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Geochemical characterisation of northern Norwegian fjord surface sediments: a baseline for further paleo-environmental investigations

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Abstract

Norwegian fjord sediments are promising archives for very high resolution records of past environmental changes. Recent investigations of the modern depositional environment within fjords revealed that the accurate quantification of the inputs, sources, and sedimentary preservation of organic and inorganic material is crucial to decipher long term past climate signals in the sedimentary record of a certain fjord. Here, we investigate the elemental composition, bulk mineral assemblage and grain size distribution of forty-one surface sediment samples from a northern Norwegian fjord system. We reveal modern geochemical and sedimentological processes that occur within the Vestfjord, Ofotfjord and Tysfjord. Our results indicate a very heterogeneous sediment supply and intricate sedimentation processes. We propose that this is related to the complex fjord bathymetry, a low hydrodynamic energy environment, differences in the hinterland bedrock composition and a relatively small drainage area causing a rather diffuse freshwater inflow. Moreover, we show that marine carbonate productivity is the main calcite and Ca source in all three fjords.

1 Introduction

Sediments accumulating in fjords have the potential to be one of the best high-resolution archives of climate and local environmental changes (Howe et al., 2010). High sedimentation together with the possibility to quantify the fjord's hydrological cycle

(freshwater input and marine water exchange) offer an excellent opportunity for studying land-ocean interactions and can provide ultra-high-resolution records of local responses to short-term climate variability (Faust et al., 2016; Forwick and Vorren, 2007; Hald and Korsun, 2008; Howe et al., 2010; Husum and Hald, 2004; Kristensen et al., 2004; Mikalsen et al., 2001; Paetzel and Dale, 2010; Syvitski, 1989).

In general, it is assumed that changes in precipitation and temperature alter the constitution of fluvial sediment flux, generated by weathering and erosion of bedrock and soils, from land towards ocean basins (e.g. Govin et al., 2012; Lamy et al., 2001; White and Blum, 1995). However, a detailed knowledge of the controlling transport mechanisms of the particle supply is required to explore the relationship between terrigenous input and changes in environmental conditions (Zabel et al., 2001). Sediment characteristics, accumulation and distribution vary with climate, seafloor topography, basin geometry, size of the drainage area, oceanographic regime, and distance to river outlets (e.g. Syvitski et al., 1987). Thus, identifying the provenance of the sediment components is the key factor to determine and reconstruct (1) sea-level changes, (2) hinterland weathering processes, (3) climate variability and (4) anthropogenic influences. For this reason numerous studies have focused on the contribution of organic carbon (e.g. Goñi et al., 1997; Knies and Martinez, 2009; Sargent et al., 1983; Stein and MacDonald, 2004; Winkelmann and Knies, 2005) and trace elements (e.g. Calvert et al., 1993; Cho et al., 1999; Govin et al., 2012; Hayes, 1993; Hirst, 1962; Karageorgis et al., 2005; Kim et al., 1999; Mil-Homens et al., 2014; Pe-Piper et al., 2008) in continental shelf sediments.

Fjords comprise a substantial part of the coastal environments and are important sites for carbon burial due to their high inorganic and organic sedimentation rates (Hedges et al., 1997; Knies, 2005; Knudson et al., 2011; Ludwig et al., 1996; Raymond and Bauer, 2001; Sepúlveda et al., 2011; Smith et al., 2015; St-Onge and Hillaire-Marcel, 2001; Syvitski et al., 1987) but only a very few studies exist using surface sediments to investigate modern fjord environmental settings. Studies from fjords in Chile (Bertrand et al., 2012; Sepúlveda et al., 2011; Silva et al., 2011), New Zealand (Hinojosa et al., 2014; Knudson et al., 2011; Smith et al., 2015), east Greenland (Andrews and Vogt, 2014) and Svalbard (Winkelmann and Knies, 2005) reported a significant influence of freshwater inflow on their geochemical composition and suggest a common decreasing gradient of terrigenous-derived organic- and inorganic material from the inner fjords towards the open ocean. In contrast to these

findings, Munoz and Wellner (2016) found terrigenous deposits to occur predominantly in the outer bay of an Antarctic fjord. This indicates that the processes controlling the supply and composition of the inorganic sediment fraction of fjords sediments may vary from fjord to fjord. More investigations of fjord sediments are therefore required to recognize and better understand these differences.

Overall, little is known about seasonal and bathymetry-related changes in sedimentation of particulate material in Norwegian fjords. Recent investigations of surface sediment samples from the Trondheimsfjord, mid-Norway, revealed that not only does the input of terrigenous material vary in proximity to their source but also geochemical composition of the material changes with regard to the hinterland geology (Faust et al., 2014b). Moreover, as shown by Faust et al. (2016; 2014a) a detailed study of the modern environmental fjord setting provides fundamental knowledge necessary to interpret climatic signals in long term fjord sediment sequences.

Similar to the Trondheimsfjord, the regional climate of Vestfjord, Ofotfjord and Tysfjord in northern Norway is strongly influenced by the relatively warm northward flowing North Atlantic Current (NAC) and the atmospheric circulation pattern is dominated by the North Atlantic Oscillation (NAO) (Hurrell, 1995). Thus, sediments from these fjords may contain valuable information about regional past climate changes caused by NAO and NAC variability. Moreover, although the study area was an important pathway for ice-sheet drainage during the late Weichselian (Ottesen et al., 2005), ice-sheet dynamics during the Younger Dryas period in the Vestfjord, Ofotfjord and Tysfjord area is still under debate (Bergstrøm et al., 2005; Fløistad et al., 2009; Knies et al., 2007; Rasmussen, 1984). Hence, the identification of geochemical or mineralogical provenance proxies could help to better understand the deglaciation history in this area. The hinterland geology, bathymetry and oceanography of the Vestfjord, Ofotfjord and Tysfjord are overall similar to the intensively investigated Trondheimsfjord in mid Norway. Our hypothesis is that these similarities of the environmental settings make the Vestfjord, Ofotfjord and Tysfjord sediments a promising archive for paleo-environmental studies. To test our hypothesis, we investigate geochemical, mineralogical and sedimentological data obtained from forty-one surface sediment samples from the Vestfjord, Ofotfjord and Tysfjord in northern Norway (Fig. 1). Our goal is to acquire a better understanding of the modern processes that control the

supply and composition of the inorganic sediment fraction of the fjords. We discuss the general trends within these deposits, assess how local variations affect sediment distribution and provide implications for paleo-environmental interpretations.

2 Regional setting

The Vestfjord, Ofotfjord and Tysfjord are the three main fjords of a fjord system between the Norwegian mainland and the Lofoten archipelago in northern Norway (Fig. 1). With a length of about 180 km and its cone shape the Vestfjord is an "*atypical*" fjord and has characteristics more similar to a coastal bay (Fig. 2, Mitchelson-Jacob and Sundby, 2001). The fjord becomes shallower and widens from about 15 km at its junction with the Ofotfjord and Tysfjord in the NE to about 70 km at the entrance in the SW. Moreover, the boundaries between the deeper Vestfjord basin and its shallower coastal areas on the SE and NW sides are marked by up to 300 m high side-edges (Fig. 1). This is interpreted to be the result of enhanced glacial erosion of the downfaulted Vestfjord basin as the Vestfjord functioned as a major ice-sheet drainage route during the last glacial period (Ottesen et al., 2005). The Ofotfjord and Tysfjord morphologies are, as is typical for fjords, characterised by narrow trenches, steep slopes and an entrance sill with varying water depths of 140–350 m (Fløistad et al., 2009). The fjord basins on both sides of the sill are elongated and remarkably deep (500–725 m, Fig 1).

The total drainage area of the three fjords spans about 7,100 km² (Fig. 2) and is marked by a relatively sparse vegetation cover and an alpine landscape. Mountains in this region are frequently higher than 1000 m and several small glaciers are present in the drainage area of the Tysfjord and Ofotfjord (Fig. 2). February air temperatures (monthly average) are around 0°C at the coast and minus 5 to minus 10°C in the hinterland. During August, hinterland air temperature (monthly average) rises to about 14–15°C and around 11°C at the coast. Precipitation varies strongly over short distance with topography (500–2000 mm/a) and is highest during summer/autumn and lowest in spring (The Norwegian Meteorological Institute (met.no)). No large river exists and the runoff is generally low during winter when inland water is frozen and high during summer due to snow melt and rainfall. On average two thirds of the annual runoff occurs from June to August (Mitchelson-Jacob and Sundby, 2001). For more detailed information of the topography, rivers and further hydrological

information of the drainage area, we refer to the Norwegian Mapping Authorities (<http://kart.statkart.no>) and the Norwegian Water Resources and Energy Directorate (<http://atlas.nve.no>).

The oceanography of the fjord system is driven locally by wind and bathymetry and regionally by tides and the adjacent North Atlantic and Norwegian Coastal Current systems (Furnes and Sundby, 1981; Mitchelson-Jacob and Sundby, 2001). Due to the seasonal variation of freshwater supply, temperature and salinity of the surface water layer (up to 150 m deep) vary between 2–4°C and 33–34 (PSU) during winter and about 14°C and 28 (PSU) during summer. The surface layer overlies an Atlantic water layer, which has constant temperatures and salinity of 6.5–7°C and 34.7–35 (PSU) throughout the year. There are no observations of anoxic conditions in the fjords (Gitmark et al., 2014). The general surface circulation can be described by inflowing Atlantic water along the southeast side (mainland) and an outflow current along the northwest side (Lofoten) with cyclonic circulation in between (Mitchelson-Jacob and Sundby, 2001). Yet, this major current regime is strongly affected by the dominant wind direction. SW winds reverse the flow direction and may induce upwelling on the Lofoten side and downwelling on the mainland side (Fig. 2). Additionally, the SW winds cause an enhanced flow of upper water masses into the Vestfjord, Ofotfjord and Tysfjord, which presses the underlying Atlantic water out of the fjords (Furnes and Sundby, 1981). NE winds cause the opposite effect. They force the upper water layer out of the fjords which results in an inflow of Atlantic water and may induce downwelling on the Lofoten side and upwelling on the mainland side (Fig. 2).

The bedrock geology in the drainage areas of the Vestfjord, Ofotfjord and Tysfjord can be subdivided into Precambrian basement units and overlying Caledonian nappes (Fig. 2). The basement is largely composed of Paleoproterozoic plutonic rocks of the anorthosite-mangerite-charnockite-granite (AMCG) suite intruding older metamorphic rocks (Corfu, 2004). The Caledonian nappes predominantly contain metamorphosed Ordovician-Silurian sediments such as micaschist, metasandstone and subordinate marble (Andresen and Steltenpohl, 1994; Corfu et al., 2014).

3 Methods and Data

3.1 Fjord surface sediments: sampling and preparation

In June 2014, forty-one surface sediment samples were collected at water depth between 59 and 634 m across the Vestfjord, Ofotfjord and Tysfjord ($67^{\circ}40'N$, $13^{\circ}00'E$, $68^{\circ}40'N$, $17^{\circ}40'E$) (Fig. 1 and ES-1). In general, sediments are mainly transported by rivers into fjords and the main controlling factors of their distribution are the bathymetry of the fjord and oceanography. Therefore, sampling locations were selected based on water depth, distance to river inlets and coastal water inflow in order to cover the entire sedimentary regime of the three fjords. Moreover, each sampling site has been investigated using a TOPAS (Topographic Parametric Sonar) prior to coring to avoid turbidites or other large disturbance in the sediment structure. The first two centimetres of two 5.5 cm wide multicores were sampled at every station aboard the research vessel "FF Seisma" (Geological Survey of Norway) and stored in plastic bags at minus $18^{\circ}C$. Prior to analyses, all samples were freeze-dried and, except for grain size measurements, homogenised through grinding.

3.2 Bulk elemental geochemistry and grain size analyses

Four combined analytical methods were used to quantify major and trace elements at the ALS Geochemistry Laboratories in Loughrea, Ireland. A subsample of 0.9 g was added to 9 g of Lithium Borate Flux, well mixed and fused between 1050 to $1100^{\circ}C$. A molten disc was prepared from the resulting melt and analysed for the major elements (Al, Ca, Fe, K, Mg, Mn, Na, P, Si, Ti) by X-Ray Fluorescence Spectroscopy (XRF). For the analysis of Ba, Ce, Cr, Cs, Dy, Er, Eu, Ga, Gd, Hf, Ho, La, Lu, Nb, Nd, Pr, Rb, Sm, Sn, Sr, Ta, Tb, Th, Tm, U, V, W, Y, Yb and Zr a 0.2 g subsample was added to 0.9 g of lithium metaborate flux and fused at $1000^{\circ}C$. The resulting melt was cooled and dissolved in 100 ml 4% HNO_3 /2% HCl and the solution was subsequently analysed by inductively coupled plasma mass spectrometry (ICP-MS). Determination of Ag, Cd, Co, Cu, Li, Mo, Ni, Pb, Sc and Zn was performed by inductively coupled plasma atomic emission spectroscopy (ICP-AES) following a four-acid total digestion method. 250 mg of each sample was heated ($200^{\circ}C$) in a mixture of H_2O -HF-HClO₄-HNO₃ (2:2:1:1) and the residue was dissolved in hydrochloric acid (50%) and analysed.

The determination of the grain size distribution was performed by laser diffraction using a Coulter LS 13320 instrument at the Geological Survey of Norway. The analysis was carried out on material within a particle diameter range of 0.4–2000 µm and the results are presented as cumulative volume percentage. Prior to the grain size analyses, sediment samples were decarbonated using 10% (vol.) hydrochloric acid (HCl), organic matter was removed using hydrogen peroxide (H₂O₂) and to prevent particles becoming charged and agglomerated all samples were treated with 5% sodium pyrophosphate (Na₄P₄O₇ • 10H₂O, Merck PA). Samples were then placed in an ultrasonic bath until analysis. We assume that biogenic silica has a negligible effect on the grain size measurement because (I) the biogenic silica content in North Atlantic sediments is generally very low (e.g. Schlüter and Sauter, 2000), (II) biogenic silica content in Trondheimsfjord sediments (Mid-Norway) was found to be very small (Faust et al., 2014b) and (III) our XRD analysis reveal only traces of amorphous silica (<0.3%) in just six samples.

3.3 *Bulk mineral assemblage*

Bulk mineral assemblages were measured via X-ray diffraction (XRD) using a Bruker D8 Advance diffractometer with Cu K_α radiation and a Lynxeye XE detector at the Geological Survey of Norway, Trondheim, Norway. XRD scans were carried out for 3–75° 2θ and a step size of 0.02°. Signal acquisition time was 1 s per step. The optical system was equipped with soller slits (2.5°) and fixed divergence and antiscatter slits (0.6 mm). Quantification of the mineral content was carried out with Quantitative Phase-Analysis with X-ray Powder Diffraction (QUAX) (details are given in Vogt et al., 2002). Bulk mineral assemblages from station 29 are missing due to insufficient sediment material.

4 Results and Discussion

4.1 Grain size and mineral assemblages

Grain size distribution in marine sediments is generally a good indicator for sediment erosion, transport and deposition and mineral assemblages. Changes in sediment grain size may also provide information about the sediment origin. The bulk mineral assemblages of the Vestfjord, Ofotfjord and Tysfjord sediments consist mainly of phyllosilicates (23%) and

plagioclase (22%) followed by quartz (14%), calcite (13%) and potassium feldspars (9%) (ES-1 and ES-2). However, the mineral content varies considerably from sample to sample (7%–50% phyllosilicates, 2%–36% plagioclase, 1%–31% quartz, 1%–51% calcite). Moreover, sediments are generally fine-grained (<63 µm) in the inner part of the Ofotfjord (station 1–3, 5) and in the deepest part of the Vestfjord (station 17, 39, 30) (Fig. 3). Samples from the inner part of the Tysfjord (station 23–26), the sill (station 14, 15) and partly from the shelf areas of the Vestfjord (station 38, 41, 28) are more coarse-grained (>125 µm). On average, all surface sediments samples consist mainly of the 2–63 µm (average 74%) and 63–125 µm (average 15%) fractions. However, the amount of the 2–63 µm and 63–125 µm fraction varies between 27%–93% and 1%–40%, respectively (Fig. 3). Moreover, the mean grain size distribution resembles the 2–63 µm fraction ($r = 0.95$, $n = 41$) and all other fractions are on average ≤5%.

The variable mineral- and grain size distribution in the surface sediments indicates a very heterogeneous sediment supply and complex sedimentation processes. In general, it is assumed that the distribution of sediments within a fjord is largely controlled by its bathymetry, depth and hydrographic regime (Howe et al., 2010). However, in our study area neither grain size nor specific mineral assemblages show any characteristic spatial distribution pattern (Fig. 3 and ES-2). Exceptions are the clay fraction (<2 µm) as well as phyllosilicates, expandable- and mixed layer clays, which reveal statistical relevant Pearson correlation coefficients with water depth ($r = 0.5$ – 0.7 , $p < 0.01$, $n = 40$, ES-1). This relationship is in accordance with the general assumption that the deepest parts of a fjord are in general the lowest energy environments (Syvitski, 1989) and therefore, favour the settlement of fine-grained material. Due to longer transport distances, we would further expect the portion of the fine-grained material to increase with increasing distance from the coast. However, grain size and quartz content in the Vestfjord sediments increase towards the open ocean (Station 30–33). We assume that this indicates that stronger bottom water currents caused winnowing and sediment redistribution in the outer part of the fjord.

The local mineral and grain size distribution shows a complex distribution pattern which can be explained by the physiographic setting of the study area. Considerable changes in land topography cause irregular precipitation over very short distances and the relatively small drainage area of the three fjords prevent longer river systems to form. Hence, freshwater

inflow is often rather diffuse and we assume that these environmental conditions may cause irregular erosion and a very inhomogeneous sediment supply from each drainage area, which results in strong variations of the sediment composition even over very short distances.

4.2 Geochemistry

The inorganic geochemical composition of the surface sediments from the Vestfjord, Ofotfjord and Tysfjord reveals a very heterogeneous pattern. Simple and multivariate statistical data analysis failed to provide a better understanding of the association of the analysed parameters or the spatial distribution pattern. We found that an individual examination of the geochemical characteristics of each fjord is required because in our study area the element distribution is not necessarily related to a specific grain size fraction or mineral. As such, in the following, we discuss each fjord individually and depict similarities and differences between the analysed parameters. We focus our discussion mainly on the elements of Al, Si, Hf, Fe, Ti, and Zr because most of these elements are frequently used as indicators of terrestrial sediment supply in sediment core studies (e.g. Bertrand et al., 2012; Brendryen et al., 2015; Faust et al., 2014a; Wehrmann et al., 2014; Zabel et al., 2001). Moreover, to avoid dilution and grain size effects, we discuss the spatial distribution of these elements in the fjords as Al-based ratios (Bertrand et al., 2012; Faust et al., 2014b; Loring, 1990).

4.2.1 Ofotfjord

Linkage between terrigenous elements, grain size and water depth

In the Ofotfjord, Si correlates strongly with Hf and Zr and these elements are the only elements which are positively related to quartz ($r = 0.7\text{--}0.8$; $p < 0.01$, $n = 11$, ES-1). Moreover, they are related to the $>63 \mu\text{m}$ grain size fraction and show a clear negative correlation with the water content of the samples, water depth and with other terrigenous elements like Al, Mg, Fe, K, Ti, and Ni (ES-1). These elements (Al, Mg, Fe, K, Ti, Ni) correlate positively with the water content of the sediment samples and the $<63 \mu\text{m}$ grain size fraction. Additionally, they have a strong positive relationship to kaolinite and the sum of phyllosilicates (ES-1). The spatial distribution of Si/Al in the Ofotfjord (Fig. 4) shows that the

fine-grained material, which is rich in Al and clay minerals, settles in the middle and deeper part of the fjord. Closer to the shore and at the entrance sill, sediments show higher percentages of coarse-grained material, which are enriched in quartz and Si (Fig. 3 and ES-2). This finding is in accordance with the typical sediment distribution pattern in fjords, where coarser and heavier sediment components are usually deposited at the shore and in river estuaries and the grain size decreases with depth (Holtedahl, 1975; Skei, 1983). The higher amounts of Si and quartz together with the larger grain sizes at the entrance sill (Fig. 3 and ES-2) are probably related to winnowing caused by higher bottom current velocity.

4.2.2 Tysfjord

Narrow and long fjord arms, deep basins (up to 725 m) and large changes in depth over very short distances (Fig. 1) result in complicated geochemical and mineralogical signatures in the Tysfjord. Moreover, we note that the interpretation of the Tysfjord sediments composition is biased by the fact that sediment samples from the deepest part of the fjord are missing and that the number of sediment samples (ten) is relatively low to obtain reliable statistical analysis.

4.2.2.1 Connection between terrigenous elements and water depth

Similar to the Ofotfjord, Si, Hf, Zr and Ti are negatively correlated with the water content of the sample, water depth and the <63 µm grain size fraction (ES-1). Thus, Si concentrations are higher close to the shore and in shallower areas. The strong relationship between Si and the water depth explains the varying Si/Al values in the innermost fjord arms and over very short distances (Fig. 4 and ES-2). For example, station 23 and 25 are both located in the inner arms of the Tysfjord but station 23, which is at 120 m water depth shows much higher Si/Al values (3.9) than station 25 (2.7), which is at 444 m water depth (Fig. 4). Also the neighbouring stations 19 and 20 show very different Si/Al values (4 and 3.2, respectively) likely due to a change in water depth of 322 meters (Fig. 4). In accordance with the environmental setting in Patagonian fjords (Bertrand et al., 2012) these results suggest that the distribution of Si, Zr, Hf and also Ti in the surface sediments of the Tysfjord (and Ofotfjord) are controlled by their association to heavy and coarse-grained minerals such as quartz, zircon, amphibole, pyroxene and rutile. Furthermore, Bertrand et al. (2012) showed that Zr/Al and Ti/Al are sensitive to changes in the energy of the terrestrial supply to the

fjords. In a low energy system, values are highest in deltaic and proximal fjord environments of the tributaries. Moreover, Hinojosa et al. (2014) investigated several New Zealand fjords and concluded that differences in down-fjord geochemical gradients were related the presence or absence of a major river outlet in the inner fjord and amount of freshwater inflow. Thus in the Tysfjord, the clear relationship of Si, Hf, Zr and Ti with the water depth and the separation of fine and coarse-grained material even on a relatively short distance (e.g. station 19 and 20), indicates that the energy of the hydrodynamic system in the Tysfjord is relatively low. This is likely due to the small drainage area and the lack of large rivers entering the fjord.

4.2.2.2 *Origin of potassium*

Similar as in Ofot- and Vestfjord sediments, K has a strong, positive correlation with the <63 µm grain size fraction, the mixed and expandable clay minerals and Na (ES-1). However, in contrast to both other fjords, K shows no significant positive correlation to any other element or mineral. The relationship of K to the fine-grained material and the lack of correlation with almost any other element and mineral phase suggest that the analysed K originates potentially from a mixture of different (clay) minerals and may be a sign of an individual K source in the hinterland. The spatial distribution of the grain-size independent K/Al ratio reveals a clear inside-outside trend (Fig. 5) with highest values at the entrance of the fjord, which points to a possible K source in the middle to outer Tysfjord. Potassium is generally not associated with a specific mineral in sediments. Although, K-feldspar, kaolinite and illite are known as possible K sources in marine sediments (Martinez et al., 1999; Shimmield, 1992; Yarincik et al., 2000), these minerals do not correlate with K in the Tysfjord sediments (ES-1). Parts of the Precambrian basement in the mid-Tysfjord drainage area consist of biotite and K-feldspar rich gneisses (Andresen and Tull, 1986; Karlsen, 2000; Müller, 2011) and complementary airborne radiometric data indicate Precambrian rocks in northern Norway to be enriched in K (Nasuti et al., 2015). The Caledonian rocks in our study area are enriched in muscovite. However, since K does not show a significant correlation with the muscovite content ($r = -0.3$, $p > 0.1$, $n = 10$) in the Tysfjord sediments the Caledonian rocks can probably be excluded as potential K source. Thus, we conclude that Precambrian rocks are the main source of K in the Tysfjord sediments and we suggest that temporal changes of K/Al can be applied as an indicator of the variable supply of sediments

from the Precambrian rocks. Hence, K/Al in sedimentary records from the Tysfjord may be used to investigate past changes in terrestrial input and, thus, reconstruct variable freshwater inflow, weathering and glacial erosion.

4.2.3 Vestfjord

Two different sedimentary regimes

Similar to the sediments from Ofot- and Tysfjord, Si shows a positive correlation with Hf, Zr and quartz in the Vestfjord (ES-1). However, unlike the other fjords there is also a strong positive affiliation of Si to Al ($r = 0.9$, $p < 0.01$, $n = 20$) and to other terrigenous elements such as Ti, K, Fe (ES-1). These elements (Ti, K, Fe), however, show only a very weak correlation with quartz, but they are related to the phyllosilicates, chlorite, illite and micas (ES-1). A closer investigation of the relationship between quartz, Fe and Si in the surface sediments from the Vestfjord reveals two different sedimentary regimes. Considering all samples from the Vestfjord, quartz is strongly related to Si ($r = 0.8$, $p < 0.01$, $n = 19$) but not to Fe ($r = 0.2$, $p > 0.1$, $n = 19$) and both, Fe and Si are related to the sum of illites and micas ($r = 0.7$, $p < 0.01$, $n = 19$) (Fig. 6). Together with Al, K, Mg, and Ti, Fe is strongly related to the water depth of the sample ($r = 0.9$, $p < 0.01$, $n = 20$, Fig. 6), and the highest Fe concentrations (>3.5%) are found in the middle of the fjord along an inside-outside trend (stations 17; 30–33; 39; 42; 43, Fig. 1). By examining the samples from the deeper and the shallower areas (shelf and sill) separately, we found that Fe and quartz are positive correlated on the shelf areas ($r = 0.6$, $p = 0.05$, $n = 11$, Fig. 6) but negatively correlated in the deeper and outer fjord basins ($r = -0.9$, $p < 0.01$, $n = 8$, Fig. 6). This division by the two territories is also present in many other geochemical and mineralogical parameters in the Vestfjord surface sediment samples. For example, Al versus plagioclase reveals a clear positive correlation ($r = 0.9$, $p < 0.01$, $n = 11$) for the shelf and sill sediment samples and a clear negative correlation ($r = -0.8$, $p < 0.05$, $n = 8$) for the samples from the deeper Vestfjord basin (Fig. 6). Moreover, Al versus the sum of phyllosilicates (Fig. 6) shows an increase of the Pearson correlation coefficient from $r = 0.4$ at the shallow parts to $r = 0.9$ in the deeper fjord areas. Sediments at the Vestfjord shelf and sill area contain higher amounts of coarse-grained material (Fig. 3) and the positive correlation, for example between Al and plagioclase, and Si and quartz, shows that the sediment material is relatively fresh and of

local origin. The samples from the deeper basins are rich in fine-grained material (Fig. 3), and Al and Si are negatively correlated to plagioclase and quartz and positively correlated to clay minerals. Due to the relatively small catchment area of the Vestfjord, about 90% of the freshwater inflow originates from the adjacent fjords in the east (Mitchelson-Jacob and Sundby, 2001; Sundby, 1982). Thus, the fine-grained sediments in the deep basin of the Vestfjord may be transported over longer distances and may derive also from the Ofot- and Tysfjord. Indications of sediment transport from these two fjords into the Vestfjord are also found in the distribution of biotite and illite. Compared to the other fjords, the Vestfjord is enriched in illite (Fig. 7) which shows no relationship to the water depth and is present in the shallow and the deep parts of the fjord. Yet, apart from one sample (station 15; 0.6%), biotite is only found in the deep areas of the Vestfjord (station 17, 30–33, 39, 42, 43, (ES-1)). This indicates that biotite does not originate from the Vestfjord drainage area but it originates from the Ofotfjord or the biotite-rich Tysfjord area (Fig. 7). Thus, the surface sediments forming the deep Vestfjord basin probably contain a sediment mixture of all adjacent fjords.

To summarize, we found a very heterogeneous spatial distribution pattern of most analysed elements. In general, this can be explained by the complex fjord bathymetry with large changes in water depth over very short distances, a low hydrodynamic energy environment and a relatively small drainage area causing a rather diffuse freshwater inflow. Similar to the sediments from Ofot- and Tysfjord, elements associated with coarser and heavier sediment components (Si, Hf, Zr) are enriched at the shallower areas and higher values of elements often associated with the fine grain material (Al, Fe, K) are found in the deeper fjord basins. Potassium in the Tysfjord reveals a solitary behaviour which points to an individual K source in the hinterland. This source is probably biotite and K-rich Precambrian rocks enclosing the middle to outer Tysfjord. Similar as in the Tysfjord and Ofotfjord, Vestfjord sediments at the shelf are relatively fresh and of local origin. But in contrast to the two other fjords the surface sediments forming the deep Vestfjord basin probably contain a sediment mixture of all adjacent fjords. The differences of the elemental distribution in the Vestfjord are caused by a different origin of the sediment material and differences in the grain size/mineralogical association of certain elements. Hence, the distribution of a certain element in fjord sediments is not necessarily related to a specific grain size fraction or mineral.

4.3 Terrigenous provenance proxies: biotite, muscovite and illite

Most phyllosilicates are detrital and their distribution in shelf sediments depends largely on the diversity of geology and weathering processes of the contributing source areas in the hinterland (e.g. Petschick et al., 1996; Velde, 1995). We found that the Ofotfjord sediments are enriched in muscovite, the Tysfjord in biotite and the Vestfjord in illite (Fig. 7). The different mineral assemblages of the three fjords may reflect differences in the bedrock composition in the drainage areas. Muscovite is very common in the Caledonian rocks surrounding the Ofotfjord. On the contrary, Precambrian rocks in the drainage areas of the Vest- and Tysfjord are relatively poor in muscovite. Moreover, biotite-rich gneisses in the Tysfjord drainage area (Andresen and Tull, 1986; Karlsen, 2000; Müller, 2011) probably contribute to the elevated biotite concentrations in the Tysfjord (Fig. 7). However, we note that biotite, muscovite and illite are very common minerals and typically found in marine sediments in boreal regions where weathering is largely physically controlled (e.g. Vogt and Knies, 2009). It is, therefore, very challenging to identify a particular source of these minerals in the hinterland. Nevertheless, the occurrence of large amounts of biotite and chlorite, which are usually unstable under hydrolysis (Petschick et al., 1996; Wilson, 2004) in the fjord sediments, and the low concentrations of secondary clay minerals such as kaolinite (2–3%) and smectite (0–2%), indicate that the fjord sediments are relatively fresh and of very local origin. We suggest that these findings are probably related to the relatively small drainage areas of the three fjords. Therefore, distance and time between erosion and sedimentation is short and additionally the temperate climate in northern Norway favour physical- over chemical weathering.

4.4 Ca and calcite: marine productivity proxies

Their strong relationship to marine productivity and, thus, water temperature, salinity, nutrient supply and degree of ice coverage make Ca, calcite and CaCO_3 very appropriate proxies to reconstruct climate and environmental changes (e.g. Schneider et al., 2006). The CaCO_3 content in surface sediments usually reflects the modern surface water oceanography and was, for example, successfully applied as a paleoceanographic tool to reconstruct northern Hemisphere glacial/interglacial cycles (e.g. Bond et al., 1992; Ruddiman and McIntyre, 1981; Zamelczyk et al., 2014). Furthermore, Faust et al. (2016)

recently revealed that Ca/Si and CaCO₃ in fjord sediments can provide detailed reconstructions of atmospheric circulation changes. In this study, we attempt to identify the controlling factors of the Ca and CaCO₃ distribution in the Vestfjord, Ofotfjord and Tysfjord to assess their future applicability as paleo-environmental proxies in these three fjords.

Ca anti-correlates to all other elements or minerals other than calcite, aragonite and Sr ($r \geq 0.9$, $p < 0.01$, $n = 40$) in all three fjords. As elemental ratios are insensitive to dilution effects, in the following we also focus on the Ca/Al distribution which shows a strong correlation with Ca and calcite ($r \geq 0.9$, $p < 0.01$, $n = 40$).

Ca/Al in the Tys- and Ofotfjord sediments shows a clear inside-outside trend and highest values are found at the entrance of each fjord and in their deepest areas (Fig. 8). Moreover, calcite is present in all samples, but aragonite is only found in station 8, 13, 27 and 25 (ES-1 and 2). This is probably related to the occurrence of cold water corals which have been found during the sampling procedure and are known to grow at and around the entrance sill (Fossa et al., 2002 and references therein). Samples close to the marble formation in the hinterland of the Ofot- and Tysfjord (Fig. 2) contain lowest Ca/Al concentrations in the entire study area (Fig. 8 and ES-2). This indicates that the marble rocks in the fjord drainage areas do not have a significant impact on the Ca/Al distribution. We assume that either the terrigenous carbonates are dissolved during weathering or the marine carbonate input is so much larger than the carbonate signal from the marble rocks that the terrigenous carbonates are heavily diluted and are therefore barely discernible. An indication for the dissolution of the dolomite rich marble during erosion is the strong association of Mg with the water content of the sample, Na and NaCl in the Ofotfjord samples ($r \geq 0.9$, $p < 0.01$, $n = 11$, ES-1). Mg and NaCl may be precipitated from the seawater during the freeze drying of the samples prior to the geochemical analysis. As Mg shows this relationship only in the Ofotfjord, this may indicate that the Ofotfjord water column is relatively enriched in Mg due to the weathering of dolomite-rich marble in the drainage area. The Vestfjord and especially the adjacent shelf areas are well known to be areas of high marine productivity, probably sustained by nutrient-rich coastal waters and upwelling along the steep side-edges of the Vestfjord (Espinasse et al., 2016; Furnes and Sundby, 1981; Sundby and Solemdal, 1984). The highest Ca/Al values of the entire study area were found along the shallow coastal areas on the north-western and the south-eastern fjord margins (Fig. 8). In accordance with this

finding, high concentrations of calcite (up to 51%) in these areas are related to large numbers of shell and coral fragments in the sediment samples.

In accordance with findings from the Trondheimsfjord in mid-Norway and in northern Chilean fjords (Bertrand et al., 2012; Faust et al., 2014b), the strong positive correlation between Ca and calcite ($r = 0.96$, $p < 0.01$, $n = 40$) and a strong negative correlation ($r \leq -0.8$, $p < 0.01$, $n = 40$) to any terrigenous element (as e.g. Al, Fe, Ti and Si) indicates marine carbonate productivity to be the main calcite and Ca source in all three fjords. However, we note that to confirm this assumption it is necessary to investigate the organic components (such as C_{org} and $\delta^{13}C_{org}$) of the sediments samples to distinguish the marine and terrigenous origin of Ca (Faust, 2014; Knies et al., 2003). Nevertheless, the increase in Ca/Al from the inside towards the entrance of the Ofot- and Tysfjord fjords (Fig. 8) and the high values along the upwelling areas in the Vestfjord are most likely caused by enhanced primary productivity due to the inflow of Atlantic water. These findings express that Ca/Al is a well-suited proxy for changes in carbonate marine productivity versus terrigenous sediment supply and may serve as indicator for changes in the inflow of Atlantic waters versus river discharge, nutrient supply and sea surface temperature changes.

5 Conclusion

The inorganic geochemical composition of Vestfjord, Ofotfjord and Tysfjord sediments in northern Norway reveals that, as in many other fjords, Ca/Al and calcite are good indicators for marine carbonate productivity versus terrigenous sediment supply. However, the distribution patterns of the mineral assemblages, grain size and the elemental composition are overall complex and variations on very short distances are large which indicates a very heterogeneous sediment supply. Besides the Ca/Al proxy for the inflow of north Atlantic water and river discharge, the geochemical signatures in the three fjords are very different to the Trondheimsfjord in mid Norway. Moreover, even though the hinterland sediment source areas of the three investigated fjords are relatively similar in terms of bedrock type and drainage area size, our results indicate that differences in bathymetry and the hydrodynamic energy can cause considerably different sedimentary regimes for each Norwegian fjord. Thus, to identify the modern sedimentological and environmental setting,

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it is important to examine each fjord basin separately. Furthermore, different erosion and transport systems in the hinterland of each fjord may be responsible for the diverse distribution pattern of the investigated parameters presented here. To evaluate this, further investigations of the onshore sediment transport and bedrock weathering processes are necessary. This will also help to better identify the sources of biotite, muscovite and illite of the three fjords, as well as the origin of the K anomaly in Tysfjord sediments.

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Appendix

Electronic supplementary data associated with this article (ES-1 (Tab. S1–S6) and ES-2 (Fig. S1–S6)) can be found in the online version.

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Figures

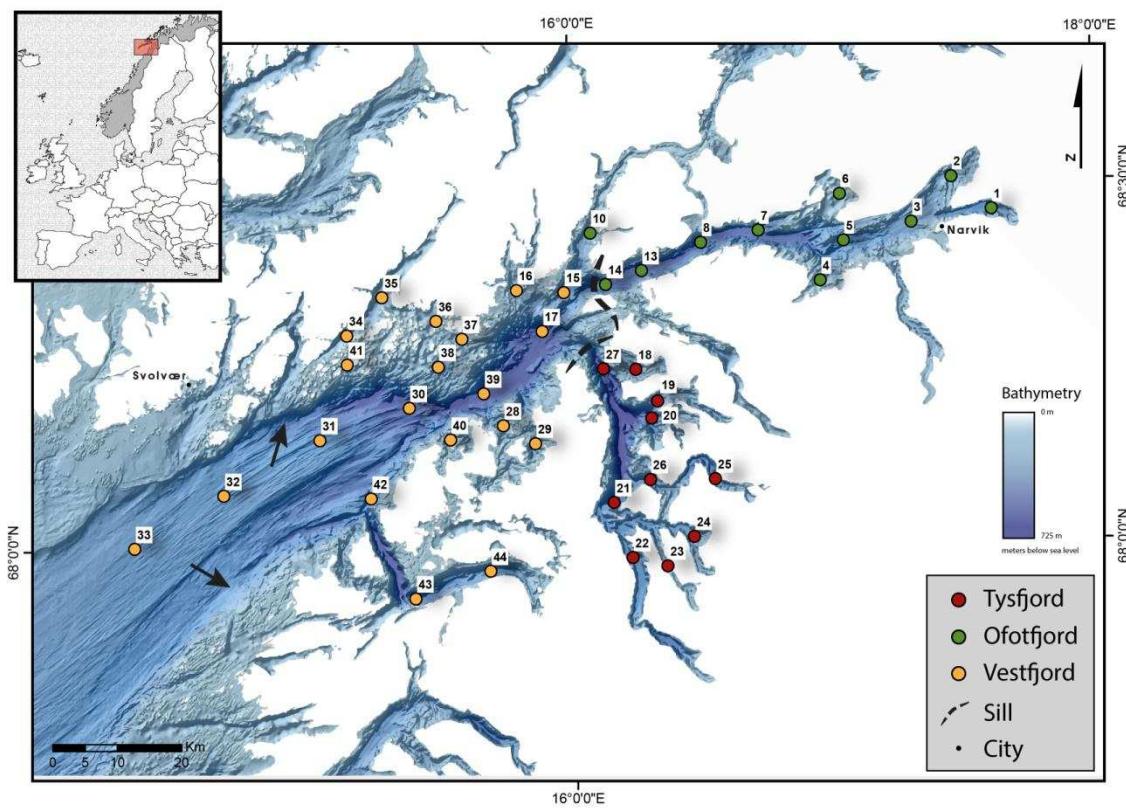


Fig. 1: Location of the study area in northern Norway (upper left inlet). Map of the Vest-, Ofot- and Tysfjord showing the bathymetry (from mareano.no) and sampling positions of each fjord (ES-1). Broken black line indicates the sill between the three fjords. Black arrows indicate the up to 300 m high side-edge between the deeper Vestfjord basin and its shallower coastal areas.

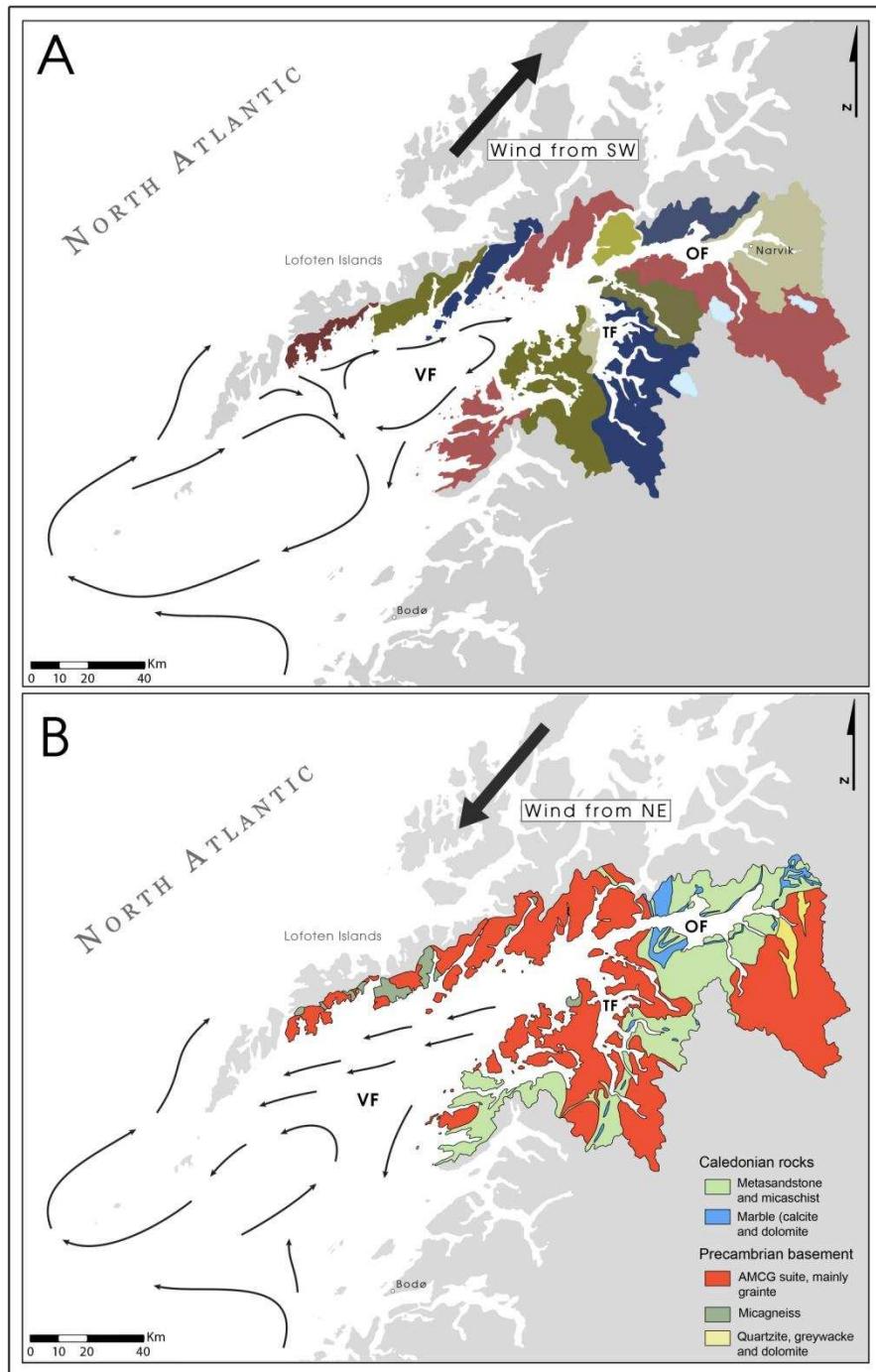


Fig. 2: The Vestfjord (VF), Ofotfjord (OF) and Tysfjord (TY) are the three main fjords of a complex fjord system between the Norwegian mainland and the Lofoten archipelago in northern Norway. **A)** Total catchment area of the three fjords. Each coloured field represents a sub-drainage basin. The three main glaciers present in the drainage area today are indicated by white fields in the south of the Ofot- and Tysfjord. Additionally, black arrows indicate the surface circulation pattern during SW winds. **B)** Simplified geological map of the study area and surface circulation during NE winds (black arrows).

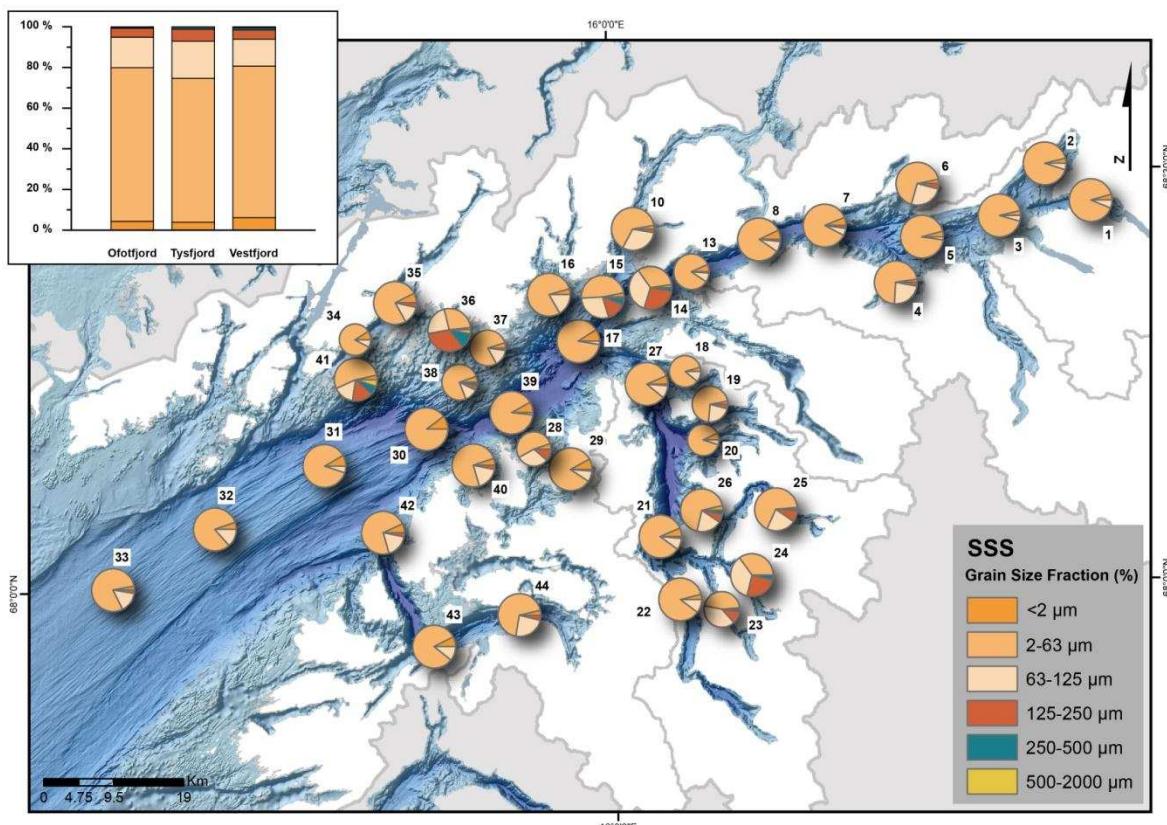


Fig. 3: Grain size distribution in the Vestfjord, Ofotfjord and Tysfjord surface sediment sample (SSS) and average grain size of each fjord (bar chart, upper left). To prevent the pie diagrams to overlap they vary in sizes.

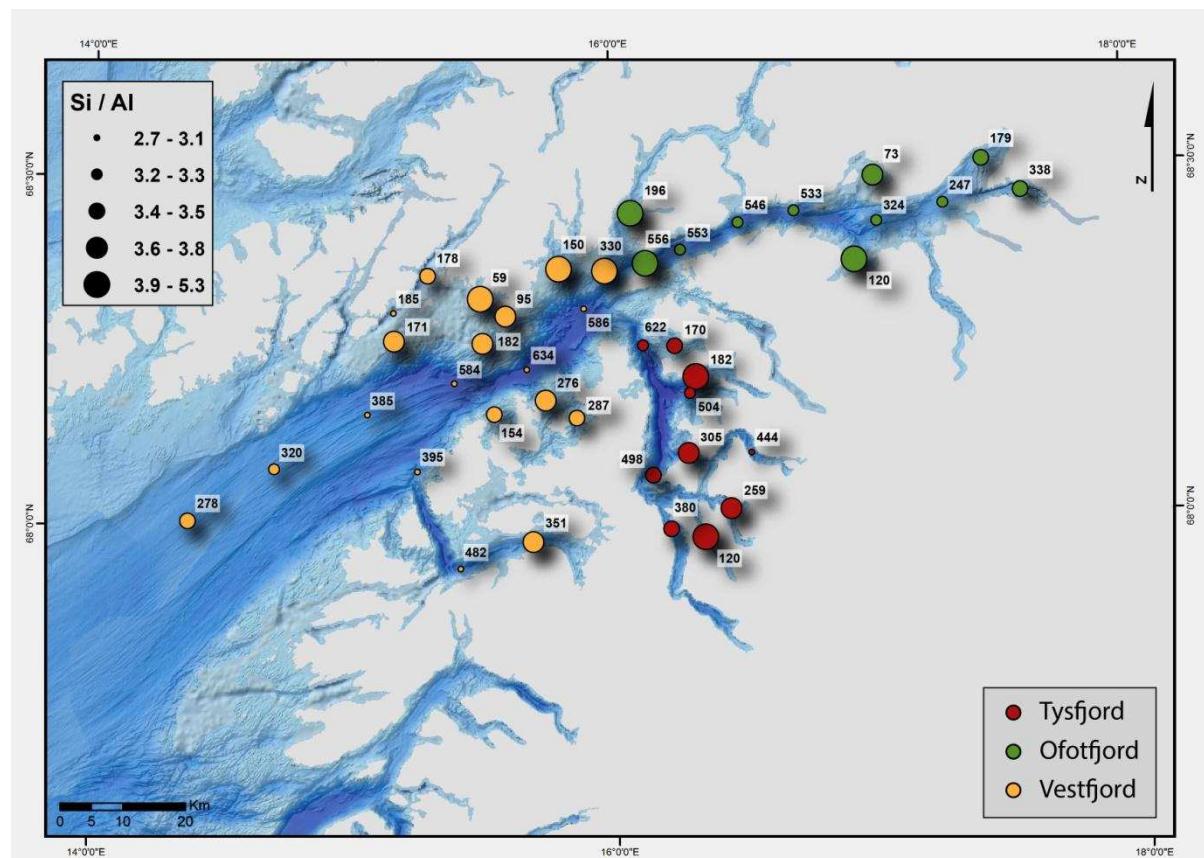


Fig. 4: Water depth and spatial distribution of Si/Al of the surface sediments samples from the Vestfjord (yellow), Ofotfjord (green) and Tysfjord (red).

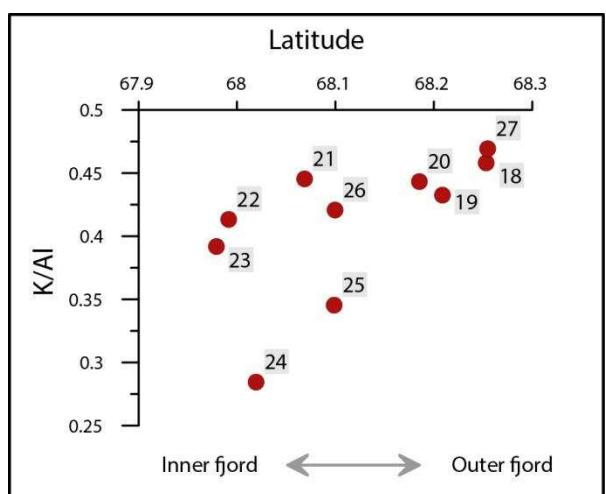


Fig. 5: K/Al in the Tysfjord surface sediment samples with station numbers. K/Al reveal a clear inside outside trend with highest values at the entrance of the fjord.

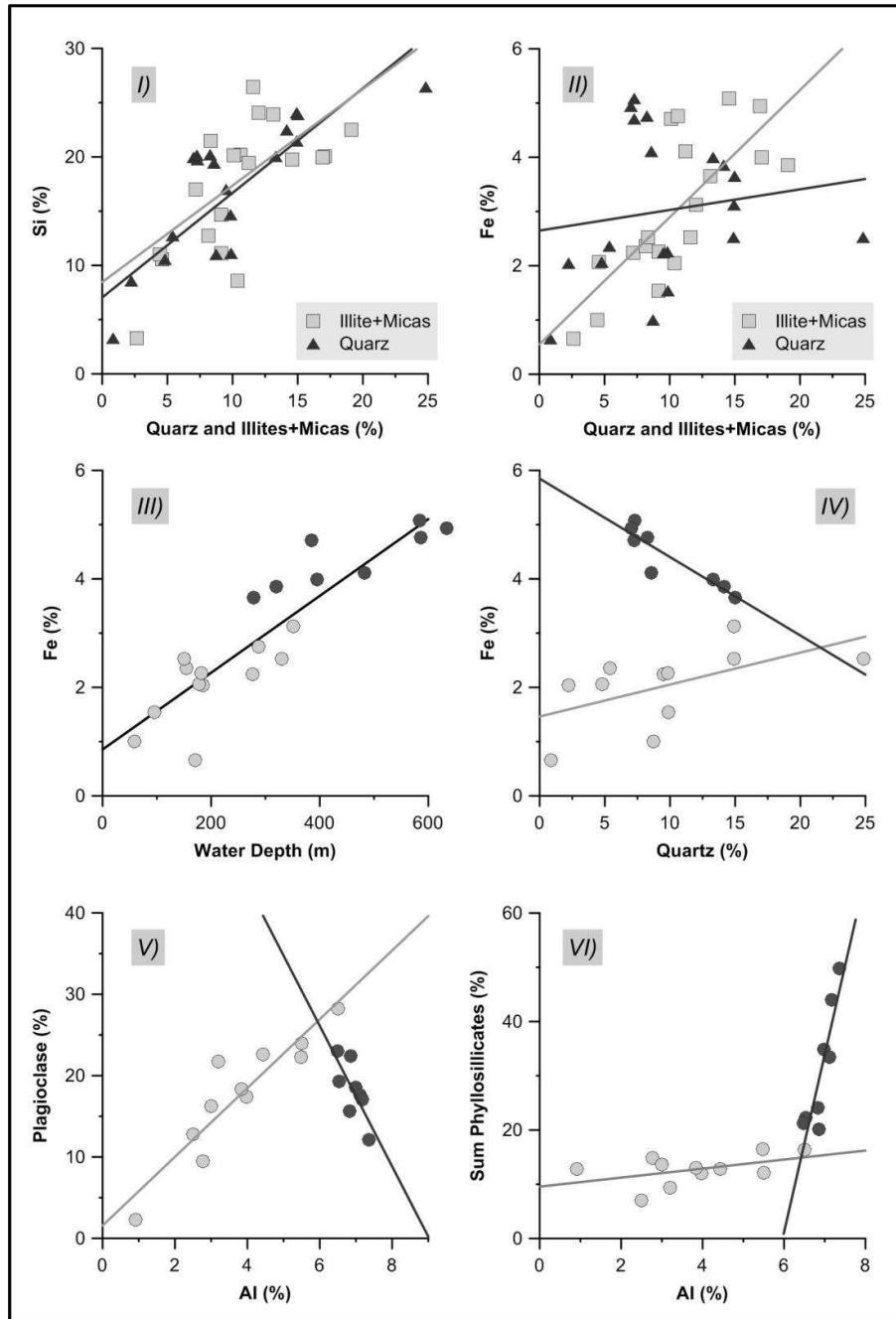


Fig. 6: (I) Considering all samples from the Vestfjord Si correlates with quartz ($r = 0.8$) and the sum of illites and micas ($r = 0.7$). (II) Fe correlates with the sum of illites and micas ($r = 0.8$) but not with quartz ($r = 0.2$). (III) Fe is strongly related to the water depth of the sample ($r = 0.9$). Light grey points: samples from the shelf and sill (stations 15; 16; 28; 29; 34–38; 40; 41; 44, Fig. 1), dark grey points: samples from the deep basin (stations 17; 30–33; 39; 42; 43, Fig. 1). (IV) Fe and quartz are positive correlated on the shelf areas ($r = 0.6$) and negative in the deeper and outer fjord basins ($r = -0.9$). (V) Al versus plagioclase reveals a clear positive correlation ($r = 0.9$) for the shelf and sill sediment samples and a clear negative correlation ($r = -0.8$) for the samples from the deeper Vestfjord basin. (VI) Al versus the sum of phyllosilicates shows an increase of the Pearson correlation coefficient from $r = 0.4$ at the shallow parts to $r = 0.9$ in the deeper fjord areas.

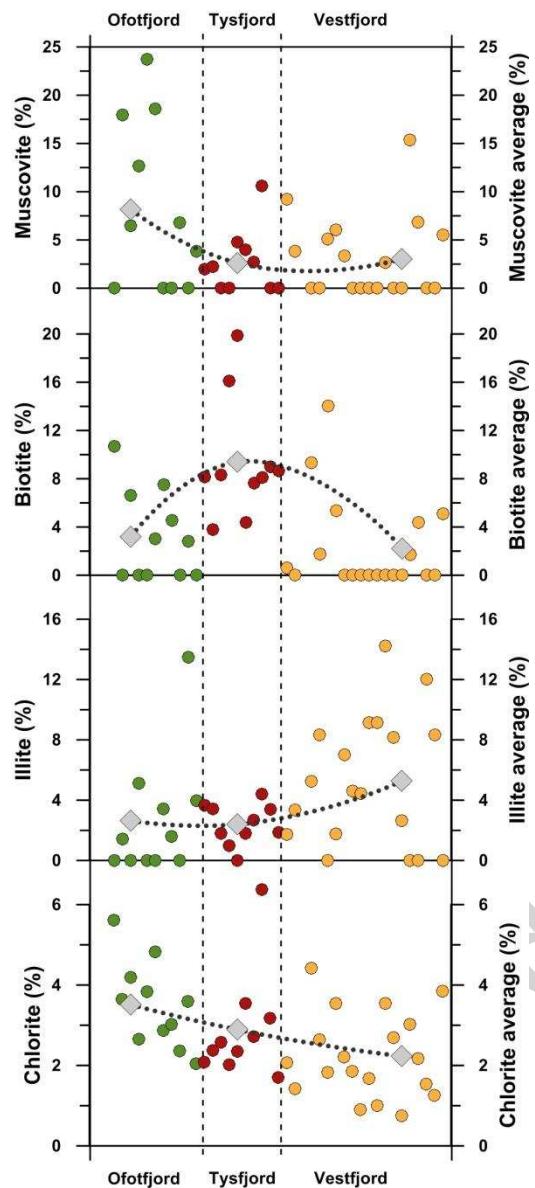


Fig.7: Muscovite, biotite, illite and chlorite concentrations (%) in the surface sediments samples from the Ofotfjord (green), Tysfjord (red) and Vestfjord (yellow). Average content is indicated by grey diamonds.

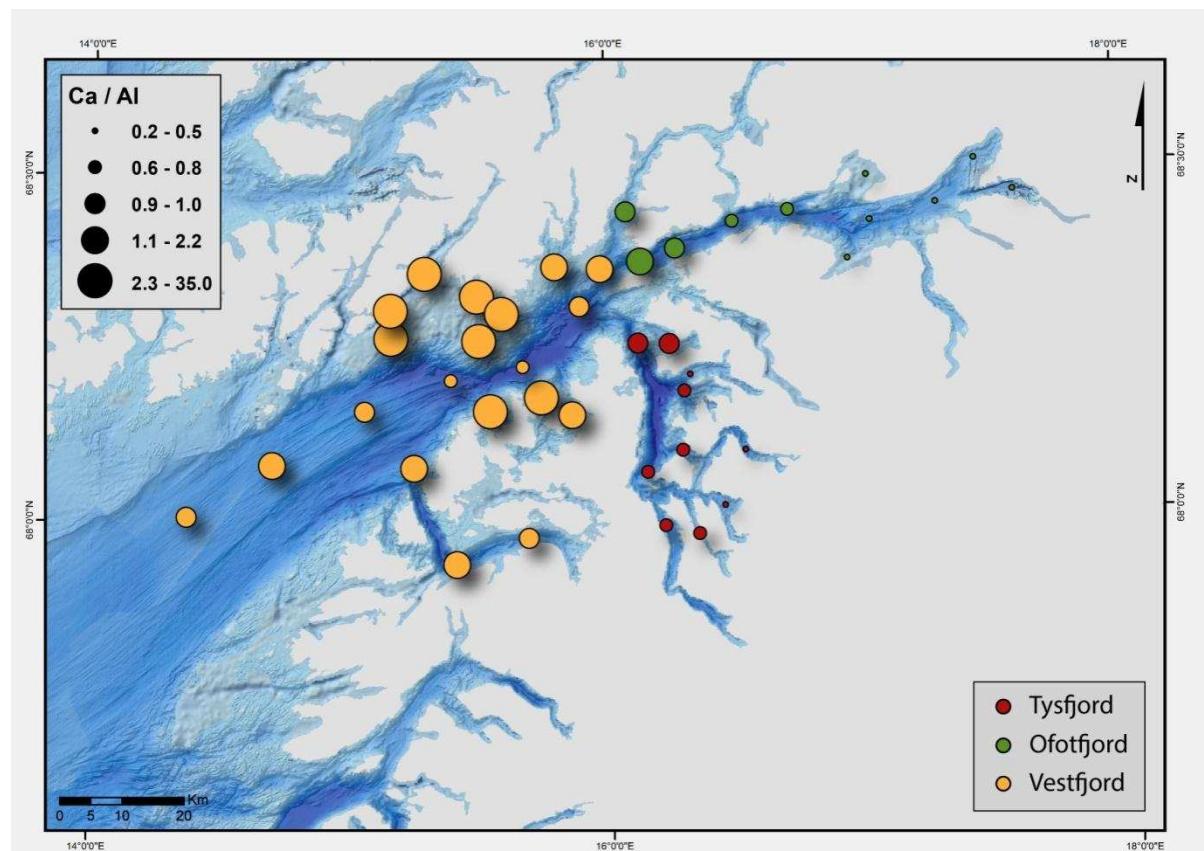


Fig. 8: Ca/Al distribution in the surface sediments of the Ofotfjord (green), Tysfjord (red) and Vestfjord (yellow). Highest values are found close to the sill and at the shallow shore areas of in the NW and SE of the Vestfjord.

Highlights

- The distribution of a certain element in fjord sediments is not necessarily related to a specific grain size fraction or mineral.
- Ca/Al and calcite are good indicators for marine carbonate productivity versus terrigenous sediment supply in fjords.
- Fjord sediment compositions can vary considerably even over very short distances if major river outlets are absent.
- Differences in bathymetry and the hydrodynamic energy can cause considerably different sedimentary regimes for each Norwegian fjord.