Particle shuttling and oxidation capacity of sedimentary organic carbon on the Baltic Sea system scale

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ABSTRACT

Continental margin sediments receive most of the particulate organic carbon (POC) deposited on the global seafloor, making them crucial locations in the carbon cycle. However, the complex environments in coastal oceans make it challenging to predict the fate of sedimentary organic carbon (OC) in these areas. Here we use data from 21 sites in the Baltic Sea, representing different biological and physiochemical regimes, to explore controls on sedimentary OC cycling. To this end, we combine in situ measured benthic fluxes of dissolved inorganic carbon (DIC; proxy for OC oxidation) with data on sediment properties. In the Gulf of Bothnia, low sedimentary OC oxidation capacities (yearly DIC flux divided by sedimentary POC inventory) were likely caused by a large fraction of terrestrial material in the POC pool, indicated by low sedimentary chlorophyll a content and high (> 10) carbon:nitrogen ratios. The highest OC oxidation capacities were measured at shallow, permanently oxic sites in the Baltic Proper, where bioturbation likely stimulates OC oxidation. The other sites in the Baltic Proper and all stations in the Gulf of Finland displayed increasing OC oxidation capacities with increasing normalised water depth (station depth divided by maximal depth in the basin). This pattern suggests that substantial quantities of POC are shuttled, through repeated cycles of resuspension-redeposition, from shallow erosion-transport (ET) areas to deep accumulation (A) areas. This interpretation was supported by decreasing sediment age and increasing sedimentary inventories of POC and chlorophyll a with normalised water depth. Our calculations indicate that particle shuttling redistributes almost half of the deposited export production from ET areas to A areas in the Baltic Proper, and that substantial amounts of terrestrial organic material are transported through particle shuttling to the deeper parts of the Gulf of Finland and Gulf of Bothnia. Depositional setting and POC origin can thus be central factors in predicting the distribution and fate of OC in coastal and shelf sediments.

1. Introduction

Coastal oceans play a paramount role in the global carbon (C) cycle (Bauer et al., 2013; Bianchi et al., 2018; Gruber, 2015). Continental margin sediments only comprise 7–8% of the global seafloor area but receive on average 86% of the oceanic particulate organic C (POC) deposition (Sarmiento and Gruber, 2006). There are several reasons for the high POC deposition rates in these areas. First, continental margins are relatively shallow, enabling a large proportion of settling particles to escape pelagic degradation due to a short residence time in the water column (Dunne et al., 2007). Second, rivers export some 200 Tg of terrestrial POC to the coastal ocean (Cai, 2011), which amounts to about 10% of the POC flux to the continental margin seafloor (Sarmiento and Gruber, 2006). Third, coastal and shelf areas are generally nutrient rich and support high rates of primary production, resulting in elevated export production and a high deposition rate of organic matter on the

seafloor (Dunne et al., 2007). Since such a high fraction of marine POC is deposited on coastal and continental margin sediments, the fate of this POC (i.e. burial or recycling) is of importance for C cycling globally.

At the same time, coastal systems are complex and our understanding of their impact on the C cycle is constantly developing (Bauer et al., 2013). Coastal areas were previously thought to be strong sinks for atmospheric carbon dioxide (CO_2) (Tsunogai et al., 1999; Walsh et al., 1985), but are now emerging as much weaker sinks (Laruelle et al., 2014; Liu et al., 2000) or even sources of CO_2 to the atmosphere (Chen et al., 2013). The influence of estuaries and coastal seas on atmospheric CO_2 concentrations is also believed to have changed due to anthropogenic activities (Bauer et al., 2013) in combination with an increased flux of POC from rivers to the ocean (Schlesinger, 1997). Furthermore, the fate of terrestrial organic carbon (OC) that is brought to coastal areas by rivers is a long-standing question (Hedges et al., 1997). Although terrestrial material generally is less reactive in marine environments than material with a marine origin (e.g. Bianchi et al., 2018; LaRowe et al., 2020), on a global scale only half of the riverine OC is buried in marine sediments (Hedges et al., 1997). The "missing" terrestrial OC has led to the suggestion that recycling rates of this material are higher than previously anticipated (Bianchi, 2011).

The extent of OC oxidation versus burial in marine sediments is influenced by biological and physicochemical factors as well as the origin and pre-depositional degradation of the material. The main basins of the Baltic Sea in northern Europe differ substantially with regards to environmental conditions as well as input rates and sources of OC (Gustafsson et al., 2014; Snoeijs-Leijonmalm et al., 2017). Therefore, the Baltic Sea is an interesting area for studies of factors that affect the fate of deposited POC. Several studies have investigated POC burial in the Baltic Sea, based on measurements of the sedimentary POC pool and specific compounds (e.g. Josefson et al., 2012; Kaiser and Arz, 2016; Leipe et al., 2011; Miltner and Emeis, 2001). However, benthic OC recycling rates have been less studied (although see Almroth-Rosell et al., 2009; Hall et al., 2017; Schneider et al., 2002; Sommer et al., 2017).

In a recent paper, we presented benthic fluxes of dissolved inorganic C (DIC; used as a proxy for OC recycling or oxidation rates) and sedimentary POC concentrations from all main basins in the Baltic Sea (Nilsson et al., 2019). The measurements were used to construct a benthic OC budget, which showed that the OC recycling rates were
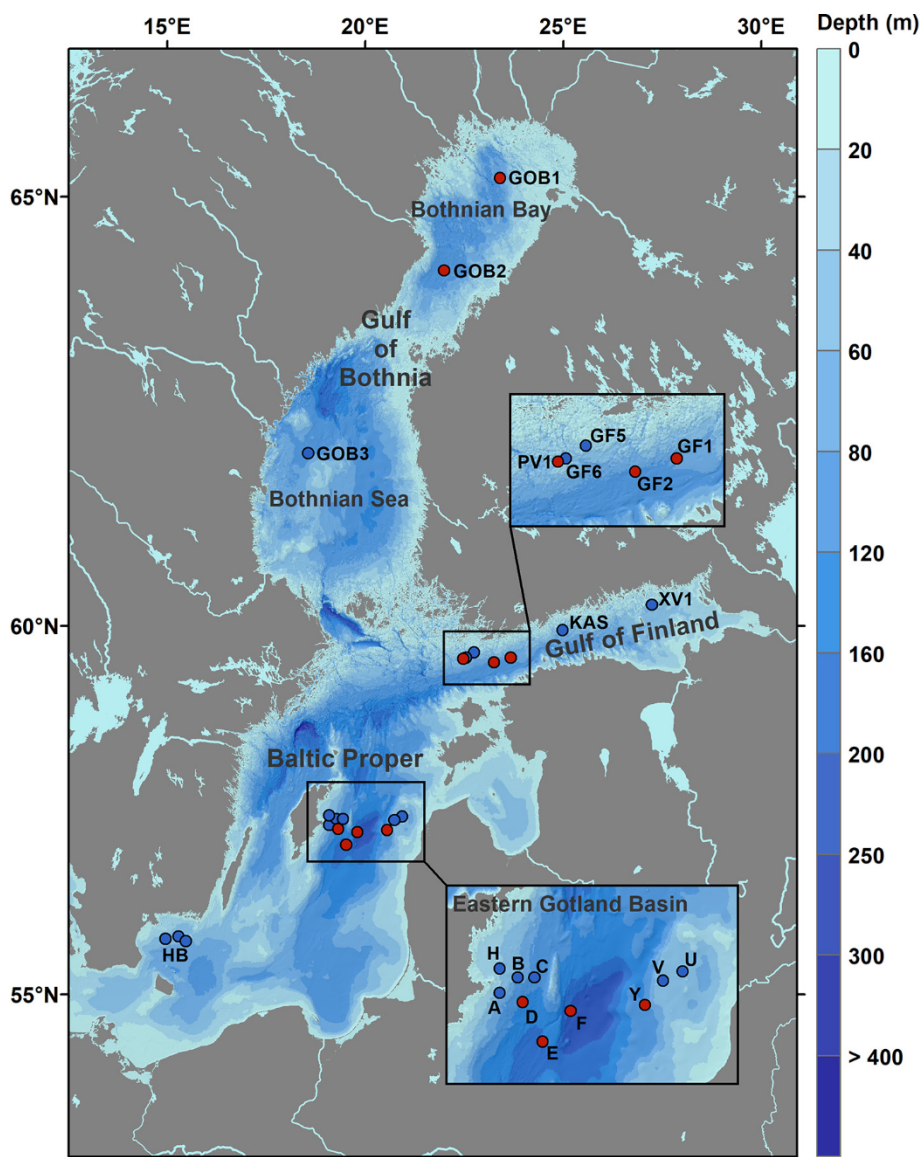
Fig. 1. Map of the Baltic Sea study area. Individual stations are marked (red = accumulation areas, blue = erosion-transport areas), modified from Nilsson et al. (2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

much higher than previously thought. Here we combine that data set with data of sediment characteristics to explore controls on OC cycling and redistribution in different sedimentary regimes and environments. Specifically, we investigate how (1) the origin of OC (terrestrial or marine) and (2) depositional setting (accumulation vs erosion-transport bottom) influence OC recycling rates and oxidation capacities.

2. Methods

2.1. Study area

The Baltic Sea is a large (392,978 km^2) brackish, semi-enclosed sea in northern Europe (Fig. 1). Its shore spans across nine countries and the drainage basin covers 1.7 million km^2, inhabited by ~85 million people (Leppäranta and Myrberg, 2009; Snoeijs-Leijonmalm et al., 2017). The southern basins, Baltic Proper (BP, maximum depth 459 m) and Gulf of Finland (GOF, maximum depth 123 m), are heavily influenced by eutrophication; oxygen depleted bottom water covers around 33% of the total area (Hansson et al., 2019). The BP and GOF can be classified as mesotrophic (Nixon, 1995) and primary production amounts to approximately 100–200 g C m^{-2} yr^{-1} (Wasmund et al., 2001). The salinity ranges from 6 to 12 in the surface water and 8–15 in the bottom water, but is considerably lower in the inner parts of the GOF due to intense river runoff (Snoeijs-Leijonmalm et al., 2017). There is a strong halocline at 60–80 m depth, separating the surface layer from the bottom water. The BP and GOF are not separated by a well-defined sill, which allows for close interactions between the areas. The northern part of the Baltic Sea, the Gulf of Bothnia (GOB), is substantially different from the BP and GOF. The salinity ranges from almost zero in the northernmost part to approximately 5 at the boarder to the BP and the halocline is weak. The GOB is oligotrophic with a primary production of 20–70 g C m^{-2} yr^{-1} and is much less affected by eutrophication (Lundberg et al., 2009). The bottom water is permanently oxygenated.

This study includes data from 21 stations in the Baltic Sea, visited in 2001–2014 (Fig. 1, Table 1). The GOF was visited in 2002–2005 and measurements were conducted at seven stations (GF1, GF2, GF5, GF6, KAS, PV1 and XV1). The BP was visited in 2001 and 2008–2010,

covering ten stations in the Eastern Gotland Basin (stations A–F, H, U, V and Y; maximum basin depth 249 m) and three stations in the Bornholm Basin (maximum basin depth 105 m). The Bornholm Basin stations were situated close to each other and at similar water depth, and were therefore grouped to one station (HB) in the data analysis. Three stations in the GOB were visited in 2013–2014 of which two were located in the Bothnian Bay (GOB1 and GOB2; maximum basin depth 146 m) and one in the Bothnian Sea (GOB3; maximum basin depth 293 m).

2.2. Benthic DIC fluxes measured in situ

The big and small benthic chamber landers from the University of Gothenburg were used for in situ measurements of benthic DIC fluxes (Ekeroth et al., 2016; Kononets et al., 2021; Nilsson et al., 2019). We use the DIC fluxes as a proxy for OC recycling or oxidation rates, since the dissolution of calcium carbonate is negligible in Baltic sediments (Leipe et al., 2011; Nilsson et al., 2019). The small lander was deployed using a drag line or on a mooring with a buoy, while the big lander was deployed fully autonomously at the seafloor during incubations, without any connection to the surface. The big lander had four incubation chambers and the small one had two. The chambers each enclosed a 400 cm^2 square area of sediment together with overlying water, in which concentration changes of solutes over time were detected by sensors (Aanderaa Data Instruments, Norway) or through analysis of discrete water samples taken by nine syringes per chamber at pre-set times. Oxygen optodes (model 3830) mounted in the chambers and on the frame of the lander monitored the bottom water oxygen concentrations. Turbidity and salinity were monitored with sensors (model 3612A and 3919A, respectively) to detect any sediment resuspension or chamber leakage. From 2007, salinity was also used to determine the volume of the incubated water; a necessary parameter to determine the benthic flux. This was done by initial injection of a small, known volume (ca. 60 mL) of de-ionized (Milli-Q) water to the chamber water and recording of the corresponding salinity change. Before the installation of conductivity sensors (i.e. 2001–2005), a weak bromide solution was injected instead. The bromide concentration in the water samples was analysed after recovery and the dilution was used to calculate the chamber

Table 1
Station information.

	Station	Bottom type	Water depth	Norm. water depth	Bottom water O$_2$	DIC flux	POC inventory	Oxidation capacity	LSAR	Sediment age
			m		μM	mmol m^{-2} d^{-1}	mol C m^{-2}	yr^{-1}	mm yr^{-1}	yr (top 2 cm)
BP	A	ET	60	0.24	178–279	10.1 ± 0.4 (4)	6.7	0.55	0.5	40
	B	ET	75	0.30	2–125	5.7 ± 2.3 (2)	11.6–13.8	0.16	0.3	67
	C	ET	90	0.36	6	4.3 (1)	9.1	0.17	0.2	100
	D	A	130	0.52	0	28.3 ± 6.9 (7)	41.7–42.3	0.25	0.5	40
	E	A	170	0.68	0	43.3 ± 13.1 (5)	32.6–40.9	0.43	0.8	25
	F	A	210	0.84	0	43.5 ± 7.1 (4)	33.1–36.1	0.46	1.0	20
	H	ET	44	0.18	248	8.2 ± 2.2 (10)	5.2	0.58	0.5[a]	40
	HB	ET	48	0.46	374	10.9 ± 3.1 (9)	7.7	0.52	0.5[a]	40
	U	ET	50	0.20	191	8.2 ± 0.3 (2)	1.3–3.3	1.30	0.37	54
	V	ET	65	0.26	14–36	13.1 ± 5.7 (4)	6.0–10.0	0.64	1.1–1.4[b]	16
	Y	A	120	0.48	0	16.1 ± 4.8 (3)	47.1–50.9	0.12	0.6–1.4[b]	20
GOF	GF1	A	78	0.63	0	15.5 ± 0.0 (2)	26.9	0.21	3.1	6
	GF2	A	85	0.69	0	27.3 ± 8.2 (3)	27.4	0.36	1.4	14
	GF5	ET	51	0.41	330	4.0 ± 1.7 (2)	18.2	0.08	2.8	74
	GF6	ET	48	0.39	328	2.3 ± 0.1 (2)	12.2	0.07	0.3	47
	KAS	ET	50	0.41	150–251	10.4 ± 2.8 (14)	14.2–37.6	0.15	0.4	32
	PV1	A	75	0.61	5–143	27.2 ± 8.7 (14)	23.0–23.9	0.42	0.6	7
	XV1	ET	37	0.30	85–285	6.0 ± 2.1 (3)	14.9–21.0	0.12	0.3	73
GOB	GOB1	A	86	0.59	303–341	7.5 ± 1.4 (7)	46.9–49.1	0.06	1.0	20
	GOB2	A	110	0.75	287–294	9.1 ± 2.3 (6)	31.7–42.8	0.09	1.5	13
	GOB3	ET	91	0.31	179–208	4.2 ± 0.9 (4)	5.5–6.3	0.26	0.5	40

Normalised water depth (Norm. water depth) is the station water depth divided with the maximum water depth of the basin, oxidation capacity is the yearly DIC flux divided by the POC inventory. DIC fluxes are given as average values ± standard deviation (nr. of chambers). Ranges are given for bottom water O$_2$ concentrations and POC inventory, which are often based on two values.

[a] No sample, average value for ET areas in the BP.
[b] Two values available for the station.

volume.

DIC concentrations in the discrete water samples were analysed in a customized CO_2 stripping system with a precision of 0.2% (relative standard deviation). Samples were injected in triplicates via a volumetric loop (~7 mL), which ensured a precise injection volume and kept the system airtight. Samples were automatically transferred into a strip-tower and acidified with 8.5% phosphoric acid, converting all carbonate species into gaseous CO_2. The CO_2 was stripped out with dinitrogen (N_2) gas and carried to the analyser (Li-COR6262) where it was detected by IR spectrometry (Goyet and Snover, 1993; Sullivan and Millero, 1998). The DIC concentrations in the samples were calculated from a two-point calibration based on certified reference material (Dickson Laboratory, Scripps Inst. of Oceanography, San Diego) and de-ionized (Milli-Q) water. Repeated measurements of certified reference material during the analyses were used to identify and correct for any instrument drift. Linear regression models (simple or quadratic) were fitted to the DIC concentration change over time in the chambers using the open-source software R (R Core Team, 2017), function 'lm' (CRAN:stats). The DIC flux (mmol m^{-2} d^{-1}) was calculated from the slope (simple regression) or the initial slope (quadratic regression) of the regression line and the volume of the incubated water (Kononets et al., 2021; Nilsson et al., 2019).

2.3. Sediment sampling and analysis

Sediment cores were collected using a custom-made multiple corer (Barnett et al., 1984). The cores were sliced in a glove bag filled with N_2 gas instantly after collection to minimize recovery artefacts and ensure oxygen free conditions. Slicing was done in intervals of 0.5 cm down to a sediment depth of 2 cm and 1–2 cm down to the maximum depth (≤ 20 cm). From each slice, the solid phase was sampled for analysis of water content and porosity, OC content, chlorophyll *a* (Chl a) fluorescence and linear sediment accumulation rates (LSAR).

Sediment samples for determination of water content and porosity were dried at 78 °C to constant mass (>48 h). Porosity was obtained from samples with a known wet volume. For samples with an unknown volume, the porosity was calculated from the water content and a sediment dry density of 2.65 g cm^{-3} (Blake and Hartge, 1986; Boyd, 1995). Sediments were classified as belonging to accumulation (A) or erosion-transport (ET) areas based on the water content and the shape of the OC depth profile in the sediment as described by Nilsson et al. (2019) and Håkanson and Jansson (1983). In general, sediments with surface layer water content >75% were classified as A areas while those with values <75% were classified as ET areas.

Sediment samples for measuring solid phase POC and nitrogen (N) were dried and homogenized using a mortar and pestle prior to analysis. Samples were treated with acid fumes (HCl, 37%) before analysis with an elemental analyser (Carlo ERBA N1500g, precision ±1%) as described by Verardo et al. (1990). We used the POC inventory as a measure of the readily available OC pool that is oxidised during initial stages of early diagenesis, and thus contributes to the measured DIC flux. The POC inventory (mol C m^{-2}) was calculated from the dry bulk density (g cm^{-3}), POC content (% dwt) and thickness of the reactive layer (cm), defined as the surficial sediment layer in which the POC content distinctly decreased before approaching a stable value down-core.

Chlorophyll a concentrations in the sediment, used as an indicator of fresh and labile organic material, were determined by fluorescence using the acidification method (Strickland and Parsons, 1972). This method is known to overestimate the Chl a concentration due to the presence of other fluorescent substances (Falkowski and Sucher, 1981), but was used to allow continued comparisons in our long records of measurements. A known volume (1–2 mL) of sediment was frozen in the dark at −20 °C for at least 24 h to induce cell lysis. Extractions were made with 96% ethanol (10 x the sediment volume) by shaking and refreezing in −20 °C in the dark for at least 24 h. Ethanol extracts were filtered through cellulose acetate filters (0.45 μm) and were diluted if needed

before measurement on a Turner TD-700 fluorometer. Sample fluorescence was read at 680 nm before and after acidification with a few drops of 1 M HCl. The Chl a concentration was obtained from a 5-point standard curve produced from a stock solution of the algae *Anacystis nidulans* dissolved in ethanol. The Chl a inventory was calculated in the same way as the POC inventory, using the sedimentary Chl a concentration (μg cm^{-3}) instead of the POC content.

Linear sediment accumulation rates were obtained from the excess ^{210}Pb activity, analysed by a 46 keV gamma line using a low energy, high purity germanium detector. The gamma emission was corrected for self-absorption (Cutshall et al., 1983). Samples were packed in gas-tight foils and stored for 3 weeks before measurements to obtain equilibrium ^{226}Ra-^{222}Rn. For the GOF, LSAR was calculated from the mass sediment accumulation rate assuming a dry sediment density of 2.65 g cm^{-3} (Blake and Hartge, 1986; Boyd, 1995). The mean age for the top 2 cm of sediment was calculated from the thickness of the layer divided by the LSAR. It should be noted that "age" in this context describes how long ago the sediment was deposited; it does not refer to the age of the material itself.

2.4. Statistical analyses

Statistical tests were conducted in the software R (R Core Team, 2019). Since most parameters were not normally distributed, Spearman's rank correlation coefficient (r_s) was calculated using the function 'cor.test' (package 'stats'). Differences between bottom types were tested using Welch's *t*-test (function 't.test', package 'stats').

3. Results and discussion

Results from the benthic flux measurements and sediment investigations are presented in Table 1. In the following sections, we demonstrate differences in benthic OC recycling rates between and within the Baltic Sea basins and suggest mechanistic explanations. Firstly, within-basin differences are discussed in relation to water depth and depositional settings. Secondly, contrasts in benthic OC recycling dynamics between the basins are presented.

3.1. Particle shuttling redeposits material

3.1.1. Geochemical signals in the sediment

The benthic DIC flux increased with water depth ($r_s = 0.50$, p = 0.021; Fig. 2). We hypothesize that this pattern was caused by resuspension and lateral transport of particles, shuttling POC from shallow to deeper areas (e.g. Almroth-Rosell et al., 2011; Christiansen et al., 2002). Increased recycling rates with depth, or when approaching the maximum depth of a basin, is a known phenomenon that has been observed in other depo centres (e.g. Cathalot et al., 2015; Glud et al., 2013). Previous studies have indicated that this process also takes place in the Baltic Sea; the finest particle layer, known as the "fluffy" layer, is
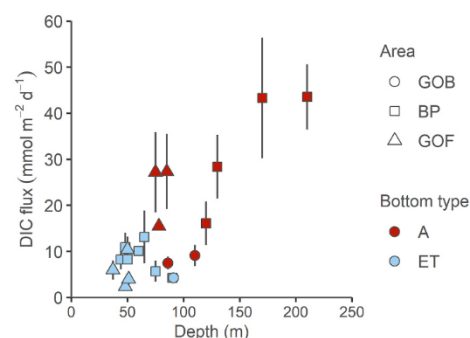
Fig. 2. Benthic fluxes of DIC versus water depth. Fluxes are average per station, with standard deviation ($r_s = 0.50$, $p = 0.021$).

resuspended on average 1–7 days per months in the BP (Danielsson et al., 2007). This resuspension and lateral transport in turn affects the distribution of bulk organic matter as well as isotopes and specific compounds (e.g. Almroth-Rosell et al., 2011; Christiansen et al., 2002; Jonsson et al., 1990; Leipe et al., 2011; Struck et al., 2004).

Particle shuttling results in a gradient of increasing inventories of sedimentary material from shallow areas toward the deepest point in the basin. Thus, the deepest point of each individual basin in the Baltic Sea can be assumed to act as a focal point, or 'end station', for suspended and redepositing sediment particles. Since the maximum depths in the studied basins differ, we will use normalised water depths in the following discussion. Normalisation was done by dividing the station water depth by the maximum water depth of the corresponding basin (Table 1). The normalised water depth thereby ranged from zero (at the sea surface) to one (at the maximum water depth of the basin).

Both the POC and Chl a inventories increased with normalised water depth, as predicted by the shuttling hypothesis. There was a strong, positive correlation between the POC inventories and normalised water depth ($r_s = 0.81$, $p < 0.001$; Fig. 3a). The Chl a inventories also showed an increasing trend with normalised water depth, however, this trend was not statistically significant when the low values at stations in the GOB were included ($r_s = 0.40$, $p = 0.125$; Fig. 3b). Since these low Chl a values likely are a result of high inputs of terrestrial OC (see section 3.2), they were excluded from the regression analysis and a significant trend was obtained ($r_s = 0.88$, $p < 0.001$). The degradation of Chl a is enhanced in oxic and bioturbated sediments (Bianchi et al., 2000), which could contribute to the pattern of higher Chl a inventories in the deeper anoxic basins. Taken together, however, the positive trends suggest that particle shuttling along the bathymetric continuum strongly influences deposition of POC, and organic matter in general, on a basin scale.

A further indication of particle shuttling was the elevated LSAR values in A areas compared to ET areas (Table 1). This difference in LSAR shows that the sediment, integrated over a given sediment thickness (in this case 2 cm), is older in ET areas than in A areas (Fig. 4). The age of the top sediment layer in ET areas ranged from 25 to 74 years (with one exception of 14 years) compared to 7 to 16 years in A areas. The age of the top sediment layer decreased significantly with normalised water depth ($r_s = -0.63$, $p = 0.002$; Fig. 4), indicating that sediment in the deeper parts of the basins were deposited more recently compared to those in shallow areas.

3.1.2. Particle redistribution on a basin scale

The effect of redistribution of POC from ET to A areas can be calculated using a mass balance approach (Table 2) based on numbers from Nilsson et al. (2019) and references therein. Assuming steady state, deposition rates of POC at a certain bottom type and basin is here defined as the sum of our measured OC recycling and burial rates. The deposition rates can then be compared to estimates of marine export
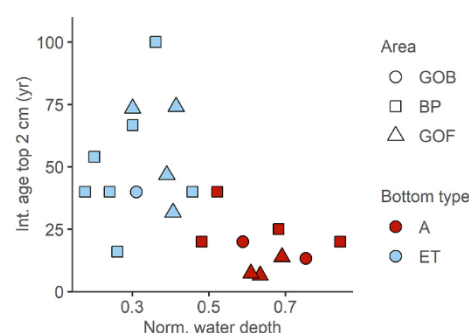

Fig. 4. Sediment age versus normalised water depth. The sediment age is integrated over the top 2 cm ($r_s = -0.63$, $p = 0.002$).

production. Although these relatively coarse generalisations create large standard deviations in some cases, this exercise gives insight into major movements of sedimentary OC in the Baltic Sea.

To correctly interpret the mass balance, however, uncertainties around the values of OC recycling rates require attention. Whereas the DIC fluxes presented here are in the same range as other benthic chamber lander measurements from the area (2.2–98 mmol m^{-2} d^{-1}; Ekeroth et al., 2016; Hall et al., 2017; Sommer et al., 2017), they are generally higher than OC recycling rates calculated from porewater DIC and ammonium profiles (1.3–8.8 mmol m^{-2} d^{-1}; Kuliński and Pempkowiak, 2011; Noffke et al., 2016) and calculated accumulation of total CO_2 in the deep Eastern Gotland Basin based on repeated water column profile measurements (5.5 mmol m^{-2} d^{-1}; Schneider and Otto, 2019). Part of the discrepancy is probably a result of different methodologies. Calculations of accumulation or fluxes of DIC based on concentration gradients can underestimate OC recycling rates due to limited vertical resolution of the concentration profiles, especially close to the sediment-water interface where concentration gradients generally are steepest (e. g. Sundby et al., 1986). Accumulation rates of DIC in the water column can also underestimate DIC production rates if lateral intrusions (causing dilution) take place (Schneider and Otto, 2019), i.e. if the assumption of stagnancy of the relevant part of the water column is not completely fulfilled. At the same time, it has been shown that at sites with stagnant water, sediment incubations where the overlying water is being stirred (i.e. in benthic chambers) can erode the diffusive boundary layer thickness and thereby increase the oxygen uptake (Glud et al., 2007; Hall et al., 1989). It is possible that changes to the diffusive boundary layer also affect DIC and nutrient fluxes, although this has to be confirmed in future studies. Another plausible explanation to the high measured fluxes is seasonality. The samplings in this study were conducted during the summer months (May to September), when OC recycling rates tend to be highest (Berelson et al., 2003; Schneider and Otto, 2019). When integrated over a year, our values may thus
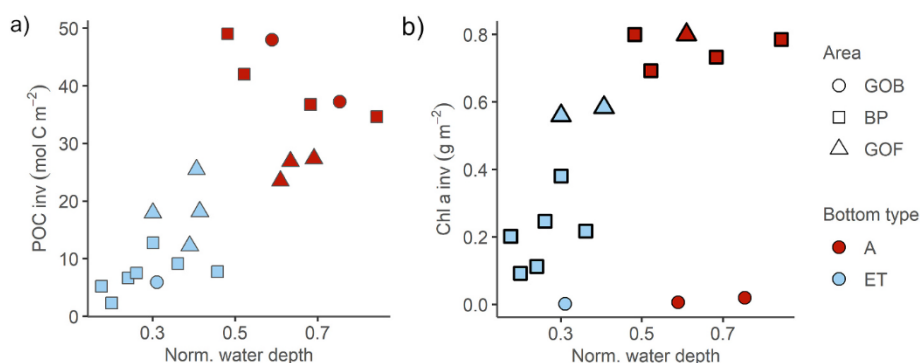

Fig. 3. Sedimentary inventories. a) POC ($r_s = 0.81$, $p < 0.001$) and b) Chl a in the reactive sediment layer versus normalised water depth ($r_s = 0.88$, $p < 0.001$; only markers with thick borders were included in the analysis).

Table 2
Local sedimentary organic carbon budgets for the basins of the Baltic Sea.

Area	Bottom type	Recycling[a]	Burial[a]	Deposition (recycling + burial)	Marine export[a] production (MEP)	Difference (MEP - deposition)
GOB	ET	1.36 ± 0.28		1.36 ± 0.28	0.39 ± 0.003	−0.97 ± 0.28
	A	1.19 ± 0.39	0.21 ± 0.03	1.40 ± 0.39	0.17 ± 0.002	−1.29 ± 0.39
BP	ET	5.54 ± 4.49		5.54 ± 4.49	9.63 ± 0.003	4.06 ± 2.37
	A	12.00 ± 6.27	0.45 ± 0.29	12.45 ± 6.27	5.69 ± 0.002	−6.76 ± 4.78
GOF	ET	0.43 ± 0.29		0.43 ± 0.29	0.63 ± 0.003	0.20 ± 0.29
	A	1.26 ± 0.64	0.32 ± 0.10	1.58 ± 0.65	0.44 ± 0.002	−1.14 ± 0.65

[a] Values from Nilsson et al. (2019) and references therein. Average rates ± standard deviations in Tg C yr^{-1}.

somewhat overestimate the total OC recycling rate.

In the BP, the mass balance calculations result in a surplus input of OC to ET areas (4.06 ± 2.37 Tg C y^{-1}), i.e. the export production is higher than the calculated deposition rate based on our measurements. However, the OC input surplus in ET areas is approximately matched by an OC input deficit to A areas (−6.76 ± 4.78 Tg C y^{-1}), showing that more material is deposited than can be explained by the export production. The discrepancy between export production and deposition to the different bottom types suggests that material is resuspended in ET areas and transported laterally to A areas in the basin. Thus, we suggest that almost half of the annual gross POC deposition in ET areas emanating from the photic zone could be redistributed through particle shuttling to A areas in the BP.

In the GOB, the calculated deposition was roughly an order of magnitude higher than estimates of the export production in both ET and A areas (Table 2). Only three samplings were conducted in the area, which naturally will introduce uncertainty in the upscaling calculations. Furthermore, the large difference between calculated deposition and export production may result partly from an underestimation of primary production rates, as rate measurements in the area differ substantially (Savchuk et al., 2012 and references therein). Yet, the discrepancy

suggests that another substantial source contributes to the OC deposition in the GOB, namely rivers. Since riverine terrestrial OC is required to close the budget in A areas, shuttling via the coastal ET areas to the central A areas is likely to be an important process in the basin. In the GOF, the surplus marine export production in ET areas was not large enough to explain the surplus deposition in A areas. This again suggests that input of riverine OC is important in the basin. The large standard deviations for values in the BP also allow room for input of riverine material; however, marine primary production is the predominant source of OC to the area (see section 3.2.1).

The "missing" OC in the GOB and GOF (~2 and 1 Tg C yr^{-1}, respectively) corresponds to the total estimated inputs of dissolved and particulate OC from rivers to these basins (Dahlgren Strååt et al., 2018; Gustafsson et al., 2014). Although it is unlikely that all of the riverine OC is deposited on the seafloor and our budget contains large uncertainties, these numbers point to a possible underestimation of input and deposition of riverine material to some Baltic Sea basins (Nilsson et al., 2019). Previous studies have indeed pointed out the need for more field measurements to reliably asses the size and seasonal pattern of the riverine OC input (Dahlgren Strååt et al., 2016; Gustafsson et al., 2014). Local studies have also shown that flocculation of dissolved riverine
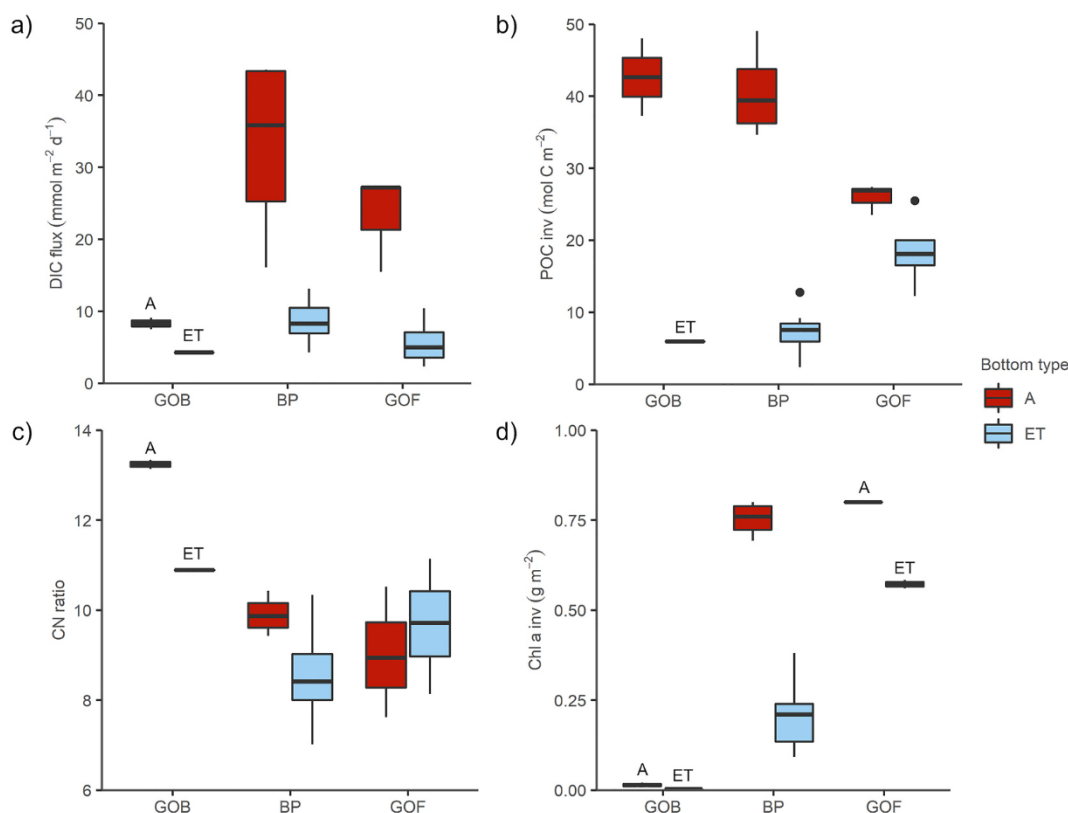
Fig. 5. Geochemical parameters in different depositional areas in the basins of the Baltic Sea. a) Sedimentary flux of dissolved inorganic carbon (DIC, used as a proxy for OC recycling), modified from Nilsson et al. (2019). b) Inventory of particulate organic carbon (POC) in the reactive layer of the sediment. c) Carbon:nitrogen ratio (CN ratio) in the reactive layer of the sediment. d) Inventory of chlorophyll a (Chl a) in the reactive layer of the sediment.

material is a source of OC to the sediment, but we lack general information about under what circumstances and to what extent this process takes place (Asmala et al., 2014; Gustafsson et al., 2000; Skoog et al., 2011). Nevertheless, our budget (Table 2) indicates that particle shuttling is an important mechanism that redistributes both marine and terrestrial OC in the Baltic Sea.

3.2. Controls on benthic OC dynamics

3.2.1. The effect of OC origin

On a Baltic Sea system scale, there was a significant difference between DIC fluxes in A and ET areas ($t = 3.71$, df $= 8.77$, $p = 0.005$; see also Fig. 5a). However, rates of OC recycling and burial have been shown to vary substantially between basins and bottom types in the Baltic Sea (Nilsson et al., 2019). The highest DIC fluxes were measured in the A areas of the BP. As the OC in the BP is primarily of marine origin and the OC content in the sediment is higher than most marine areas (10–16%; Leipe et al., 2011; Miltner and Emeis, 2001; Nilsson et al., 2019), high DIC fluxes are expected.

A striking difference between basins was the considerably lower sedimentary OC recycling rates (benthic DIC flux) in the GOB compared to the BP and GOF (Fig. 5a). Yet the reactive layer POC inventories in the GOB did not differ from those in the BP (Fig. 5b). The comparatively low recycling rates observed in the GOB can thereby not be explained by OC limitation, but rather by a lower reactivity of the available POC pool than in the other basins. The low OC recycling rates in the GOB can likely be attributed to a larger proportion of terrestrial material in the POC pool, as terrestrial material generally is less reactive in the marine environment than material of marine origin (e.g. Bianchi et al., 2018; LaRowe et al., 2020). Elevated carbon:nitrogen (CN) ratios in the GOB (Fig. 5c) are consistent with this explanation, since terrestrial matter is characterized by high CN ratios (Burdige, 2006). A high contribution of terrestrial matter to the sedimentary POC pool is also indicated by low Chl a inventories in the GOB (Fig. 5d; Josefson et al., 2012). The low Chl a inventory is likely caused by lower primary productivity in the GOB compared to the BP and GOF (Gustafsson et al., 2014; Wania et al., 2001), as well as dilution of the marine POC by the high load of riverine terrestrial organic matter. That the sedimentary POC pool in the GOB contains a high proportion of terrestrial material has been shown in studies of biomarkers and lignin oxidation products (Kaiser and Arz, 2016; Miltner and Emeis, 2001). The material is also believed to have undergone substantial pre-depositional degradation (Miltner and Emeis, 2001), which likely decreases the reactivity further.

While the GOF also receives a considerable input from rivers, the sediment did not show the clear signals from terrestrial POC that were observed in the GOB (Fig. 5). The difference in terrestrial signal between the two basins is likely a reflection of land use in the drainage area. Whereas the drainage area of the GOB is dominated by forests and non-productive land, more of the GOF drainage area consists of arable land and pastures (Miltner and Emeis, 2001). A large riverine nutrient input to the GOF (HELCOM, 2015), as well as high amounts of regenerated nutrients, support a relatively high primary production in the basin which likely dilutes the terrestrial signal in the sediment. At the same time, the burial efficiencies amount to 10–20% in both the GOB and the GOF, compared to 3.6% in the BP (Nilsson et al., 2019). The relatively high burial efficiency in the GOB can be explained by the sizeable contribution of refractory terrestrial material to the POC pool, whereas the lack of a clear terrestrial signal in the GOF sediment suggests that a substantial part of the buried matter is of marine origin. Studies of lignin oxidation products have further indicated that the terrestrial POC in the surface sediment of the GOF is considerably less altered by pre-depositional degradation than material in the GOB (Miltner and Emeis, 2001), possibly meaning that the material is more reactive. Our data do not allow us to elucidate why relatively large amounts of seemingly reactive OC is buried in the GOF, but oxygen deficiency and association with mineral particles (Bianchi et al., 2018; LaRowe et al., 2020) are plausible causes.

3.2.2. Oxidation capacity and the effect of particle shuttling

The correlation between OC recycling rates and reactive layer POC inventory was moderate ($r_s = 0.49$, $p = 0.023$), suggesting that the available pool of POC in the reactive sediment layer per se had a rather limited influence on the recycling rates. Interestingly, the OC oxidation capacity (yearly DIC flux divided by the reactive layer POC inventory) varied substantially and indicated differences in POC reactivity between stations (Fig. 6). The OC oxidation capacities at stations in the GOB were low compared to in the BP and GOF, especially in A areas. These low oxidation capacities were likely related to the high contribution of terrestrial material to the POC pool, as discussed above.

The highest OC oxidation capacities were found at permanently oxygenated sites in the BP (stations A, H, HB, U and V; Table 1). Values on the order of 0.5–1 yr^{-1} suggest that the pools of available OC at these stations could be turned over within a year. The OC reaching these relatively shallow stations is mainly fresh, marine material, which is expected to be highly reactive. Bioturbating macro- and meiofauna at these permanently oxygenated sites could be another reason for the efficient oxidation of OC, since fauna is known to stimulate degradation of organic matter in sediments (e.g. Aller and Aller, 1986; Nascimento et al., 2012; Welsh, 2003). Interestingly, the OC oxidation capacity at permanently oxic ET stations in the GOF was low (Fig. 6, Table 1). Possible reasons for this could be POC association with mineral particles, as discussed above, and lower or different types of bioturbation (Snoeijs-Leijonmalm et al., 2017).

Apart from the permanently oxic ET stations in the BP, the OC oxidation capacity increased significantly with normalised water depth in the BP and GOF ($r_s = 0.82$, $p < 0.001$, Fig. 6). A likely explanation for this pattern is particle shuttling. Fresh, reactive organic matter particles are easily resuspended in high-energetic ET areas and are kept in suspension for a long time, making them especially prone to be transported to and accumulate in low-energetic A areas (Almroth-Rosell et al., 2011). The material deposited in these A areas after lateral and vertical transport is thus relatively young (Fig. 4) and emanates mainly from marine primary production (Leipe et al., 2011; Miltner and Emeis, 2001). This is further seen in the increasing Chl a inventory with normalised water depth (Fig. 3b). Fresher, or diagenetic 'immature' organic matter is generally degraded efficiently (Burdige, 2007; Cowie and Hedges, 1994), which likely explains the high OC oxidation capacities in these A areas. Shuttling of material would further cause mixing of old (refractory) and fresh (reactive) sedimentary material. It is thus possible that the high OC oxidation capacity in A areas partly is a priming effect, i.e. enhanced remineralisation of refractory OC after input of fresh OC (van Nugteren et al., 2009), leading to high oxidation rates. Whether priming has an effect in aquatic environments is still under debate (Bengtsson et al., 2018), but it could potentially be an important positive
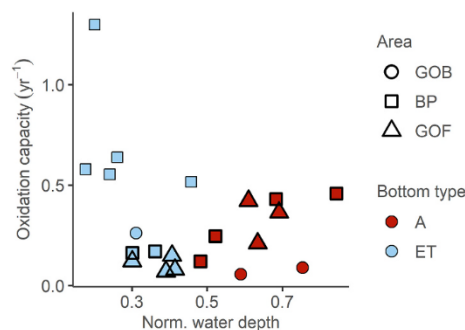

Fig. 6. Organic carbon oxidation capacity versus normalised water depth. Organic carbon oxidation capacity = yearly DIC flux / sedimentary POC inventory ($r_s = 0.82$, $p < 0.001$; only markers with thick borders were included in the analysis).

feedback mechanism between eutrophication and OC oxidation.

Sediments underlying anoxic water columns, such as the deeper parts of the BP and GOF, are often associated with increased OC burial since oxygen increases the degradation of certain types of organic matter (Arndt et al., 2013; Bianchi et al., 2000; LaRowe et al., 2020). Oxygen deficiency could indeed act to preserve organic matter in A areas of the BP. Hylén et al. (2021) observed a temporary increase in the degradation of sedimentary organic matter after an oxygenated deep-water inflow (a so called Major Baltic Inflow) to the Eastern Gotland Basin, suggesting that oxygen affects degradation rates. Yet the OC burial efficiency in these oxygen-depleted A areas is low, especially in the BP (3.6%; Nilsson et al., 2019), probably due to the focusing of reactive OC. Oxygen depletion below the permanent halocline could also play an important role in the transport of reactive POC to the sediment. The anoxic water column in the BP and GOF has decreased the zooplankton abundance, which protects suspended matter from pelagic mineralisation (Le Moigne et al., 2017; Lee, 1992; Ploug and Bergkvist, 2015). We did not observe a correlation between the OC oxidation capacity and bottom water oxygen concentrations ($r_s = -0.24$, $p = 0.302$), indicating that oxygen did not directly affect the fate of sedimentary OC. However, since other environmental factors such as depth (and thereby OC age) and diversity and abundance of bioturbators follow the oxygen gradient, laboratory based experiments are required to fully disentangle the role of oxygen in sedimentary OC recycling in the Baltic Sea.

4. Conclusions

In this study, we present in situ-measured sediment-water DIC fluxes and sediment properties from 21 stations across the major basins of the Baltic Sea. Using DIC fluxes as a proxy for OC recycling or oxidation, we discuss mechanisms behind the large differences in OC oxidation rates and burial efficiencies between the three major basins: the oligotrophic GOB and the mesotrophic BP and GOF.

The observed OC recycling rates were considerably lower in the GOB than in the other basins. The main reason behind this difference was low reactivity of the sedimentary POC in the GOB, displayed as low OC oxidation capacity (yearly DIC flux divided by POC inventory). The cause of the low reactivity was presumably a large contribution of terrestrial material to the sedimentary POC pool, indicated by high organic CN ratios and low Chl a content.

The highest OC oxidation capacities were found in permanently oxygenated ET areas in the BP. These high values were likely due to stimulation of OC oxidation by bioturbation or the presence of oxygen. However, apart from those stations, the OC oxidation capacity increased significantly with normalised water depth in the BP and GOF. This pattern was attributed to shuttling of particles from energetic ET areas to depositional A areas, transporting fresh and reactive OC to deeper parts of the basins. Consistent with this hypothesis, there was an increase in the sedimentary inventories of Chl a and POC with normalised water depth, while the age of the top sediment layer decreased. In addition to focusing of POC, it is possible that shuttling increases the OC oxidation capacity through priming.

By comparing POC deposition rates (the sum of measured OC recycling and burial rates) with values of marine export production in the different depositional areas, we estimate that almost half of the gross POC deposition emanating from the photic zone in the BP is redistributed through shuttling from ET areas to A areas. Similar comparisons in the GOB and GOF indicate that a substantial amount of terrestrial organic material is transported to the deeper parts of the basins.

Our results demonstrate that particle shuttling is an important process in all major basins of the Baltic Sea, transporting POC of different origins and reactivity toward deposition centres. The combination of origin and shuttling will thereby have strong effects on the patterns of OC oxidation and burial. Similar mechanisms are likely to be occurring in other coastal seas. The reactivity of organic matter depends both on the nature of the compound and its environment (Arndt et al., 2013;

Bianchi et al., 2018; LaRowe et al., 2020). Future studies should therefore consider situations where particles ultimately are deposited under very different conditions from where they originally reached the seafloor.

Declaration of Competing Interest

None.

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