

This paper provides an airborne mass balance method to quantify both carbon dioxide and methane emission rates from four liquefied natural gas terminals in Australia. The authors demonstrate the performance of this mass balance method by comparing to operator-reported carbon dioxide emissions. In addition, this paper provides an alternative approach to estimate methane emission rates based on measured CH₄:CO₂ mole fraction compared to the previous direct mass-balance based methane measurements. However, some of the arguments regarding the feasibility of using CH₄:CO₂ ratio for estimating methane emissions are not well-documented and lack strong supporting discussion and should be strengthened. The current paper needs to be revised before it is publishable in EGU sphere.

Additionally, the language in this paper needs to be improved. Some of the sentences in this paper are too long, which increases the difficulty for reader to follow. Some of the sentences are ambiguously phrased, leading to potential misinterpretation and should be improved. Specific problems are outlined in the detailed comments below.

The detailed comments are summarized below:

(1) Most of the CO₂ emissions from the LNG terminals originate from the combustion of fuel gas or flare gas. However, the combustion efficiency is time-varying parameter related to the intermittent flaring events and combustion efficiency changes and that is difficult to quantify precisely. In practice, operators often assume a constant combustion efficiency when estimating CO₂ emissions, which can mask significant daily or hourly variation of CO₂ emissions. The approach of using measured CH₄:CO₂ ratio for estimating methane emissions in this study heavily depends on the accuracy of operator estimated CO₂ emission. The inherent inaccuracy of operator estimated CO₂ emissions therefore limits the reliability of using CH₄:CO₂ ratio for estimating site-level methane emissions.

In addition, the non-coinciding plume sources of CH₄ and CO₂ would also increase the difficulty of using tracer correlation method to estimate CH₄ emissions. For example, in the LNG terminals, the flare stack is usually a few hundred meters above the ground-level equipment, where fugitive methane emissions often originate. This vertical separation violates the co-emission assumption underlying the ratio method, as the CH₄ and CO₂ plumes may not be well-mixed or co-located in the sampled air mass. As a result, such condition can significantly undermine the validity of CH₄ estimates based on the tracer correlation approach.

Please provide additional documentation or explanation on how the study accounts for the two issues described above.

(2) The sentence in lines 50-52 and lines 60-62 are too long. It is recommended to break them into short sentences.

(3) In this study, the measurements were only conducted at a single screen downwind of each facility. However, there are usually some co-located nature or anthropogenic emission sources, such as wetlands and natural gas compressor stations, that do not belong to LNG terminals. How to make sure that there are no emission sources from the three unmeasured sides of surrounding each LNG facility entering the measured screen?

(4) For better understanding, please provide the units of parameters in all equations and figure coordinates titles. Specifically, does the site-level emission rate E in equation (1) refer to mass emission rate (e.g. kg/hr) or volume emission rate (e.g. Scf/hr)?

(5) In the first step of determining the background emission rate, why the 10th percentile not other percentile was selected? In addition, why two times the measurement precision not one time was selected? Please provide some clarification.

(6) For Equation (3), the text in line 317 mentioned that any CH₄-only or CO₂-only plumes were ignored. Is it “CO₂-only plumes” or “CO₂-only measurements points at each transect”?

(7) For Figure 3, it is recommended to use different colors to represent different LNG terminals for better understanding the variations across different sites. In addition, there are three cluster of site-level measurements with almost constant operator estimated CO₂ emissions, but with one cluster operator estimated CO₂ emissions ranging from 600-700 t/hr. What is the reason for this discrepancy? Can you clarify it in the main text?

(8) In lines 366-368, the text shows that the 20% of relative difference between operator estimate and mass-balance measurements is impact by the monthly aggregated total emissions at two sites. I do not totally agree with this point, because the case where some mass-balance measurements are larger or smaller than operator estimate exists for every site, not only at the two sites that use monthly aggregated total emissions. The authors should dig deeper into the reasons for these larger discrepancy between measurements and operator estimates and make a clarification.

(9) In lines 450-458, the authors made a conceptual shift from the daily variation of CO₂ emissions measurements to operator estimated CO₂ emissions. The Figures 5 shows a similar random variation between CH₄ measurements and CO₂ measurements, but it does not mean the operator estimated CO₂ emissions and measured CH₄ emissions have similar random variation. I think there should be a larger daily variation than 3% in the measured CO₂ emissions based on the observation from Figure 3. Please make a clarification here.

(10) In Figure 7 (b)-(d), the y-axis OLS CH₄:CO₂ anomaly is calculated as relative to the daily mean ratio, so it only represents the daily variation. So, these figures can only represent the relationship between sampling variables and daily CH₄:CO₂ ratio variation, not the relationship between sampling variables and measured CH₄:CO₂ ratio. Please clarify it.

(11) In line 557, it should be “Fig. 8”, not “Fig.7”. It is recommended to add the explanations about the difference of Fig 7 (a) and (b) in the figure caption.

(12) what is the meaning of the bracket in the title of y-axis of Figure 9? Please correct it. Same problem in both the x-axis and y-axis title of Figure 10.

(13) Please correct the typo error of “CH₄:CO₂” in the caption of Figure 9.

(14) In line 655, it should be “Figure 10”, not “Figure 9”.

(15) In lines 716-718, the authors argue that one of the benefits of tracer correlation is that the measured site-level ratios on different days could be extrapolated to reporting timescales without the need for detailed knowledge of CO₂ emissions at the time of sampling. However, caution must be exercised when extrapolating these site-level ratios from snapshot measurements to longer timeframes, as the CH₄:CO₂ emission ratio is inherently time-variable. The tracer correlation measurements on a few days capture only brief snapshots of a site’s emission profile. Without a comprehensive understanding of the underlying CH₄ and CO₂ emission sources, including their duration, frequency, and their magnitude, such extrapolation risks producing highly inaccurate, and potentially misleading annual emission estimate.

(16) The paragraph beginning on line 731 should be moved to the end of the above paragraph.

(17) The overall coefficients of determination shown in the Figure S4 in the supporting information are not consistent with the results shown in Figure 6 in the main text. Please correct it.