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Aerobic and anaerobic mineralisation of sediment organic matter in the tidal River Elbe

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Abstract

Purpose The share of microbially degradable sediment organic matter (SOM) and the degradation rate depend, among others, on the intrinsic properties of SOM as well as on the type and concentration of terminal electron acceptors (TEA). Next to its role as TEA, molecular oxygen enhances SOM decay by oxygenase-mediated breakdown of complex organic molecules. This research investigated long-term SOM decay (> 250 days) under aerobic and anaerobic conditions to (1) provide a basis for sediment carbon flux estimates from the River Elbe estuary and (2) assess the potential for carbon burial in relation to redox conditions and dredging interventions.

Methods Long-term aerobic and anaerobic SOM decay in fluid mud, pre-consolidated and consolidated sediment layers was investigated over three years along a transect of ca. 20 km through the Port of Hamburg, starting at the first hydrodynamically determined hotspot of sedimentation after the weir in Geesthacht. Absolute differences between aerobic and anaerobic cumulative carbon mineralization were calculated, as well as their ratio. Findings were correlated to a suite of solids and pore water properties.

Results SOM decay followed first order multi-phase exponential decay kinetics. The ratio between C release under aerobic and anaerobic conditions ranged around 4 in the short-term, converging to a value of 2 in the long term. Strong gradients in absolute C release along the upstream–downstream transect did not reflect in a corresponding gradient of the aerobic–anaerobic ratio. C release was most strongly correlated to the water-soluble organic matter, in particular humic acids. Contact of anaerobically stabilized sediment with the oxygenated water phase induced significant release of carbon.

Conclusion SOM degradability in the study area exhibited strong spatial gradients in relation to the organic matter source gradient but was mainly limited by the high extent of organic matter stabilization. Under these conditions, molecular oxygen as TEA provides little thermodynamic advantage. Carbon-sensitive sediment management, considering SOM reactivity patterns in stratified depositional areas, is a powerful strategy to reduce environmental impacts of dredging measures.

Keywords Organic matter degradability · Sediment geochemistry · Oxygen consumption · Carbon-sensitive dredging

1 Introduction

The Port of Hamburg, Germany, located within the tidally influenced section of the lower Elbe River, has been identified as a hotspot of microbial metabolism (Norbisrath et al.

2022). Microbial degradation of sediment organic matter (SOM) follows either aerobic or anaerobic metabolic pathways, depending on the presence and type of terminal electron acceptors. The predominantly fine-grained sediments in the investigated sedimentation areas show high oxygen consumption rates, inducing oxygen-depleted zones and hence negative redox potentials conditions just a few mm below the sediment–water interface (Zander et al. 2020; Spieckermann et al. 2021). Under these conditions, the potent greenhouse gas methane (CH₄) is released as terminal product of carbon mineralization in addition to carbon dioxide (CO₂). When molecular oxygen is available as terminal electron acceptor, SOM decay provides a higher energy yield and enables the activity of oxygen-dependent enzymes (such as oxygenases)

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for SOM breakdown. These conditions have therefore been assumed to allow for higher degradation rates and a higher share of degradable OM (Bastviken et al. 2003). On the other hand, it has been found that degradation under oxic conditions was only faster for older, refractory OM, but provided no advantage over anoxic conditions in the decay of fresh, labile OM (Westrich and Berner 1984; Henrichs 2005).

Dredging and relocation of sediment can bring anoxic sediment layers in contact with the oxygenated water phase. The ensuing chemical and biological oxidation of reduced pore water components such as Fe^{2+} , Mn^{2+} , NH_4^+ , of solid phase organic matter, or of the metabolic products released by the previous anaerobic decay, can contribute to the development of oxygen minimum zones or aggravate those that develop naturally from die-off and decay of riverine phytoplankton (Geerts et al. 2017). Man-made low-flow areas (such as harbor basins or groin fields) have been shown to contribute to SOM burial and hence C sequestration (Dean and Gorham 1998; Mendonça et al. 2014) but can also function hot-spots of CH_4 fluxes (Delsontro et al. 2010; Jacinthe et al. 2012; Maeck et al. 2014; reviewed in Ward et al. 2017). CH_4 emissions from freshwaters represent an under-researched carbon flux that could play an important role in the revision of continental greenhouse gas fluxes (Bastviken et al. 2011).

The purpose of this study was to: (i) quantify differences between aerobic and anaerobic SOM decay in space and over time; and to (ii) investigate the susceptibility of SOM decay to changing redox conditions. To this end, long-term (> 500 days) cumulative C release during aerobic and anaerobic incubations of > 200 samples collected on a transect through the port of Hamburg were compared regarding their temporal dynamics, and the ratio of and absolute differences between aerobic and anaerobic C release. It was hypothesized that SOM decay in sediment containing a higher share easily degradable organic matter, originating from planktonic biomass entering the investigated river section from upstream (Mendonça et al. 2012; Ward et al. 2016; Zander et al. 2022), is more similar under anaerobic and aerobic conditions, while towards downstream locations and increasing recalcitrance, SOM degradability under aerobic conditions would clearly exceed degradability under anaerobic conditions.

In this paper, carbon release by microbial decomposition of SOM is equated with carbon mineralization, degradability is assessed by the mass of carbon released by microbial degradation normalized to the concentration of total organic carbon (TOC), aerobic decay of SOM is equated with the availability of molecular oxygen, and anaerobic decay of SOM is related to the absence of molecular oxygen.

2 Methods

2.1 Investigation area and sampling approach

Between 2018 and 2020, sediment samples were collected from nine locations along an upstream–downstream gradient through the Port of Hamburg, Germany (Fig. 1), using a 1-m core sampler ('Frahmlot'). On board, the core was divided into three layers based on differences in visual consistency and strength: fluid mud (FM), pre-consolidated sediment (PS), and consolidated sediment (CS), from top to bottom.

2.2 Sediment properties

The analysis of solids properties included, amongst others, total nitrogen (TN, DIN EN 16168 2012), total organic carbon (TOC, both DIN ISO 10694 1995), water content (WC, DIN ISO 11465 1993), redox potential (Eh, DIN 38404-6 1984), particle size distribution (DIN ISO 11277 2009), pH-value (pH, DIN EN 15933 2012), electrical conductivity (EC, DIN EN 27888 1993), in the filtrated pore water ammonium (NH_4^+ , DIN ISO 11732 2005) and silicic acid (as SiO_2 , DIN 38405-D21:1990–10 1990).

Organic matter fractions were analysed with an acid–base fractionation method that uses the aggregation/precipitation and dissolution properties of natural organic matter established by the International Humic Substances Society (IHSS) to determine acid–base-extractable DOM humic acid (HA), fulvic acid (FA), hydrophobic neutrals (HoN) and hydrophilic acids (Hi) as described in Van Zomeren and Comans (2007), Straathof et al. (2014) and Zander et al. (2023).

The $\delta^{13}\text{C}$ -values of the sediment organic matter were determined on bulk fresh and incubated samples, using an isotope-ratio mass spectrometer (Delta V; Thermo Scientific, Dreieich, Germany) coupled to an elemental analyser (Flash 2000; Thermo Scientific). Prior to analysis, samples were treated with phosphoric acid (43%, 80 °C for 2 h) to release inorganic carbon. Values are expressed relative to Vienna Pee Dee Belemnite (VPDB) using the external standards IAEA IAEA-CH7 (-31.62 ‰ vs. VPDB) and IVA soil 33,802,153 (27.46 ‰ vs. VPDB).

2.3 Degradation of sediment organic matter

Long-term (> 500 days) release of CO_2 -C and CH_4 -C under aerobic and anaerobic conditions was analyzed under standardized conditions in the laboratory. The conditions (temperature) and the duration of the experiment were chosen to capture the total amount of degradable carbon as accurately as possible within the time available for the study. This required to carry out the measurements until

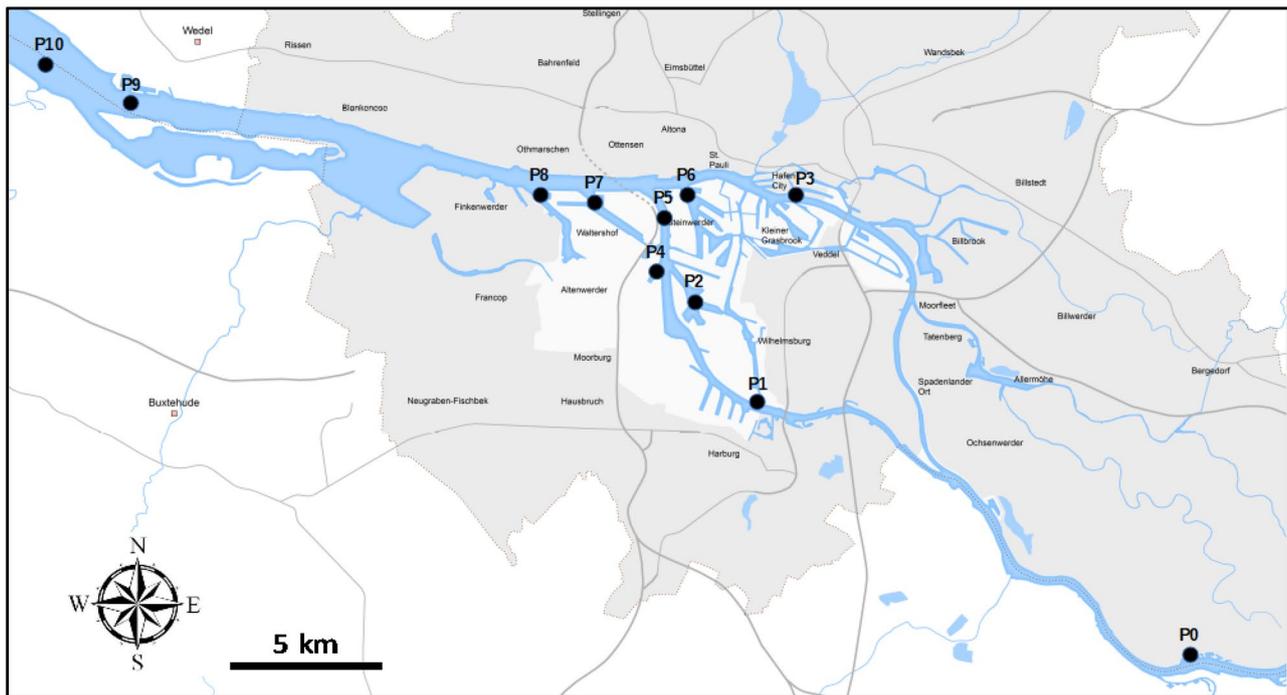


Fig. 1 Investigation area around the Port of Hamburg (adapted from Hamburg Port Authority) with sampling locations between river km 598 (P0, upstream) and 646 (P10, downstream)

the reaction rates were declining and hence cumulative carbon release was plateauing (compare Fig. 5), enabling the extrapolation of the final cumulative carbon release by multi-phase exponential curve fitting. In both the aerobic and the anaerobic setup a balance was made between (anticipated) sample reactivity, sample weight and volume of bottle as well as frequency of measurement to detect changes in headspace concentration.

For quantification of degradability under anaerobic conditions, approximately 200 g of freshly sampled sediment were placed into 500 ml glass bottles in triplicate, sealed with a butyl rubber stopper and secured with a screw cap. The bottle headspace was flushed with N_2 to establish anaerobic conditions and bottles were incubated at 36 °C in the dark. Anaerobic carbon release was calculated from the increase in headspace pressure in combination with gas chromatographic (Da Vinci Laboratory Solutions) analyses of headspace composition.

SOM degradability under aerobic conditions was quantified by placing around 15 g of sediment in 1000 ml glass bottles sealed with a butyl rubber stopper and incubated at 20 °C in the dark. Carbon was calculated from the increase in headspace CO_2 concentration over time. To avoid inhibition of respiratory activity by high CO_2 concentrations, the bottle headspace was flushed with air when concentrations exceeded 3 vol.%. Possible loss in moisture of the water-saturated sediment samples was monitored by regular

weighing of the bottles and replenishing water (if necessary at all) to restore the original moisture content.

In all cases, the share of CO_2 -C dissolved in the aqueous phase was calculated using the CO_2 concentration and the pressure in the bottle headspace as well as the temperature- and pH-corrected solubility of CO_2 in water (Carroll et al. 1995) as given by Henry's constant (Sander 2015). In order to compare C release by anaerobic and aerobic degradation, data from the anaerobic incubation were normalized to a temperature of 20 °C based on the known temperature dependency of both processes (Zander 2022).

To assess the effect of exposure to oxygenated conditions following anaerobic conditions, anaerobically incubated sediments where SOM decay had nearly terminated (after 700–900 days) were washed with original Elbe River water to elute accumulated pore water components and then either exposed to aerobic conditions or re-incubated under anaerobic conditions as described above.

3 Results

3.1 Selected sediment properties

Figure 2 highlights selected parameters of sediment and water column properties along the investigated transect. All sampling locations were characterized by a high share of the

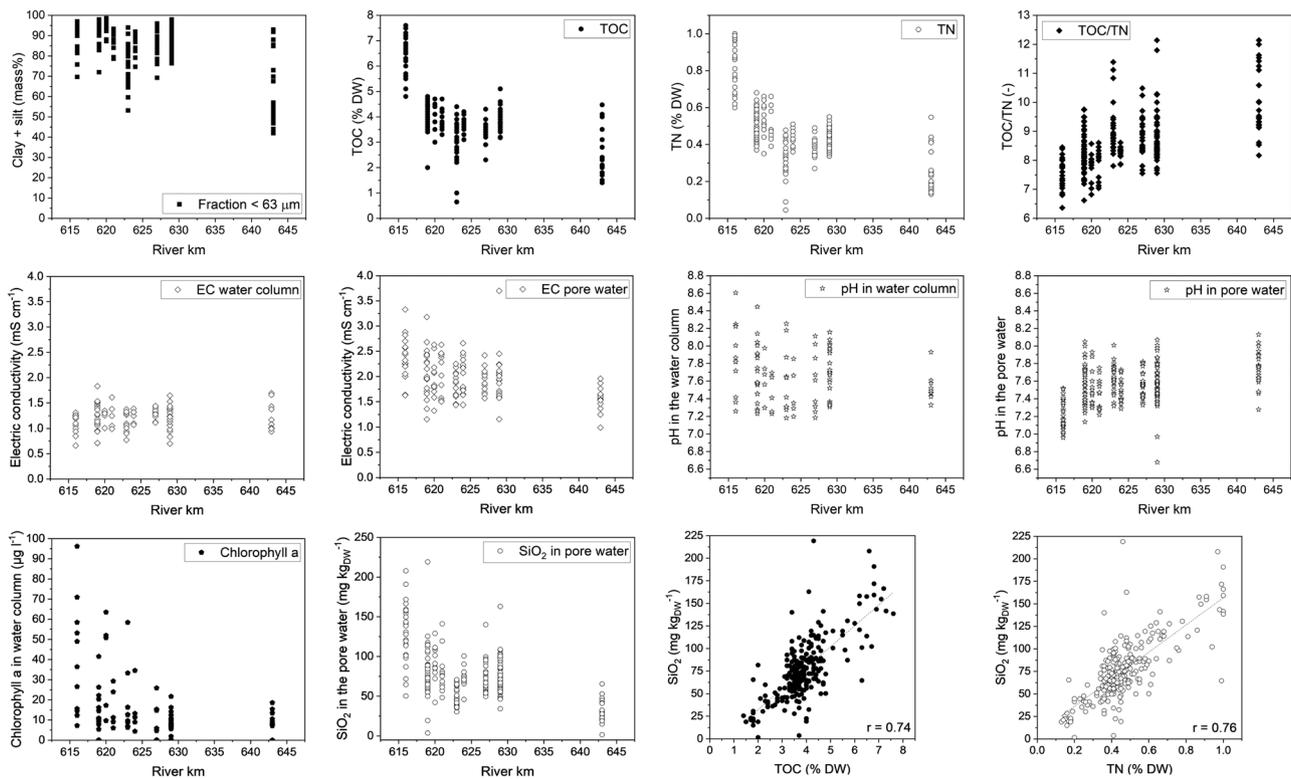


Fig. 2 Share of fines ($< 63 \mu\text{m}$), concentrations of total organic carbon (TOC), total nitrogen (TN) and ratio of TOC to TN (total nitrogen), electrical conductivity (EC), pH, chlorophyll a in the water col-

umn, and concentration of silicic acid (SiO_2) in the pore water along the investigated transect in fluid mud (FM), pre-consolidated (PS) and consolidated (CS) sediment layers

fine fraction ($< 63 \mu\text{m}$), only the most downstream site was more coarsely textured. The salinity of the water column, approximated by the parameter electrical conductivity, was more or less equal at all sites, whereas the electrical conductivity of the sediment pore water was clearly elevated at locations P1 to P8, especially at the most upstream location P1 (km 616). pH values of the water column decreased slightly in downstream direction but remained well above seven at all times. Pore water pH was lower, especially at location P1. A clear gradient was detected for total organic carbon (TOC), total nitrogen (TN) as well as silicic acid (given as SiO_2) in the pore water and chlorophyll a in the water column, all decreasing in downstream direction, whereas the ratio of TOC to TN was increasing. The concentration of silicic acid in the pore water was strongly positively correlated with TOC and TN. The concentration of total inorganic carbon (TIC) averaged between 1.2 and 1.0 for river kilometer 619 to 643, respectively, but was significantly higher at the most upstream location (km 616) with an average of 2.4% (data not shown), reflecting the spatial pattern also visible for the parameters TOC and TN (Fig. 2).

Redox potential upon sampling was negative for the majority of the samples and decreased with depth and hence age of sediments (Fig. 3, left). Concomitantly, pore water

ammonium concentrations increased with depth, reaching values of close to 400 mg l^{-1} in consolidated (CS) layers (Fig. 3, right). Pore water ammonium contributed an average of 1.2% and a maximum of 10.6% to the total concentration of nitrogen in the sediments, indicating that the largest part either resides within the (mineral-associated) organic matter and microbial biomass.

Fresh bulk density of all sediment layers was strongly and inversely related to TOC (Fig. 4) with each layer following a similar exponentially decaying trend of density with increasing TOC content. The respective level of bulk densities plausibly increased with depth, reflecting the increasing level of material consolidation and hence reducing water content.

3.2 Relationship between aerobic and anaerobic carbon release

Over time of experimental incubation, both cumulative aerobic and anaerobic carbon release increased non-linearly and eventually approached a plateau, due to progressively declining degradation rates (exemplary data given in Fig. 5 left panel; see also Zander et al. 2022). Using first order multi-phase exponential decay fitting (G model; Westrich and Berner 1984; reviewed in Arndt et al. 2013), up to three differently

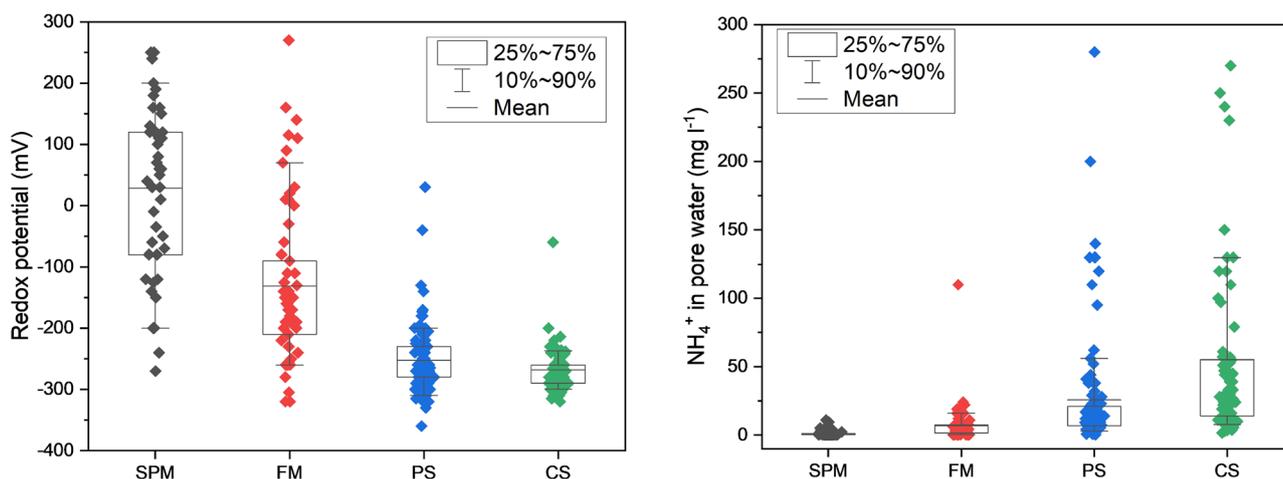


Fig. 3 Redox potential (left) and pore water ammonium (NH₄⁺) concentrations of SPM, FM, CS and PS layers, reflecting a depth and therefore age gradient from left to right. Updated data set from Zander et al. (2020)

degradable organic matter pools and the recalcitrant, non-degradable pool 4 were quantified. In the case of the example given in Fig. 5, around 150 mg C g_{TOC}⁻¹ were degradable under anaerobic conditions, hence 85% of organic matter were considered recalcitrant. More carbon was released when oxygen was available as terminal electron acceptor, as shown in Fig. 5 (right panel) for the cumulative carbon release over 21 days for 363 samples from suspended particulate matter, fluid mud, pre-consolidated and consolidated sediment layers.

The non-linearity of cumulative C release is also seen from the cumulative values aggregated for the benchmark

incubation time of 10, 21, 100 and 250 days (Fig. 6, left panel). When plotting the ratio between cumulative aerobic and anaerobic C release over time, it is seen that aerobic conditions enhanced SOM decay by an average factor of 4 in the short term. In the long-term, this factor progressively declined towards a value of 2 (Fig. 6, right panel), indicating that degradation rates converged to more similar values. Also, the range of the factor was highest in the beginning and approached a lower variability in the long term. However, at all benchmarks high ratios of > 6 up to factor 18 were observed.

3.2.1 Variability with sediment depth (age)

The benchmark value of 21 days was used to illustrate differences between aerobic (R21) and anaerobic (G21) decay over depth and therefore age of sediment. Both decline from the suspended particulate matter phase towards the underlying fluid mud phase and the settled pre-consolidated and consolidated sediment layers (Fig. 7, left panel), indicating progressively less available easily degradable organic matter as the sediment ages. The ratio between aerobic and anaerobic carbon release over 21 days did not vary with depth, averaging around the value of 4 for all layers/depths (Fig. 7, right panel).

3.2.2 Spatial variability

SOM decay followed a strong spatial trend with higher C release upstream and lower C release downstream, both when normalized to dry weight (Fig. 8, left panel) and to total organic carbon (TOC, Fig. 8, right panel). Higher values of the latter indicate increased lability of the organic matter present in upstream and increased stability in

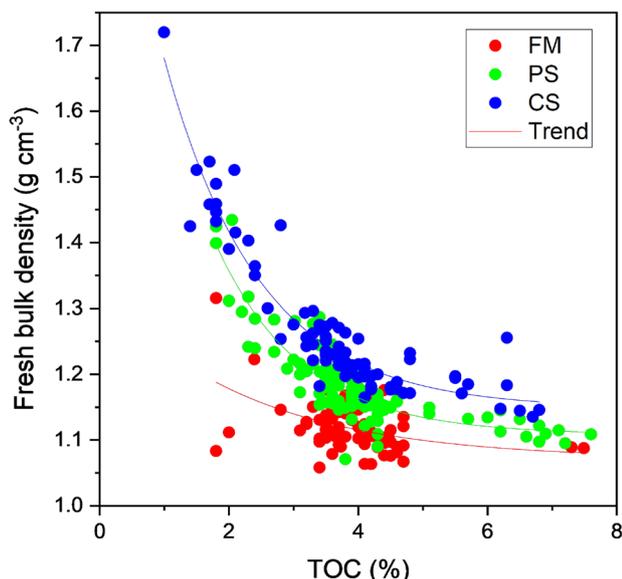


Fig. 4 Relationship between total organic carbon (TOC) and fresh bulk density

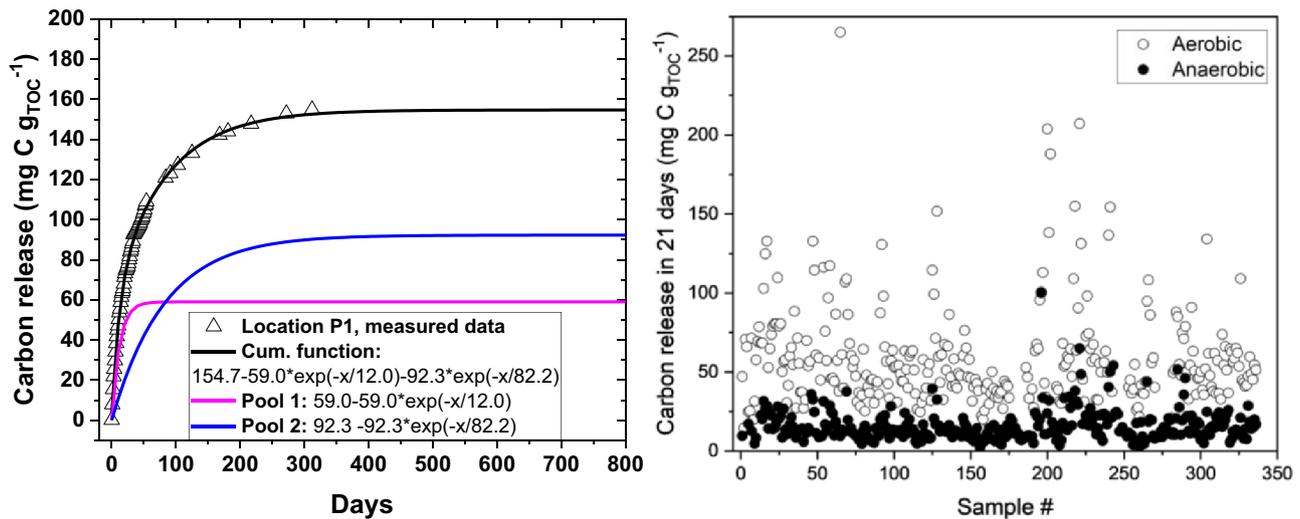


Fig. 5 Exemplary course of cumulative anaerobic carbon release (left), including a 2 phase exponential fit, and cumulative carbon release in 21 days under aerobic and anaerobic conditions (right).

downstream sediment. The ratio of aerobic to anaerobic carbon release (ratio of R21 to G21), however, revealed no clear spatial trend, although a minimum was observed at river km 624 with consistently increasing values in downstream direction (Fig. 9, left panel). To further elucidate the difference between aerobic and anaerobic C release in absolute terms, cumulative anaerobic C release in 21 days was subtracted from aerobic C release (Fig. 9, right panel). The largest differences were found on upstream site at river km 616 mostly owing to the high C release under aerobic

Data include SPM (suspended particulate matter), FM (fluid mud), PS (pre-consolidated sediment) and CS (consolidated sediment) layers

conditions. The lowest differences were clearly observed at the most downstream site.

3.2.3 Reactivation of organic matter degradation by aerobic conditions

In order to analyze effects of dredging and relocation of sediments and therefore the renewed contact with the oxygenated water column on SOM degradation, samples from pre-consolidated and consolidated layers that had reached

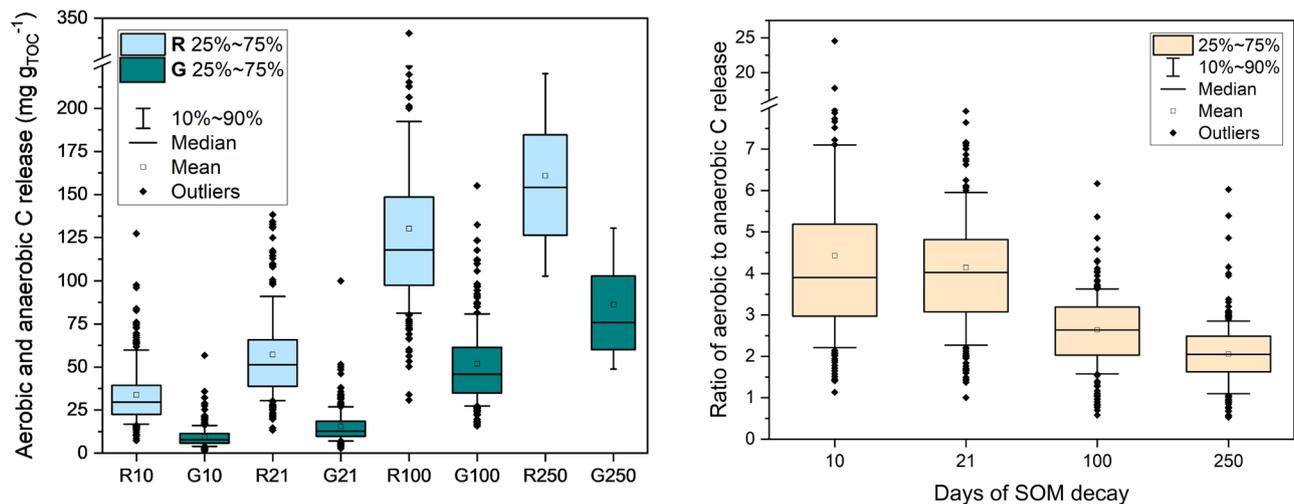


Fig. 6 Aerobic (R) and anaerobic (G) cumulative C release (left) and their ratio (right) over time of experiment (10, 21, 100, and 250 days). Data include layers FM (fluid mud), PS (pre-consolidated sediment) and CS (consolidated sediment)

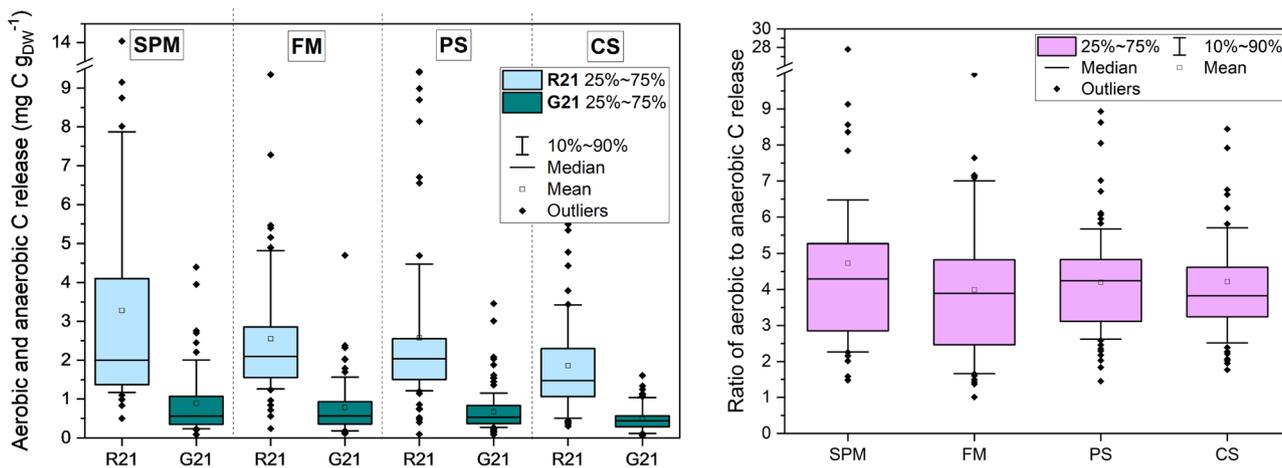


Fig. 7 Aerobic (R) and anaerobic (G) cumulative C release (left) and their ratio (right) in 21 days for sediment layers SPM (suspended particulate matter), FM (fluid mud), PS (pre-consolidated sediment) and CS (consolidated sediment)

the plateau phase during long-term anaerobic incubation were further incubated under aerobic conditions. This led to a swift and marked increase in C release in all samples (Fig. 10), which approached a new plateau during the ensuing around 200 days of aerobic incubation. Although the anaerobic decay had terminated mostly after about 700 to 900 days (seen from the flattened cumulative decay curve), a large carbon mobilization potential (between 33 and 42% of the anaerobically degraded carbon) was observed within 100 days after changeover from anaerobic to aerobic conditions. This means that long-term anaerobically degraded samples, an analogue to sediments buried under anaerobic conditions at the river bottom, still contain aerobically degradable SOM. At upstream location P1, the additional C release was larger than at downstream location P8; however, the relative

share was similar. In both PS layers, the absolute amount of aerobically decayed SOM was larger in the (upper) PS layer than in the (deeper) CS layer.

3.3 Correlation of SOM degradability with sediment properties

Carbon release under aerobic and anaerobic conditions over time of experiment, normalized to dry weight and to total organic carbon, was cross-correlated with all sediment parameters analysed in this study. Table 1 summarizes the most striking results for the cumulative long-term C release (after 250 days).

The long-term (250 days) mineralization of organic matter increased with increasing content of organic carbon and

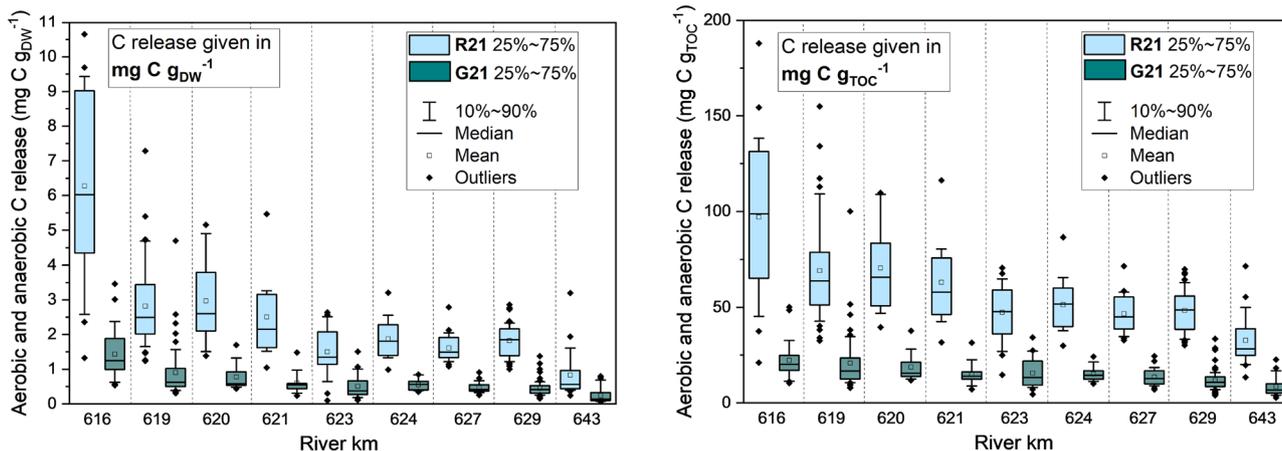


Fig. 8 Aerobic (R) and anaerobic (G) cumulative C release in 21 days along the sampled transect from river kilometer 616 (P1) to 643 (P9, see Fig. 1). Left=C released normalized to unit dry matter (DM).

Right=C release normalized to total organic carbon (TOC). Data include layers FM (fluid mud), PS (pre-consolidated sediment) and CS (consolidated sediment)

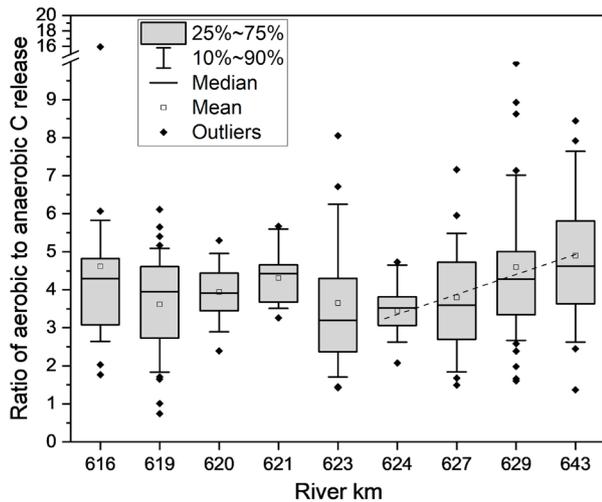
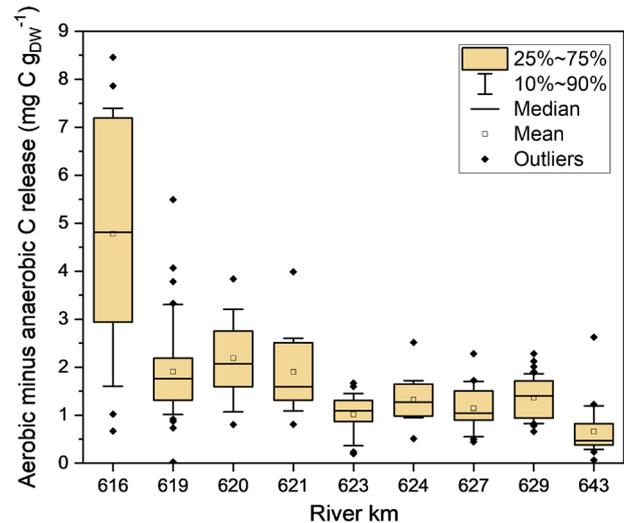


Fig. 9 Aerobic (R) and anaerobic (G) cumulative C release in 21 days along the sampled transect from river kilometer 616 (P1) to 643 (P9, see Fig. 1). Left=C release normalized to total organic carbon



(TOC). Right=absolute difference in C release between R and G. Data include layers FM (fluid mud), PS (pre-consolidated sediment) and CS (consolidated sediment)

nitrogen, an increasing share of organic carbon in relation to sulfur, an increasing share of nitrogen in relation to phosphorus, and an increasing amount of microbial carbon and silicic acid in the pore water. In general, the relation of the listed parameters was stronger for aerobic organic matter decay (respiration, R) than for anaerobic decay (gas production, G). A very strong positive relationship was found

with water-extractable SOM as well as its fractions (hydrophilics, humic and fulvic acids). The higher the share of water-extractable SOM, the higher also the absolute difference between the amount of carbon mineralized under aerobic conditions versus the amount mineralized under anaerobic conditions (R250-G250) and their ratio (R250/G250). C release was higher with an increased mass of sediment in the light density fraction LF ($< 1.4 \text{ g cm}^{-3}$) and consequently inversely related to the share of sediment in the heavy density fraction HF. The inverse relationship of SOM mineralization to the stable carbon isotopic signature $\delta^{13}\text{C}$ reflects a higher C release from organic matter with a higher share of ^{12}C . In general, the values for Pearson's r indicated a stronger relationship between the listed parameters and the absolute SOM mineralization, expressed per mass unit dry matter, than for the SOM degradability, here defined as C release per mass unit TOC.

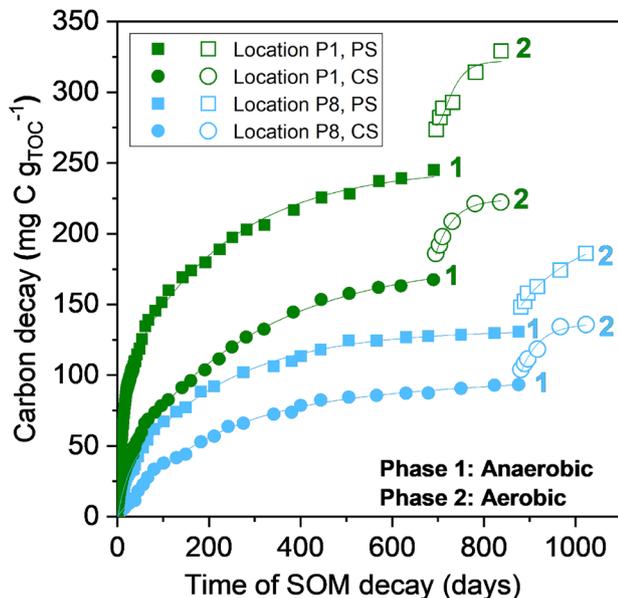


Fig. 10 Effect of changeover to aerobic conditions after long-term incubation under anaerobic conditions for pre-consolidated (PS) and consolidated (CS) sediment from upstream location P1 and downstream location P8 (see Fig. 1)

4 Discussion

The investigation area is characterized by a spatial gradient in nutrients as well as biological indicators such as chlorophyll *a* and SiO_2 in the water column (Fig. 2), the latter being strongly correlated to sediment TOC and TN. Elevated electrical conductivity in pore water compared to the water column evidences the release of solutes due to the low redox potentials (Fig. 3) and the accumulation of metabolic products from sediment microbial activity (see NH_4^+ in Fig. 3), especially seen for the most upstream point at river-kilometer 616. High rates of organic matter breakdown and hence

Table 1 Pearson's coefficients of linear correlation (r) for SOM degradable within 250 days (R = aerobic, G = anaerobic) with selected properties of sediments (layers FM, PS, CS) and the water column. All values significant on a level of 1% error probability for a 2-sided test. nc = no correlation. Bold = highest absolute value for Pearson's r

Parameter	Number of samples n	C release related to dry weight (mg C g _{DW} ⁻¹)			C release normalised to TOC (mg C g _{TOC} ⁻¹)			Ratio of R to G R250/G250
		R250	G250	R250-G250	R250	G250	R250-G250	
TOC	235	0.86	0.70	0.59	0.60	0.36	0.45	nc
TN		0.89	0.69	0.64	0.67	0.36	0.48	nc
TOC/TN		-0.56	-0.40	-0.43	-0.59	-0.30	-0.40	nc
TOC/S		0.74	0.50	0.60	0.51	0.18	0.32	nc
TN/TP		0.72	0.57	0.51	0.55	0.34	0.43	nc
LF, -HF	97	0.70	0.58	0.63	0.70	0.56	0.58	0.18
$\delta^{13}\text{C}$	40	-0.70	-0.66	-0.66	-0.52	-0.36	-0.41	nc
C_{mic}	73	0.68	0.60	0.56	0.55	0.50	0.33	nc
$\text{SOM}_{\text{H}_2\text{O}}$	16	0.94	0.84	0.96	0.85	0.51	0.70	0.66
$\text{Hi}_{\text{H}_2\text{O}}$		0.93	0.89	0.92	0.92	0.68	0.73	0.55
$\text{HA}_{\text{H}_2\text{O}}$		0.93	0.81	0.97	0.80	0.42	0.68	0.71
$\text{FA}_{\text{H}_2\text{O}}$		0.86	0.89	0.81	0.91	0.81	0.73	0.39
SiO_2	235	0.66	0.56	0.44	0.49	0.34	0.42	nc

TOC Total organic carbon (%), TN Total nitrogen (%), S Sulphur (%), P Phosphorus (%), LF, -HF Share of sediment in light and heavy fraction < 1.4 g cm⁻³ (%), $\delta^{13}\text{C}$ ratio of ¹³C to ¹²C in relation to standard VPDB (‰ VPDB), C_{mic} carbon in microbial biomass (mg kg_{DW}⁻¹), $\text{SOM}_{\text{H}_2\text{O}}$ total water-extractable SOM (g C kg_{DW}⁻¹), $\text{Hi}_{\text{H}_2\text{O}}$ water-extractable hydrophilics (g C kg_{DW}⁻¹), $\text{HA}_{\text{H}_2\text{O}}$ water-extractable humic acids (g C kg_{DW}⁻¹), $\text{FA}_{\text{H}_2\text{O}}$ water-extractable fulvic acids (g C kg_{DW}⁻¹), SiO_2 silicic acid in pore water (mg kg_{DW}⁻¹)

dissolution of CO₂ at this location are assumed to reflect in the observed lowered pore water pH. Sediment bulk density was strongly inversely related to organic matter content, due to diminished sediment particle packing by the coating and spacing of mineral particles with hydrated organic matter (Avnimelech et al. 2001). The depth pattern of the relationship indicates that the generic relationship proposed by Avnimelech et al. (2001) is only valid for sediment deposited in similar depths. Deeper, more consolidated layers exhibit higher bulk densities at the same organic carbon content in relation to the increased load (Fig. 4).

Organic matter supply also follows a pronounced spatial gradient. The upstream, non-tidal and shallow river section with peak primary production feeds more labile, easily degradable SOM (Mendonça et al. 2012; Ward et al. 2016) into the investigation area. Increased depth and turbulence of the Port of Hamburg leads to decay of algal biomass and zooplankton grazing (Schoel et al. 2014). As a result, phytoplankton-derived N rich and easily degradable dead biomass accumulates in the sediment, also indicated by the high correlation between chlorophyll in the water column (Spieckermann et al. 2021) and porewater SiO₂ (Fig. 2) with total N, and by the lowest ratio of TOC to TN (average TOC/TN = 7.5) at the most upstream location P1, corresponding to the range of 4 to 10 for algal biomass, as reviewed by Meyers (1994). Consequently, TOC and even more so TN are strong predictors of long-term carbon release (Table 1), as also found for methane generation in sediment from nine different German federal waterways (Gebert et al. 2006)

and from partially dewatered, landfilled sediment of different age (Gebert et al. 2019) as well as oxygen consumption from sediment in the port of Hamburg (Spieckermann et al. 2021). Hydrodynamically, location P1 represents the first sedimentation area for suspended material entering the port area from upstream. In addition to highest SOM contents (Fig. 2), SOM at P1 is also more labile as clearly seen when C release is normalized to total organic carbon (Fig. 7, right) with higher values indicating higher degradability of the organic matter present in the sediment, irrespective of the actual organic matter content. Analytically, the higher upstream share of more labile SOM is not only seen in more carbon release as CO₂ and CH₄, but also in the higher share of water-extractable carbon (Table 1), a lower ratio of acid-base to water-extractable SOM, a higher share of organic carbon in the low density fraction and lower $\delta^{13}\text{C}$ values, reflecting a less progressed decay of organic matter and a lower extent of stabilization in organo-mineral associations (Straathof et al. 2014; Zander et al. 2023).

Sediments are also imported into the investigation area from downstream due to the asymmetry of flood and ebb tide velocity distribution (Schoer 1990), also referred to as tidal pumping (Schwartz et al. 2015). Analytically, the phenomenon is detectable by the concentration of zinc (Zn), originating from upstream industrial polluting sources (Groengroeft et al. 1998), in the grain size fraction < 20 μm . These data indicate that marine particles are currently transported approximately up to river kilometer 619–620 (Reese et al. 2019; Zander et al. 2023), adding downstream

allochthonous organic matter to the investigation area, bound to mineral particles originating from the North Sea (Kappenberg and Fanger 2007) or the tidal marsh areas. Labile SOM of marine origin (phytoplankton) hardly reaches the investigation area as it is already degraded during its upstream-directed passage through the zone of maximum turbidity (Wolfstein and Kies 1999). The spatial patterns reflect in, for example, decreasing concentrations of chlorophyll *a* in the water column and silicic acid in the pore water (Fig. 2) along the investigated transect, but also of microbial biomass and extracellular polymeric substances in the sediment (Zander et al. 2022).

Increasing shares of acid–base to water-extractable organic matter, used as a dimensionless solid–liquid partitioning coefficient, support the findings that in downstream direction a higher proportion of organic matter is stabilized in organo-mineral associations and that aerobic and anaerobic SOM decay rates decrease accordingly (Zander et al. 2020, 2022). This share of organic matter is less accessible for microbial decay (for studies on terrestrial soils see Marschner et al. 2008; Six and Paustian 2014; Gao et al. 2019; Baldock and Skjemstad 2000) due to changes the steric conditions of SOM, reducing accessibility to degrading enzymes, as hypothesized by Hedges and Keil (1995) for marine sediments. Inversely, organic matter mineralization was highly correlated to water-extractable and hence not mineral-bound SOM (Table 1), which reflects the properties of natural dissolved organic matter (Olk et al. 2019). Pearson's *r* was highest for the relationship of aerobic and anaerobic SOM decay to water-extractable SOM, in spite of the fact that the sample number of this subset of investigations was comparatively low, suggesting a stronger relationship than with the other investigated parameters.

It was hypothesized that the ratio between SOM decay under aerobic conditions and SOM decay under anaerobic conditions would change along the transect and with depth. Lower ratios were assumed for the upstream direction and in the upper layers, as here more labile organic matter was expected to be accessible to enzymatic attack independent of the presence or absence of oxygen (Bastviken et al. 2004). Conversely, higher ratios were expected in downstream direction and in the deeper layers, assuming that aerobic conditions would enhance the degradation of the more refractory SOM, as found by Bastviken et al. (2003) for aerobic and anaerobic SOM decay in lake sediments. While the results indeed suggested a slight increase in ratios from km 624 to km 643 (Fig. 7, left), an overall gradient did not clearly emerge, also not when the data set was subdivided by sample depth (not shown). Also, the ratio did not change with depth (Fig. 6). This can be explained by the distribution of differently degradable SOM pools: only the fast and moderately degradable SOM varies along the transect (higher upstream, lower downstream), while the slowly

degradable organic matter pool is approximately equal in the entire investigation area, constituting 10–15% of total organic carbon under anaerobic and 15–20% under aerobic conditions (Zander et al. 2022). Differences in cumulative carbon mineralization were shown to be governed by the additional presence (or absence) of the fast and moderately degradable organic matter pools, for which oxygenated conditions provide little advantage over oxygen-deficient conditions (Westrich and Berner 1984; Henrichs 2005; McKew et al. 2013).

Ratios between aerobic and anaerobic SOM decay decreased over time of incubation to a factor of around 2 (Fig. 5, right). Bastviken et al. (2003) reported short term (~300 h, corresponding roughly to the R10 and G10 values in this study) anoxic SOM degradation rates to be between 44 and 78% of oxic rates in for the upper 5 cm of sediments from lake sediments with a very high organic matter content (13%–50%), corresponding to ratios of ~1.3 to 2.3. These are in the lower range of values found in this study and correspond more to the long-term ratios (compare Fig. 5, right). The authors related the lower share to the fact that in those samples organic matter was more refractory and hence degradation less benefitted from oxic conditions. Chapman et al. (2019) in a meta-study presented ratios of 4.3 to 13.4 for total carbon mineralization from various wetland soils, and Kristensen (1995) reported ratios of ~10 in 40 day incubations of marine sediment.

The long-term factor of 2 observed in this study is lower than would be expected from strictly thermodynamic considerations. This suggests that the higher the degree of partial decomposition and hence the more the remaining organic matter transitions into organo-mineral associations, the lower the enhancing effect of oxygen. Oxygen serves both as terminal electron acceptor (TEA) and as reactant in oxygenase-mediated enzymatic attack of complex substrate molecules (Kristensen 1995). While the role of oxygen as TEA can be replaced (at lower energy yields) by oxidised ion species such as NO_3^- , Fe^{3+} or SO_4^{2-} , its function as reactant for oxygenase-mediated SOM decay cannot. This consideration corroborates the above-discussed reasons for the absence of a clear gradient in the ratio of aerobic to anaerobic carbon mineralization, assumed to be due to the relatively equal share of poorly degradable SOM along the transect. In the system under study, energy yields are therefore not the only governing factor for microbial decay of SOM, but limitations imposed by substrate accessibility and, possibly, microbial community structure, affect the balance between anaerobic and aerobic carbon mineralization and hence also the potential for carbon sequestration (La Rowe and Van Capellen 2011).

While the ratio between aerobic and anaerobic C release was similar along the investigated transect and with depth, their absolute difference showed a pronounced

upstream–downstream declining trend (Fig. 8, right) and a declining trend with depth (data not shown). This explained by decline in absolute SOM content along the transect in relation to the input gradient explained above, reflected by the TOC content (Fig. 2) along the transect and at any location, with depth, as fresh organic matter is only provided from the top. Also, SOM decay in sediment that had been exhaustively degraded under reducing conditions could be re-initiated by changeover to aerobic conditions (Fig. 10). Up to 10% of the originally present TOC and up to 42% of the previously anaerobically released carbon were additionally liberated, showing that more complex SOM (e.g., lignins, complex lipids) remaining after advanced partial degradation and transitioning to organo-mineral complexes resists microbial degradation under anaerobic but can be attacked under aerobic conditions (Kristensen 1995). Bastviken et al. (2004) showed that initially anoxic conditions favored subsequent oxic mineralization of organic matter.

In addition to enhancing SOM decay, dredging measures, particularly those that re-suspend sediment in the water column such as water injection dredging, can release considerable amounts of reduced pore water solutes such as NH_4^+ (see also Fig. 3), NO_2^- , Fe^{2-} , Mn^{2-} , S^{2-} . Biochemical oxidation of these components increases sediment oxygen uptake and contributes to oxygen depletion in the water column (Steinsberger et al. 2019), possibly creating or aggravating oxygen minimum zones. While in the investigation area the biochemical oxidation of reduced species dominates oxygen consumption in the first few hours, long-term consumption is governed by mineralization of organic matter (Spieckermann et al. 2021). Conversely, these results show that if sediments remain undisturbed, anaerobic conditions support considerable sequestration of so-called blue carbon in aquatic habitats (Chapman et al. 2019). The magnitude of this sink and the potential for the mineralization of buried carbon (in the marine environment also referred to as legacy carbon), will therefore be impacted by all processes governing oxygen availability in sediment deposits (Carey et al. 2017).

5 Conclusions

The area under study is characterised by pronounced gradients in organic matter supply and degradability, reflected in corresponding gradients of carbon mineralization rates. Absolute differences between aerobic and anaerobic carbon release decreased in downstream direction, i.e. with increasing distance from the upstream regions of primary production and at any one location with depth, hence reflecting a source gradient. The relatively low ratio between aerobic and anaerobic carbon release for most samples and the absence of a trend in this ratio shows that oxygen supply

did not provide a thermodynamic advantage to the decay of the slowly degradable organic matter pool, which along the transect was equally distributed. Carbon fluxes from sediments in the River Elbe estuary are therefore not primarily governed by the energy yields but by substrate accessibility.

Mineralisation of organic matter can be re-initiated when exhaustively degraded anaerobic sediment is exposed to oxygenated water, potentially leading to significant additional release of carbon. The findings illustrate the high potential of a ‘carbon-sensitive’ sediment management in a port infrastructure with strongly patterned deposition of sediment with different SOM content and SOM lability to minimise environmental risks such as the possible creation or aggravation of oxygen minimum zones. When coupled to in situ temperature and the temperature response of SOM degradation and, the data provide the basis for C foot printing and C balancing questions.

Sediment bulk density and all connected properties, such as those relevant for safe navigation close to the nautical bottom, are strongly influenced by organic matter content and can therefore vary significantly within a port or waterway.

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Data availability Original data can be made available upon request.

Declarations

Competing interest The authors declare that they have no competing interests.

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