Reply to comments on "Locating and quantifying CH$_4$ sources within a wastewater treatment plant based on mobile measurements" by Yang et al.

Reply to Anonymous Referee #2

In "Locating and quantifying CH$_4$ sources within a wastewater treatment plant based on mobile measurements" by Junyue Yang, et al., the authors describe an approach to quantify methane emissions from a wastewater treatment plant using a multi-point Gaussian plume model optimized by a genetic inversion algorithm. Atmospheric emissions from WWTPs come from a variety of source types and can be quite variable. This work makes a notable effort to combine previously tested methods to generate plant-level estimates. Overall, this manuscript offers a clear and comprehensive description of the experiment and attempts to position the work in context of the current literature in a compelling manner.

Responses:

We truly appreciate the constructive comments and suggestions raised by the reviewer. Those comments are valuable and very helpful for improving our paper, as well as the important guiding significance to our studies. Below, we provide a point-by-point response to individual comment. And we also polish the content of the manuscript. The responses are shown in plain font, and the added/rewritten parts are presented in italics.

Comments (general)

1. Mobile measurements are described as "circling the functional area 1-2 times" for a given experiment. Was one experiment performed per day during the two campaign

periods? Is this a sufficient amount of data to generate statistically-based conclusions distinguishing the different sources or seasonal effects? Given the nature and size of the sources, it seems feasible that numerous passes could be necessary to capture any variability in the operations. Some evidence of this is noted in the text (L365) where "the substantial variation in the emissions […] suggests a degree of emission instability" for the aeration tank emission estimates. Based on Figure 5, all of the experiments are within the uncertainty of each other.

Responses and Revisions:

Thank you for the suggestion. The description of "circling the functional area 1-2 times" refers to 1-2 rounds of the entire road measurements along external roads and internal functional areas roads of the WWTP. In the actual measurements, we conducted repeated mobile measurements and fixed measurements on roads with high concentrations to calculate average concentration data, investigating the different sources and seasonal effects of CH_4 emissions from the WWTP. Each day of the experiment eventually forms a set of complete data. Based on the conclusion that there is a certain degree of emission instability in the aeration tank emission estimates, we agree that as more days of mobile measurement experiments are added, the monitoring accuracy will be elevated. The following text has been added in the text (Line128-132):

"We performed one monitoring experiment per day, with each experiment entailing 1-2 rounds of mobile measurements along external roads and internal functional areas roads. Additionally, we conducted repeated mobile measurements and fixed measurements on roads with high concentrations to calculate average concentration data".

2. The characterization of the different sources (i.e., line and point) could be further detailed. Specifically, it would be helpful to see more about how the line source was determined beyond the note in the text ("substantiated by through model validation", L221), perhaps in the Supplement? Also, it seems like the primary clarifiers resemble area sources more than line or point sources. How were they characterized in this work? As multiple point sources?

Responses and Revisions:

Thank you for the suggestion. The determination of line sources was previously described in section 2.4.2 "General Finite Line Source Model" and supplemented with Figure S1. By comparing the actual measurement data and model simulation data, we identified multiple point sources and line source diffusion patterns. The comparison is depicted in the revised Figures S1, with Figure S1(a) showing the comparison between CH_4 concentration measurements for Aeration Tank ① and point source model simulation on 29th June, and Figure S1(b) comparing CH_4 concentration measurements with the line source model simulation on a road between the Screen ① and the Primary Clarifier ①. Figure S1 illustrates that the CH_4 concentration curve presents distinct peak distributions in point source diffusion patterns, while the CH_4 concentration distribution in line source diffusion patterns consistently maintains higher levels. Considering the CH_4 emissions from the primary clarifiers of the WWTP as point and line source diffusion patterns rather than area sources allows for a better integration with mobile measurements to pinpoint the specific locations of CH_4 leaks in the WWTP, supporting the development of targeted CH_4 reduction strategies. The additional content is as follows (Line193-198):

"Based on the comparison of measurement and model simulation results (Fig. S1), it is determined that the plant exhibits multiple point sources and line source diffusion patterns. Fig. S1 illustrates that the CH_4 concentration curve presents distinct peak distributions in point source diffusion patterns, while the CH_4 concentration distribution in line source diffusion patterns consistently maintains higher levels".
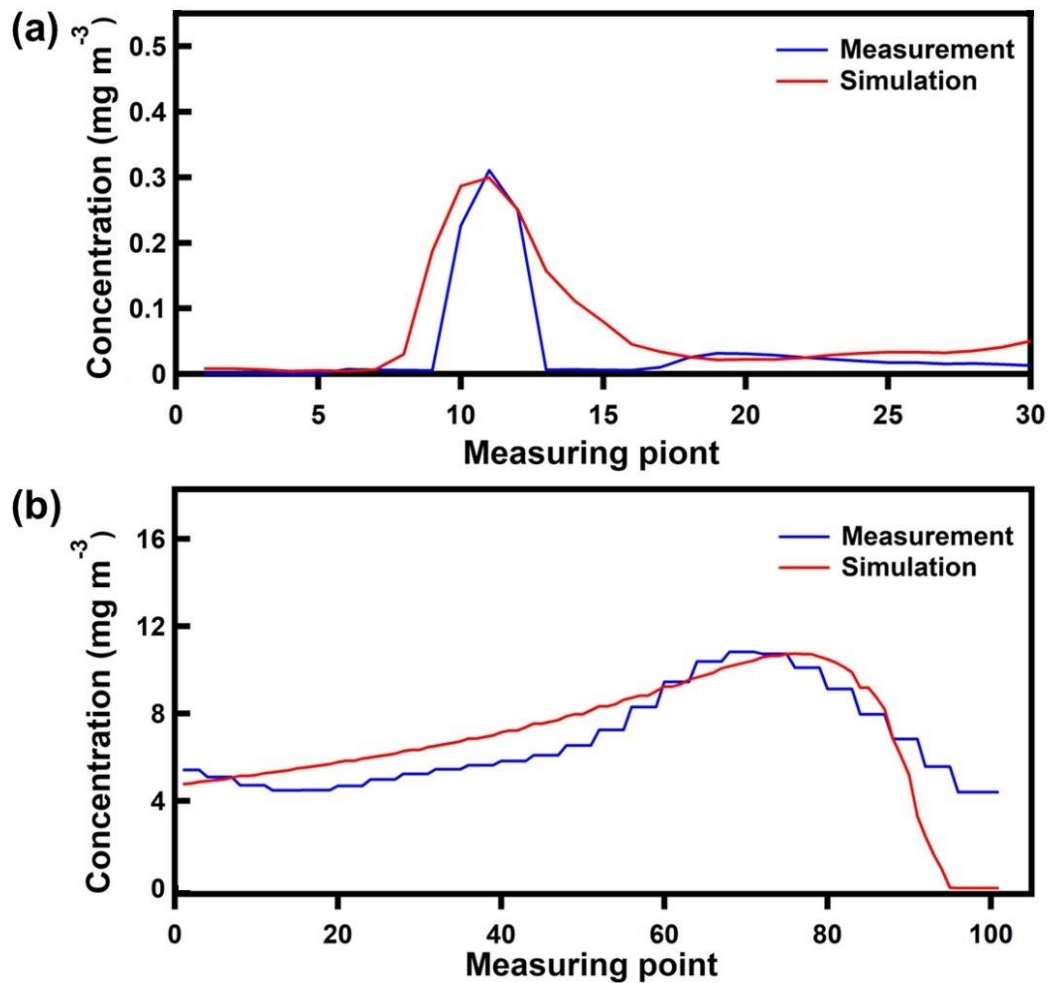
Figure S1. Comparison of CH$_4$ measurement and model simulation. (a) CH$_4$ concentration measurements for Aeration Tank ① and point source model simulation on 29th June (b) CH$_4$ concentration measurements and the line source model simulation on a road between the Screen ① and the Primary Clarifier ①.

3. There appears to be no discussion on the possible effect of interferences from surrounding areas. Is there another WWTP neighboring the site? The satellite images show similar equipment or process areas in the adjacent lot outside of the "exterior road". Also, the wind direction in Figures S5, S6, and S7 comes from an area on land with fields and some buildings, also outside of the subject area, which seem worth identifying. If the model or optimization algorithm address these interferences, then it should be explained in text.

Responses and Revisions:

Thank you for the suggestion regarding external source interference. There are expansion facilities of the WWTP near the monitoring site. However, we were unable to conduct internal roads measurements due to restricted access to the expanded area. We performed mobile measurements along the perimeter of the expansion and found no significant CH_4 emissions. Therefore, we concluded that the expansion facilities do not impact the inversion results. When employing the multi-source Gaussian diffusion model combined with a genetic algorithm framework for iterative optimization to pinpoint point sources, we were able to locate external sources. We have considered external sources during the prior inversion process, which were not explicitly mentioned in the previous manuscript. We have recognized the necessity for a detailed explanation of this aspect, and we have added Fig. S2 related to external sources in the supplement. Fig. S2 (a) and (c) depict the emission sources localization on 29th June and 13th December, revealing that external sources are primarily located along the main roadway to the south of the WWTP. Moreover, the estimated emissions vary across different days, which indicates that external source emissions are influenced by traffic vehicles. Fig. S2 (b) and (d) compare measured and simulated CH_4 concentrations before and after accounting for external sources, demonstrating that simulations are significantly closer to the measurements when external sources are included. Therefore, we conclude that considering external sources during the inversion process can enhance the accuracy of the results. The additional content is as follows (Line379-387):

"In addition, when employing the multi-source Gaussian diffusion model combined with a genetic algorithm framework for iterative optimization to pinpoint point sources, we were able to locate external sources. As shown in Fig. S2 (a) and (c), external sources are primarily located along the main roadway to the south of the WWTP. Moreover, the estimated emissions vary across different days, which indicates that external source emissions are influenced by traffic vehicles. Fig. S2 (b) and (d) compare measured and simulated CH4 concentrations before and after accounting for external sources, demonstrating that simulations are significantly closer to the measurements when external sources are included".
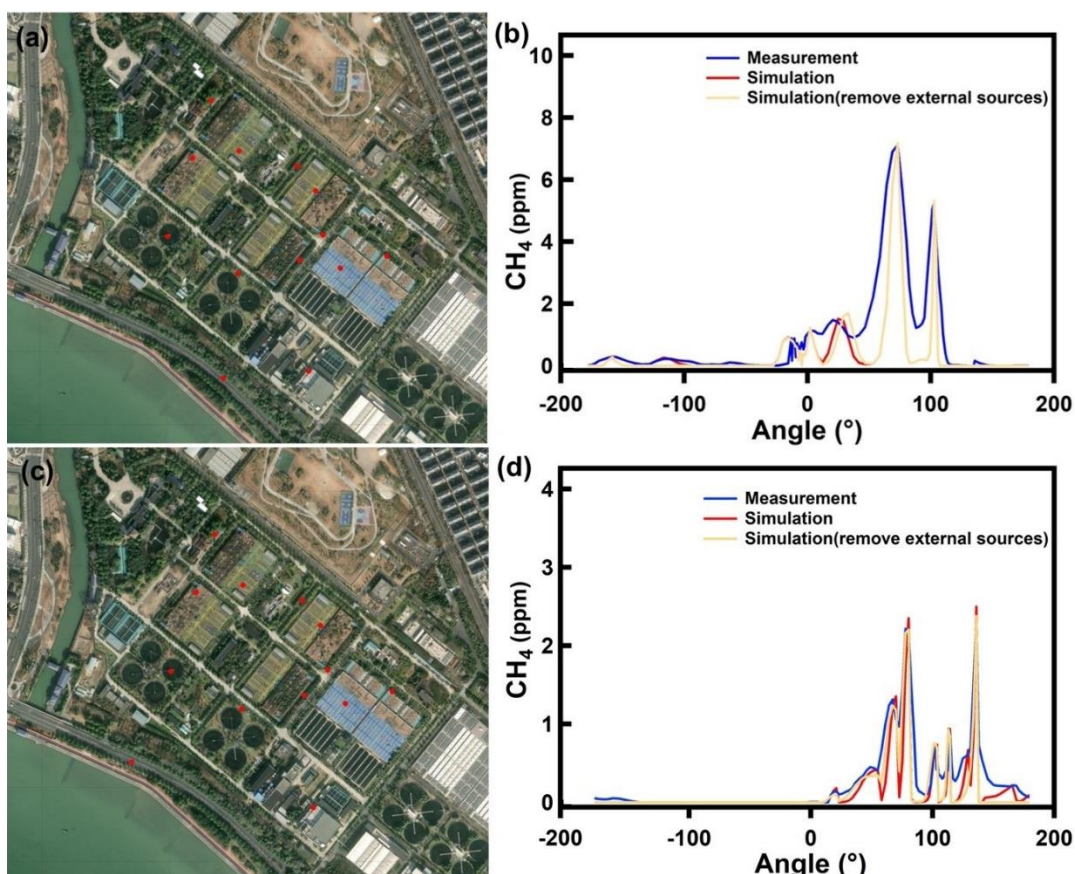
Figure S2. The emission sources localization on 29th June (a) and 13th December (c). And the comparison of CH₄ measurement and model simulation (considering external sources) on 29th June (b) and 13th December (d).

4. In Table 1, wind speed on "0711" was listed as 0.9 m s⁻¹. Presumably, this is an average value. Was there a trend in the emission estimates relative to wind speed, specifically low speed winds? Are there datapoints collected when wind speeds were below 0.7 m s⁻¹?

Responses and Revisions:

Thank you for raising the issue regarding wind speeds. Wind speed data in Table1 are the average wind speed during the monitoring time. We compared the emission estimates at different wind speeds during the summer and winter seasons. In the summer data, the lowest wind speed recorded was 0.9 m s⁻¹ on 11th July, with emission estimates higher than those on 1st June but lower than those on 29th June. In the winter data, the lowest wind speed was 1.2 m s⁻¹ on 14th December, and the estimated emissions were lower than those on the other days. Thus, we conclude that there is no significant trend

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in emission estimates under low wind speed conditions. Furthermore, we have reviewed and organized the wind speed data and found that datapoints were collected when the wind speeds were below 0.7 m s^{-1}. This was due to the fact that the mobile monitoring system was deployed for internal functional area monitoring, where the monitoring vehicle was in close proximity to the functional area facilities. Consequently, concentration data could still be detected even at the conditions of low wind speeds. The additional content is as follows (Line416-419):

"At lower wind speeds, the CH$_4$ emissions show only slight differences when compared to emissions on days with higher wind speeds. This suggests that the inversion results are less influenced by wind speed and are primarily associated with seasonal variations".

5. Discussion on how uncertainty was calculated is brief and non-specific. Further detail on the contributing factors and how the uncertainty was calculated is needed in the text (see L424 for context).

Responses and Revisions:

Thank you for the suggestion. We have incorporated detailed information in the paper to enhance the understanding of the uncertainty analysis. The added text is as follows:

Line288-299: *"To quantify the uncertainty in the inversion results, we have considered the uncertainties associated with the input parameters of the inversion model, including wind speed, wind direction, and instrument measurements. The uncertainty in the CH$_4$ emission fluxes (ε_t) is derived using the error propagation formula as follows:*

$$\varepsilon_t = \sqrt{\varepsilon_s{}^2 + \varepsilon_d{}^2 + \varepsilon_m{}^2} \qquad (7)$$

ε_s and ε_d denote the uncertainties in wind speed and direction, which are determined by the standard deviation of the wind speed and direction measurements from two fixed meteorological stations during the observation period. ε_m represents the uncertainty in instrumental measurements. This uncertainty is derived from data

provided by the manufacturer Picarro, which indicates a concentration measurement uncertainty of approximately 1 ppb for a 10-second integration time (Picarro 2010)".

Line468-472: *"The uncertainty of the inversion results was determined by accounting for the uncertainties in wind speed, wind direction, and instrument measurements, following the method presented in Section 2.4.5. The uncertainties in emission fluxes inversion range from 33% to 63% on individual days. Notably, the uncertainty associated with wind speed contributes approximately 44% to 94% of the uncertainty range".*

6. Consider emphasizing discussion of tracer flux ratio, also known as "tracer release", as another top-down method that employs mobile measurements. This method was mentioned specifically in text as an example, but it is noted throughout other publications (Delre et al., 2017; Moore et al. 2023; Yacovitch, et al. 2015; von Fischer et al. 2017), including use for validation or comparison with other modelling techniques for similar applications.

Responses and Revisions:

Thank you for the suggestion. We have added tracer gas dispersion method content to Section 3.2. The added text is as follows (Line437-460)：

"An alternative top-down approach known as the tracer gas dispersion method (TDM), which has been applied to estimate CH_4 emissions from city streets (von Fischer et al., 2017; Weller et al., 2018), WWTPs (Yoshida et al., 2014; Delre et al., 2017; Delre et al., 2018), biogas plants (Reinelt et al., 2017; Scheutz et al., 2019; Fredenslund et al., 2023) and landfills (Rees-White et al., 2019; Kissas et al., 2022). TDM involves releasing tracer gases like nitrous oxide and acetylene near the source and measuring their concentrations along with CH_4 downwind using a mobile platform. The similar diffusion patterns of CH_4 and the tracer gases result in a stable concentration ratio after atmospheric mixing, enabling the calculation of CH_4 emission rates with a better accuracy (Mønster et al., 2014).

Moreover, the TDM was employed to validate and compare other model inversion methods. Moore et al. (2023) proposed that the Gaussian dispersion modeling demands

less experimental equipment, site access, and manpower than the TMD, which allows for swifter data gathering. Yacovitch et al. (2015) utilized a five-day dataset of tracer gas release in the Barnett shale region to evaluate the Gaussian dispersion flux quantification method. The results indicated a 95% confidence interval, with a lower bound factor of 0.334 and an upper bound factor of 3.34. von Fischer et al. (2017) performed three controlled release experiments to validate the reliability of leak rate algorithm in Fort Collins, CO, which indicated a very significant correlation between known and estimated leak rates (p < 0.0001, r^2 = 0.43). Compared to the method employed in the study, the TDM offers advantages such as simpler formula calculations. Nonetheless, it also presents several drawbacks, including complex experimental procedures, safety hazards associated with the release of tracer gases, which require access permits from industrial facilities".

7. Using units of t a^{-1} could be useful for comparison-sake (e.g., with the IPCC inventory method), but given the limited number of measurement days, it seems like a significant extrapolation. Other studies present findings in kg h^{-1} (as noted in the introduction) or also otherwise normalized by other factors. Perhaps consider presenting the results in kg h^{-1} (or similar) with mention of the extrapolation (t a^{-1}) to offer the reader a quick comparison to other relevant figures.

Responses and Revisions:

Thank you for the suggestion on the emission flux unit. We have revised the unit of emission flux of the WWTP with kg h^{-1}, including a mention of the extrapolation to (t a^{-1}). The units of t a^{-1} have been maintained for the figure and text in section 3.3 to facilitate a clearer contrast with the emission inventory.

Comments (line-by-line)

1. L23: replace "emission" with "emission<u>s</u>".

Responses and Revisions:

Thank you for the suggestion. We have revised it to *"emissions"*.

2. L29: consider changing the verb tense of "are" to "is".

Responses and Revisions:

Thank you for the suggestion. We have changed the verb tense to *"is"*.

3. L57: remove " 's" from "(IEA) 's" or change sentence.

Responses and Revisions:

Thank you for the suggestion. We have revised it to *"(IEA)"*.

4. L63: clarify the meaning of "actual emission factors activity data" depending on the intent of the sentence. For example, "activity data used for actual emission factors". Or if that's not the intention, then perhaps, "activity-based emission factors".

Responses and Revisions:

Thank you for the suggestion. We have acknowledged that our previous phrasing was ambiguous and could lead to misinterpretation. We intended to convey that the activity data are based on actual emission factors. We have revised it to *"activity data used for actual emission factors"*.

5. L137: change "Spectroscopy" to "Spectrometer" or add "monitoring system" as done in the Instrumentation section and elsewhere.

Responses and Revisions:

Thank you for the suggestion. We have changed it to *"Spectrometer"*.

6. L147: is this "1-2 times" per day? Does each day comprise of one experiment? This should be addressed in the text.

Responses and Revisions:

Thank you for the suggestion. we have recognized the necessity for a more detailed experimental description. We performed one monitoring experiment per day, with each experiment entailing 1-2 rounds of monitoring along external roads and internal

functional areas roads. During each experiment, we conducted repeated mobile measurements and fixed measurements on roads with high concentrations to enhance the accuracy of concentration monitoring. The following text has been added in the text (Line128-132):

"We performed one monitoring experiment per day, with each experiment entailing 1-2 rounds of mobile measurements along external roads and internal functional areas roads. Additionally, we conducted repeated mobile measurements and fixed measurements on roads with high concentrations to calculate average concentration data".

7. L148: is a "set" equivalent to an experiment?

Responses and Revisions:

The term "a set of data" is composed of external road concentration data and internal functional area concentration data. During each experiment, we conducted repeated mobile measurements and fixed measurements on roads with high concentrations to enhance the accuracy of concentration monitoring. Each day of the experiment eventually forms a set of valid data.

We have recognized that this expression can be confusing. Thus, we have changed this in the revised manuscript (Line132-134): *"Over 10 days of experiments from June to December 2023, we obtain 8 days of complete monitoring data, including 3 days in summer and 5 days in winter".*

8. L148: what deems these datasets "valid" in this context?

Responses and Revisions:

Thank you for the suggestion. Valid data refers to the complete CH_4 concentration data from the WWTP, including both external road data and internal functional areas data of the WWTP.

We recognize that this expression can be confusing. Thus, we have changed this in the revised manuscript (Line132-134): *"Over 10 days of experiments from June to*

December 2023, we obtain 8 days of complete monitoring data, including 3 days in summer and 5 days in winter".

9. L148: what happened to the other two days/experiments?

Responses and Revisions:

Thank you for the suggestion. The other two days of experiments were conducted on 28th June and 12th December However, due to the internal maintenance of the wastewater treatment facilities, which rendered certain roads inaccessible, we could not obtain comprehensive concentration data from the plant, and the quantification of emission sources was not carried out. The additional content is as follows (Line134-135):

"On the other two experimental days, internal facility maintenance restricted access to certain roads, resulting in incomplete monitoring data".

10. L157: were the less abundant isotopologues of CH_4 or CO_2 used in the data analysis? If not, it should be stated.

Responses and Revisions:

Thank you for the suggestion. CH_4 and CO_2 isotopologues data were not used in the data analysis. *The sentence "The CRDS analyzer can be used to measure $^{12}CO_2$, $^{13}CO_2$, $^{12}CH_4$, $^{13}CH_4$ and H_2O"* has been deleted. We have changed this in the revised manuscript (Line150-151):

"In this study, the CRDS analyzer was placed inside the monitoring vehicle to measure CH_4 concentrations in the WWTP".

11. L163: how was the probe mounted "on" the roof and how did that position "mitigate the effect of vehicular emissions"? This should be addressed in the text.

Responses and Revisions:

Thank you for the suggestion on the instrument system description. We have realized that the installation position of the sampling probe was not accurately described. We have changed this in the revised manuscript (Line151-152):

"The sampling probe was placed near the roof along the window of the car to avoid interference from vehicle exhaust due to the low position".

12. L166: similar to the previous question, how was the meteorological unit mounted to the roof? Given the additional on-site wind stations, this may not be a critical point, but positioning of the wind unit close to the vehicle body will affect the measurement accuracy.

Responses and Revisions:

Thank you for the suggestion on the description of the instrument system. The meteorological instrument was adsorbed on the roof of the vehicle through the magnet at the bottom. We agree that positioning of the wind unit close to the vehicle body will affect the measurement accuracy. Consequently, the meteorological instrument on the vehicle is mainly regarded as a reference for the instantaneous wind direction during mobile measurements. The meteorological data used as input for the model simulations are obtained from portable meteorological stations.

13. L169: specific the manufacturer of the SWS-500.

Responses and Revisions:

Thank you for the suggestion. We have added specific content for SWS-500 (Hangzhou Pengpu Technology).

14. L242: make mg/s into mg s^{-1} for consistency.

Responses and Revisions:

Thank you for the suggestion. We have revised it to "mg s^{-1}".

15. L308: what does "the similarity of concentration measurement methods" mean? Wasn't the same method applied to the whole dataset? Or is this a reference to the roads driven? The wind direction is clearly different. This should be addressed in the text.

Responses and Revisions:

Thank you for pointing out the error in the sentence. The method for measuring concentration is the same. We have changed this in the revised manuscript (Line311):

"Due to the consistency of concentration measurement methods, we chose 29th June and 13th December as a typical example".

16. L414: are these average values? Or is there only one estimate determined per day? This should be addressed in the text.

Responses and Revisions:

Thank you for the suggestion. Wind speed and direction data are the average wind speed and direction during the monitoring time, and the CH$_4$ emission fluxes are obtained by the inversion of the average concentration, wind speed and direction data as input of the model, which are one estimate determined per day. The following text has been revised and added in the text (Line398-403):

"Table 1 displays the CH$_4$ emission fluxes (Q), Wind speed and direction data (W$_s$, W$_d$), the horizontal diffusion coefficient (γ$_1$, α$_1$) and the vertical diffusion coefficient (γ$_2$, α$_2$) from the 8-day monitoring experiment. Wind speed and direction data are the average wind speed and direction during the monitoring time, and the CH$_4$ emission fluxes are obtained by the inversion of the average concentration, wind speed and direction data as input of the inversion framework".

17. L414: while it's relatively clear from context what the labels in this table mean (i.e., Q, Ws, Wd, γ, α) some more detail in the caption would be helpful.

Responses and Revisions:

Thank you for the suggestion to improve the caption for Table 1. We have revised the title (Line433-435) to *"CH$_4$ emission fluxes (Q), Wind speed and direction data (W$_s$, W$_d$), the horizontal diffusion coefficient (γ$_1$, α$_1$) and the vertical diffusion coefficient (γ$_2$, α$_2$) from the 8-day monitoring experiment".*

18. L424: need more detail on the uncertainty estimates in the text. Is this a confidence interval?

Responses and Revisions:

Thank you for the suggestion regarding the section on uncertainty analysis. We have incorporated additional details to enhance the understanding of the uncertainty analysis. The determination of the numerical values is not based on confidence intervals, but rather calculations derived from wind speed, wind direction, and instrumental uncertainties. The added text is as follows:

Line288-299: *"To quantify the uncertainty in the inversion results, we have considered the uncertainties associated with the input parameters of the inversion model, including wind speed, wind direction, and instrument measurements. The uncertainty in the CH_4 emission fluxes (ε_t) is derived using the error propagation formula as follows:*

$$\varepsilon_t = \sqrt{\varepsilon_s{}^2 + \varepsilon_d{}^2 + \varepsilon_m{}^2} \tag{7}$$

ε_s and ε_d denote the uncertainties in wind speed and direction, which are determined by the standard deviation of the wind speed and direction measurements from two fixed meteorological stations during the observation period. ε_m represents the uncertainty in instrumental measurements. This uncertainty is derived from data provided by the manufacturer Picarro, which indicates a concentration measurement uncertainty of approximately 1 ppb for a 10-second integration time (Picarro 2010)".

Line468-472: *"The uncertainty of the inversion results was determined by accounting for the uncertainties in wind speed, wind direction, and instrument measurements, following the method presented in Section 2.4.5. The uncertainties in emission fluxes inversion range from 33% to 63% on individual days. Notably, the uncertainty associated with wind speed contributes approximately 44% to 94% of the uncertainty range".*

19. L509-511: it is an excellent point that collaborative monitoring offers significant

Responses and Revisions:

Thank you for the suggestion. We have changed this in the revised manuscript (Line550-552):

"We believe that it is an excellent point that collaborative monitoring offers significant improvements in the accuracy of emission fluxes and source inversion".

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