

**Reviewer n°2:****Summary**

The manuscript by Geilfus and colleagues presents the results of an extensive campaign devoted to investigate the source/sink dynamics of the major greenhouse gases (GHG) CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O along a temperate estuarine system. The authors aimed to quantify GHG fluxes across the sea-air interface under a salinity gradient, as well as to compare those fluxes between sites with contrasting circulation/atmospheric forcing features. Despite inherent heterogeneity in GHG distribution and sea-air fluxes, the main finding is that while CH<sub>4</sub> and N<sub>2</sub>O mostly offset CO<sub>2</sub> uptake in the estuary, N<sub>2</sub>O might also counteract CH<sub>4</sub> emissions.

**General assessment**

Detailed studies of GHG dynamics across the land-ocean continuum are, despite the admittedly increasing attention of the topic over the last decade, still scarce. From that perspective, there is a great value in having such a comprehensive survey of distribution and air-sea fluxes of all three gases simultaneously. An important aspect shown by these measurements/analyses, is that establishing the overall radiative balance of coastal ecosystems is challenging, as departures from the classical view (CO<sub>2</sub> uptake coupled to offset from non-CO<sub>2</sub> gases) is not necessarily a given, when local-scale environmental variability is considered. While the extent at which, for instance, N<sub>2</sub>O might offset CH<sub>4</sub> emissions in this and other systems, as well as its temporal variability over longer time scales remains to be seen, I do think that it is important to show the complex interactions as it has been done here.

→ Thank you.

Overall, the manuscript is well written, the structure is mostly clear and the figures are of good quality. That being said, I spotted a few issues which I would invite the authors to address. The perhaps major issue I see, is the difficulty of combining two scales of variability, namely, the spatial gradient due to salinity, and the variability associated to exposed and sheltered areas. While I appreciate that it is challenging to put these different aspects together, I think combining them makes the manuscript less clear than it could be based on the observations. On the one hand, the data presented in this manuscript shows a very clear salinity-dominated gradient which is reflected in other chemical parameters measured along with the gases. On the other, separating areas in exposed and sheltered (which partially overlap with the salinity gradient), adds a complexity that is not necessarily relatable to the fresh-seawater continuum. Another issue I see is that the definitions of exposed and protected are not fully explained in terms of how consistent they are spatially. For instance, while stations 1-4 and 14-21 are clustered together under the exposed category, the GHGs observations do show that the patterns can be radically different (see e.g. Figs. 2 and 7).

→ We re-evaluated our definitions of exposed and sheltered sites. Lammerant et al (2025) studied the same sites and categorized the sampling locations into four exposure-salinity groups: outer (exposed), middle (semi-sheltered), inner area (sheltered), and Pojo Bay (see their fig. 1).

The following table displays our original classification, Lammerant's, and the new classification used in our manuscript.

Site number	Initial classification	Lammerant et al (2025)	New classification
1	Exposed	Pojo Bay	Exposed
2	Exposed	Pojo Bay	Exposed
3	Exposed	Pojo Bay	Exposed
4	Exposed	Pojo Bay	Exposed
5	Sheltered	Pojo Bay	Sheltered
6	Sheltered	Pojo Bay	Semi-sheltered
7	Sheltered	Sheltered	Sheltered
8	Sheltered	Sheltered	Sheltered
9	Sheltered	Sheltered	Sheltered
10	Sheltered	Sheltered	Sheltered
11	Sheltered	Sheltered	Sheltered
12	Exposed	Semi-sheltered	Semi-sheltered
13	Sheltered	Semi-sheltered	Semi-sheltered
14	Exposed	Semi-sheltered	Semi-sheltered
15	Exposed	Semi-sheltered	Semi-sheltered
16	Exposed	Semi-sheltered	Semi-sheltered
17	Exposed	Exposed	Exposed
18	Exposed	Exposed	Exposed

19	Exposed	Exposed	Exposed
20	Exposed	Exposed	Exposed
21	Exposed	Exposed	Exposed

To align with Lammerant's study, we adapted our manuscript to Lammerant's classification, except for Pojo Bay, which is a geographic location and not indicative of exposure level. Sites within Pojo Bay (sites 1 to 4) are broadly open to the bay and are directly influenced by wind and water input from both the Karjaajoki River and the archipelago. Consequently, those sites will remain labeled as 'exposed'. Sites 5 and 6 are not actually situated in Pojo Bay.

Site 5 is situated within a small bay and connected to the main water channel through a relatively narrow opening. As a result, it will remain labeled as 'sheltered'. Site 6 is somewhat protected from the main channel with features similar to those of sites labeled 'semi-sheltered' in Lammerant's study. Consequently, we decided to change its label from 'sheltered' to 'semi-sheltered'.

We modified the text, and now L 93 reads:

"Sampling sites (N = 21) were selected to encompass a wide range of soft-sediment habitats (e.g., including both vegetated and non-vegetated sediments with grain size ranging from coarse sand to clay, silt, and mud) and to represent a spatial gradient (50 km) from the outer to innermost archipelago. We used the classification by Lammerant et al. (2025), who categorize sampling sites based on their salinity and sediment characteristics to indicate the degree of exposure to environmental forces. Exposed sites (sites 1–4 and 17–21) are likely to experience stronger wave, wind, and current energy, as well as higher water exchange, compared to sheltered sites. Sites within Pojo Bay (sites 1–4) are broadly open to the bay and are directly influenced by wind and water input from the Karjaajoki River and the archipelago. Therefore, those are included with the exposed sites. The sheltered sites (sites 5 and 7–11) are more enclosed and likely to have limited water circulation. Semi-sheltered sites fall between exposed and sheltered environments (sites 6 and 12–16)."

From a GHG perspective, even though sites 1–4 and 17–21 are labeled as "exposed" due to the clear salinity gradient, their GHG patterns can still vary substantially, as the reviewer noted. We hope that our revised explanation of the classification—from exposed to sheltered—and how it relates to water exchange and exposure, now makes our approach clearer.

Figures 3, 5, and 6 have been modified to reflect this change of categories.

All in all, my feeling is that splitting the spatial variability in two "chapters" (e.g. i) salinity gradient, ii) sheltered water ways), would help the authors to convey their arguments more clearly.

→ Thank you for the suggestion. We divided the discussion into different sections, and added a new one based on the recommendation from reviewer n°3.

4.1 Salinity gradient

4.2 Biological drivers

4.3 Exposed and semi-sheltered vs sheltered sites

4.4 Air-sea flux densities

Another aspect is the discussion on the drivers explaining the observed GHG variability. Due to the nature of the data sets gathered as part of the study, there is an imbalance between the possible depth of interpretation of pelagic vs benthic processes. I appreciate that the authors remain careful in that they suggest benthic sources/sinks are "likely" drivers. However, given the weight of this argument throughout the manuscript, I have to agree with the other reviewer (Truong An Nguyen) in that this part could be improved.

→ Here is what we answered to reviewer 1 (Truong An Nguyen):

This is correct; we don't have direct measurements of benthic processes and/or methanogenesis. To avoid misunderstanding, we have now modified both the abstract and the introduction to place less emphasis on these unmeasured processes. We do, however, still use Apparent Oxygen Utilization (AOU) to discuss the potential importance of benthic processes relative to pelagic contributors. We recognize that AOY may not be perfect because it reflects all processes that affect O<sub>2</sub> concentration in the water column. Nonetheless, we believe AOY can provide valuable insights, given the poor correlation ( $R^2 = 0.03$ ) between chl-a and AOY. Using chl-a as a proxy for pelagic primary production, this suggests that primary producers from the pelagic realm have a limited impact on the AOY. If pelagic processes alone cannot explain the observed changes in GHGs, we suggest that processes beyond the pelagic realm, such as benthic community production and

organic-matter degradation, are likely contributing to the observed surface-water pCO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O concentrations. This is further supported by studies from Attard et al. (2019) and Roth et al. (2023) (performed at similar water depth and in the same area), who highlight how benthic processes such as community production, organic matter degradation, and sediment-water interactions are likely to influence both CO<sub>2</sub> and CH<sub>4</sub> dynamics.

The discussion has been adjusted to better reflect our approach while maintaining a more measured tone in both the discussion and the conclusion.

Here I wonder whether the authors could make usage of physical information they collected during their survey to compute e.g. a stratification index that could be used to see at what extent the different areas could have been stratified or well mixed. I noticed that mixing was referred to based on literature, but perhaps it would help the interpretation if we have an understanding of water column oxygen and stratification as potential conditions affecting gas production/consumption/fluxes to the atmosphere.

→ Thank you for the inputs. Unfortunately, we measured only surface seawater properties, which makes it difficult to investigate stratification or mixing processes at the time of sampling.

Lastly, I noticed that the authors gave a relatively high weight to AOU as a tracer for explaining GHGs distribution. The problem I see with that is that AOU is not a compelling predictor of e.g. N<sub>2</sub>O in shallow systems, where air sea gas exchange is dominant. Furthermore, since the connection between primary production (using chlorophyll as a proxy) and N<sub>2</sub>O is not direct, so I would not expect this to necessarily provide a mechanistic explanation.

→ AOU is mainly used to discuss the role of biological processes and highlight the potential importance of benthic over pelagic contributors. We recognize that AOU may not be ideal, but it offers an interesting perspective (see earlier answer above).

The relationship between AOU and N<sub>2</sub>O, initially reported in the manuscript, was incorrect due to an issue in our code, which caused the figure to be incorrect. We have rectified it, and it seems that AOU is quite well correlated with N<sub>2</sub>O ( $R^2 > 0.7$ ).

We modified Figure 6, and we can now read at L 330:

“Despite the lack of correlation between N<sub>2</sub>O and chl-a or O<sub>2</sub> concentrations (Fig. 4), N<sub>2</sub>O exhibited a clear positive correlation with AOU ( $R^2 > 0.7$ ). This correlation is known to reflect the coupling between O<sub>2</sub> consumption (from organic matter remineralization) and microbial nitrogen cycling processes that produce N<sub>2</sub>O (Kock et al., 2016; Carrasco et al., 2017).”

In the following I list other comments, some of which substantiate my assessment above, and some of which correspond to specific issues I spotted while reading the manuscript, and that I hope are useful for the authors when preparing their revision.

#### Specific comments:

**I. 14 – 21:** Given that the crux of the manuscript is illustrating the variability in GHG sources and sinks, it would make sense then to express N<sub>2</sub>O and CH<sub>4</sub> in saturations or Delta values (allowing also the reader to directly draw comparisons with global ocean estimates).

→ Those lines read: “Surface water pCO<sub>2</sub> and N<sub>2</sub>O concentration ranged from undersaturated (160 ppm and 9 nmol L<sup>-1</sup>, respectively) to supersaturated (2521 ppm and 25 nmol L<sup>-1</sup>, respectively), compared to the atmosphere, resulting in an uptake of -36 and -0.0021 mmol m<sup>-2</sup> d<sup>-1</sup>, and a release up to 220 and 0.0383 mmol m<sup>-2</sup> d<sup>-1</sup>, respectively. CH<sub>4</sub> concentrations were consistently supersaturated (19 to 469 nmol L<sup>-1</sup>) compared to the atmosphere, resulting in a net source to the atmosphere from 0.014 to 1.39 mmol m<sup>-2</sup> d<sup>-1</sup>.”

These lines effectively show the observed concentrations of the three gases, highlight their under- and supersaturation relative to the atmosphere, and are enough for the abstract section.

However, we modified the Results section, where we included the complete range of supersaturation for CH<sub>4</sub>, L 216 now reads:

“Surface water CH<sub>4</sub> concentrations were consistently supersaturated (from 636 to 14609 %, data not shown)...”

For N<sub>2</sub>O, L 222 now reads:

“N<sub>2</sub>O concentration ranged from 9 to 25 nmol L<sup>-1</sup>, with saturation level ranging from 93 to 255 %. Higher concentrations and maximum saturation were observed at the mouth of the Karjaanjoki River (Fig. 2c).”

**I. 28 – 30:** The connection between benthic processes and the statement above (as indicated by “therefore”) is not clear.

→ We deleted the word “therefore”.

**I. 88 – 100:** I would suggest revisiting the categories of exposed and sheltered, in particular because areas grouped within the former seem to behave differently (see also comment above).

→ See our previous answer above. We revised the categories of exposed and sheltered sites based on published work from Lammerant et al (2025).

**I. 102 and ff:** Information on calibration and drift of the Li-COR analyzer is missing.

→ You are correct. The LI-COR was factory calibrated before the fieldwork, and standard gases for CO<sub>2</sub> (150, 420, and 1500 ppm) and CH<sub>4</sub> (1, 20, and 150 ppm) were passed through the IRGA both before and after deployment.

This precision has been added to the manuscript L 126, now reads: “The IRGA was factory-calibrated, and standard gases for CO<sub>2</sub> (150, 420, and 1500 ppm) and CH<sub>4</sub> (1, 20, and 150 ppm) were passed through the IRGA both before and after deployment. The data were correct for potential drift.”

**I. 111:** Here and in other instances the authors refer to 45 min as the time needed to reach equilibrium. However, from the formulation, it is not fully clear whether this is the response time of their equilibrator and how it was quantified. Here, a more detailed description of the measurement procedure would be useful for the reader.

→ While most parameters could be recorded quickly, gas measurements, especially for CH<sub>4</sub>, took noticeably more time. Our setup relies on continuous measurement of the gas phase, which equilibrates with water through an equilibrator. Therefore, the speed of measurements depends, among other factors (such as the size and type of the equilibrator), on the gas solubility. While CO<sub>2</sub> equilibrium was reached within minutes, CH<sub>4</sub> solubility caused equilibration to take much longer, particularly at high concentrations, such as those observed at Site 1.

This issue occurs not only when moving from low to high concentrations but also when shifting from high to low concentrations. Since our study examines the spatial variability of GHG concentrations, it was essential to ensure that each measurement truly reflected an equilibrium state. Therefore, to provide high-quality data, we allowed enough time—up to 45 minutes—for the system to reach complete equilibrium with the water.

The 45-minute timeframe is flexible and depends on how quickly we achieve equilibrium. While on site, we could monitor the changes in concentrations of both CO<sub>2</sub> and CH<sub>4</sub> on a laptop. Once CH<sub>4</sub> reached a plateau, we assumed equilibrium had been achieved. It could take up to 45 minutes at sites with high concentrations.

We added this precision at the L 121, which now reads:

“Equilibration between the seawater and gas phases was monitored in real time using a laptop connected to the IRGA. Equilibrium was considered reached when both CO<sub>2</sub> and CH<sub>4</sub> concentrations stabilized at a clear plateau. Depending on the concentration gradient between sites, this equilibration period could vary substantially. While CO<sub>2</sub> typically reached equilibrium within a few minutes, CH<sub>4</sub> required longer times (up to 45 minutes) to reach a stable plateau.”

**I. 130 – 131:** The agreement between chamber-derived and calculated flux densities seems to be remarkably good. Since such chambers are known to suffer from artefacts under turbulent conditions, it would be good for the readers (potential users) to learn about the sampling conditions.

→ We modified the text as follows (L 141):

“Air-sea exchanges of CO<sub>2</sub> and CH<sub>4</sub> were measured using the accumulation chamber technique (Frankignoulle, 1988). The chamber consists of a polyethylene container (internal diameter: 34 cm, height: 14 cm, total volume = 11.4 L) connected in a closed loop to the IRGA with 1.5 m long Tygon tubing, which allows the chamber to move freely on the water surface.”

Our main precaution was to avoid taking measurements when conditions were too rough to prevent water from entering the IRGA, which is a standard and essential step to protect the instrument.

**I. 155:** Here, it is not clear whether the standardised wind speeds were computed from the mean of wind speeds at 2 and 3.2 m height.

→ The line 172 now reads “Wind speed (in m s<sup>-1</sup>, METEK, uSonic-3 Scientific) and air temperature (in °C, Vaisala, HMP155) are measured at the 3.2 and 2 m height, respectively, above sea level at the newly established Integrated Carbon Observation System (ICOS) coastal site at TZS (ICOS code FI-Tvm; Fig. 1).”

We added the word ‘respectively’ to avoid confusion about the anemometer’s height (3.2m), which is extrapolated to the standardized height of 10m.

**I. 157 and ff.:** I think it would be important, for the sake of clarity, to mention how well the in-situ atmospheric measurements fit/are comparable to the atmospheric measurements at this site. For instance, I noticed that the mean value reported for CO<sub>2</sub> (406 ppm; l. 195), is considerably lower than values reported at PAL station at the time of sampling:

[https://gml.noaa.gov/data/data.php?site=PAL&parameter\\_name=Carbon%2BDioxide&frequency=Discrete](https://gml.noaa.gov/data/data.php?site=PAL&parameter_name=Carbon%2BDioxide&frequency=Discrete). The values publicly available for PAL oscillate between ca. 409 and 416 ppm for the same time period in 2023. Admittedly, these values are flagged as “preliminary”, but even after taking the last year of calibrated (final) measurements and applying the global annual rate of increase as an index (see: <https://gml.noaa.gov/ccgg/trends/>), the mean atmospheric xCO<sub>2</sub> value would be ca. 414 ppm. I did not check for the other gases (and certainly would not expect to change much the overall distribution), but it still would be good to clearly state which values were used and why.

→ The average CO<sub>2</sub> (406 ppm) and CH<sub>4</sub> (2.05 ppm) atmospheric concentration reported from this survey is the average value from all the atmospheric measurements obtained while we were flushing the IRGA before the air-sea flux measurements (i.e., while establishing the baseline).

We added this precision in the text, L 147:

“Before conducting the chamber measurements, the IRGA baseline was established by flushing atmospheric air through the analyser. Based on these measurements, we estimated average atmospheric concentrations of 406 ppm for CO<sub>2</sub> and 2.05 ppm for CH<sub>4</sub> during the survey.”

We are unsure that comparing with the PAL station will be most relevant, as the station is located in Lapland on land covered with Alpine tundra, about 200 km north of the Arctic Circle and 1200 km north of the Tvärminne Zoological Station (Tzs). A more suitable reference might be the ICOS marine station Utø, in the Baltic Sea, approximately 200 km west of Tzs. In Utø, the atmospheric concentrations of CO<sub>2</sub> and CH<sub>4</sub> range from 404 to 440 ppm and 1.98 to 2.14, respectively, over the same period as our study. The two datasets seem to align well, considering the variability observed in both cases.

The Tvärminne Zoological Station is also an ICOS station and measures CO<sub>2</sub> and CH<sub>4</sub> concentrations. However, the CH<sub>4</sub> sensor was not installed in 2023. Therefore, only CO<sub>2</sub> data are available on the ICOS data center, and for the same period of this study, reported atmospheric concentrations ranged from 398 to 442 ppm. Here as well, our reported values match quite well with the ICOS atmospheric data.

<https://data.icos-cp.eu/portal/#%7B%22filterCategories%22%7B%22project%22%5B%22icos%22%5D,%22level%22%5B1,2%5D,%22stationclass%22%5B%22ICOS%22%5D%7D%7D>

**I. 188:** This should read “coloured dissolved organic matter”.

→ Thank you for the correction.

**I. 208 and ff.:** Some parts of Fig. 3 are addressed during the results section, while others are mentioned only later during the discussion. Since some of the results are referred to in Fig. 4, it is confusing for the reader. I kindly suggest considering to split Fig. 3 in separate figures that appear then in sections 3 and 4.

→ Thank you for the suggestion. We could split figure n°3 into two separate figures. The new figure n°3 could focus solely on the conservative behavior of both total alkalinity (TA) and dissolved inorganic carbon (DIC) with salinity (the current figure 3a from the manuscript). Figures examining the TA:DIC ratio in relation to surface water pCO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O concentrations (Figures 3b, c, and d in the current manuscript) could then be presented as a separate figure. As we discuss those figures later, as pointed out by the reviewer, this figure will then be numbered figure n°6.

While we understand the reasoning behind the request, we prefer to keep all figures related to TA and DIC in a single plot (current figure 3). This way, all information on those variables is consolidated in one place, reducing the total number of figures in the manuscript and preventing the reader from having to navigate between figures 3 and 6.

**I. 210 – 211:** I think “air-sea flux densities” would be a more adequate term here (instead of “exchanges”).

→ We changed the title of the section to ‘Air-sea GHG flux densities’ and used similar wording in the section: “Measured air-sea CO<sub>2</sub> flux densities ranged from...”, “... source of CH<sub>4</sub> to the atmosphere, with flux densities ranging from...

**I. 232 – 241:** My impression is that this analysis would be better suited for the discussion. Also, it would perhaps help the line of argumentation if variables which are mechanistically expected to be related are analysed. An example of this would be N<sub>2</sub>O and CO<sub>2</sub>.

→ We believe the reviewer is requesting to move the section related to the statistical analysis from the Discussion to the Results section, as Truong An Nguyen (reviewer 1) did. This section has now been relocated to section 3.4.

All variables that are mechanistically expected to be related have been included in the analysis, as L 237 reads:

“The Kendall’s  $\tau$  coefficient has been calculated to investigate the correlation between surface water pCO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O concentration, as well as between physical and biogeochemical parameters and all three GHGs (Fig. 4).”

So this includes the correlation between N<sub>2</sub>O and CO<sub>2</sub>, shown in Figure 4.

**I. 344 – 365:** I think comparing the radiative budget calculated for this system with global estimates (e.g. works by Rosentreter et al and Resplandy et al), would strengthen the arguments laid in this manuscript and emphasize its relevance.

→ Thank you. We followed the suggestion by modifying the conclusion to include work by Rosentreter et al. (2021) and Resplandy et al. (2024) to highlight the interest and need for this work, both at small and large scales. It would have been interesting to compare our GHG budget with the estimate from Resplandy et al. (2024). However, our budget is only valid for one month in late summer, while theirs covers an entire year, including spring and summer blooms—both events we did not observe during our survey. Therefore, a proper comparison would not be very relevant.

The end of the conclusion now reads:

“When translated into CO<sub>2</sub>-eq, air-sea GHG fluxes were dominated by CO<sub>2</sub>, while CH<sub>4</sub> and N<sub>2</sub>O contributed comparably but in different ways. CH<sub>4</sub> consistently acted as a source, whereas N<sub>2</sub>O partially offset the CH<sub>4</sub> release through uptake. This interaction highlights that the balance between production and consumption processes, especially within different seafloor habitats, is critical for understanding coastal contributions to the global carbon budget. There is a critical need to quantify CH<sub>4</sub> and N<sub>2</sub>O exchanges more accurately and to deepen our understanding of the environmental and management factors that control their production and consumption. This would help make global estimates less sensitive to statistical assumptions and reduce uncertainties in blue carbon potential estimates (Rosentreter et al. 2021a).

While coastal ecosystems are often recognized as CO<sub>2</sub> sinks, recent work shows that they are also significant sources of CH<sub>4</sub> and N<sub>2</sub>O that can offset a substantial portion of the climate benefit of taking up CO<sub>2</sub> (Rosentreter et al., 2021a; Roth et al., 2023; Resplandy et al., 2024). Given their extent, northern temperate coastal ecosystems represent a relevant but overlooked source of GHGs, with the potential to amplify the global ocean carbon budget by increasing net greenhouse gas emissions to the atmosphere. Our results highlight the urgent need for research that integrates GHG with biodiversity, benthic-pelagic interactions, and microbial processes, and that resolves temporal variability across seasonal cycles. Such knowledge is essential to improve predictions of how coastal ecosystems will mediate carbon-climate feedback under future environmental change.”

**I. 392:** It is not clear what is meant by “amplify” the global ocean carbon budget.

→ We changed the sentence as follows: “Given their extent, northern temperate coastal ecosystems represent a relevant but overlooked source of GHGs, with the potential to amplify the global ocean carbon budget by increasing net greenhouse gas emissions to the atmosphere.”

Kind regards,  
Damian L. Arévalo-Martínez