Response to referee comment Anonymous Referee 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.

We thank the Reviewer for carefully evaluating our manuscript and for the helpful comments and suggestions about our work. We will do our best to revise the manuscript accordingly. Below we respond to each point and outline the specific revisions planned.

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 will be added to the Methods section.
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 will expand 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.

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.
Other references that we propose to add:
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.

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

Answer: We will rephrase the sentence. We will use “hypoxia and anoxia” in this context, e.g.: 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: As noted above, CNS 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. We will add this information in the Methods section.

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‰. Standard LSVEC was included by mistake and was not used.

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. We plan to add the following information to section 3.2 in the Results: "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)." The values we obtained (on average 7.60±0.22) are close to literature data; for example, Carman and Rahm (1997), for six stations in the accumulation bottom of the Baltic Proper, reported pH values for overlying water from 7.22 to 7.41 (7.28±0.08) and for pore waters in the range 6.98–8.26 (7.70±0.34). Data for the Gdańsk Basin can be found, for example, in Ehlert von Ahn (2024); the authors report pore-water pH ranging from 6.2 to 8.3.

We are aware that suction through a microporous tube inserted into the sediment (Rhizon samplers) can lead to partial CO_2 degassing and drive pH changes and carbonate equilibrium shifts; however, we believe that Rhizon samplers represent a reasonable compromise for our work. Below we explain why. 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.

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 δ13C-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.

References:

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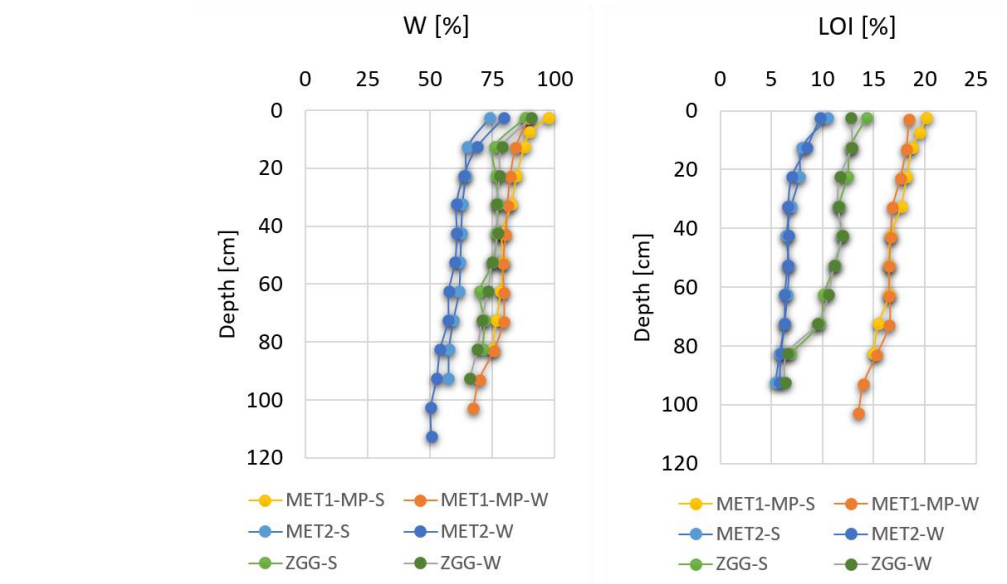
L142: The samples were only dried at 40-50°C, so the samples were not completely water-free. How to refer then the analytical results to the necessary dry weight %?
and
L145: Data are given in 'wt.%'? But according to the analytical protocoll, CNS was only measured in decarbonatized samples. Therefore, 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. 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. We can add this information in the text, along with the relevant references.
In this study, we use data on the % C, N, and S content and isotopic data (δ13C, δ15N) 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 (needed for correction of methane concentration). We describe this in lines 156–160, and porosity profiles are shown in Fig. S2 (Supplement). We considered presenting the LOI values unnecessary because organic carbon concentration is reported and serves as the measure of organic matter content; however, we will add LOI and water content profiles to the Supplement.



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 178–181.

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 the sediment pore water was used for modelling (see Figure S2). pH in pore water was measured spectrophotometrically using m-cresol purple as indicator (line 139). The results presented in S9 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 (see Figure S2). The pe values were then used for calculating dissolved Fe(II) and Fe(III) species.

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 will be corrected.
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.
The statement that these are potential rates appears in the Results (line 285) and the Discussion (lines 423–429); we propose clarifying in line 423 that these are potential rates: “Incubation-derived potential R_{OSR} values…”.

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

Answer: In some ‘dolomites’ 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. Therefore, we will supplement the manuscript with a more precise explanation of this issue, while retaining the term dolomite for relevant carbonates in the text.

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 will be corrected as follows: 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 compairng the d13C and d15N data of OM with very general sources, the data should be compared with extensive measuremements done by the Struck and Voss laboratories.

Answer: Thank you, we will extend the discussion section as suggested by comparing our data with those obtained in the Struck and Voss studies in the Baltic Sea area, for example Struck, U., Emeis, K. C., Voss, M., Christiansen, C., & Kunzendorf, H. (2000). Records of southern and central Baltic Sea eutrophication in δ C-13 and δ N-15 of sedimentary organic matter. Mar. Geol., 164, 157-171.

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: In this paragraph 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.

L618: Are the given decimals significant?

Answer: We will evaluate decimal values for significance and correct values of rates of microbiological processes if necessary.

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 will clarify that we meant incubations of discrete sediment slurry samples, not whole-core incubation techniques. We applied the radiotracer to discrete sediment samples (lines 105–107) 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. Moreover, other results also indicate that methane sediments 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 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 sulphur 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).