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Organic Matter Sources in North Atlantic Fjord Sediments

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Key Points:
- Organic matter sources in North Atlantic fjords are much more heterogeneous than previously estimated
- Ongoing climate change is likely to have a pronounced effect on carbon burial in North Atlantic fjords

Supporting Information:
- Supporting Information S1
- Data Set S1

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Abstract To better constrain the global carbon cycle fundamental knowledge of the role of carbon cycling on continental margins is crucial. Fjords are particularly important shelf areas for carbon burial due to relatively high sedimentation rates and high organic matter fluxes. As terrigenous organic matter is more resistant to remineralization than marine organic matter, a comprehensive knowledge of the carbon source is critical to better constrain the efficiency of organic carbon burial in fjord sediments. Here we investigated highly productive fjords in northern Norway and compare our results with both existing and new organic carbon to organic nitrogen ratios and carbon stable isotope compositions from fjords in mid-Norway, west Svalbard, and east Greenland. The marine organic carbon contribution varies significantly between these fjords, and the contribution of marine organic carbon in Norwegian fjords is much larger than previously suggested for fjords in NW Europe and also globally. Additionally, northern Norwegian fjords show very high marine carbon burial rates (73.6 $gC \cdot m^{-2} \cdot year^{-1}$) suggesting that these fjords are probably very distinct carbon burial hotspots. We argue that the North Atlantic Current inflow sustains these high burial rates and changes in the current strength due to ongoing climate change are likely to have a pronounced effect on carbon burial in North Atlantic fjords.

1. Introduction

It is well known that deposition and burial of marine organic matter (OM) in sediments have played a key role in controlling atmospheric carbon dioxide and oxygen concentrations as well as fossil fuel formation over the past 500 million years (Berner, 2003). Compelling evidence for the sensitivity of marine OM burial to global climate change during the last 150,000 years has recently been published (Cartapanis et al., 2016). They show that pulses of marine OM burial in deep-sea sediments correlate with sea level fall and ice volume increase, and ultimately atmospheric CO_2 decrease on a global scale. Still, critical components of the carbon cycles in coastal and shelf regions remain unresolved (Bauer et al., 2013). In particular, the natural ability of shelf regions including fjords to sequester CO_2 through the burial of marine OM in sediments is not well understood. As a result, uncertainties persist between estimates of carbon burial in modern sediments and those derived from global Holocene sediment accumulation rates. This implies that some depositional environments may be under-represented and not included in the overall modern carbon budget. In this respect, an important but so far understudied region of the continental margins are fjord systems. Even though temperate fjords represent only a small fraction (<0.1%) of the total volume of continental margin sediments, they contain an estimated 12% of the total margin sediment deposited during the last 100.000 years (Nuwer & Keil, 2005; J.P.M. Syvitski et al., 1987). A recent study on a new global OM burial assessment, including the accumulation of OM in fjords for the first time, indicates that the potential of fjords to naturally sequester CO_2 through OM burial in sediments is highly underestimated (Smith et al., 2015). According to Smith et al. (2015) about 18 million tons of organic carbon (marine and terrigenous) is buried in fjord sediments each year, equivalent to 11% of annual marine carbon buried globally.

Few studies exist using surface sediments to investigate the environmental processes that control the organic geochemistry of fjord sediments. Studies from fjords in Chile (Bertrand et al., 2012; J. Sepúlveda et al., 2011; Silva et al., 2011), New Zealand (Hinojosa et al., 2014; Knudson et al., 2011; R. W. Smith et al., 2015) and Norway (Johan C. Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014; Winkelmann & Knies, 2005) reported a significant influence from freshwater inflow on their geochemical composition and suggest a common decreasing gradient of terrigenous organic material from the inner fjords toward the open ocean. In contrast, recent findings of predominantly terrigenous OM in the outer area of an Antarctic bay (Munoz & Wellner, 2016) put the latter inferences into question and emphasize the need for

an improved understanding of regional to local effects controlling OM sedimentation in mid-latitude and high-latitude fjord systems. This is especially important since remineralization of particulate terrestrial OM in the oceans is also much less efficient than that of marine OM and is therefore more likely to enter the long-term carbon cycle (Burdige, 2005; Mayer et al., 2007). Smith et al. (2015) found evidence that a minority of the total OM pool in fjord sediments is of marine origin and a follow-up study by Cui et al. (2016) suggest that the average percentages of marine OM in fjord sediments are 38-45% globally and 24% in NW Europe. However, these assumptions are mainly based on studies from New Zealand fjords with dense vegetation cover in the drainage area and fjords with low oxygen bottom water concentration in NW Europe (Cui et al., 2016; Hinojosa et al., 2014; Smith et al., 2015).

Compared to fjords in the Southern Hemisphere, Arctic and sub-Arctic fjords in the North Atlantic region are located at higher latitudes and have a sparser vegetation cover in their drainage areas. Moreover, the environmental setting of fjords located on the eastern side of the North Atlantic, in Norway and Scotland, is strongly influenced by the North Atlantic Current (NAC), the northern extension of the Gulf Stream. The NAC transports heat to much higher latitudes than in any other ocean and disbands in the Svalbard region. Thus, Greenland fjords on the western side of the North Atlantic are not affected by the warm water current (Figure 1). Therefore, fjords in Norway are mostly ice-free during winter, while Svalbard and Greenland fjords are seasonally (winter) sea ice covered and glaciated in the drainage area. The different climate settings of these fjords have a large effect on their biological processes, influencing primary productivity and the input of terrigenous OM from the fjord drainage areas (Syvitski et al., 1987). However, the effect of these different environmental settings on burial rates in fjords is not well investigated. Much research in shelf areas focuses on the mechanisms of OM preservation and remineralization. OM sources (marine versus terrigenous) are rarely examined in North Atlantic fjords. But this information is very important to better constrain and understand remineralization processes in fjord sediments, and the role of the coastal ocean in regulating atmospheric CO_2 levels over variable timescales (Bianchi et al., 2018).

To better define the efficiency of carbon burial in fjords, we here provide a detailed study of OM sources in three fjords from off the Lofoten Islands, northern Norway. By comparing these fjords with several glaciated and nonglaciated fjords from Norway, Svalbard, and Greenland we show that the fraction of marine versus terrigenous OM varies significantly from fjord to fjord. The main explanatory factors appear to be (a) the drainage area versus fjords surface area ratio and (b) the strength of the NAC inflow, which provides nutrients and the physical conditions (salinity and temperature) to sustain marine organisms to flourish. This indicates that even though North Atlantic fjords are generally characterized by high sedimentation rates and large OM fluxes, their carbon sequestration efficiency may vary drastically in accordance to their distinct environmental setting.

2. Materials and Methods

2.1. Study Area

The Vestfjord, Ofotfjord, and Tysfjord are the three main fjords between the Norwegian mainland and the Lofoten archipelago in northern Norway (Figures 1 and 2). The Vestfjord is an "*atypical*" fjord with a length of about 180 km, and its cone shape gives it the character of a coastal bay (Mitchelson-Jacob & Sundby, 2001). The fjord widens from about 15 km at its junction with Ofotfjord and Tysfjord in the NE to about 70 km at the entrance in the SW. Moreover, the boundary between the deeper Vestfjord basin and its shallower coastal area on the east and west side is marked by an up to 300-m high edge (Ottesen et al., 2005; Figure 2). The Ofotfjord and Tysfjord are "*typical*" fjords (Syvitski & Shaw, 1995), with a complex morphology characterized by narrow trenches, steep slopes, and an entrance sill (water depth 140-350 m) where they merge with the Vestfjord (Fløistad et al., 2009). The fjord basins before and behind the sill are elongated and very deep (500-725 m; Figure 2).

The total drainage area of all three fjords spans about 7,100 km^2 (Figure 3) and is characterized by a relatively sparse vegetation cover and an alpine landscape. Precipitation is highest during summer/autumn and lowest in spring. No larger river exists. Runoff is low during winter when inland water is stored as snow, and high during summer due to snow melt and rainfall. The oceanography of the fjord system is very complex as it is locally driven by wind and bathymetry and externally by tides and the adjacent NAC and Norwegian Coastal Current (Furnes & Sundby, 1981; Mitchelson-Jacob & Sundby, 2001). The fjord

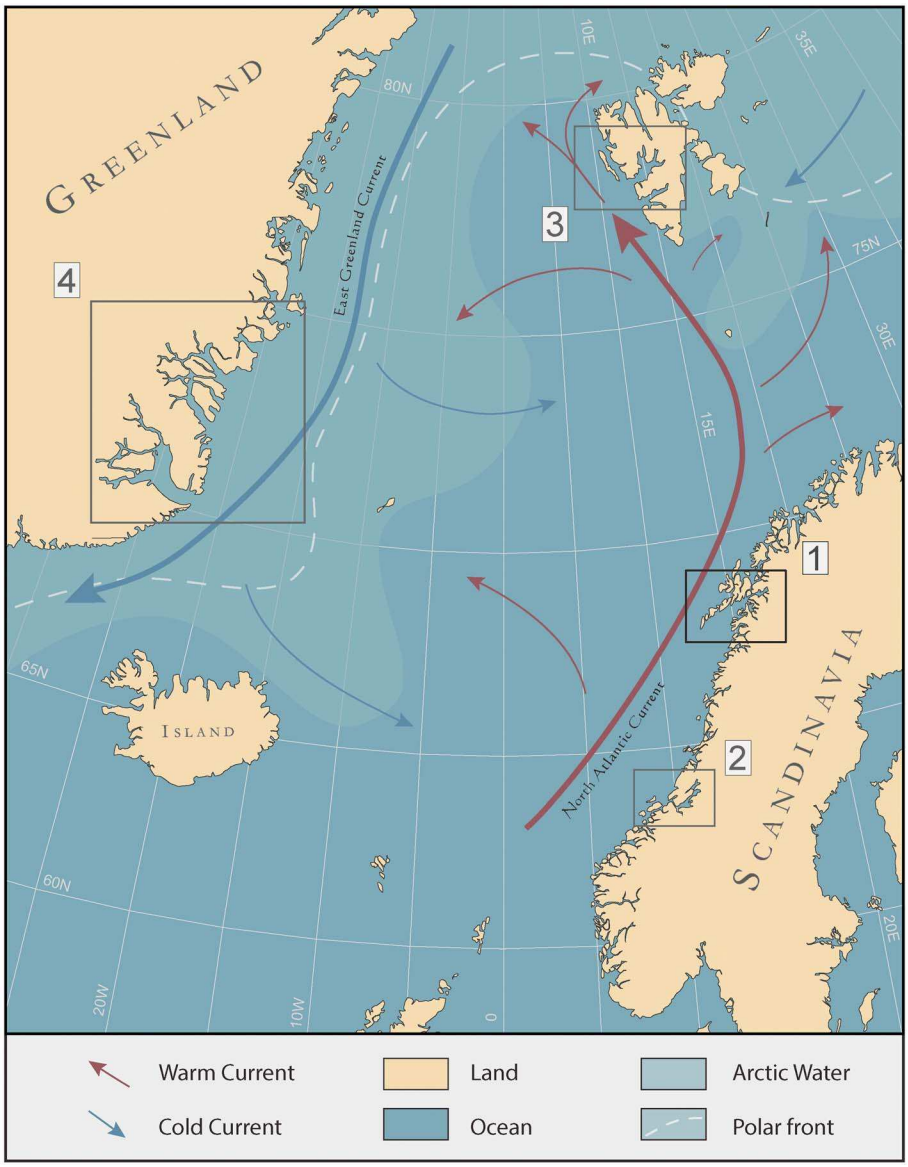

Figure 1. Overview map of the investigated Atlantic fjords: (1) Northern Norway: Vestfjord, Ofotfjord, and Tysfjord; (2) Mid Norway: Trondheimsfjord; (3) Svalbard: Kongsfjord, Isfjord, and Van Mijenfjord; and (4) East Greenland: Hochstetter Bugt, Kong Oscar Fjord, and Scoresby Sund. Sample locations are shown in Figure 2 and in Figure S1. The red arrows indicate the relatively warm northward flowing North Atlantic Current, and the blue arrows represent the colder southward flowing East Greenland Current. The white dotted line displays the atmospheric polar front. The coastline is based on the International Bathymetric Chart of the Arctic Ocean V 3.0 data set (Jakobsson et al., 2012).

estuarine circulation is characterized by an up to 150-m-deep surface water layer and an Atlantic water layer below. There have been no observations of anoxic conditions in these fjords (Gitmark et al., 2014). The general surface circulation can be described by inflowing Atlantic water along the east side (mainland) and an outflow current along the west side (Lofoten) with cyclonic circulation in between (Mitchelson-Jacob & Sundby, 2001). Upwelling and downwelling can be induced by prevailing SW and NE winds on the Lofoten and mainland side along the steep edges in the Vestfjord (Furnes & Sundby, 1981). For further details of the study area we refer to Faust et al. (2017).

2.2. Fjord Surface Sediments: Sampling and Preparation

In June 2014, 39 surface sediment samples were collected at water depths between 59 and 634 m across the Vestfjord, Ofotfjord and Tysfjord (67°40′N, 13°00′E, 68°40′N, 17°40′E; Figure 2 and Table S1 in the

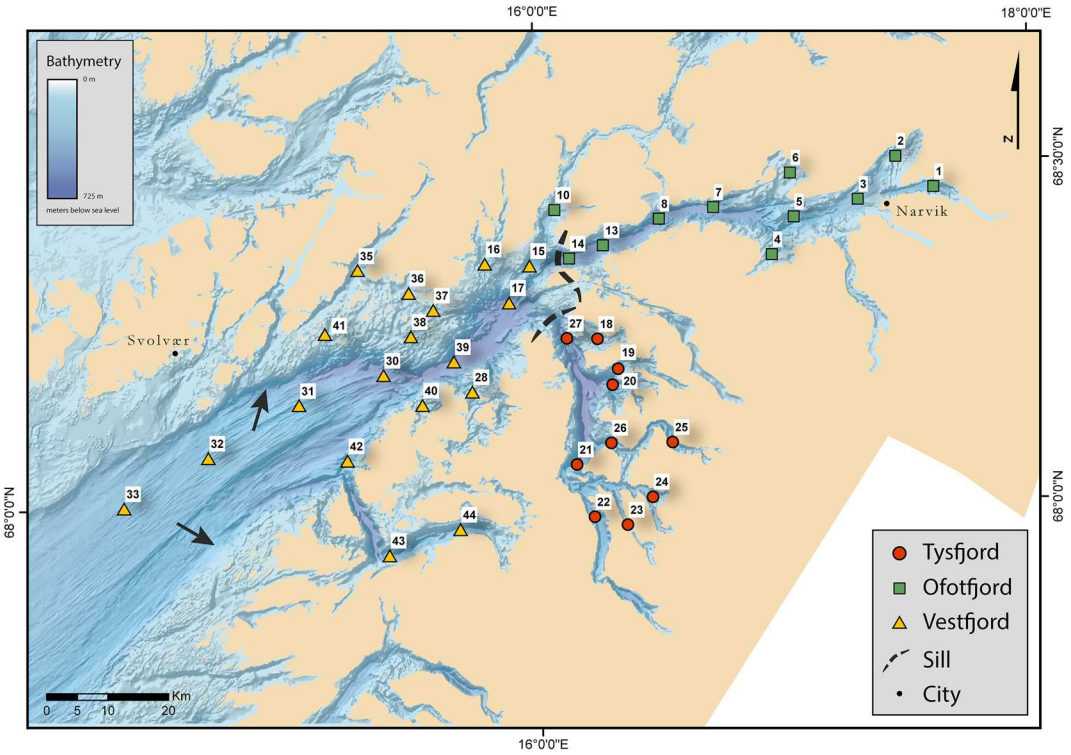

Figure 2. Bathymetry (from mareano.no) and sampling locations in Vestfjord, Ofotfjord, and Tysfjord. The broken black line indicates the position of the sill between the three fjords. The black arrows indicate the up to 300-m high edge between the deeper basin and its shallower coastal areas in the Vestfjord.

supporting information). The first two centimeter of two 5.5-cm-wide multicores were sampled at every station aboard the research vessel "FF Seisma" and stored in plastic bags at -18°C. Prior to further analyses, all samples were freeze-dried and homogenized through gentle grinding.

This new data set is supplemented by additional results from sub-Arctic and Arctic fjord systems in the Nordic Seas: the Trondheimsfjord (see Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014, for details), selected fjords of western Svalbard (see Kumar et al., 2016, and Winkelmann & Knies, 2005, for details), and east Greenland. All unpublished stations are listed in Table S2, and sampling position are shown in Figure S1 in the supporting information. An identical analytical approach as described below has been applied for all samples.

2.3. Organic Carbon Analysis

Analyses for total organic carbon (C_{org}) were performed at the Laboratory of the Geological Survey of Norway (NGU). Weight percentages (wt.%) of C_{org} were determined using the LECO SC-444 (Table S1). Prior to the analysis of C_{org}, sediment subsamples (approximately 200 mg) were transferred into carbon-free pervious ceramic combustion boats. To remove inorganic carbon (carbonate), combustion boats were placed on a heating plate at 50°C (±5°C) and samples were treated with 10% (vol.) hydrochloric acid (HCl). Subsequently, samples were rinsed 10 times with distilled water.

2.4. Nitrogen and Stable Isotope Analysis of Nitrogen and Carbon

Total nitrogen and stable nitrogen isotopes (N_{tot} [wt.%], $\delta^{15}N_{tot}$ [‰versus air]) were determined using an elemental analyzer isotope ratio mass spectrometer (EA-IRMS; Iso-Analytical Ltd., UK). Duplicate measurements of about 20% of the samples produced a standard deviation of 0.002% for N_{tot} (1 sigma, $n = 8$) and 0.07‰ for $\delta^{15}N_{tot}$ (1 sigma, $n = 8$). The inorganic nitrogen (N_{inorg}) and stable inorganic nitrogen isotope ($\delta^{15}N_{inorg}$) content was analyzed on 40mg sediment subsamples treated with KOBr-KOH solution to remove organic nitrogen (see Knies et al., 2007, for details) prior to the analysis using an EA-IRMS (Iso-Analytical Ltd., UK). Precision of the N_{inorg} measurement was 5.35% ($n = 7$) and 6.46‰ for $\delta^{15}N_{inorg}$. The organic

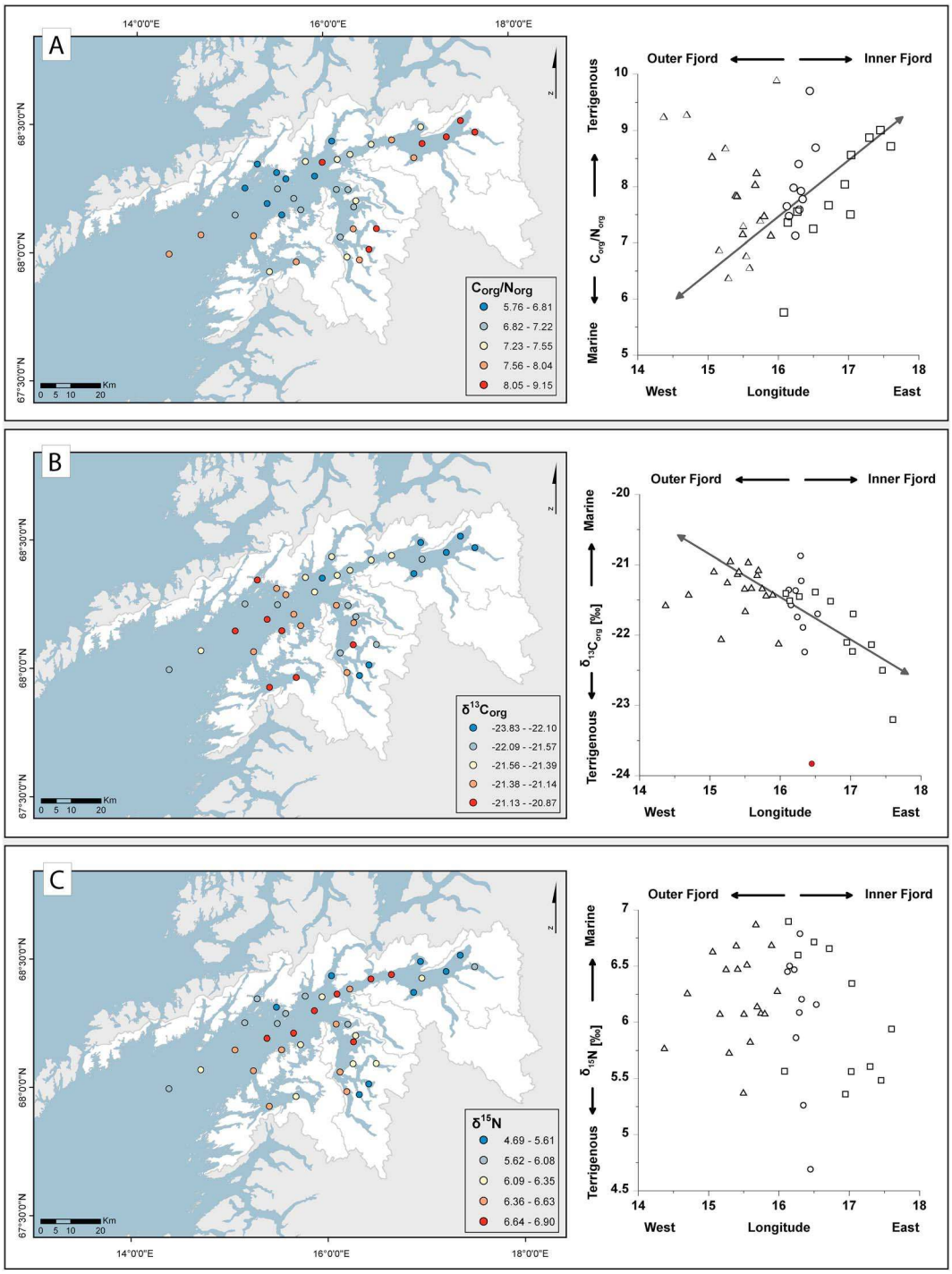

Figure 3. Spatial distribution of (a) C_{org}/N_{org}, (b) $\delta^{13}C_{org}$, and (c) $\delta^{15}N_{org}$ in the Ofotfjord (squares), Tysfjord (circles), and Vestfjord (triangles) surface sediment samples. The white area around the fjord indicates the drainage area of all three fjords.

proportion of the total nitrogen (N_{org}) and stable nitrogen isotope ($\delta^{15}N_{org}$) content were calculated from the measured amounts of N_{tot} and N_{inorg} and the isotopic values of $\delta^{15}N_{tot}$ and $\delta^{15}N_{inorg}$ using a simple isotope mass balance (Schubert & Calvert, 2001). However, after the removal of organic nitrogen almost all of the nitrogen was removed. This meant that the nitrogen available in the samples for isotope analysis was too low for a reliable analysis and the $\delta^{15}N_{inorg}$ data should be used with discretion. As $\delta^{15}N_{tot}$ and $\delta^{15}N_{org}$

show the same signal ($r = 0.97$; Figure S8) we assume that the reliable $\delta^{15}N_{tot}$ equals $\delta^{15}N_{org}$. The results from the nitrogen analyses are shown in Table S1.

Stable carbon isotopes of the C_{org} fraction ($\delta^{13}C_{org}$) were measured on decarbonated (10% HCl) aliquots using an EA-IRMS (Iso-Analytical Ltd., UK). $\delta^{13}C_{org}$ values are given in per mil versus Vienna Peedee belemnite (PDB; Table S1). The applied reference standards were IA-R005 (Beet sugar) with a $\delta^{13}C_{V\text{-}PDB}$ value of -26.03‰, IA-R001 (wheat flour) with a $\delta^{13}C_{V\text{-}PDB}$ value of -26.43‰, and IA-R006 (sugar from cane) with a $\delta^{13}C_{V\text{-}PDB}$ value of -11.64‰. The mean standard deviation for $\delta^{13}C$ of IA-R005, IA-R001, and IA-R006 is 0.24% ($n = 4$), 0.18 % ($n = 8$), and 0.33 % ($n = 4$), respectively.

2.5. *Fraction of Marine Versus Terrigenous* OM *and Organic Carbon Accumulation Rates*

To better estimate the contribution of terrigenous versus marine OM, we combined our $\delta^{13}C_{org}$ and N_{org}/C_{org} values from this study with results from Trondheimsfjord surface sediments located in mid-Norway (see Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014, for further details). This reveals a clear mixing line between marine and terrigenous OM (Figure S5; $r = 0.8$, $n = 99$). Consistent with previous studies (Knies & Martinez, 2009; Winkelmann & Knies, 2005), we used the systematic relationship of these two proxies to define marine and terrigenous $\delta^{13}C_{org}$ end-member values of -19.3‰ and -26.5‰, respectively (see Figure S6 for further details). These values are in accordance with $\delta^{13}C_{org}$-based end-member values in the Arctic region, e.g., -20.1‰ and -26.1‰, respectively, in western Barents Sea surface sediments (Knies & Martinez, 2009). Subsequently, a simple two-end-member mixing model was used to calculate the percentage of allochthonous versus autochthonous OM contribution (Thornton & McManus, 1994):

$$OC_{terr}(\%) = \left(\frac{\delta^{13}C_i - \delta^{13}C_M}{\delta^{13}C_T - \delta^{13}C_M}\right) \cdot 100$$

OC_{terr} is the terrigenous fraction of the OC (%), $\delta^{13}C_i = \delta^{13}C_{org}$ of a given sample, $\delta^{13}C_T$ = terrigenous $\delta^{13}C_{org}$ end-member, and $\delta^{13}C_M$ = marine $\delta^{13}C_{org}$ end-member. The percentage of the marine organic carbon (OC_{mar}) is 100-OC_{terr}.

$\delta^{13}C_{org}$ and N_{org}/C_{org} in surface sediments from the Kongsfjord, Isfjord, and Van Mijenfjord in Svalbard (Figure 1 and Table S2; Kumar et al., 2016; Winkelmann & Knies, 2005) fall along the $\delta^{13}C_{org}$ versus N_{org}/C_{org} mixing line from the Trondheimsfjord and Ofotfjord, Tysfjord, and Vestfjord (Figure S5). As N_{org}/C_{org} measurements from the Hochstetter Bugt, Kong Oscar Fjord, and Scoresby Sund in Greenland are not available, we calculated OC_{mar} for the Greenland and Svalbard fjords by applying the same end-member mixing model as before.

Mass accumulation rates in the Ofotfjord, Tysfjord, and Vestfjord; the Trondheimsfjord; Scoresby Sund; and the Kangerlussuag Fjord region were calculated by using the same porosity (0.77) and bulk density (1.85 g/cm^3) values as by Cui et al. (2016). Sedimentation rates for the Ofotfjord, Tysfjord, and Vestfjord are based on a sediment core recovered at sampling location 31 (Figure 2; Knies & Elvenes, 2018). Sedimentation rates for the Trondheimsfjord, Scoresby Sund and the Kangerlussuag Fjord region are published in Faust, Knies, Milzer, et al. (2014); Marienfeld (1992); and Smith et al. (2002). All values are present in Table S3.

3. Results and Discussion

In order to estimate the relative contributions of marine versus terrigenous OM in the surface sediments of the Ofotfjord, Tysfjord, and Vestfjord, we examine the stable isotope composition of organic carbon ($\delta^{13}C_{org}$) and nitrogen ($\delta^{15}N_{org}$) as well as the organic carbon (C_{org}) versus organic nitrogen (N_{org}) ratio (C_{org}/N_{org}). These parameters have been extensively investigated and successfully utilized in previous studies to differentiate marine from terrigenous OM in fjord and ocean surface sediments (Bertrand et al., 2012; Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014; Goñi et al., 1997; Karageorgis et al., 2005; Knies et al., 2007; Knies & Martinez, 2009; Knudson et al., 2011; Perdue & Koprivnjak, 2007; Sepúlveda et al., 2011; Sepúlveda et al., 2009; Stein & MacDonald, 2004; Winkelmann & Knies, 2005). Previous investigations of fjord surface sediments from Chile, New Zealand, and Norway found clear gradients of

terrigenous versus marine OM from the inner fjords toward the open ocean (Duffield et al., 2017; Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014; Faust et al., 2017; Knudson et al., 2011; Sepúlveda et al., 2011; Silva et al., 2011). These geochemical gradients were associated with two opposing and fundamental processes: the inflow of oceanic water versus the inflow of freshwater from the fjord drainage area (Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014; Faust et al., 2017). As autochthonous and allochthonous OM have different levels of reactivity a geochemical characterization of OM sources is needed to evaluate the cycling of organic carbon in fjord systems. In the following section we first discuss the spatial distribution of C_{org}/N_{org}, $\delta^{13}C_{org}$, and $\delta^{15}N_{org}$ in Ofotfjord, Tysfjord, and Vestfjord surface sediments. We show that the contribution of terrigenous OC is very small even in the innermost parts of the fjords and only a minor inside-outside gradient of $\delta^{13}C_{org}$ and C_{org}/N_{org} can be identified. We then compare our results with OC sources in the sub-Arctic Trondheimsfjord and in Arctic fjords from west Svalbard and east Greenland and discuss the role of the Polar Front system with northward flowing, warm Atlantic water in the east, and southward flowing cold polar surface waters in the west Atlantic (Figure 1).

3.1. OM Sources in the Ofotfjord, Tysfjord, and Vestfjord

3.1.1. C_{org}/N_{org} and $\delta^{13}C_{org}$

C_{org} and N_{org} content in the surface sediment samples varies between 0.47–4.43% and 0.06–0.65%, respectively (Table S1). Despite a very heterogeneous spatial distribution pattern in both parameters, C_{org} and N_{org} are highly correlated ($r = 0.99$, $n = 39$; Figure S2) and show a close to zero intercept. This clearly indicates a common source. Compared to aquatic plants, terrestrial vegetation contains higher proportions of nonprotein materials, that is, cellulose and lignin; hence, C_{org}/N_{org} ratios for terrigenous OC are typically >15 and values for marine OC are <10 (Bordovskiy, 1965; Rullkötter, 2006; Stein & MacDonald, 2004). C_{org}/N_{org} in the Ofotfjord, Tysfjord, and Vestfjord sediments is always <10, even in the inner most parts of the three fjords (Figure 3). Despite the overall low C_{org}/N_{org} values, still a small but clear decreasing gradient from the inner parts of Tysfjord and Ofotfjord toward the open ocean can be identified (Figure 3).

$\delta^{13}C_{org}$ in marine sediments reflects the isotopic composition of the carbon source and the fractionation between ^{12}C and ^{13}C during photosynthesis (Hayes, 1993). As the admixture of C_4 plant types is insignificant in the Arctic region (Collins & Jones, 1986; Still et al., 2003) typical terrigenous OC values of C3 plants in our study area range between -22‰ and -30‰. Marine OC is isotopically enriched in ^{13}C compared to terrestrial C_3 plant material (Arthur et al., 1985). Hence, values for marine OC range between about -17‰ and -22‰ (Descolas-Gros & Fontugne, 1985). Our results show that $\delta^{13}C_{org}$ values in the surface sediments increase slightly from the inner Ofotfjord (-23.2‰, sample 1) toward the central and outer Vestfjord (-21.1‰, sample 31). Highest and lowest $\delta^{13}C_{org}$ values (-20.9‰ [sample 26] and -23.8‰ [sample 24], respectively) of all analyzed samples were found in the Tysfjord sediments (Figure 2 and 3 and Table S1). Except for station 21, samples on the east-side of the Tysfjord are more depleted in $\delta^{13}C_{org}$ than samples on the west-side. Nevertheless, $\delta^{13}C_{org}$ values reveal overall an increasing gradient toward heavier $\delta^{13}C_{org}$ values from the inner fjords toward the outer fjord areas and therefore, an increase in the contribution of marine OM toward the open ocean (Figure 3).

Both the C_{org}/N_{org} and $\delta^{13}C_{org}$ results show that the origin of the sedimentary OM in all parts of the three fjords is predominantly marine. In comparison to other fjords around the world the inside-outside gradients of these parameters are very weak, and a fjord so entirely dominated by marine OM has to our knowledge not been found before.

3.1.2. C_{org}/N_{org} Versus $\delta^{13}C_{org}$

Provenance discrimination is substantially improved by the simultaneous application of two or more organic parameters as potential bias from single analysis is minimized (Jasper & Gagosian, 1990; Sepúlveda et al., 2009; Thornton & McManus, 1994). Since C_{org}/N_{org} and $\delta^{13}C_{org}$ are good indicators for the OC source, several studies from fjords in Chile, New Zealand, and Norway found a strong correlation between both parameters (Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014; Knudson et al., 2011; Sepúlveda et al., 2011; Silva et al., 2011; Winkelmann & Knies, 2005). Yet, C_{org}/N_{org} and $\delta^{13}C_{org}$ in the Ofotfjord, Tysfjord, and Vestfjord sediments show only a weak correlation ($r = 0.4$, $n = 39$; Figure S4; for mathematical reasons we use N_{org}/C_{org} instead of C_{org}/N_{org} for all calculations provided in this manuscript; Perdue & Koprivnjak, 2007). It has been shown that the acid treatment of sediment samples prior to the analysis of C_{org} and $\delta^{13}C_{org}$ may alter the content of OC leading to the production of unreliable data

(Brodie et al., 2011). But the comparison of the C_{org} analysis of two different laboratories (NGU [LECO] and Iso-Analytical [EA-IRMS]) reveals very similar C_{org} results ($r = 0.99$, $n = 39$). Moreover, nitrogen and carbon values can vary considerably with grain size, for example, due to the adsorption of OM on fine particles (Leithold & Hope, 1999). However, no clear relationship between any grain size fraction and C_{org} and N_{org} was found ($r < 0.3$, $n = 39$; grain size data are published in Faust et al., 2017). Additionally, the strong correlation between C_{org} and N_{org} indicates no individual dilution or grain size effect. Only the inorganic nitrogen fraction is strongly related to the clay fraction ($r = 0.8$, $n = 39$) indicating a land-derived origin as suggested earlier (Knies et al., 2007). We believe that the poor correlation between C_{org}/N_{org} and $\delta^{13}C_{org}$ is caused by the low contribution of terrigenous OC. Except for sample 1 and 24 from the innermost parts of the Ofotfjord and Tysfjord (Figures 2 and 3), all $\delta^{13}C_{org}$ and C_{org}/N_{org} results reflect typical $\delta^{13}C_{org}$ results within the range of expected marine OC end-member values in the North Atlantic region (Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014; Knies, 2005; Knies & Martinez, 2009; Winkelmann & Knies, 2005). Moreover, the variability of both parameters is very small. By excluding sample 1 and 24 with the lightest $\delta^{13}C_{org}$ values (-23.2‰ and -23.83‰), all C_{org}/N_{org} and $\delta^{13}C_{org}$ values lie in the range of 7.9±2 and -21.69±0.81‰, respectively. The small nonlinear variations between C_{org}/N_{org} and $\delta^{13}C_{org}$ may simply be caused by factors such as phytoplankton growth rate, cell size, metabolism, variations in ^{13}C content of the carbon source, and also diagenetic alteration (Hayes, 1993; Laws et al., 1995; Rau et al., 1997; Rullkötter, 2006; Talmy et al., 2014). These findings are in agreement with previous investigations, which show that the three fjords and especially the adjacent shelf areas of the Vestfjord are areas of high marine productivity, probably sustained by the inflow of nutrient-rich Atlantic waters as well as upwelling along the steep side-edges of the Vestfjord (Figure 2; Espinasse et al., 2016; Furnes & Sundby, 1981; Höffle et al., 2014; Similä et al., 1996; Sundby & Solemdal, 1984).

3.1.3. *$\delta^{15}N$ of Sedimentary* OM

Nitrogen is an essential element for all organisms and nitrate a major nutrient required by all marine and terrestrial photoautotrophs. The ^{15}N to ^{14}N ratio of most photosynthesizing organisms depends on the isotopic composition of the nitrogenous substrate (e.g., NO_3^-) and the isotopic fractionation during the process of nitrogen assimilation (Wada & Hattori, 1991). In situations when physical supply of nitrate exceeds biological demand, $\delta^{15}N$ of particulate OM is lower than the nitrate source and vice versa (Farrell et al., 1995). $\delta^{15}N$ of marine OM from phytoplankton typically ranges between 3 and 8‰ and clearly differs from lighter terrigenous OM with an average value of 0.4‰ (Peters et al., 1978). In many shelf regions the isotopic signal of the overlying water column is transferred to the seafloor by sinking OM (Sigman et al., 2009). Therefore, $\delta^{15}N$ in marine sediments was successfully applied as a proxy for nutrient utilization in surface waters and OM sources (terrigenous versus marine). However, in places where significant denitrification occurs $\delta^{15}N$ values can be highly depleted or where atmospheric nitrogen fixation is important they can be highly enriched.

$\delta^{15}N$ in Arctic shelf sediments has been successfully applied as tool for tracing allochthonous versus autochthonous OM (Knies et al., 2007; Schubert & Calvert, 2001). Also, in fjord surface sediments from Patagonia, New Zealand, and Scotland $\delta^{15}N$ variations are interpreted as an indicator of OM source, which often shows an inside-outside trend with lower (terrigenous) values in the inner fjord and higher values (marine) at the fjord entrance (Hinojosa et al., 2014; Sepúlveda et al., 2011; Smeaton & Austin, 2017). In the Ofotfjord, Tysfjord, and Vestfjord surface sediments $\delta^{15}N$ content ranges between 4.69‰ and 6.90‰. These values are in the typical range of marine OM (Knies et al., 2007; Peters et al., 1978; Schubert & Calvert, 2001; Sepúlveda et al., 2011) and $\delta^{15}N$ values from shelf surface sediments, which are strongly affected by the NAC (4.0‰–7.0‰; Knies et al., 2007). However, in the Ofotfjord, Tysfjord, and Vestfjord $\delta^{15}N$ neither shows an inside-outside trend nor any other spatial distribution pattern (Figure 3). Moreover, $\delta^{15}N$ is only weakly associated with $\delta^{13}C_{org}$ ($r = 0.6$, $n = 39$) and shows no relationship to C_{org}/N_{org} ($r = 0.1$, $n = 39$). As inorganic nitrogen concentrations are extremely low (<0.02%) the effect of terrigenous nitrogen input on the $\delta^{15}N$ distribution should be negligible. Remarkable though is a strong relationship between $\delta^{15}N$ and water depth ($r = 0.8$, $n = 39$; Figure S7). However, as $\delta^{15}N$ variations occur with depth on very short distances, especially in the Vestfjord, it seems unlikely that the $\delta^{15}N$ distribution reflects changes in the nutrient conditions during OM formation in the surface water layer. The poor relationship of C_{org}/N_{org} and $\delta^{13}C_{org}$ could be caused by variant taxa and growth conditions shifting the degree of

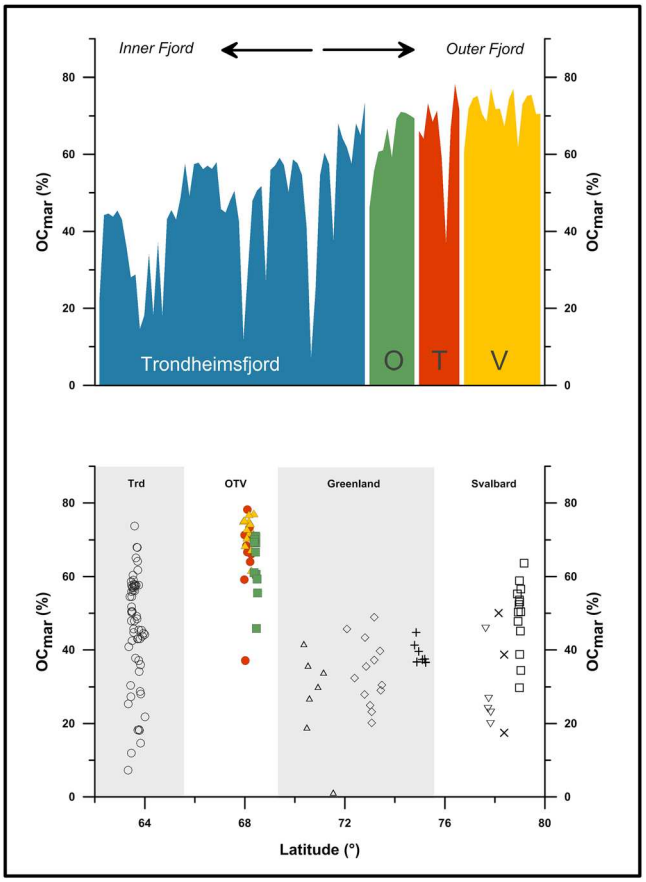

Figure 4. (top) Fraction of marine organic matter (OC_{mar}) in surface sediments from the Ofotfjord (O), Tysfjord (T), and Vestfjord (V) and for comparison from the Trondheimsfjord in mid-Norway. Samples are sorted by the distance to the fjord entrance. (bottom) Fraction of marine organic matter (OC_{mar}) for the Trondheimsfjord (Trd); Ofotfjord, Tysfjord, and Vestfjord (OTV); East Greenland fjords: Scoresby Sund (triangle), Kong Oscar Fjord (open diamond), and Hochstetter Bugt (plus); Svalbard fjords: Van Mijenfjord (triangle), Isfjord (cross), and Kongsfjord (open square).

isotopic fractionation associated with primary productivity (Bickert, 2006). However, Robinson et al. (2012) compared $\delta^{15}N$ from sediment traps and surface sediments on a global scale and found that the alteration of $\delta^{15}N$ toward higher values appears to be a function of water depth. They attributed this relation to different oxygen exposure times between the shallower and deeper deposited nitrogen. When oxygen concentrations are low bacterial reduction of nitrate to N_2 (denitrification) occurs, which strongly increases the $\delta^{15}N$ in the remaining OM (nitrate) pool (Altabet & François, 1994; Sigman et al., 2009). If the oxygen exposure time is responsible for the strong relationship between $\delta^{15}N$ and water depth in the Ofotfjord, Tysfjord, and Vestfjord, this would require rapid changes of the bottom water oxygen concentration, sedimentation rates, and/or OM accumulation rates on very short spatial distances (Figure 3). But, water masses of the fjords investigated here are well mixed. Suboxic or anoxic conditions in the deeper fjord basins have never been reported or observed during fieldwork. Therefore, it is also unlikely that bacterial OM denitrification already in the water column causes the increase of $\delta^{15}N$ with water depth (Bickert, 2006; Robinson et al., 2012; Sigman et al., 2009). Nevertheless, Faust et al. (2017) investigated the inorganic composition of the Ofotfjord, Tysfjord, and Vestfjord sediments and found strong indication that distance and time between erosion and sedimentation are short, and therefore, sedimentation may be very low in the deeper centre of the fjords. A longer exposure time of the OM could increase $\delta^{15}N$ alteration by denitrification.

In summary, our C_{org}/N_{org}, $\delta^{13}C_{org}$, and $\delta^{15}N_{org}$ results show that the entire Ofotfjord, Tysfjord, and Vestfjord are dominated by marine OM. The contribution of terrigenous OM is very low but can still be observed from the slight inside-outside trend of C_{org}/N_{org} and $\delta^{13}C_{org}$.(Figure 3). In the following section we show that in comparison to other sub-Arctic and Arctic fjords the Ofotfjord, Tysfjord, and Vestfjord seem to be more strongly dominated by marine OM.

3.2. Source of OM in North Atlantic Fjords

The primary allochthonous OM source to fjords is from the surrounding drainage area (Burrell, 1988). Hence, differences in topography and climate have often been interpreted to be responsible for variations in the amount of terrestrial versus marine OM in different fjords. Our results show that the percentage of marine OC (OC_{mar}) in the Ofotfjord, Tysfjord, and Vestfjord sediments varies between 37% and 78% with an average value of 68%. In comparison, Trondheimsfjord OC_{mar} values vary between 7% and 74% with an average value of 46% (Figure 4 and Table S1; Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014). These findings indicate that the relative contribution of marine OC to fjords in Norway can be much larger than previously suggested for fjords in NW Europe (24%) and also globally (38–45%; Cui et al., 2016). One potential explanation is that all fjords selected by Cui et al. (2016) from Norway and Sweden (NW Europe) are characterized by a stratified water column and anoxic sediments (Huguet et al., 2007; Müller, 2001; Nordberg et al., 2001; Nordberg et al., 2009; Skei, 1983; Velinsky & Fogel, 1999). Thus, the low contribution of marine OC in the fjords studied by Cui et al. (2016) could be related to lower primary productivity in these fjords due to water column stratification caused by a relatively weak inflow of oceanic water or intense river runoff (Inall & Gillibrand, 2010). To further evaluate these findings, we integrated additional new and published OC_{mar} data from Arctic fjords in Svalbard and Greenland (Table S2; Kumar et al., 2016; Winkelmann & Knies, 2005).

We found that in east Greenland fjords OC_{mar} values range between 19 and 49% (Figure 4), except for one sample with OC_{mar} = 0.9% from the Scoresby Sund (PS1939-1; Table S2 and Figure S1). OC_{mar} in the

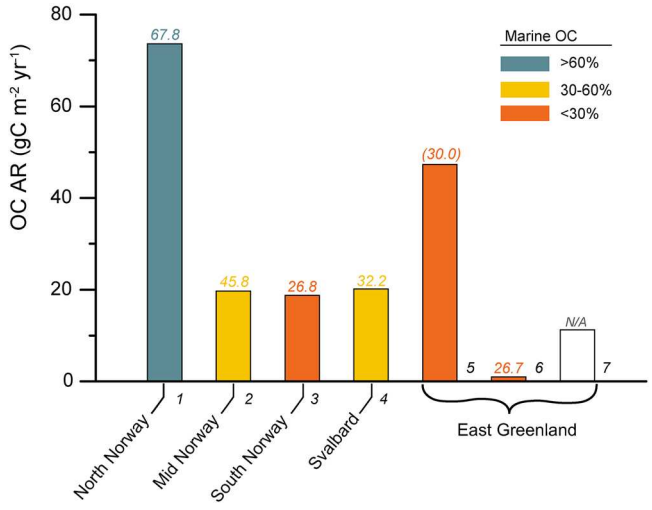

Figure 5. Organic carbon accumulation rates (OC AR) and dominant OC source for the (1) Ofotfjord, Tysfjord, and Vestfjord (this study); (2) Trondheimsfjord (Johan C. Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014); (3) Nordaesvannet Fjord, Kyllaren Fjord, Drammensfjord, and Framvaren Fjord (X. Q. Cui et al., 2016); (4) Hornsund, Von Keulenfjord, Kongsfjord, Storfjord, and Smeerenburgfjord (X. Q. Cui et al., 2016 and this study); (5) Yound Sound, Kangerlussuaq, Miki Fjord, and Nansen Fjord (X. Q. Cui et al., 2016; % of marine OC only available for the Yound Sound); (6) Scoresby Sund (Marienfeld, 1992, and % of marine OC from this study); and (7) Kangerlussuaq Fjord region (L. M. Smith et al., 2002).

Isfjord and Van Mijenfjord in Svalbard vary between 18% and 50% and are slightly higher than OC_{mar} values of 19–31% recently reported from the Hornsund fjord in the south-west of Svalbard (Koziorowska et al., 2016). This shows that the marine OC contribution in Greenland fjords, Isfjord, and Van Mijenfjord is overall lower than in the Ofotfjord, Tysfjord, and Vestfjord and is similar to the inner Trondheimsfjord and its river deltas (Figure 4). Compared to the other Arctic fjords the contribution of marine OC is highest in Kongsfjord surface sediments with OC_{mar} values of up to 64% (Figure 4). Indeed, this may be related to its unusual physical properties for a fjord in such high latitudes (79°N). In opposition to other Arctic fjords the Kongsfjord is strongly affected by the inflow of the relatively warm and saline northern most extension of the NAC (Figure 3). This increases water temperature and can induce enhanced vertical water mixing, transporting nutrients to the photic zone, which can enhance marine primary productivity. Consequently, the Kongsfjord features many sub-Arctic environmental characteristics, which leads to unusual presence of different boreal species in the fjord (Hodal et al., 2012; Hop et al., 2002; Svendsen et al., 2002; Willis et al., 2006). Also, the Ofotfjord, Tysfjord, and Vestfjord experience substantial inflow of oceanic water by trapping the northward flowing NAC between the Lofoten and the Norwegian mainland (Figure 1; Furnes & Sundby, 1981; Mitchelson-Jacob & Sundby, 2001).

3.3. Estimation of Carbon Burial Rates

A comparison of organic carbon accumulation rates form the Ofotfjord, Tysfjord, and Vestfjord and Trondheimsfjord with accumulation rates from North Atlantic fjords in Svalbard and east Greenland (Cui et al., 2016; Smith et al., 2015) reveals extremely high marine carbon burial rates (73.6 $gC \cdot m^{-2} \cdot year^{-1}$) in the Ofotfjord, Tysfjord, and Vestfjord (Figure 5 and Table S3). In these Arctic fjords primary productivity is strongly affected by the annual seasonal cycle and is therefore close to zero during winter due to very low solar irradiance and short day length. Thus, the high burial rates of mainly marine OC indicate a fast and direct carbon sequestration from the atmosphere into the sediments. This makes these fjords probably to a very efficient carbon burial hot spot.

In contrast, carbon accumulation rates are considerably lower in the Trondheimsfjord (19.7 $gC \cdot m^{-2} \cdot year^{-1}$) but still in the same order of magnitude as in fjords from Svalbard and south Norway (Figure 5 and Table S3). However, even though burial rates are very similar the efficiency of the carbon storage may be different between these fjords as the Trondheimsfjord shows a strong marine versus terrigenous OC gradient and the dominant OC source in Svalbard fjords varies from fjord to fjord (Figure 4). Hence, further investigations are required to better understand the impact of marine versus terrigenous OC on the efficiency of carbon burial in these fjords. Estimating carbon burial rates in east Greenland fjords is challenging due to extreme seasonal and spatial variations in sedimentation rates (Cui et al., 2016). The global fjord carbon burial data set from Cui et al. (2016) and Smith et al. (2015) indicates carbon burial rates in east Greenland to be more than twice as high as in Norwegian or Svalbard fjords (Figure 5). However, based on publications from Smith et al. (2002) and Marienfeld (1992), we found carbon burial rates in east Greenland (Scoresby Sund and Kangerlussuaq region) to be much lower than in Norway or Svalbard. This indicates that the inflow of warm and nutrient-rich seawater is a strong factor for marine primary productivity and has a pronounced impact on carbon burial in Atlantic fjords, particularly in fjords with low OM content in the drainage area. However, this needs to be confirmed by further investigations, especially of Greenland fjords, to gain more and better data for carbon burial rate calculations.

3.4. Fjord Oceanography Is an Important Control on OM Composition

It is generally assumed that changes in marine OM input in fjords are mostly controlled by two opposing and fundamental processes: (a) the inflow of freshwater and (b) the inflow of oceanic waters sustaining marine organisms via nutrient supply and its physical conditions. This becomes apparent by the comparison of the

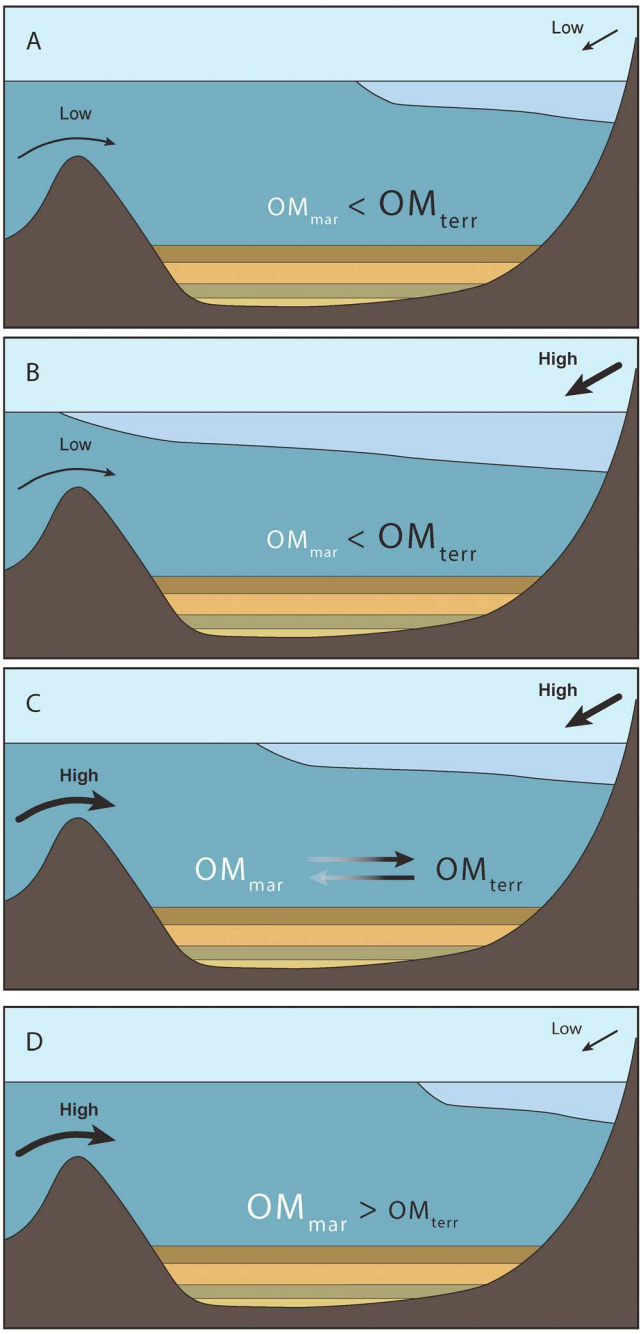

Figure 6. Sketch of fjords typical bathymetry and estuarine circulation pattern where oceanic water enters the fjord across an entrance sill (left) and the freshwater input in the inner part (right) creates a brackish surface water layer. (a and b) In fjords with low marine and low freshwater inflow as well as in fjords with high river runoff but relatively low marine inflow are dominated by terrigenous organic matter (OM; OM_{terr}). (c) Fjords were both, marine inflow and freshwater runoff are high, have a substantial inside-outside gradient of terrigenous versus marine OM. (d) And if the marine inflow is high and river runoff is low, fjord sediments are dominated by marine OM (OM_{mar}).

marine OC distribution in the Ofotfjord, Tysfjord, and Vestfjord with the Trondheimsfjord. In contrast to very low contribution of terrigenous OC in the northern Norwegian Ofotfjord, Tysfjord, and Vestfjord, recent investigations of surface sediments from the Trondheimsfjord in mid Norway revealed a clear trend of marine versus terrigenous OC contribution from the inner to the outer fjord (Figure 4; Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014). One of the main differences between these two fjord systems is the ratio of total drainage area to fjord surface area. With a value of 1.3 this ratio is very small in the Ofotfjord, Tysfjord, and Vestfjord compared to the value of 14 for the Trondheimsfjord. In accordance to this finding Hinojosa et al. (2014) suggested that the lack of a geochemical gradient in a New Zealand fjord (Nancy Sound) could be related to its small catchment size to fjord area ratio of 6.6 causing low freshwater inflow. Remarkably, the Nancy Sound is dominated by terrigenous OM, which was interpreted to be caused by low marine water intrusion. These results highlight the importance of the marine water inflow versus freshwater runoff as important controlling factors of the OM composition in fjord sediments, which affects the OM stability and therefore the carbon burial efficiency in fjord sediments. We suggested that based on these findings fjords can generally be categorized in four settings illustrated in Figure 6: OM in fjords with low marine and low freshwater inflow is terrestrial dominated as well as in fjords with high runoff and relatively low marine inflow (Figure 6a and 6b). Examples for this setting are the Greenland fjords used in this study, the fjords from NW Europe used by Cui et al. (2016), and maybe also the Nancy sound in New Zealand (Hinojosa et al., 2014). These fjords are also likely to reveal anoxic conditions in the bottom water layer. Fjords where both marine inflow and freshwater runoff are high (Figure 6c) have a substantial inside-outside gradient of terrigenous versus marine OM, for example, the Trondheimsfjord (Faust, Knies, Milzer, et al., 2014; Faust, Knies, Slagstad, et al., 2014) and fjords from Patagonia (Sepúlveda et al., 2011). As shown here for the Ofotfjord, Tysfjord, and Vestfjord, if the NAC inflow is high and river runoff is low (Figure 6d) fjord sediments are dominated by marine OM. This implies that changes in fjord oceanographic settings, for example, due to changes in the NAC strength induced by ongoing climate change are likely to have a pronounced effect on carbon accumulation in fjords.

4. Concluding Remarks

Recently, fjords have been recognized as an important area for carbon burial and therefore as a major component of global carbon cycles and budgets. Thus, it is essential to understand the origin, transport, and character of OM entering fjords to accurately constrain carbon burial rates. Fjords are transitional regions connecting terrestrial with oceanic systems, which typically leads to a gradient of terrigenous versus marine related geochemical parameters from the inner to the outer fjord in sediments and in the water column. As autochthonous and allochthonous OM have different levels of reactivity a geochemical characterization of OM sources is needed to evaluate the cycling of organic carbon in fjord systems. The investigation of the provenance of the OM in fjords from middle and northern Norway, Svalbard, and east Greenland, in this study, reveals that the fjord oceanographic setting has a strong impact on the fraction of sedimentary marine OM in North Atlantic fjords. Moreover, in contrast to previous evidence, our findings show that not in all fjords the

majority of OM is terrigenous and that OM sources in North Atlantic fjords are much more heterogeneous than previously estimated. Fjords with high inflow of relatively warm and nutrient-rich Atlantic currents in Norway and partly in Svalbard can feature very high fractions of marine OM compared to fjords, for example, from Greenland. This indicates that even though fjords are generally characterized by high sedimentation rates and large OM fluxes, their carbon sequestration efficiency may vary in accordance with their environmental setting.

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