Author's point-by-point response

The revised manuscript according to suggestions by the two referees.

Response to referee #1:

Comments by referee #1

Review of the revised manuscript: "Methane dynamics in the Baltic Sea: investigating concentration, flux, and isotopic composition patterns using the coupled physical, biogeochemical model BALTSEM-CH4 v1.0" by E. Gustafsson, B. G. Gustafsson, M. Hermans, C. Humborg, and C. Stranne, under review for Geoscientific Model Development

The authors have responded well to most review comments, and the manuscript improved substantially. The discussion about methane production in oxic water has been improved. The methane oxidation rates are compared to previous field studies, and in general, there is more consideration for previous biogeochemical research. The introduction became more informative. In response to the comments of the other reviewer, the model description has been expanded.

The most questionable part remains the overall methane budget for the Baltic Sea. The authors explain well that the model is not well-suited to constrain ebullitive emissions. The text also states: "Estimates by Weber et al. (2019) indicate that ebullitive fluxes contribute a major fraction of CH4 released to the atmosphere from shallow coastal areas" (lines 671-672). To be more precise, Weber et al. (2019) estimate that ebullitive fluxes account for roughly 50% of total global ocean methane emissions. Therefore, I agree with the following assessment: "This indicates that the simulated CH4 outgassing is likely underestimating the real outgassing from the Baltic Sea" (lines 679-680).

The average depth in the Baltic Sea is ~50 m. The model is fitted to data from the Gotland Basin, which has a maximum depth of ~460 m. Currently, it seems extremely difficult, if not impossible, to obtain empirical data needed to constrain a methane budget for the shallow parts of the Baltic Sea. Additionally, the model cannot resolve point sources and simulate methane ebullition. Coastal sediments are highly diverse in terms of organic matter loading, sedimentation rates, and substrates. These factors, which can only be distinguished for different locations by a 3-D modeling approach, significantly influence methane production rates. It is very likely that the methane production in shallow parts greatly differs from that in deeper parts.

My concern is that the model has been designed for deeper regions and fitted only to data from the Gotland Basin. Extrapolating these results to the entire Baltic Sea leads to highly unreliable estimates. Although the authors acknowledge the large uncertainties and describe their work as a preliminary budget, the current estimate is so uncertain that it does not enhance the quality of the paper. In the worst case, other studies may uncritically adopt these emission rates, potentially propagating significant errors.

In my opinion, the paper would be much stronger if it focused on the Gotland Basin, for which the model has been fitted. The simulated dynamics related to the interplay between physical and biogeochemical processes are sufficiently interesting. The budget for the entire Baltic Sea, on the other hand, is speculative at best. The text itself already contains many warnings. The authors may find it regrettable in the future if this part turns out to be significantly inaccurate.

I could agree with a minor revision if the quality of the paper is improved by reducing the emphasis on the preliminary budget. This could be achieved by removing the estimates of CH4 release from

sediments and CH4 emissions to the atmosphere for the entire Baltic Sea from the abstract and graphical abstract and instead providing values specifically for the Gotland Basin. Additionally, if the text focused more on the Gotland Basin, the analysis would be more robust, requiring less caution from the reader. The model output for the entire Baltic Sea could still be discussed in the text as part of ongoing model development, but should not be presented as robust scientific findings.

Response: The Baltic Sea scale budget has now been replaced by a budget just for the Baltic Proper (including in addition to the Gotland Sea also the Bornholm and Arkona basins). Measured profiles of both CH4 concentration and isotopic composition are available not only for the Gotland basin, but also from the Bornholm and Arkona basin, indicating that the model (reasonably well) represents the central Baltic Sea area. On the other hand, the Gulfs of Bothnia, Finland, and Riga as well as the Kattegat basin are no longer included in the budget analyses. Furthermore, the graphical abstract was removed to reduce the emphasis on the preliminary budget calculations.

Consequently, the text has been modified on several places in the manuscript:

- *The abstract has been updated and now reads:*

"Methane (CH4) cycling in the Baltic Sea is studied through model simulations that incorporate the stable isotopes of CH4 (12C-CH4 and 13C-CH4) in a physical-biogeochemical model. A major uncertainty is that spatial and temporal variations of the sediment source are not well known. Further, the coarse spatial resolution prevents the model to resolve shallow-water near-shore areas for which measurements indicate occurrences of considerably higher CH4 concentrations and emissions compared to the open Baltic Sea. A preliminary CH4 budget for the central Baltic Sea (the Baltic Proper) identifies benthic release as the dominant CH4 source, which is largely balanced by oxidation in the water column and to a smaller degree by outgassing. The contributions from river loads and lateral exchange with adjacent areas are of marginal importance. Simulated total CH4 emissions from the Baltic Proper correspond to an average ~1.5 mmol CH4 m-2 y-1, which can be compared to a fitted sediment source of ~18 mmol CH4 m-2 y-1. A large-scale approach is used in this study, but the parametrizations and parameters presented here could also be implemented in models of near-shore areas where CH4 concentrations and fluxes are typically substantially larger and more variable. Currently, it is not known how important local shallow-water CH4 hotspots are compared to the open water outgassing in the Baltic Sea."

- *The text on line 99-100 now reads:*

"... 2. set up a preliminary CH4 budget for the Baltic Proper (where measured profiles of CH4 concentration and isotopic composition are available),..."

- *The text on line 536-543 reads:*

"To allow a preliminary assessment of the relative importance of different processes, total CH4 sources (river load, import from adjacent sub-basins, and sediment release) and sinks (outgassing, export to adjacent sub-basins, and pelagic oxidation) were aggregated over the Baltic Proper (sub-basin 7-9, Figure S1), representing the area where the model has been fitted based on available observations. The CH4 sources were largely dominated by benthic release which amounted to an average 4155 Mmol y-1 over the 2001-2020 period (Table 2). This source was mainly balanced by oxidation in the water column (3816 Mmol y-1, 92% of the sinks) and to a smaller degree by emission to the atmosphere (348 Mmol y-1, 8% of the sinks). The river load (11 Mmol y-1) and net exchange (import - export) with adjacent sub-basins (8 Mmol y-1) were comparatively small."

- *Table 2 (line 546) has been updated with numbers for the Baltic Proper.*
- *The text on line 547-552 reads:*

"Figure 6 illustrates simulated monthly fluxes, net accumulation as well as the total amount of CH4 in the Baltic Proper. The total CH4 stock amounted to almost 1800 Mmol over the ~2010-2014 period, which exceeded the stock before and after that period by a factor 3 (Fig. 6). This comparatively large CH4 stock was the result of a large anoxic deep-water volume and thus low oxidation rates (Fig. 2). There was an average net accumulation of 10 Mmol y-1 over the 2001-2020 period (Table 2), but net changes of the total CH4 stock between individual years varied considerably, which largely reflected oxygen dependent changes in CH4 oxidation rates (Fig. 6)."

- *The original Figure 6 (line 554) has been replaced by a corresponding figure for the Baltic Proper.*
- *The text on line 559 reads*

"This study presents a first quantification of key CH4 fluxes in the Baltic Proper"

- *The text on line 698-701 reads:*

"The calculated average total CH4 emission of 348 Mmol y-1 from the Baltic Proper corresponds to approximately 1.5 mmol CH4 m-2 y-1, and constitutes only about 8% of the fitted sediment source (~18 mmol CH4 m-2 y-1). The model includes both shallow- and deep water sediment areas, but the fitted sediment source is in the lower range of rates reported for a shallow-water coastal area (~21-34 mmol CH4 m-2 y-1; Roth et al., 2023),..."

Minor comments

Lines 449-450: "the intention... Gotland Sea"

Please, make explicit what is meant by "existing observations" by spelling out the fitted parameters.

Response: The sentence has been rewritten and now reads (line 449-451): "These parameters (i.e., CH4 oxidation rates and fractionation values, CH4 sources from the sediments, rivers, and the North Sea, as well as the isotopic compositions of these sources) are mostly fitted values, where the intention was to reasonably well reproduce existing observations of both CH4 concentration and isotopic composition from the Gotland Sea"

Line 53: "250 µmol m-2 day-1":

Here µmol is used. In other parts of the manuscripts, both grams and moles are used. Please, use the same units throughout the manuscript.

Response: Mole units are now consistently used for CH4 fluxes throughout the manuscript.

"Calibration" versus "fitting" in the entire manuscript:

Fitting is more appropriate in the context of this study. There is a difference in meaning between the words. Fitting means adjusting the model to match known data as well as possible. Calibrating is a

broader process that includes fitting but also ensures the model performs well in different situations and against additional validation data.

Response: The word "calibrated" has now been replaced by "fitted" throughout the manuscript.

Response to referee #2:

Comments by referee #2

In the first paragraph, when describing the estimates of global CH4 emissions by the two approaches, can they authors include uncertainty range as well? The current description reads like too accurate.

Response: The ranges have now been included on line 29-31: "... global CH4 emissions have been estimated to be 576 Tg CH4 y-1 (range 550-594), whereas bottom-up approaches (process-based modeling of land surface emissions and data on anthropogenic emissions) yield a total of 737 Tg CH4 y-1 (range 594-881; Saunois et al., 2020)"