atmospheric dispersion estimates"), Gifford 1961 ("Use of Routine meteorological observations for estimating atmospheric dispersion"), etc.
 - ii. Weighting is performed b/w 2 parameters with different operational ranges ($z_0 \sim 0-0.6$ versus $1/L \sim -.15 - 0.1$). Is one value weighted more than the other? Is it simple Euclidean distance between stability classes? What is being weighted, the dispersion coefficients or the resulting Q of each class?
- g. How do the authors account for the averaging time of their measurements?

Most stability classifications are based on average distribution of the emission across multiple hours (see Briggs 1972, Pasquill 1971, etc). This will result in a much broader plume distribution than would be observed if one could instantaneously sample the complete transect. A quote from Briggs 1972 exemplifies this: "However, the comparison made with the range of β values observed by Hoult et al (1969) is not a fair one since these values represent best

fits to many individual data, while the Schwartz and Tulin β values are best fits to several sets of *averaged* data.” Below is a figure from Seinfeld and Pandis demonstrates the increasing plume width with increasing measurement time.

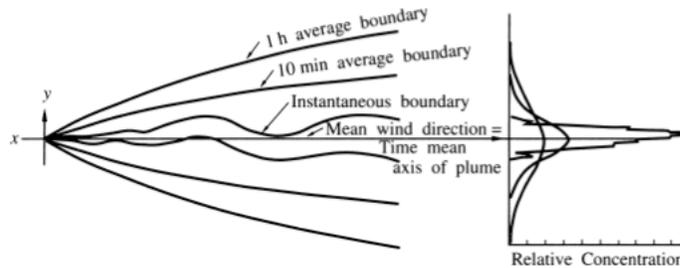


FIGURE 18.2 Plume boundaries and concentration distributions of a plume at different averaging times.

Figure 1: Seinfeld & Pandis, "Atmospheric Chemistry and Physics"

Using dispersion constants that are derived from long sampling periods will overestimate the dispersion and width of the Gaussian shape used to fit the data. The result will be an overestimate of emissions as the model thinks those emissions are spread over a larger area.

- h. How do the authors account for the non-instantaneous measurement of the transect? As the vehicle proceeds along the transect, the wind is also changing direction meaning that the mobile platform is never sampling the same plume distribution. This is a major hurdle for all mobile platforms and needs to be discussed. Long sampling periods with many transects will be essential, but the sampling time and number of transects are never discussed.
4. Results & Discussion: This section should be restructured and address critiques provided in General Comment 3.
 - a. Presenting more of the transects in figures would improve communication of the technique and provide more transparency (see comment about Figure 2). While the authors show data from transects of 3 successful uses of their approach (Fig 2, 3, 5), they do not show any data from the other transects. There are 14 datasets and several are discussed (l242-257) that did not behave as those simple, easy to interpret datasets.
 - b. Discussion of fitting of source location is not particularly impactful and, in my opinion, does not justify the amount of space that is discussed here. It is intuitive that the plume axis (center) is observed at a different location than the wind data implies. Significant uncertainty in wind direction arises from using the HRRR datasets: 1) Winds are rarely uniformly linear as implied by using 1 hr HRRR wind data, and 2) the 3km average likely does not represent ground-level winds that are influenced by local features like buildings, trees, etc.

- c. Recommend restructuring the results with subsections for 1) Controlled releases, 2) several possible sections discussing limitations of approach, 3) application to facility data.

Specific Comments:

- Pg 1 L 38-57 This paragraph discusses how global methane budget is estimated using top-down or bottom-up approaches. This seems tangential to this work and doesn't promote the development of Gaussian Plume Models. Suggest removing.
- Pg 3 L 65-66 "...stead-state solution with constant emission rates as well as constant and homogenous winds and terrain."
Technically, the model provides a multi-hour averaged dispersion. It is not really a "steady state" and doesn't assume "homogenous winds". See references in General Comment 3g.
- L 87 None of the listed sites in section 2.3 are described as landfills?
- L 87-89 Please help describe the measurement process in more clarity. Are some of these sample sets only a single transect? See general comment 1.
- Pg 4 L 96 The sampling system is on top of a car and only 2.5 m agl? That seems really low and only ~1 m above the car. At this height, wouldn't the car affect the windflow and measured concentration? How tall is the vegetation and other structures in the vicinity?
- Pg 5 L 125 "... stability class, accounting for 48% of the uncertainty, followed by wind direction (38%), windspeed (28%), and source location (19%)."

The sum of these adds up to >100% of uncertainty. How is that possible? Do the authors mean that there is a 48% uncertainty in the source estimate due to stability class?
- L135-136 Please provide complete list of model variables used and be specific to what they are called in the datafiles.
- L144 Technically, stability class does not affect plume-rise. Plume rise is affected by wind fields, buoyancy (e.g. composition and temperature of plume), solar heating and albedo, etc. Stability class is a qualitative description of things like plume rise and turbulence, etc.
- L144 "...Briggs plume-rise scheme..."
Please provide a reference!
- L146 Define which set of stability classes you use. Provide a reference!
- L153 Surface roughness and Obukhov length were not listed as variables from HRRR earlier? Please explain where these parameters come from and how they are derived (see General Comment 2). This allows the reader to assess validity and limitations of using the HRRR data without digging through other literature.

- Oh, and provide a reference to the HRRR model and MYNN model used by HRRRv4.
- P6 L154 How was the weighting performed? See General Comment 3f.
- L165 "...translated along (parallel to) the transect path..." both of these descriptions are a little confusing.
- L166 This is not what autocorrelation describes. Autocorrelation describes how a signal or time-series correlates with a delayed version of itself (i.e. 'auto'). Not the phasing between two separate signals (observed and modeled concentration).
- L167 How is skewness incorporated into a Gaussian Plume Model? The angle of the transect with relation to the wind direction is not discussed anywhere in this manuscript.
- P7 L170 "Autocorrelation values of 0.66 to 0.955 were chosen as cutoffs..." Again, incorrect use of 'Autocorrelation'. Assuming that the authors mean 'correlation', how were a range of values chosen as a cutoff? Did they test different cutoffs affected their interpretation of Q? Maybe using the controlled release? That would be good to do and discuss.
- Wouldn't a simple Least-Squares approach achieve the same thing without tuning a cutoff?
- L187-189 Were the other datasets only single transects? Describe how these transects were average together. "Overlaying the measurements on top of each other" is not a precise description.
- Sect 2.3 Really need to know parameters like the distance of transects downwind of source, number of transects for each experiment/measurement set, speed of the mobile station, amount of time sampling the plume. These are essential parameters in the Gaussian Plume Model (see General Comment 3).
- P8 L205 Correlation (R^2) is a poor metric for determining performance.
- Hypothetically, assuming two Gaussian shapes with the same mean but different standard deviations and amplitudes that represent our measured and model transects. These two profiles will always have a high R^2 value because they always increase and decrease together. They may not even have a slope near 1 since correlation doesn't care about that. Maybe χ^2 metric better describes the predictiveness of the model.
- L211 "...converged to give physical and meaningful emission estimates..." This does not really provide meaning without defining what a *physical* and a *meaningful* emission is. Is it within a certain % of the true emission rate?
- L222 "...extremely sensitive to..." This doesn't seem true. It is insensitive to z_0 , but is also insensitive to Obukhov length in areas with $z_0 > 0.2$, (e.g. $z_0 = 0.4$, $1/L = -0.10$).

- Sect 3.2 See general comment 3 and 4.
- L226 “The source translation algorithm generally resulted in higher correlation, lower RMSE, and higher emission estimates.”
- By definition, aligning the peak in the measured concentration with the model plume center will maximize the estimate of the Gaussian Amplitude and emission estimate. This should not need discussion.
- L228 What was the angular offset between the HRRR wind direction and the measured plume axis?
- P9 Fig 2 The 3 panels should be combined into a single panel with the different posterior estimates having different line styles. This would reduce redundancy of the figure and allow better comparison of the blended dispersion estimate to the stability classes.
- It would also be good to see the uncertainty bands of the measured data.
- Provide information on interpreted emission rate from each posterior and the known emission rate of the controlled release experiment.
- L238-239 “This is supported by the fact that the overestimation was present before the source translation took place.” I’m confused by why this supports your hypothesis of uncertainty in downwind distance from the source.
- L242 What is meant by “real” observation data?
- L242-258 It would be good to show some of these transects, similar to what is suggested for Fig 2. Maybe with a subplot for each of the non-ideal datasets discussed here.
- P10 Fig 3 See General Comment 4. This figure is probably not needed. Showing the transect before aligning the model to the data is not very interesting.
- L250-251 “Note that...thus emissions are not expected to agree with each other.” I am really confused by this statement. Maybe better description of the data collection would help.
- L255 “...size of the signal...” → “emission rate”
- L257 “...returned meaningful and physical results” What is a meaningful result? What is a physical result?
- L260 “...plume shape is the same whether the source is farther away and emitting less, or closer but emitting more...” This isn’t true. The source that is farther away would have a much smaller amplitude and wider distribution than the nearer+larger source.
- L261-270 This section doesn’t make much sense to me
- P11 Sect 3.3 See General comment 3 regarding derivation of dispersion parameters.
- P12 Fig 4 Add a line showing the transects.
Add heat map for measured CH4 concentration

		Move legend over map in an area that doesn't conflict with location and concentration data.
		Mark a priori wind direction and plume locations.
L276		What is the time delay for air to move through the tube? What size tube? How long? It can't be that long but that may depend on speed of the mobile station.
L281		Why remove this point? It is hard to classify this as an outlier since it still falls within the 95% CI of the regression.
P13	Fig 5	<ul style="list-style-type: none"> • Add axis labels for subplots c and d. • Use a constant font size. The subplot labels are much larger than other fonts. The font used for the axis ticks and colorbar are too small. • It doesn't really seem hard to fit a second Gaussian curve to the residual of the primary Gaussian. This would allow identification (and quantification) of multiple peaks observed along the transect.
L289-291		<p>"This is likely because the averaging..."</p> <p>Again, this is the expected outcome of averaging transects and shifting model peak location.</p>
P14	Table 2	<ul style="list-style-type: none"> • R^2 is not a good performance metric. • Add columns for known emission rates of experiments for controlled release experiments. • The "before" columns are not particularly interesting. See General Comment 4.
P15	Fig 6	Why do the ranges of both axis go below 0 kg/hr?
P16	L326-331	This still doesn't make sense to me.