Referee #2

General Comments:

The manuscript presents an interesting comparison between measured methane concentrations and reported emission factors at a large metropolitan wastewater treatment lagoon. The use of Inverse-Dispersion Modelling (IDM) with open-path lasers (OPL) is a robust approach for non-intrusive monitoring; however, the current manuscript has significant gaps regarding spatial weighting, micrometeorological artifacts, and the representativeness of the upscaled annual data. Without addressing these potential biases, the conclusion that emissions are 2–3 times higher than NGERS reports may be premature.

We thank the Reviewer for their thorough and constructive review of our manuscript. Below, we address each of the reviewer's comments.

Specific comments:

1. How does the "touchdown" coverage (mentioned as >20% in Section 2.5) vary across the pond surface? Does the weighting reflect the actual spatial concentration gradient?

It depends on the wind direction, but the source area can be seen when the wind's direction is within a range of >330° to < 40°. This is when the measurements can be considered for representing the source emission and not contaminated by nearby sources (e.g., eastern side and western side).

As the pond is particularly large and that the downwind open-path laser parallel to the pond, that is set with a typical pathlength of about ~200 m to maintain its stability and performance, we can only sample a portion of the pond emission, in addition to limited resources, we deployed two open-path lasers downwind the pond, by dividing the pond into two half, one laser can be focused on the pond area close to the anaerobic pod and the other one in contrast to the western side of the pond.

The touchdown coverage varies with wind direction and atmospheric conditions. We also analysed its spatial distribution. The footprints showing its spatial distribution indicate which parts of the lagoon contributed to the OPL-IDM estimate for each laser path.

However, the footprint does not provide spatially resolved concentrations within the sampled area. For each laser path and time step, we obtain one aggregated estimate for the area sampled by that footprint. Therefore, the footprint tells us where the measurement is coming from, but not how methane concentration varies within that area.

The weighting does not represent a measured spatial concentration gradient. Instead, it is used to scale the two aggregated estimates, one from the west footprint and one from the east footprint to the full lagoon area. We clarified this in the revised text and assessed the sensitivity of the full lagoon estimate to alternative weights.

2. Is the 8:00 AM peak a result of a biological process, or is it a micrometeorological artifact caused by the breakup of the nocturnal boundary layer? At 8:00 AM, the atmosphere often transitions from stable to unstable, which can "dump" accumulated methane toward the sensors. In the early morning, the "surface layer" might not be fully developed. Are you seeing a real biological emission peak, or is it just the "fumigation" of methane trapped near the water surface overnight being released as the sun hits the pond?

This diurnal pattern on the eastern pond area, with peak emission at ~8:00, was associated with operation activities (aerator on/off and influent flow rate high/low) rather than micrometeorological artifact. A similar diurnal pattern has also been reported in the literature (Mannina et al., 2018; Bühler et al., 2022; Guisasola et al., 2008). Ebullition (bubbling) and diffusion (dissolved methane in the effluent) are the dominant pathways of methane emissions and can be influenced by a range of locations-specific, biotic and abiotic factors.

As sewage from the anaerobic digestion pod enter the Pond 1, the influent is saturated with dissolved methane. When the aerators are switched on, they not only rapidly strip dissolved methane from the water surface to the atmosphere but also generate turbulence in the sediment-water system in the pond, contributing to diffuse emission from the lower to the up layers of the pond and ultimately to the atmosphere. The influent flow rate plays an important role in regulating methane emissions: it exhibits a similar 24-hr diurnal variation as CH_4 emissions but with a time lag of 1-2 hrs. Furthermore, other studies suggest the peak emission in the morning could be due to the sewage system, where higher methane production with longer retention time in the effluent, and methane could also accumulate in sediments (Guisasola et al., 2008). If aerators are turned off in the afternoon, methane could build up overnight and then be released the following morning when aeration resumes (Bühler et al., 2022).

3. I am wondering why a simple linear weighting (0.67/0.33) is superior to a more robust spatial interpolation. If the wind direction shifts even slightly, the "footprint" of what those lasers "see" changes drastically. Did you perform a sensitivity analysis on those weights?

We agree that wind direction affects the area represented by each laser path. This is why we used the IDM footprints to interpret the two OPL measurements. However, a full spatial interpolation of methane concentrations across the lagoon is not possible with these data.

The method gives a single aggregated estimate for the area sampled by each laser path, that is, the laser footprint. We do not have spatially resolved concentrations within the footprints. At each time step, we therefore have two aggregated estimates for two different parts of the lagoon, not many point measurements that could be used to build a concentration map.

Applying a spatial interpolation method in this setting would require assumptions about the spatial autocorrelation structure, which cannot be estimated from only two aggregated observations per time step, and about emissions in unsampled portions of the lagoon. This would mostly be extrapolation rather than interpolation. For this reason, we used a simple weighting approach to scale the two estimates to the full lagoon. This method is transparent and does not imply more spatial detail than the data can support.

We also tested how sensitive the results were to the chosen weights. The main estimate used the 0.67/0.33 weighting, but we also recalculated emissions using 0.75/0.25 and 0.5/0.5. We report the results as a range, with 0.67/0.33 used as the most representative case based on the relative areas represented by the west and east laser footprints.

4. Was the background laser (OPLC34) moved to account for different northerly wind angles (e.g., NNE vs. NNW)? If not, the $(C/Q)_{sim}$ could be biased by "dirty" upwind air that wasn't properly subtracted.

The background laser position was not moved during the summer and winter measurements. To the north of the pond (NNE-NNW) there was short grass adjacent to the pond, and a corn field a few hundred meters away; because the land cover was similar, the background condition remained stable during our measurements, and methane emissions from the corn field can be considered negligible compared with the large source of aerated pond.

Furthermore, downwind CH_4 concentrations on the western and eastern side of Pond 1 varied with time, while the background concentration on northern side of the pond remained constant at ~1.9 ppm under northerly winds. During the summer campaign, enhancement in CH_4 concentrations under northerly wind conditions were 3.6 ppm (western area) and 10.8 ppm (eastern area). During the winter campaign the averaged enhancements measured by laser 33 (western laser) and laser 1013 (eastern laser) was 2.3 ppm and 13.5 ppm, respectively. These enhanced downwind concentrations measured by both lasers were much higher than the change in background concentration.

Importantly, we applied filtering criteria to ensure that the periods with unreasonable (and high) values from the upwind, which do not represent the true background of the pond, are not included in the flux calculations: the difference in background concentration between the simulated background level and the measured one (upwind) is < 20 ppb, and wind directions >330° or < 40°.

5. Did the cross-calibration (Section 2.2) account for the difference in lower-detection limits between the two brands? If the "East" laser is less precise, the uncertainty in the "high emission" zone is actually higher than in the "low emission" West zone.

Yes, this is an important point. We deliberately matched each instrument to the expected concentration range at its location. The higher-precision Unisearch laser (< 1 ppb at 100 m) was used in the western low-emission zone and the Boreal laser (~20 ppb at 100 m) in the eastern high-emission zone.

In practice, concentrations at both sites were well above detection limits throughout the campaign.

At the western site, the Unisearch laser measured enhancements of 0.12–10.8 ppm in winter and 0.96–14 ppm in summer, at least >100 time above its detection limit of <1 ppb. At the eastern site, enhanced concentrations ranged at 2.1–34 ppm in winter and 1.2–57 ppm in summer, or at least >50 times the Boreal laser's detection limit of 20 ppb.

Ultimately, instrument precision contributed negligibly to the total uncertainty in our flux estimates. Because flux is derived from a large number of measurements, random instrument noise averages out (SE = SD/sqrt(n), where n is the number of measurements), contributing only 0.02% of total flux uncertainty, which, as reported in the manuscript, is dominated by uncertainty in the wind dispersion model (~20%).

6. You calculated an annual emission based on a 5-week summer campaign and a 7-week winter campaign. But wastewater chemistry (BOD/COD) and microbial activity aren't just seasonal; they are operational. Was the "flow rate" or "aerator schedule" during these 12 weeks truly representative of the other 40 weeks of the year? If the facility had a "high load" period during their measurement window, the "2–3 times higher than NGERS" claim might be an overestimation of the annual total.

The flow rate of wastewater and aerator schedule had significant variations on a diurnal timeframe as discussed in the paper.

While there are some minor seasonal changes between summer and winter in the flow of COD through Pond1, these variations are not very significant to the overall results of the paper. There were not changes to normal operations or unusually high loads during the 5 week and 7 week campaigns and results indicated that a longer campaign would be of little benefit. Furthermore, due to budget constraints a longer campaign period was not possible.

References

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