

GENERAL COMMENTS

The paper extensively describes a detailed data-set of CO₂ and CH₄ in surface waters of NE Baltic Sea. The figures are of good quality but the text could be improved. The terminology is in place awkward. The first part of the Introduction is a succession of unrelated statements.

MAJOR COMMENTS

Most of the paper is based on textual descriptions of changes of CO₂/CH₄ that are stated to relate to changes of salinity, temperature, or depth. It could be useful to plot CO₂/CH₄ as a function of these variables to back these statements. Such plots allow to explore possible additional features in the data-set.

Thank you very much for the comments. If the total data set is considered, simple property-property plots do not show up here. Therefore, we will select certain episodes (salinity for rivers, temperature for upwelling, water depth for sedimentary interaction) and provide some property-property plots in the supplementary material to support and illustrate our statements.

It's unclear why the mixed layer depth is used to explain patterns in CO₂ and CH₄. I suggest that the authors compute a stratification index such as potential energy anomaly (PEA) according to Simpson (1981). This is a simple computation from the density (salinity-temp) vertical profiles that allows to quantify the strength of water column stratification.

Thank you for the suggestion. However, we are of the opinion that calculating PEA will not be fully relevant in here, as it will depend on the depth. The idea here was to show whether the mixing reaches the bottom or not, stimulating enhanced bottom shear stress and thus sediment interaction. We will add a short explanation to give clearer reasoning for the choice of mixed-layer depth in this context.

SPECIFIC COMMENTS

I do not see what is the logical link between the first, second and third paragraphs of the introduction. The content is correct, but it's unclear how these statements connect together to

introduce the paper. I suggest to remove the first two paragraphs and start the introduction by the section on the Baltic.

With the Introduction's first paragraphs, our point was to provide the reader with a general context of carbon system parameters in the atmosphere, then on the air-sea interface and finally describe the processes in the seawater in more detail. To improve the coherence of the Introduction's text, after the first paragraph we will add a paragraph of the role of the marine realm and more specifically the coastal ocean in the CO₂ / CH₄ cycle and atmospheric budget and improve the logical connection along this line.

L44: There are recent papers showing long term changes in salinity and alkalinity that should also affect the "CO₂ system of surface waters in the Baltic Sea".

Thank you. We will modify the sentence: "In addition to the exchange at the air-sea interface and biological processes, the CO₂ system of surface waters in the Baltic Sea is influenced by the changes in hydrological and hydrographic conditions, e.g. river discharges, waves, currents, salinity and temperature, vertical stratification and mixing, upwelling/downwelling, fronts, etc. (e.g. Müller et al., 2016; Jacobs et al., 2021).".

L55: The collapse of phytoplankton blooms and delivery of fresh material to sediments poor in organic matter seem to stimulate CH₄ release and a seasonal peak that does not coincide with the peak in temperature (Borges et al. 2018). Temperature seems to control seasonality in sediments rich in organic matter.

We slightly extended the sentence to: "In coastal areas, dominant controlling factors for the seasonal variations of methane emission are the sediment organic matter content (Heyer and Berger, 2000), which might be modulated by seasonal deposition of fresh organic material from primary production, and temperature (Borges et al., 2018)". This is meant as a general statement in the context of CH₄ dynamics in shallow and deeper areas. We prefer not to specify the seasonal course and dependencies further in this part (Introduction).

L60: Production of methane in aerobic conditions seem to be only relevant in the deep ocean but not the coastal ocean (Weber et al. 2019), although, concentrations and emissions of CH₄ in the deep ocean are negligible compared to the coastal ocean.

This is a little difficult in the context of the Baltic Sea as the mentioned studies are from the central Gotland Basin, and though showing that the process of production by zooplankton cannot explain the observed (moderate) surface oversaturation. However, the modelling analysis suggests that other surface production pathways must play a role here. However, is correct that the contribution from aerobic production is not important in methane-rich coastal areas. We modify our sentence accordingly: “Production in the upper, oxygenated water column might also contribute to or even govern methane sea-air fluxes (Schmale et al., 2018; Stawiarski et al., 2019), but is of minor importance in the coastal ocean (Weber et al., 2019), and negligible in shallow coastal areas of high methane concentrations / emissions.”.

I suggest that the authors use the full names of the regions instead of the abbreviations (GoF, GoR, and NBP). In the journal Biogeosciences, there is no word limit, so it is unnecessary to abbreviate. For the readers that are unfamiliar with the Baltic Sea it is already difficult to follow the reasoning with these different sub-regions. The use of abbreviations leads to further confusion (letter soup).

Thank you for this suggestion, but we do not consider it necessary to use full names instead of abbreviations since there are only three abbreviations (GoF, GoR and NBP) which are used throughout the manuscript and plots. Using the full names of the regions makes the overview of already detailed plots even more confusing.

L200: why “rapid” ? in relation to what ?

We believe that this comment refers to line 209. ‘Rapid model’ is the name of the approach used for the gas flux calculations (described in detail by Woolf et al., 2016). The term is also used in the FluxEngine vocabulary (Holding et al., 2019). We will modify the sentence: “The CO₂ fluxes were calculated using a rapid model approach (Woolf et al., 2016) implemented into the FluxEngine toolbox.”.

L210: It is not necessary to use subscripts A and W for Henry’s constant (α). The same value is applied to both pCO₂ in air and water.

The ‘rapid’ equation takes into account solubilities in the skin layer and below the surface layer (in the foundation layer); therefore, different subscripts are used.

In the manuscript, after the equation (1), we will complement the sentence and explain the meaning of the subscripts more precisely.

L216: Equation (2) is incorrect. $c\text{CH}_4$ corresponds to dissolved concentration so Henry's constant (α) is not necessary and in fact chemically meaningless.

Thank you for this comment. We will correct the equation (2).

L222: This is a strange result. Please briefly explain why “negligible differences in the average net CO_2 flux were observed when using the different gas transfer parametrisations”. I guess this reflects that wind speed was generally low since all parameterisations converge at low wind speed. Please list the different gas transfer parametrisations that were tested.

We will clarify the paragraph accordingly: “In order to accurately describe the fluxes and the carbon budget, it is essential to include relevant processes to the air–sea CO_2 and CH_4 flux parametrisation. Nightingale et al. (2000) was used for the gas transfer velocity parametrisation for both CO_2 and CH_4 in our study. The sensitivity analysis of the gas transfer velocity in the Baltic Sea (Gutiérrez-Loza et al., 2021) used different parametrisations of the gas transfer velocity to evaluate the effect of other relevant processes in addition to wind speed on the net CO_2 flux at regional and sub-regional scale. In the Estonian sea area, they observed negligible differences in the average net CO_2 flux when using the different gas transfer parametrisations relative to the wind-based parametrisation.”.

L 320: it's water that is under-saturated not CO_2 itself.

We will correct our sentence accordingly: “The surface waters were undersaturated in CO_2 in most areas of the GoF except the Narva Bay, where the water column was well-mixed down to the seabed, oversaturated in the Väinameri Sea and Pärnu Bay, and undersaturated in the NBP (Fig. 8c).”.

REFERENCES

Müller, J. D., Schneider, B., and Rehder, G.: Long-term alkalinity trends in the Baltic Sea and their implications for CO_2 -induced acidification, *Limnol. Oceanogr.*, 61, 1984–2002, <https://doi.org/10.1002/lno.10349>, 2016.

Weber, T., Wiseman, N. A., and Kock, A.: Global ocean methane emissions dominated by shallow coastal waters, *Nat. Commun.*, 10, 4584, <https://doi.org/10.1038/s41467-019-12541-7>, 2019.