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 #1

This study conducted CH_4 mobile measurements in a wastewater treatment plant in summer and winter of 2023 and utilized a multi-source Gaussian plume model along with a genetic algorithm inversion framework to locate major sources within the plant and quantify the corresponding CH_4 emission fluxes. Similar to previous studies, they found that emission estimates based on their inversion framework were higher than those estimated using traditional IPCC methods. They also found that the emissions are higher during summer than winter. Given the important role of CH_4 in climate change, these types of studies are essential. So, the paper is within the scope of ACP. However, the paper lacks some key details which makes it hard to understand the experimental design and the inversion framework.

Responses:

We thank the reviewer for the constructive suggestions and comments concerning our manuscript. These comments are invaluable and greatly assist in enhancing our paper. Below, we present a point-by-point response to each 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.

Major comments

1. Figure 1: Can you please explain how the numbers for different components are defined in Figure 1? I see secondary clarifiers 1, 2, and 5 but not 3 and 4. It would also be helpful to show the roads on which you drove the mobile van.

Responses and Revisions:

Thank you for the suggestion. We initially assigned numbers to the facilities in sequential order based on their quantity. The designation "Secondary Clarifier ⑤" was chosen because it is adjacent to "Primary Clarifier ⑤" and "Aeration Tank ⑤". We have acknowledged that this naming convention may cause misunderstandings. Consequently, we have revised the label in Figure 1 from "Secondary Clarifier ⑤" to "Secondary Clarifier ③". Furthermore, we have modified Figure 1 to show the roads on which we drove the mobile van.

2. Lines 147-149: Why were two days of data left out? How were the monitoring days determined? What were the meteorological conditions during the measurement days and were those conditions representative of typical summer and winter conditions?

Responses and Revisions:

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. About monitoring days，the monitoring experiment was carried out when the WWTP was conveniently opened in summer and winter and there was no significant precipitation throughout the day. Compared with the historical weather data, the meteorological conditions of the monitoring date are in line with the typical weather conditions in summer and winter. The additional content is as follows (Line132-135):

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

3. Lines 210-211: Can you please provide more details about how the initial emission estimates are derived? Are they derived in Section 2.3? Since the sources are so close to each other, there is a high possibility of plumes overlapping with each other and the observed concentrations being affected by multiple sources. Can you please explain how this overlapping issue was addressed for the 12 sources considered in the multi-source Gaussian plume?

Responses and Revisions:

Thank you for the suggestion. The initial emission estimates are not derived in Section 2.3. The initial emission estimates are mainly based on the concentration distribution and internal functional area features of the WWTP to define the initial source positions and to approximate the initial emissions using established empirical formula (Weller et al., 2019). We agree that the superposition of 12 sources may introduce discrepancies in the initial predictions. Therefore, these initial source locations and emissions serve as a reference point, with further optimization of the emission source locations and emissions through model inversion. In the implementation of a multi-source Gaussian model inversion, we have taken into consideration the overlapping plumes from point sources, ensuring that concentration levels are accurately represented through spatial superposition. The additional content is as follows (Line211-218):

"We used the improved empirical equation to estimate the initial emissions of emission sources (von Fisher et al., 2017; Weller et al.,2019). This method was primarily utilized for urban CH_4 leakage source emissions estimation (Defratyka et al., 2021; Maazallahi et al., 2020). The empirical equation is as follows:

$$ln\left(M_{CH_4}\right) = -0.988 + 0.817 \times ln\ (CH_4\ emission\ rate) \qquad (2)$$

The M_{CH4} is the maximum enhancement value of CH_4 concentration, ppm. The CH_4 emission rate represents the CH_4 emission flux, L min^{-1}".

4. Line 223: What about the emissions upwind (e.g., secondary clarifier 2, power sanitation 1 etc.) of primary qualifier 1? How are those removed from this line source?

Responses and Revisions:

In the actual measurements, different points on the road of the WWTP were selected for fixed measurements. By analyzing wind direction and corresponding concentration distribution, we ultimately determined the location of the line source. For the emissions upwind (e.g., secondary clarifier ②, power sanitation ① etc.) of primary qualifier ①, no significant line source leakage distribution was detected when it was located downwind. Thus, we infer that this location is not part of the line source.

5. Equation (2) and lines 240-253: How are the values of different parameters determined using the observations?

Responses and Revisions:

Thank you for the suggestion. The CH_4 concentrations were obtained through mobile measurements using vehicle-mounted CRDS monitoring system. The portable meteorological stations collected data on wind speed and direction, while GPS tracked the mobile paths to pinpoint emission source locations. The diffusion parameter power function expression coefficients were selected based on the atmospheric stability conditions of the day, and finally combine them with Equation (2) to obtain the horizontal and vertical dispersion parameters. The additional content is as follows (Line243-246):

"During the observation, the CH_4 concentrations were obtained through the vehicle-mounted CRDS monitoring system. The portable meteorological stations collected data on wind speed and direction, while GPS tracked the mobile paths to pinpoint emission source locations".

Minor comments

1. Line 26: What do you mean by "emission data" here? Activity data, emissions factors?

Responses and Revisions:

Thank you for the correction of the "emission data" description. The meaning of "emission data" here is activity data We have revised it to *"activity data"*.

2. Line 28: Suggest replacing "in combination with" by "using".

Responses and Revisions:

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

3. Line 35: Since measurements are done only during 10 days, I recommend reporting the emissions in tons/day rather than tons/annum.

Responses and Revisions:

Thank you for the suggestion on the emission flux units. We have revised the units of emission flux of the WWTP with kg h^{-1}, as this unit is more suitable for short-term measurements. However, 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.

4. Line 63: Suggest adding "," after "emission factors" because emission factors and activity data are different parameters.

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. Line 195-196: Please mention the TOW value deduced from the workbook.

Responses and Revisions:

Thank you for the suggestion. The content of TOW value is added as follows (Line176-178):

"*The Total Organic Waste (TOW) is calculated by the amount of treated water and COD influent concentration of the WWTP provided in the yearbook".*

6. Lines 220-221: Screen 1 and primary qualifier 1 are located diagonally from each other. Can you mark this road in Figure 1?

Responses and Revisions:

Thank you for the suggestion on Figure 1. We have modified Figure 1 to mark the line source.

Figure 1. Distribution of functional areas of the WWTP. The yellow mark represents the simulated location of the line source. Solid lines show the roads measured by the mobile vehicle. Map data are from ESRI.

7. Lines 316-317: Were higher background concentrations in December due to shallower boundary layer?

Responses and Revisions:

Thank you for the suggestion regarding background concentrations. We agree that the higher background concentrations observed in December are primarily due to the

lower atmospheric boundary layer. The variation in atmospheric boundary layer height significantly affects the CH_4 concentration. During summer, due to solar radiation and ground heating, the boundary layer height tends to be relatively high. This enhances the dispersion capacity of CH_4 in the atmosphere, leading to a corresponding decrease in the concentrations. In contrast, during winter, the boundary layer height is typically lower, which facilitates the retention of CH_4 near the surface, resulting in higher ground-level concentrations. The additional content is as follows (Line318-320):

"The analysis indicates that the shallower boundary layer in winter causes CH_4 to accumulate near the surface, resulting in a higher background concentration".

8. Lines 361-362 and Figures 3-4: How are the emission source locations determined? Are they known a priori?

Responses and Revisions:

Thank you for the inquiry regarding the location of the emission sources. The positions of the emission sources were not known a priori. We initially identified the source locations through the concentration distributions obtained from mobile measurements. Subsequently, we utilized model simulations to further refine and optimize the source locations.

References

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