Response to referees comments on "Biogeochemical controls on carbonate dynamics driven by methane and freshwater inputs in shallow sediments of a brackish continental shelf sea" by Łukawska-Matuszewska et al.

A. Response to comments by Tom Jilbert (Referee #1)

We thank Prof. Tom Jilbert for his careful review and constructive comments. We have revised the manuscript accordingly and address each point below, describing the specific changes made in response to his suggestions.

Interactive comment on "Biogeochemical controls on carbonate dynamics driven by methane and freshwater inputs in shallow sediments of a brackish continental shelf sea" by Łukawska-Matuszewska et al.

General

This paper presents an extremely comprehensive analysis of sediment biogeochemistry at three locations in the Gdansk Deep. The authors should be commended for the integrated approach to study solid-phase and porewater chemistry, which yields valuable insights into coupled primary and secondary diagenetic processes. In particular the work focuses on the factors controlling the rates of diagenetic mineral formation and burial (especially dolomite and pyrite) but touches on many other topics due to the breadth of the data.

I agree with most of the conclusions of the study but there are some interpretations that I would suggest to modify because their implications may be misunderstood if taken at face value. For example I do not think it is justified to state unilaterally that methanogenic sediments favor authigenic carbonate formation. I would also like to see some clarification of the separate measurements of del13-DIC and del 13C-CO2 included in the porewater analysis and how these results should be interpreted, and some more details about the incubation experiments and connected data processing. Overall I would say that the breadth of the approach introduces some difficulties to maintain coherence in the manuscript. Some rewriting of the introduction would help to guide the reader towards the rationale of the study in a more logical way. My Line by Line comments contain all the relevant points to be addressed. I wish the authors success in their revisions.

We made every effort to implement all changes suggested by the reviewer, including revisions to the Methods and to the interpretation of results. The original Methods section had been shortened for the main text and omitted several procedural details; we have now expanded it to restore key analytical and procedural information so that the results can be unambiguously interpreted. Below we summarize how we addressed the main and minor issues raised.

Line by Line

Line 24: "In" rather than "At" the methane-free sediment

Answer: "At the methane-free sediment" was changed to "In the methane-free sediment." (line 24).

Lines 25 and 29: DIC production rates are given here in two separate units. For the sake of readability it would be sensible to use the same unit.

Answer: Thank you for the comment. We corrected the inconsistent units for clarity. Revised sentences (included in the manuscript in lines 29-37) read: In such conditions, carbonate dynamics were markedly enhanced: methane-related processes - in particular AOM, which reached up to 1077 μmol dm^{-3} d^{-1} in freshwater-influenced sediments - produced additional DIC, contributed to carbonate supersaturation and promoted pronounced authigenic carbonate (mainly dolomite) formation. Depth-integrated DIC

production from AOM (331 mmol m^{-2} d^{-1} over 0–100 cm) substantially exceeded OSR-derived DIC (55–59 mmol m^{-2} d^{-1}) in these settings, corresponding with elevated authigenic dolomite burial rates (467–994 µmol m^{-2} d^{-1}).

Lines 34-80 (general comments on Introduction): Lines 72-80 do a good job of outlining the eventual rationale for the study, but some of the earlier text is harder to read. I suggest to try to introduce the key biogeochemical processes to be studied in a more consistent way, e.g. treat primary and secondary diagenetic processes separately, then introduce the effects of salinity and submarine groundwater discharge, and finally focus on those processes that control alkalinity production as a driver of carbonate precipitation. The current flow jumps between these concepts and is not always consistent.
Answer: Thank you for this suggestion. The Introduction was reorganized and now follows the following outline: (1) Continental shelf seas significance in the global carbon cycle. Anthropogenic nutrient loading and coastal eutrophication; (2) Input and fate of organic matter to sediments; redox cascade of sedimentary organic matter oxidation (sequence of electron acceptors and transitions from aerobic to anaerobic respiration); (3) Organoclastic sulfate reduction in coastal sediments - prevalence and contribution to organic matter oxidation; (4) Methanogenesis and methane turnover in sediments - conditions favoring methanogenesis and pathways of methane oxidation; (5) Impacts of OM remineralization on DIC and total alkalinity (production, implications for seawater buffering, DIC-driven supersaturation and long-term burial mechanisms of authigenic carbonate precipitation); (6) Role of reoxidation and authigenic mineral formation in alkalinity dynamics - alkalinity consumption by reoxidation, alkalinity gain from mineral preservation (e.g., pyrite formation); (7) Regional context - shallow shelf seas and the Baltic Sea example: close benthic–pelagic coupling, high sedimentation rates, freshening effects, enhanced anaerobic remineralization, and shallow SMT dynamics; (8) Study scope and objectives: environments compared, measurements made (pore-water chemistry, sediment geochemistry, mineralogy, isotopes), rate experiments, and conceptual model development.

Lines 45-48: OSR should not lower pH or enhance carbonate dissolution because there is 2:1 production of HCO3- vs. H+. Please check and rephrase.
Answer: The Introduction was rewritten as suggested. This sentence has been removed (line 66).

Line 91: Does this mean 7 cores in July 2023 and another 7 in February 2024? Please be specific.
Answer: We clarified that seven sediment cores were collected during each sampling campaign: seven cores in July 2023 and seven cores in February 2024 (lines 129-130).

Line 94: This should read "CH4, CO2 and their stable isotopes..." (but see later comments about needed clarifications about this methodology).
Answer: It was corrected as suggested (line 132). We also added further details in the Methods on sampling for CH_4, CO_2 and their stable isotopes (lines 156-158).
A methodological clarification is provided in our response to the comment about Supplement Fig. S3 (labelled important) on page 7 of this document.

Line 99: How was the rhizon sampling performed under anoxic conditions, was the setup placed in a glove bag?
Answer: Thank you for this remark. The description of porewater sampling was replaced with the following text: Pore waters were extracted from intact, sealed cores using Rhizon® samplers (0.15 µm pore size). For each core liner, 4 mm holes matching the Rhizon® sampler fitting were pre-drilled at 5 cm intervals and sealed with a tape. After cores retrieval aboard, the tape was pierced and samplers inserted; to minimize contact with air, cores remained sealed throughout porewater sampling. The

uppermost sampler (1–4 cm above the sediment surface, depending on a station) collected bottom water overlying the core. Porewater was drawn into attached 20 cm^3 plastic syringes. To minimize oxidation artefacts, all subsequent subsampling and treatment were performed immediately after porewater sampling (lines 138-144).

Line 107: At what stage was a headspace injected into the methane sample vials? This should be stated.
Answer: Wet sediment aliquots were subsampled directly into gastight serum vials containing 2.5% NaOH to halt microbial activity and preserve dissolved gases. Vials were then sealed and a headspace was created and equilibrated immediately by shaking; headspace gas was subsampled and analyzed for CH_4 concentration. This information was added in lines 151-154.

Line 160: The term *IG* in equation (2) is not defined.
Answer: IG is loss on ignition (LOI) in percent of wet sediment. It was defined in line 198.

Line 163-172: this section concerns porewater analyses so it is not clear why it is placed here in the sediment analyses section.
Answer: This section (CH_4 analysis plus C and H isotopes of CH_4 and C isotopes of CO_2) was relocated to section 2.2.1 (it's now in lines 191-209).

Line 168: What was the detection limit in terms of CH4 concentrations to obtain reliable delC13 data? Was a pre-concentration unit used?
Answer: The detection limit for obtaining reliable $\delta^{13}C$ data was 0.1% CH_4. No pre-concentration unit was used. This this information has been added in Methods section (line 205).

Line 178: How were thin sections produced? Was the sediment embedded and if so with which resin?
Answer: Undisturbed air-dried samples of relevant horizons were embedded in a Technovit EPOX resin. A relevant information has been added to the Methods section (line 255-256).

Line 186: Clarify that *d* in equation 3 is the wet bulk density.
Answer: We clarified that *d* in Equation (3) refers to the wet bulk density (line 196).

Line 190: Was the PXRD quantification achieved using the Rietveld method? Please comment on the uncertainty for estimates of dolomite and pyrite content in the ranges calculated in this study.
Answer: Mineral composition was quantified by Rietveld full-pattern X-ray diffraction fitting technique using SIROQUANT analysis of randomly oriented powders. A relevant information has been added to the Methods section in lines 249-205.
The uncertainty was estimated from standard deviations on individual scale factors for each phase. These uncertainties are (in wt.%) 0.3 for pyrite, 0.5 for dolomite and 0.4 for calcite (it has been reported in lines 250-251).

Line 198: If the measured product is H_2S, how does this method to determine OSR exclude H_2S produced by SO_4-AOM?
Answer: Thank you for this comment. In the organoclastic sulphate reduction assays, lactate was supplied as a dedicated electron donor for sulphate-reducing bacteria at concentrations largely exceeding the potential availability of methane introduced with the 1 cm^3 sediment slurry. The sediment was pre-diluted with sterile seawater enriched with lactate (line 276) to enable accurate handling of dense sediment, which further ensured an excess of organic substrate relative to methane. Therefore, sulphate reduction activity measured as H_2S production was driven by organoclastic sulphate reduction rather

than sulphate-dependent anaerobic oxidation of methane. The following information has been added in Methods section (lines 277-278): Lactate was added as an electron donor during sample pre-treatment to ensure that measured H_2S production primarily reflected organoclastic sulphate reduction.

Line 237: 3.2 has the same heading as 3.1 (!)
Answer: The heading was corrected (Section 3.2 reads: "3.2. Composition of pore water and bottom water" (line 324).

Line 286: This clarification of the incubation approach should also be stated in the Methods.
Answer: It has been added to the Methods (lines 273-275) and is also included in the Results (lines 376-379).

Line 296 (Table 1): The raw data used to calculate the rates needs to be included in the Supplement. Also it is interesting to note that MET-2 and ZGG are very similar in terms of experimentally-derived process rates. This does not seem to match with the porewater profiles, in which AOM appears much more important at MET-2.
Answer: Thank you for this suggestion. We included the raw data used to calculate the rates in the Supplementary Material (Table S2 and table S3). Specifically, we provided the measured H_2S concentrations obtained during incubations at 0.1 M sulphate concentration, which were used for rate calculations. In addition, we included in the Supplement the fitted kinetic parameters derived from the rate-versus-substrate concentration curves. We also added the parameters and conditions for determining CO_2 concentration as a result of methane oxidation (AOM). We refer to these data in section 3.4 (line 388).
The experimentally-derived AOM rates represent the potential activity of the microbial community under substrate-replete conditions rather than the actual in situ process rates (lines 273-275 in Methods and lines 376-379 in Results). Therefore, the similar experimentally-derived rates at MET-2 and ZGG indicate a comparable microbial potential for AOM at both sites. In contrast, the porewater profiles reflect in situ conditions, where methane availability differs substantially between the sites. At MET-2, methane is present and supports active AOM, which is reflected in the porewater profiles. At ZGG, methane concentrations are negligible, so AOM appears less significant in situ despite the presence of a microbial community capable of performing this process if methane became available.
The following text was inserted in the Discussion (lines 548-554): „It should be also mentioned that experimentally-derived AOM rates represent the potential activity of the microbial community under substrate-replete conditions rather than the actual in situ process rates. Therefore, the similar experimentally-derived rates at MET-2 and ZGG indicate a comparable microbial potential for AOM at both sites. In contrast, the porewater profiles reflect in situ conditions, where methane availability differs substantially between the sites. At MET-2, methane is present and supports active AOM, which is reflected in the porewater profiles. At ZGG, methane concentrations are negligible, so AOM appears less significant in situ despite the presence of a microbial community capable of performing this process if methane became available."

Line 308: Can this variable volume of "sediment matrix" be related to changes in water content downcore? The images imply so, but it depends partly on how the thin sections were prepared, see earlier comment and adjust accordingly.
Answer: The values presented here were obtained from planimetric analysis using an optical microscope. Up to half of the sediment was a mineral-organic matrix, grouped together due to the inability to distinguish individual components. Since thin sections were prepared from air-dried sediments, water content should not have been a significant factor.

Line 328: Where are the SEM-EDS results?
Answer: Initially, we decided not to include EDS spectra in the text due to its length. Representative spectra of pyrite, dolomite and calcite have been added to the Supplementary Materials (Figure S8).

Line 334 (Fig. 4): "c" appears to be pyrite and not calcite as stated in the caption.
Answer: Thank you for pointing this out. We noticed that there are a few mistakes here and hence the entire caption was corrected as follows (lines 446-450): Examples of SEM-BSE images of authigenic sulfides and carbonates: (a) pyrite framboids, MET2 (40–45 cm); (b) pyrite framboid and disseminated pyrite crystals, ZGG (10–15 cm); (c) pyrite framboids within a diatom shell, ZGG (40–45 cm); (d) pyrite framboid, diatom, dolomite, MET1-MP (40–45 cm: thin section); (e) euhedral and subhedral dolomite crystals, MET1-MP (10–15 cm; thin section); (f) euhedral dolomite, MET1-MP (10–15 cm; thin section); (g) dolomite, MET1-MP (40–45 cm); (h) calcite surrounded by clay minerals, ZGG (40–45 cm); (i) siderite, MET2 (40–45 cm).

Line 398: The diagenetic reactions are given different codes (e.g. R1, R2) in the main text and in the supplement. Please be consistent throughout.
Answer: We revised the Introduction and resolved the reaction-numbering inconsistency. We moved all diagenetic reaction equations to the Table S1 in Supplementary Material and referred to them from the main text (ensuring a single, consistent reference for each reaction. We also reorganized the reactions in Table S1 so they appear logically: (1) organic-matter degradation (primary redox reactions), (2) reoxidation of reduced by-products and AOM (secondary redox reactions), and (3) mineral precipitation/dissolution.

Line 426-427: Is this a reference to the incubation results of the present study? If so please be clear.
Answer: Yes, this refers to incubation results from the present study. It was clarified by revising the sentence to: "Experiments with labeled substrates in the present study indicate the potential for AOM at ZGG, with rates up to ~10 µmol dm^{-3} d^{-1}." (lines 544-545).

Line 435: For completeness should denitrification be considered here?
Answer: In the reducing sediments we analyzed, porewater nitrate concentrations were consistently below the detection limit, and nitrite occurred only at trace levels, indicating that their availability was insufficient to support measurable denitrification under the study conditions. Given the redox conditions and the absence of oxidized nitrogen forms, the contribution of denitrification is likely negligible and was therefore not included in the quantitative DIC budget. The following text was added (lines 560-564): “Besides Mn(IV) reduction, organic matter oxidation in marine sediments can proceed via denitrification. However, in the reducing sediments examined here nitrate was consistently below detection limit and nitrite occurred only at trace levels (data not shown), so denitrification was not quantified. While denitrification can produce DIC, we have no evidence for substantial availability of oxidized nitrogen species in these samples and therefore expect its contribution to be negligible.”

Line 439: Refer to Table 2 again showing the low values of dolomite at this site.
Answer: We added a reference to Table 2 in the sentence: Despite thermodynamic supersaturation, carbonate contents remain low (Table 2). (line 567).

Line 446-447: This logic is not quite correct, because as stated earlier in the manuscript, carbonate alkalinity, as HCO3- + 2(CO3 2-) is not quite equivalent to DIC. I am not against using these type of approximations to derive useful information from the dataset, but all assumptions should be made clear.

Answer: Thank you for the suggestion. Our point was to show how large is the contribution of non-carbonate inorganic bases to TA (this is important because this part of alkalinity is "reversible" for example due to oxidation of HS^- and/or NH_3, the most important non-carbonate inorganic bases in anoxic sediment). This explanation was added in lines 577-579. We also clarified that DIC = $[CO_2^*] + [HCO_3^-] + [CO_3^{2-}]$ with $[CO_2^*]$ denoting the sum of dissolved $[CO_2]$ and $[H_2CO_3]$ (Dickson et al., 2007) (lines 575-577).

Dickson, A.G., Sabine, C.L. and Christian, J.R. (Eds.) 2007. Guide to best practices for ocean CO_2 measurements. PICES Special Publication 3, 191 pp.

Line 499: Should the reference to Fig. 4 actually be to Fig. 5?
Answer: We changed the in-text reference from Fig. 4 to Fig. 5 (line 633) and verified all figure references for consistency (the order and numbering changed after adding new figures - ESD and Raman spectra in the supplement).

Line 544-545 (important): I think the interpretation is more nuanced. Generally the formation of carbonates will be a function of the overall rate of the primary diagenetic reactions producing DIC and thus raising the IAP of dolomite and calcite. There is no mechanistic reason why methanogenic sediments (in the absence of sulfate) should favor carbonate precipitation; on the contrary OSR-dominated systems would be expected to have higher rates of diagenesis generally, due to greater energy yield of OSR. However, in systems where both process occur but overall diagenesis rates are so high that sulfate is exhausted in the uppermost decimeters (e.g. MET-1, MET-2 here), DIC will be observed to be higher overall (thus favoring carbonate formation), and by coincidence a methanogenic zone will be observed in short sediment cores that would otherwise be confined to deeper horizons. So I would suggest that one of the main outcomes of this study is to highlight how topographic features such as pockmarks influence the sedimentation regime such that demand for electron acceptors is concentrated in specific areas, leading to knock-on effects in secondary processes such as mineral formation (with contrasting outcomes for sulfides and carbonates).
Answer: Thank you for this remark. We attempted to remove the implication that methanogenesis per se caused carbonate precipitation and reframed the interpretation to emphasize that carbonate formation was controlled primarily by the overall rate of primary diagenetic reactions producing DIC (higher DIC → higher IAP → greater potential for carbonate precipitation), rather than by the identity of a single terminal electron-accepting process.
In the Discussion, the explanation has been added that the physical–sedimentary character of pockmark drives the discussed processes because OM accumulation increases demand for electron acceptors (lines 720-723); as a result of intense diagenesis sulfate are exhausted in the upper decimeters and the methanogenesis zone shifts toward the sediment surface; the intense diagenesis is responsible for the high DIC. We highlighted the importance of methanogenesis and AOM as additional sources of DIC.
The sentence "These data indicate that methane-bearing sediments with active methanogenesis constitute a more effective sink for inorganic carbon than anoxic sediments dominated solely by OSR." has been removed (line 679). In lines 676-677, it has been added that: "Intense diagenesis leads to a strong increase in DIC, which raises porewater saturation with respect to carbonates and thereby promotes precipitation of authigenic phases". Then in lines 681-683 an additional explanation is provided: "Taken together, the porewater chemistry and burial rates suggest that methane-bearing sediments in our study area sequester more inorganic carbon than methane-free, OSR-dominated sediments - likely because methane-related processes generate additional DIC."
Also in Abstract and Conclusions we made similar corrections (lines 26-30 in Abstract and lines 824-826 and 831-833 in Conclusions).

Lines 693-695 - "This allows pyrite to form not only within the sediment but also in the water column (Raiswell and Berner, 1985). As a result, pyrite is more abundant at ~~methane-free~~ ZGG than at ~~methane-bearing~~ MET2 (Table 2)." was replaced with: This allows pyrite to form not only within the sediment but also in the water column (Raiswell and Berner, 1985) and, as a result, pyrite is more abundant at ZGG than at MET2 (Table 2)."

Line 973: Whiticar (1999) is missing from the bibliography. Check all references.
Answer: Whiticar, M.J., 1999. Carbon and hydrogen isotope systematics of bacterial formation and oxidation of methane. Chem. Geol. 161, 291–314. https://doi.org/10.1016/10.1016/S0009-2541(99)00092-3 was added to the bibliography (lines 1152-1153).
We checked all references for completeness and consistency and made corrections where necessary.

Supplement Fig. S3 (**important**): This point concerns the methodology for stable carbon isotopic analysis of dissolved inorganic carbon species used in the study, and how the data should be interpreted. Samples appear to have been taken in two separate ways: 1. in connection with the CH4 sampling, for which wet sediments were stored in 2.5%NaOH and then headspace gas was extracted for del13C-CO2; and 2. in connection with the DIC sampling, for which extracted porewaters were acidified with phosphoric acid and all DIC converted to CO2 for determination of del13C-DIC. A couple of queries here:

- for method 1, do the authors assume the data is reporting the del13C of only the dissolved CO2 (i.e. not the HCO3- and CO3(2-) derived fractions)?
- if so, how should those data differ from the del13C-DIC, if the entire DIC pool is in equilibrium? I checked the Whiticar (1999) paper and it seems that there is some interchangeable use of the terms del13C-CO2 and del13C-DIC in that study. For example in Fig. 7 of Whiticar the axis states "del13C-CO2" but the caption indicates "carbon isotopes of bicarbonate". But if the authors are aware of literature of isotopic fractionation within the carbonate system in porewaters they should cite it here and explain how it impacts on the observations.
- please comment on how it was possible to achieve sufficient recovery of CO2 by method 1, if the treatment with NaOH shifted the equilibrium in the carbonate system towards HCO3- and CO3(2-)? Is it a question of how quickly the headspace samples were taken?

Answer: Thank you for bringing this to our attention. We had not described the sampling procedures for isotopic analyses of gases and pore water, which understandably created uncertainty about data interpretation, and we apologize for the omission. We acknowledge that additions of NaOH or acid alter carbonate-system equilibria; therefore we collect isotope samples separately (as described in our earlier works, for example in **Brodecka-Goluch A., Łukawska-Matuszewska K.**, Kotarba M.J., **Borkowski A.**, Idczak J., Bolałek J. (2022) Biogeochemistry of three different shallow gas systems in continental shelf sediments of the South-Eastern Baltic Sea (Gulf of Gdańsk): carbon cycling, origin of methane and microbial community composition. Chem Geol 597:120799).
The description of the sampling procedures for isotopic analyses in gases and pore water was added to the Methods section.

1. For $\delta^{13}C\text{-}CO_2$ (and $\delta^{13}C\text{-}CH_4$): samples were collected in the same manner as the CH_4 samples but the procedure was carried out inside an N_2 glove box. Instead of adding NaOH, vials were pre-filled with Milli-Q water (this information was added to section 2.1 on sampling (lines 156-158), immediately after the description of methane-sampling procedure).

No NaOH was added to the vials used for the $\delta^{13}C\text{-}CO_2$ headspace samples to avoid shifting the carbonate-system equilibrium. Samples were collected in an N_2 glove box into vials pre-filled with Milli-Q water (no alkali), sealed with butyl septa, and equilibrated immediately by gentle shaking to allow CO_2 to partition into the headspace; headspace aliquots were then taken promptly for isotope

analysis. These steps (no NaOH, anaerobic handling, rapid equilibration and sampling) ensured efficient recovery of CO_2 into the headspace for isotope measurement.

2. For δ^{13}C-DIC: sampling was carried out in an N_2 glove box; porewater samples were collected into 2 mL septum vials that were filled by injecting porewater with one needle while displacing N_2 through a second needle; vials were completely filled with porewater and stored at 4°C prior to analysis (this information was added to section 2.1 (lines 147-150), right after the description of DIC-sampling procedure).

During analysis, samples were reacted with H_3PO_4 for 24 h at 25°C and the evolved CO_2 was analyzed for carbon isotope composition using a Thermo Delta V isotope ratio mass spectrometer coupled to a Thermo GasBench II under continuous helium flow (this is described in lines 177-178). Therefore the data is reporting the δ^{13}C of CO_2 as well as bicarbonate and carbonate derived fractions (δ^{13}C-DIC).

Supplement Fig. S9: There is no legend so the color coding is not clear.
Answer: We added a legend to Supplementary Figure S9 (it's Figure S11 in revised version) explaining the color coding.

B. Response to comments by Anonymous Referee (Referee #2)

We thank the Reviewer for evaluating our manuscript and for the helpful comments and suggestions about our work. We did our best to revise the manuscript accordingly. Below we responded to each point and outlined the specific revisions made.

Comment on egusphere-2026-1238
The authors present original geochemical data from the composition of porewater and selected sediment samples obtained during two cruises to the Bay of Gdansk. Besides dissolved major and selected trace elements, sediment samples were investigated for microbial variables, and standard powder X-ray diffraction and SEM-EDS was used for bulk phase analysis. Furthermore, potential net microbial sulfate reduction rates at room temperature were obtained from laboratory incubations with added substrate. Hydrochemical results are used to calculate saturation indices for selected minerals. The authors further use estimates for pyrite and CaMg carbonate (the authors call 'dolomite') contents to calculate so-called 'burial/accumulation' rates for these two minerals. Not Rieveld evaluation of the powder XRD was carried out, what makes a proper quantification difficult. CNS data are apparently given for decarbonatized samples, no TIC data were analyzed. The data are then used to state that low-T dolomitization is an important process for carbon sequestration in Baltic Sea sediments.
Answer: Mineral composition was quantified by Rietveld full-pattern X-ray diffraction fitting technique using SIROQUANT analysis of randomly oriented powders (for more details please see also an answer on comment L190). A relevant information has been added to the Methods section (lines 249-250).
The CNS data are presented in this paper for decarbonated samples because they were used in the context of identifying the origin of organic matter. Please see our explanation below (L93, L142 and L145).

A such, the data set is original and add on the existing data base for the Baltic Sea. As outlined below, however, the manuscript contains many assumptions that lead to a substantial overinterpretation of the actual results. Many data that are essential in evaluating the actual results are missing. The authors state without analytical proove, that a carbonate with a pressumable Ca:Mg stoichiometry of 1 is dolomite and that it is formed by authigenesis. As correctly mentioned in the manuscript, no ordering reflections where able to be identified due to the low mineral contents. So claiming that 'authigenic dolomite' is found is a possible overinterpretation of the actual observations, considering the known problems to

form dolomite at low temperature. Claiming both, the cationic ordering state and the formation process requiers independent proove by other methods. It has been shown previously in a not-cited study (Jakobsen & Postma, GCA, 1989) that also detrital dolomite from erosional fluxes from land is found in the Baltic Sea.

Answer: Indeed, we did not detect the presence of ordering peaks due to low carbonate contents in the sediments. Therefore, taking into account this Reviewer's concern, we expanded the discussion on this topic (see also our reply to the comment to line 328 below).

Jakobsen and Postma (1989) indeed reported dolomite crystals (or grains) in the centers of rhodochrosite aggregates in the Baltic Sea, suggesting that rhodochrosite had nucleated upon preexisting detrital dolomite (its detrital origin was inferred based on fractured appearance). This is however probably not our case because many Ca-Mg carbonate crystals found in the sediments were perfectly euhedral (see e.g. Fig. 4 e, f) and did not reveal any fractures or any transport-related features.

It should also be emphasized that our manuscript is not a mineralogical work, but rather primarily concerns the biogeochemical cycling of carbon in methanogenic sediments. From this perspective, the most important point is that carbon in these sediments can be bound in authigenic Ca-Mg carbonates, and whether these carbonates are dolomite, protodolomite, or high-magnesium calcite seems to be of secondary importance. Nevertheless, in the revised version of the manuscript we present the results of Raman microspectroscopic (Figure S9 and text in lines 424-434) analysis supporting the hypothesis that the observed Ca-Mg carbonate is dolomite.

Further detailed comments:

L39: have these references observed this for the first time?

Answer: We cited these works because they clarify the mechanisms and implications for coastal carbon budgets and provide a concise, interdisciplinary synthesis showing that coastal oceans are highly active interfaces for carbon exchange - integrating terrestrial inputs, in situ processing, and human impacts - making them valuable, up-to-date overviews of how coasts modulate the global carbon cycle.

We have cited additional works (line 48):

Berner, R.A., 1982. Burial of organic carbon and pyrite sulfur in the modern ocean: Its geochemical and environmental significance. Am. J. Sci., 282: 451-473.

Hedges JI, Keil RG. 1995. Sedimentary organic-matter preservation—an assessment and speculative synthesis. Mar. Chem. 49:81–115.

Wollast, R., 1998. Evaluation and comparison of the global carbon cycle in the coastal zone and in the open ocean. In: Brink, K.H., Robinson, A.R. (Eds.), The Sea. John Wiley, New York, pp. 213–252.

The latter two (Hedges & Keil 1995 and Wollast 1998) have been added to the reference list.

L40: Why calling this a trendy 'dead zone' when microbes are rather active?

Answer: We replaced the term “dead zone” with “hypoxia and anoxia” and implemented the following wording (lines 51-52): "Degradation of the increased OM associated with eutrophication rapidly consumes oxygen and promotes the expansion of bottom water hypoxia and anoxia (Rabalais et al., 2010; Fennel and Testa, 2019)."

L93: No TIC was analyzed

Answer: Contents of C and N, as well as their isotopic composition, were used to identify the origin of the organic matter; TIC was not informative for this purpose and thus was not discussed in this study. We performed thermal analysis (450°C for OC and 950°C for IC) on samples from summer 2023, obtaining IC values from 3.0 to 12.2%, with the highest values (5.2–12.2%) at MET1-MP and ≤6.4% at the other two sites. However, we chose not to include these data because the thermal decomposition ranges of OC and IC in sediment overlap, so OC and IC cannot be accurately quantified by difference

after combustion at different temperatures (Froelich, 1980). Additionally, a complex sediment matrix — specifically the presence of sulfides and various clay minerals — means that determining the proportions of OC and IC in the sediment by thermal methods is subject to a large error. We are working on a more accurate quantification of IC in the studied sediments and plan to combine this with isotopic analysis of the inorganic C fraction to better characterize carbonate dynamics in future studies.

Froelich P.N., 1980. Analysis of organic carbon in marine sediments. Limnol. Oceanogr. 25, 564-572.

L110: has the rim being discared?
Answer: Yes. To reduce risk of sediment contamination and sample alteration, due to e.g. mechanical smearing, oxidation, contamination from liner, we discarded the rim and collected samples from the central part of the core. This information has been added in the Methods section (line 159-160).

L127: Which d13C values were assigned to the standards used?
Answer: The values of d13C for NBS-19 was +1.95‰, and for NBS-18 was -5.014‰. This information has been added in lines 180-181.

L135: Has the pH been measured in the pre water? Show the data. How have rhizon-typical phenomena like degassing of CO2 and H2S been considered in a correction of the pH?
Answer: Thank you for pointing this out. Yes, pH was measured and the data are presented in Fig. S2 in the Supplementary Material; however, the data were not described in the text. The following information has been added in the revised manuscript (lines 344-345): "The pH at all sampling sites was from 7.00 to 8.31, with an average of 7.57±0.21 at station MET1-MP, 7.66±0.26 at station MET2, and 7.57±0.20 at station ZGG (Fig. S2)."

pH values were not corrected for possible Rhizon-related degassing because we do not have sufficient data to perform a reliable correction - the only samples which were collected using various methods are bottom-water samples (pore water sampling was performed using Rhizon samplers, we explain why below). We also noted that our mean pH (7.60 ± 0.22) falls within reported regional ranges — for example, Carman and Rahm (1997) reported overlying-water pH 7.22–7.41 (7.28 ± 0.08) and pore-water pH 6.98–8.26 (7.70 ± 0.34) for six Baltic Proper stations, and Ehlert von Ahn (2024) reported pore-water pH 6.2–8.3 for the Gdańsk Basin — indicating that our measurements are consistent with previous studies and unlikely to result from sampling or analytical artefacts.

Additionally, we would like to point out that IAP values for individual minerals, for which pH and ORP were used in calculations, are auxiliary data in our study. We do not use them to infer mineral presence (we have direct mineralogical evidence instead) or stability. The IAP data are intended to illustrate differences between stations and variability of saturation with depth. If the degassing effect on pH and IAP mentioned by the Reviewer affected all samples, results from all stations and sediment layers would carry a similar bias; however, the observed inter-station and vertical IAP differences are too large to be explained by a sampling artefact, and moreover the same pattern is observed in both seasons.

Aware that partial degassing can affect the carbonate system in pore water, in one of our previous studies we attempted to estimate its effect on the IAP of various minerals. We performed additional PHREEQC simulations of saturation indices response to pH (Fig. 1). We calculated saturation indices at different pH values to the extent that they could change as a result of partial or complete degassing of CO_2 (data from 128 pore water samples collected from 3 sediment cores collected in the Gulf of Gdańsk area were used to determine the pH range). The shifts do not apparently alter our general conclusions and

relationships between samples — i.e., supersaturated phases in pore water remain supersaturated and undersaturated phases remain undersaturated. We believe that the potential effect of sampling using Rhizons is of rather minor importance and should not significantly affect the results of our study.

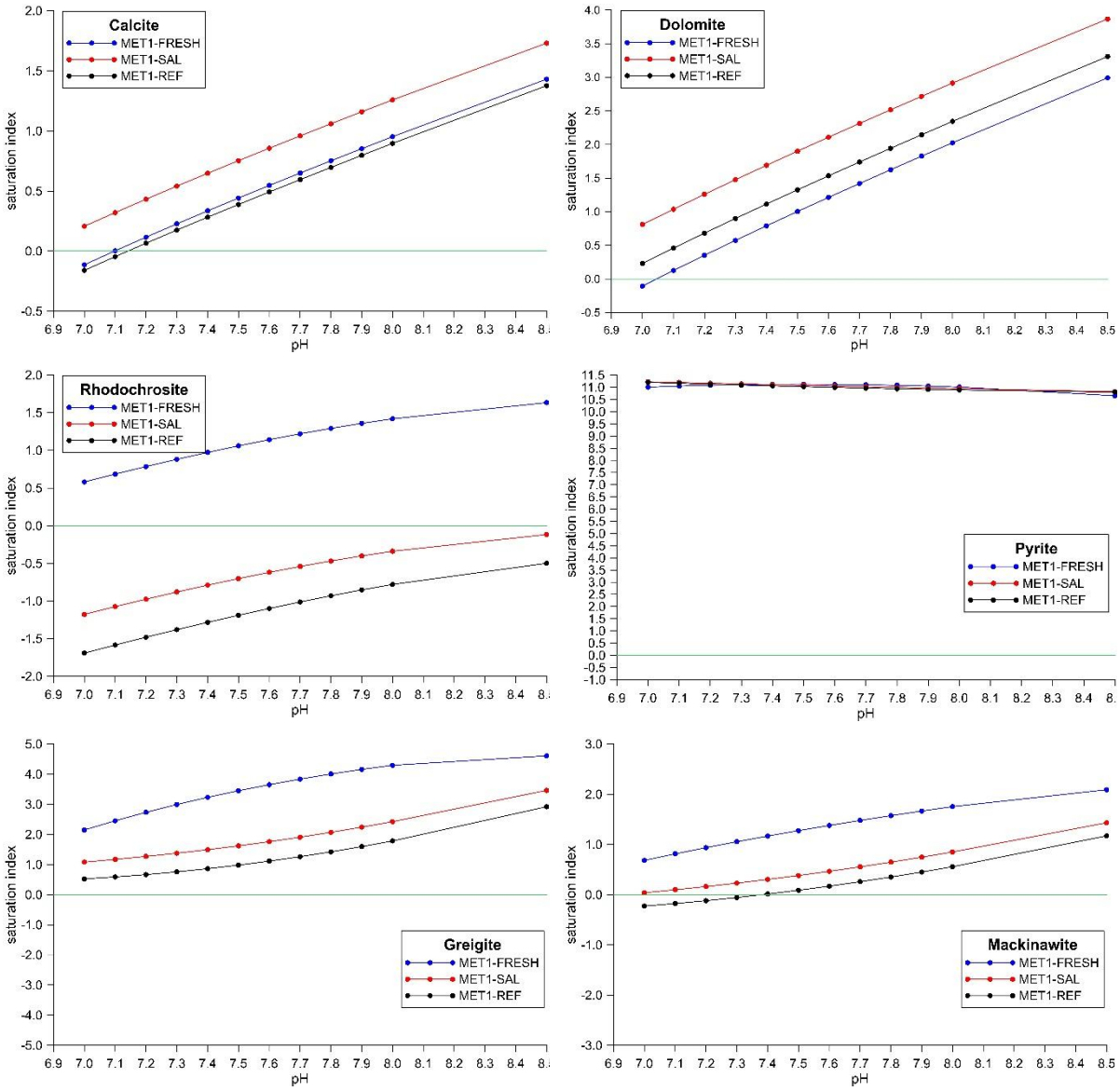

Fig. 1. Variations of saturation indices in pore water form 10-15 cm sediment layer at different pH values based on PHREEQC modeling. Green lines positioned at SI = 0 denote saturation (Rzepa et al., 2026, https://doi.org/10.5697/NDCH3019, in press).

In our view, each method of ex situ extraction of pore water from marine sediments - mechanical squeezing of a core section (Manheim, 1966), centrifugation (both requiring handling sediment samples prior to the pore water extraction), whole-core squeezing (Bender, 1987), suction through a microporous tube (Rhizon samplers) - has advantages and limitations. None of ex situ sampling methods completely avoids degassing effect, because the decompression itself during and after core recovery inevitably causes partial CO_2 loss. Decompression can shift the carbonate equilibria and increase CO_3^{2-} concentration promoting $CaCO_3$ precipitation and consequently lowering alkalinity and Ca concentrations (e.g., Murray et al., 1980). Therefore, simply retrieving cores on deck can cause the changes to the carbonate system mentioned by the Reviewer.

We would also like to point out that, although the effect of degassing during sampling with Rhizons has been observed and discussed in the literature, results remain ambiguous even within the same research programs. A good example is comparative study between Rhizon sampling and squeezing during IODP cruises (Backman et al., 2006; Schrum et al., 2012): Schrum et al. (2012) observed a relatively small and consistent decrease of alkalinity in samples collected using Rhizons compared to those collected by whole round squeezing. They suggested that the use of Rhizons resulted in CO_2 degassing, which lead to $CaCO_3$ precipitation and consequent lowering of DIC and alkalinity. These findings were inconsistent with the report of Backman et al. (2006) who found no significant difference in alkalinity between samples obtained by the two methods. In turn, Miller et al. (2017) proposed that the observed discrepancies depend on temperature and the time elapsed between core retrieval and completion of pore-water extraction, i.e., they are temperature- and pressure-dependent.

The selection of Rhizons as porewater samplers was motivated by the characteristics of the sediments studied, as well as a broad range of parameters to be analysed. We chose this technique primarily because Rhizons require no sediment handling (slicing, transferring, etc.) prior to pore-water extraction; allow minimally destructive, spatially resolved sampling from intact sediment cores; reduce physical disturbance compared with mechanical squeezing; avoid oxidation artefacts common during squeezing of sediment segments or centrifugation, as well as pressure changes associated with squeezing that can further alter solute equilibria; and are practical aboard ship for processing multiple cores and discrete depths.

The photo shows sediment from the deep-water part of the Gdańsk Basin where we conducted our study. It is not the exact station used in this paper but a nearby pockmark, MET1-BH (approximately 2 NM from the MET1-MP pockmark described here), located at the same depth (seabed depth around both pockmarks is 78 m). There are fragments of foil and vascular plant debris visible in the photo, transported into this area by currents and deposited in MET1-BH, which is relatively deep (~10 m of relative depth). We never observed such debris at MET1-MP, which is much shallower (~2 m), although the sediment itself is similar at both sites - reducing, very soft, with high water content. This is the main reason we chose Rhizon samplers for pore-water sampling. We aimed to collect samples quickly with minimal disturbance of the core to avoid gas migration. Importantly, using Rhizons allowed us for pore-water collection without exposure to air, which is crucial for most of the parameters we measured.

Image of mud from MET1-BH pockmark sampled with Van Veen grab (Foto: Olga Brocławik)

Besides parameters presented in this paper (DIC and δ^{13}C-DIC, pH, alkalinity, sulfate, chloride, calcium, magnesium, hydrogen sulfide, ammonia, phosphate, manganese, and iron), we also analyzed silicate, nitrite, aluminum, sodium, potassium, bromide, and dissolved organic carbon. For most of these parameters, Rhizons are advantageous. Oxidation artifacts can be expected for some analytes (Fe, Mn, H_2S, SO_4^{2-}, TA, phosphate, ammonia, nitrite, silicate, ORP; Bray et al., 1973; Troup et al., 1974; de Lange et al., 1992), so we avoided opening the core to minimize O_2 influx until the required sample volume was obtained (usually a several ml from each layer).

Rhizons are also advantageous for nutrients because pressure-based recovery techniques (squeezing) cause bacterial cell lysis and release of these substances into the pore water. Squeezing also alters pore-water chemistry via solid-solution reactions (adsorption/desorption and ion exchange), which

affects concentrations of major cations, ammonia, and phosphate (Rosenfeld, 1979; Santschi et al., 1984; Böttcher et al., 1997), as well as particle-reactive trace metals (Santschi et al., 1984; Bender et al., 1987). By contrast, measurements of most of these parameters (including pH) from Rhizon samples have yielded good results in other studies (e.g., Spangenberg et al., 1997; Song et al., 2003; Seeberg-Elverfeldt et al., 2005).
The above considerations guided our choice of Rhizons as a compromise to preserve a broad suite of analytes while minimizing core disturbance.

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L142: The samples were only dried at 40-50°C, so the samples were not completely water-free. How to refer then the analytical results to the necessary dry weight %?
and
L145: Data are given in 'wt.%'? But according to the analytical protocoll, CNS was only measured in decarbonatized samples. Therefore, they do not refer to the original bulk sediment(?)
Answer: Analysis of C and N content was performed according to Hedges and Stern (1984), who described carbon and nitrogen determinations of carbonate-containing solids; however, the procedure was not described in detail and citation was missing (it has been added in line 213). Low-temperature drying of organic matter (up to 60°C) before C and N isotopic analysis is a standard procedure (e.g. Hedges and Stern, 1984) to avoid degradation of organic matter. We chose a slightly lower temperature and extended the drying time to ensure that we removed water as much as possible and our samples were still suitable for isotopic analysis. From an analytical point of view, even if small amounts of H_2O remain in the sediment sample, the error in the wt. % C, N, and S determined using an elemental analyzer will be irrelevant and moreover, it does not affect the TOC/TN and TOC/TS ratios, which are crucial for our considerations.
In this study, we use data on the % C, N, and S content and isotopic data ($\delta^{13}C$, $\delta^{15}N$) in the organic fraction of sediments to determine its origin, and therefore these data refer to samples from which the carbonate fraction was removed.

Hedges J.I. , Stern J.H. , 1984. Carbon and nitrogen determinations of carbonate-containing solids, Limnology and Oceanography, 3, doi: 10.4319/lo.1984.29.3.0657

L161: For what reason LOI was analyzed (data are not shown)?
Answer: LOI was measured to calculate sediment porosity (required to correct methane concentrations). This is described in lines 193-198., and porosity profiles are provided in Fig. S2 (Supplement). LOI and water content (W) are reported in the Results (lines 306-309). We added LOI and W profiles to the Supplement (Fig. S2) and cited that figure in the Results (lines 306-307).

L173: What are the SEM-EDS and Mössbauer spectroscopy instruments used?
Answer: SEM analyses were performed using a FEI Quanta 200 FEG microscope equipped with EDS. ^{57}Fe Mössbauer spectroscopy measurements were performed using a RENON MsAa-4 spectrometer with an LND Kr-filled proportional detector and a $^{57}Co(Rh)$ source. The relevant section is in lines 256-258.

L182: Which pH was used for the modeling? The results in S9 look rather unusual for sediment pore waters. Extremely high supersaturations in some regions. How was the redox system quantified for dissolved FeIII/FeII which is needed to make an estimate for the stability of Fe(III) bearing solids?
Answer: The pH measured in the sediment pore water was used for modelling. pH in pore water was measured spectrophotometrically using m-cresol purple as indicator.
The results presented in Figure S9 (after revision S11) are the result of PHREEQC modelling (using the verified thermodynamic database wateq4f.dat) for analyses of the chemical composition of pore water collected in the profiles. Redox conditions were assumed based on pe calculated from the measured ORP data (presented in Figure S2 and line 305 in Results section). The pe values were then used for calculating dissolved Fe(II) and Fe(III) species. Additional information (thermodynamic database, pH used and redox system quantification) has been added in Methods section (lines 260-264).

L190: How to gain quantitative information about low mineral contents from powder XRD without using Rietveld? How many samples were analyzed in which way to make an estimate of the CaMg carbonate and pyrite contents from EDS scans?

Answer: We performed Rietveld fitting but this information was missing in the manuscript. It has been added in lines 249-250.

SEM–EDS elemental distribution maps were also used to independently assess the content of dolomite and pyrite. It was assumed that dolomite occurs in areas where high concentrations of Ca, Mg, C, and O overlap, while other major elements (e.g., Si, Al, etc.) are absent. Similarly, pyrite was present where high concentrations of Fe and S occurred in the absence of significant amounts of oxygen. An appropriate algorithm was written in Mathcad software. To verify the results, the obtained maps of the spatial distribution of carbonates and sulfides were overlaid on the corresponding SEM-BSE image. For each sample, two fragments of the sediment surface were analyzed.

L192: The are obviously 'potential' sulfate reduction rates, after dilution, addition of external substrate used by complete oxidizers, and at a temperature (20°C) exceeding in-situ conditions. Nothing is found in the discussion about the uncertainty of this approach and the inherent assumptions.

Answer: We agree that the reported rates represent potential sulphate reduction rates measured under controlled laboratory conditions (substrate amendment, dilution, and incubation at 20°C), rather than in situ rates. As such, a formal quantification of uncertainty in terms of environmental variability is not applicable. The purpose of these measurements is to provide a comparative assessment of microbial activity across the studied samples under standardized conditions. Sediment samples require controlled pre-treatment to ensure precise and reproducible substrate dosing, which is essential for obtaining internally consistent and comparable results.

This is clarified in the Methods (lines 273-275) and Results (line 376-379). We also mention this in the Discussion (lines 548-554): „It should be also mentioned that experimentally-derived AOM rates represent the potential activity of the microbial community under substrate-replete conditions rather than the actual in situ process rates. Therefore, the similar experimentally-derived rates at MET-2 and ZGG indicate a comparable microbial potential for AOM at both sites. In contrast, the porewater profiles reflect in situ conditions, where methane availability differs substantially between the sites. At MET-2, methane is present and supports active AOM, which is reflected in the porewater profiles. At ZGG, methane concentrations are negligible, so AOM appears less significant in situ despite the presence of a microbial community capable of performing this process if methane became available."

L328: Proto dolomite? Are there other elements in the CaMg carbonate lattice than Ca and Mg (e.g. (Mn or Fe)?

Answer: In some Ca-Mg carbonates there are trace Fe admixtures (too small to affect the position of the basal (104) peak in the diffraction patterns). Also, Mössbauer spectroscopy suggests the lack of significant Fe substitution in carbonates. According to the current recommendations of Lukoczki et al. (2026), the term 'protodolomite' should be applied to a poorly ordered dolomite with a kind of cation ordering based on the presence of attenuated PXRD ordering peaks. We cannot therefore use this term because we are unable to observe any other dolomite peaks than the basal one. On the other hand, we cannot use the term VHMC (very high magnesium calcite) either, as it refers to a carbonate containing more than 30 mol% $MgCO_3$ with no evidence of cation ordering indicated by the absence of ordering reflections. We also supplemented the manuscript with the results of Raman microspectroscopy, which apparently exclude high-magnesium calcite and indicate dolomite rather than protodolomite. Therefore, despite the lack of unambiguous PXRD evidence, the results (chemical composition, morphology, and Raman spectra) suggest that the dominant carbonate in the sediments is dolomite. Consequently we decided to retain the term dolomite for relevant carbonates throughout the manuscript.

Lukoczki G., Bish D., Gregg J.M., 2026. A best-practices guide to X-ray diffraction studies of sedimentary carbonates. Sedimentary Geology 493, 107028.

L340: In Fig.4h no calcite is marked (the text only states pyrite, 'dolomite' and diatom
Answer: You are right. We noticed that there are a few mistakes here and hence the entire caption was corrected as follows (lines 446-450): Examples of SEM-BSE images of authigenic sulfides and carbonates: (a) pyrite framboids, MET2 (40–45 cm); (b) pyrite framboid and disseminated pyrite crystals, ZGG (10–15 cm); (c) pyrite framboids within a diatom shell, ZGG (40–45 cm); (d) pyrite framboid, diatom, dolomite, MET1-MP (40–45 cm: thin section); (e) euhedral and subhedral dolomite crystals, MET1-MP (10–15 cm; thin section); (f) euhedral dolomite, MET1-MP (10–15 cm; thin section); (g) dolomite, MET1-MP (40–45 cm); (h) calcite surrounded by clay minerals, ZGG (40–45 cm); (i) siderite, MET2 (40–45 cm).

L375-392: Instead of comparing the d13C and d15N data of OM with very general sources, the data should be compared with extensive measurements done by the Struck and Voss laboratories.
Answer: We extended the discussion section as follows (lines 498-503): Increased biomass development may also lead to a modification of the isotopic composition of both nitrogen and carbon in sediments, the effect of which may also be visible in the case described. For example, a decrease in the $\delta^{15}N$ value of OM can be caused by the addition of isotopically light nitrogen to the N pool, bound by diazotrophic cyanobacteria, as in the case described by Struck et al. (2000) for the Gotland Basin. In turn, increased primary production can lead to an increase in ^{13}C content in sediment organic matter. For example, in the Oder Haff an increase of $\delta^{13}C$ in OM of 1.7 ‰ was found, while in the Gotland Basin it was 2.6 ‰ (Struck et al. 2000).

Struck, U., Emeis, K. C., Voss, M., Christiansen, C., & Kunzendorf, H. Records of southern and central Baltic Sea eutrophication in $\delta^{13}C$ and $\delta^{15}N$ of sedimentary organic matter. Mar. Geol., 164, 157-171, 2000.

L495-497: Why stating that the pore water profiles may be indicative for Fe or Mn-AOM (in 30-60 cm below seafloor), when two lines later this assumpiton is taken back?
Answer: We decided to retain this paragraph to provide a complete description of AOM processes. We aimed to discuss the results as comprehensively as possible. We acknowledged that changes in Mn^{2+} and Fe^{2+} concentrations below the sulfate reduction zone may have resulted from Mn(IV)- or Fe(III)-driven AOM and that these processes may have constituted an additional source of DIC; however, based on the Mn^{2+} and Fe^{2+} concentrations, their contribution was likely minor, which partly explains why we did not include them in the quantitative DIC budget. In our opinion, this aspect is worth highlighting, especially given the lack of such data for the studied sediments. Considering the characteristics of the area (low salinity, along with the local influence of groundwater discharge), this represents an interesting and important direction for future research on the methane cycle.
This keeps the manuscript consistent with Reviewer 1's suggestion — since we referred to denitrification when discussing organic-matter degradation (this is described in lines 560-564), we likewise wished to discuss the range of possible AOM pathways and the potential for their occurrence in the studied sediments.

L618: Are the given decimals significant?
Answer: We rounded the microbial process rates and the DIC production rates calculated from them to whole numbers (Table 1 and text).

L627: What is ment with 'similar radiotracer methods'? The whole-core incubation using 35SO42- is clearly not similar to the approach applied in this study
Answer: We clarified the sentence to state that we referred to methods in general (not any single specific protocol); the text now makes explicit that even broadly similar methodological approaches can yield substantially different rates due to microbiological and sedimentary variability. This change was implemented in the manuscript in lines 770-772.
We applied the radiotracer to discrete sediment samples because our sediments are methane-rich, and all sampling had to be done immediately after pulling the cores onto deck to avoid gas expansion and sediment disturbance; thus a prolonged whole-core incubations are impractical.

L684: How is the formation process of this carbonate phase ('authigenesis') prooven?
Answer: The authigenic nature is suggested by SEM observations showing euhedral, automorphic rather than detrital morphology of well-formed carbonate crystals (a relevant explanation has been added in lines 424-439). Moreover, other results also indicate that sediments with intense diagenesis, impacted by freshwater input with high rates of DIC production from various microbiological processes are places where the formation of authigenic carbonates is favoured by specific physicochemical conditions (see the discussion in the manuscript).

S9: What was the base for calculating the saturation indices? How was the pH estimated? How was the FeII/III distribution in the solution calculated? The results for greigite are completely different to the those obtained by Kulik et al. (2000; Aqu. Geochem.) Why are these discrepencies not discussed?
Answer: Saturation indices were calculated using PHREEQC software based on chemical composition of pore water collected in individual sediment profiles (a verified thermodynamic database wateq4f.dat of Parkhurst and Appelo (2013) was applied). The measured pH values (see Fig. S2) were used for modelling. The Fe(II)/Fe(III) distribution was calculated based on the calculated pe.
The supersaturation values we obtained for greigite may indeed differ significantly from those obtained by Kulik (2000). This is due to the very high solubility product value assumed in that paper, many times higher than that found in the wateq4f database. Turney et al., 2023 (Greigite formation in aqueous solutions: Critical constraints into the role of iron and sulfur ratios, pH and Eh, and temperature using reaction pathway modeling, Chem Geol 635) pointed to the possibility of a wide range of logK for greigite depending on the conditions of a given system (and the logK value adopted in the wateq4f.dat database is consistent with the parameters given there).

Parkhust, D.L., Appelo, C.A.J.: Description of Input and Examples for PHREEQC Version 3—A Computer Program for Speciation, Batch-Reaction, One-Dimensional Transport, and Inverse Geochemical Calculations. US Geological Survey Techniques and Methods, Book 6, Chapter A43, 497 p. http://pubs.usgs.gov/tm/06/a43, 2013.