

Reconstruction of the provenance of detrital sediments in the Skagerrak region over the past 150 years

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Abstract

The North Sea acts as an efficient natural sink for organic carbon, sequestering atmospheric CO₂ derived from anthropogenic emission. Sedimentary processes play a crucial role in the burial and long-term storage of particulate organic carbon (POC), as they directly control its accumulation in marine sediments and removal from the carbon cycle, with sediment transport, erosion, and deposition being the primary influencing factors. However, since the onset of industrialization, sediment dynamics have been significantly affected by both human activities and climate-driven changes. The Skagerrak basin, as the major depocenter for fine-grained sediments transported through the North Sea, provides valuable insights into sediment transport pathways and the evolution of the sedimentary system over time.

To trace the provenance of Skagerrak sediments and reconstruct the main sedimentary processes in the North Sea, radiogenic Sr, Nd, and Hf isotope signatures and clay mineral compositions of the detrital clay fraction of surface sediment samples from the North Sea, the Scandinavian fjords and the Baltic Sea were measured. Additionally, to investigate the temporal variability in sediment supply from the predominant source areas and the changes in sedimentation over the past 150 years, Skagerrak sediment cores were analyzed using a time series of radiogenic isotopes, clay mineral compositions, grain-size distributions, and X-ray fluorescence (XRF) core scans.

The spatial distribution of the geochemical characterization identifies the northern North Sea, southern North Sea, and Scandinavian fjords as the main sources of fine-grained sediments deposited in the central part of the Skagerrak. Seabed erosion at the Fladen Ground, located in the northern North Sea, supplies the Skagerrak with large amounts of clay-sized sediments, primarily controlled by the inflow of the Atlantic Current into this region. Contributions from mid-European rivers such as the Elbe, Weser, and Ems, as well as the Baltic Sea, are only minor contributors. Seabed erosion within the English Channel and coastal erosion along the southeastern coast of England are assumed to supply large amounts of suspended particulate matter (SPM) to the southern North Sea. This material is predominantly deposited in the depositional environments of the German Bight and partly trapped in the Wadden Sea before being transported further into the Skagerrak basin.

Radiogenic isotope ratios and XRF data from Skagerrak sediment records indicate a shift since the second half of the 20th century, with reduced sediment supply from the southern North Sea and increased contributions from the northern North Sea. This shift is caused by an increase in

storm frequency and intensity in the 1980s and 1990s, associated with a positive North Atlantic Oscillation (NAO) index that amplified hydrodynamic conditions in the North Sea region. Strong westerly winds intensified the Atlantic inflow into the northern North Sea and Skagerrak basin enhancing seabed erosion in the Fladen Ground and increasing sediment supply from the northern North Sea. Additionally, intensified bottom trawling in this region during the 1980s and 1990s increased seabed erosion in the Fladen Ground, further amplifying sediment supply to the Skagerrak. Strong bottom current velocities within the Skagerrak basin, evidenced by the sedimentation of coarser deposits, intensified erosional processes along the Scandinavian coast, increasing sediment supply from the northern North Sea to the Skagerrak. These more dynamic hydrodynamic conditions in the second half of the last century may have hampered the settling of clay and fine silt fractions in the inner part of the Skagerrak, potentially explaining the decline in sediment accumulation over the past 60 years. The reduced sediment supply from the southern North Sea can be attributed to increased trapping efficiency in the Wadden Sea under energetic hydrodynamic conditions. In contrast, intensified deepening of large mid-European rivers has not significantly influenced the sediment contribution to the Skagerrak, as indicated by the radiogenic isotope signatures.

These findings emphasize that climate variability primarily influence the provenance and deposition of Skagerrak sediments due to changes in current velocity and circulation dynamics forced by the storm activity. While human interventions, such as bottom trawling, have influenced the sediment dynamics and contributions to the Skagerrak to a lesser extent.

Kurzfassung

Die Nordsee fungiert als effiziente natürliche Senke für organischen Kohlenstoff, indem sie atmosphärisches CO₂ aus anthropogenen Emissionen aufnimmt. Sedimentäre Prozesse spielen eine entscheidende Rolle bei der langfristigen Speicherung von partikulärem organischem Kohlenstoff (POC), da sie direkt dessen Anreicherung in marinen Sedimenten und Entfernung aus dem Kohlenstoffkreislauf steuern, wobei Sedimenttransport, Erosion und Ablagerung die Hauptkontrollfaktoren sind. Seit Beginn der Industrialisierung sind die Sedimentdynamiken jedoch sowohl durch menschliche Aktivitäten als auch durch klimabedingte Veränderungen erheblich beeinflusst worden. Der Skagerrak stellt das Hauptablagerungsgebiet für feinkörnige Sedimente dar, die durch die Nordsee transportiert werden, und bietet wertvolle Einblicke in die Sedimenttransportwege sowie die Entwicklung des sedimentären Systems im Laufe der Zeit.

Um die Herkunft der Skagerraksedimente zu bestimmen und die wichtigsten sedimentären Prozesse in der Nordsee zu rekonstruieren, wurden radiogene Sr, Nd und Hf Isotopensignaturen sowie die Tonmineralzusammensetzungen der detritischen Tonfraktion von Oberflächensedimentproben aus der Nordsee, den skandinavischen Fjorden und der Ostsee gemessen. Zusätzlich wurden Sedimentkerne aus dem Skagerrak untersucht, um die zeitliche Variabilität der Sedimentversorgung aus den Hauptliefergebieten und die Veränderungen in der Sedimentation in den letzten 150 Jahren zu analysieren. Hierfür wurde eine Zeitreihe von radiogenen Isotopen, Tonmineralzusammensetzungen, Korngrößenverteilungen und Röntgenfluoreszenz (XRF)-Kernscans verwendet.

Die räumliche Verteilung der geochemischen Charakterisierung identifiziert die nördliche Nordsee, die südliche Nordsee und die skandinavischen Fjorde als die Hauptliefergebiete für die im zentralen Skagerrak abgelagerten feinkörnigen Sedimente. Die Bodenerosion am Fladen Ground in der nördlichen Nordsee versorgt den Skagerrak mit großen Mengen an tonigen Sedimenten, deren Transport hauptsächlich durch den Zufluss des Atlantikstroms gesteuert wird. Die Sedimentzufuhr von mitteleuropäischen Flüssen wie der Elbe, Weser und Ems sowie der Ostsee ist hingegen eher zu vernachlässigen. Die Bodenerosion im Ärmelkanal und die Küstenerosion entlang der englischen Küste versorgen die südliche Nordsee mit großen Mengen an suspendiertem partikulärem Material. Diese feinen Partikel sedimentieren überwiegend innerhalb der Ablagerungsgebiete in der Deutschen Bucht und werden zum Teil im Wattenmeer zurückgehalten, bevor diese weiter in das Skagerrak-Becken transportiert werden.

Die radiogene Isotopenverhältnisse und XRF-Daten aus den Sedimentkernen des Skagerraks zeigen einen reduzierten Sedimentbeitrag aus der südlichen Nordsee und eine erhöhte Zufuhr aus der nördlichen Nordsee in der zweiten Hälfte des 20. Jahrhunderts. Diese Verschiebung wurde durch eine Zunahme der Sturmhäufigkeit und -intensität in den 1980er- und 1990er-Jahren verursacht, die mit einem positiven Index der Nordatlantischen Oszillation (NAO) einhergeht und die hydrodynamischen Bedingungen in der Nordseeregion verstärken. Dabei intensivieren starke Westwinde den Atlantikzufluss in die nördliche Nordsee bis hin zum Skagerrak, wodurch die Bodenerosion im Fladen Ground verstärkt und die Sedimentversorgung aus der nördlichen Nordsee erhöht wurde. Zudem führte die verstärkte Grundschleppnetzfisherei in dieser Region während dieser Zeit zu einer weiteren Verstärkung der Bodenerosion, was die Sedimentversorgung zum Skagerrak zusätzlich anstieg ließ. Intensivierte Geschwindigkeiten in den Bodenströmungen des Skagerraks, die sich in der Ablagerung gröberer Sedimente zeigen, intensivierten erosive Prozesse entlang der skandinavischen Küste, wodurch die Sedimentzufuhr aus der nördlichen Nordsee in den Skagerrak weiter verstärkt wurde. Diese hydrodynamischen Bedingungen in der zweiten Hälfte des letzten Jahrhunderts könnten das Ablagern von Ton- und feinen Siltfraktionen im inneren Teil des Skagerraks erschwert haben, was möglicherweise den Rückgang der Sedimentakkumulation in den letzten 60 Jahren erklärt. Der reduzierte Sedimentzufluss aus der südlichen Nordsee lässt sich durch eine erhöhte Effizienz in der Sedimentaufnahme des Wattenmeers aufgrund der hydrodynamischen Bedingungen erklären. Im Gegensatz dazu hat das Ausbaggern großer mitteleuropäischer Flüsse die Sedimentzufuhr zum Skagerrak nicht signifikant beeinflusst, wie die radiogenen Isotopensignaturen deutlich zeigen.

Diese Ergebnisse verdeutlichen, dass die Klimavariabilität die Herkunft und Ablagerung der Skagerraksedimente hauptsächlich beeinflussen, bedingt durch Änderungen in der Strömungsgeschwindigkeit und den Zirkulationsdynamiken, die durch die Sturmaktivität gesteuert werden. Im Gegensatz dazu haben menschliche Eingriffe wie die Grundschleppnetzfisherei die Sedimentdynamik und die Beiträge zum Skagerrak in geringerem Maße beeinflusst

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1. Introduction

1.1 APOC project

This thesis was conducted in the framework of the project APOC ("Anthropogenic impacts on particulate organic carbon cycling in the North Sea") funded by the Federal Ministry of Education and Research, Germany (BMBF) as part of the MARE:N Coastal, Marine and Polar Research for Sustainability program. It was a collaborative project involving the Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research (AWI), Helmholtz Centre Hereon (Hereon), University of Hamburg (UHH), GEOMAR Helmholtz Centre for Ocean Research Kiel (GEOMAR), as well as the non-governmental organization German Federation for the Environment and Nature Conservation (BUND). This collaboration focused on the cycling of particulate organic carbon (POC) and its role in carbon sequestration within the North Sea region, while also investigating the impacts of anthropogenic activities and climate change on the ecosystem.

1.2 Motivation and objectives of APOC

The greenhouse gas carbon dioxide (CO₂) causes atmospheric warming and plays a key role in controlling the climate on earth (IPCC, 2023). Since the beginning of industrial times, the atmospheric CO₂ concentration has risen from 280 to 424 ppm (NOAA, 2024), an increase of almost 50% over the last 260 years. Both, the atmosphere and the ocean absorb substantial amounts of anthropogenic CO₂ (Zeebe and Wolf-Gladrow, 2007), with the ocean acting as a major carbon sink, sequestering around 25% of the global CO₂ emissions (DeVries, 2022; Friedlingstein et al., 2023). Within the marine carbon cycle, excess CO₂ in the atmosphere leads to an elevated carbon flux to the ocean, where it is redistributed by biological and physico-chemical cycling. Phytoplankton in surface waters fixes CO₂ and converts it into POC, which is the foundation of the biological carbon pump (Volk and Hoffert, 1985; Longhurst and Harrison, 1989). While sinking through the water column, POC undergoes partial remineralization releasing CO₂ back into the water (Longhurst, 1991). However, a proportion of the POC settles onto the seafloor, where it is embedded into marine sediments, while other proportions undergo diagenesis and dissolution in the sediments and are released back to the water column via benthic fluxes (Emerson and Hedges, 2003). The POC sediment fixation effectively removes carbon from the active carbon cycle and leads to its storage in the sediments for millennia or even on geological time scales (Menon et al., 2007; Ciais et al., 2014). This

mechanism highlights the crucial role of the ocean as an effective long-term carbon sink and its importance in mitigating atmospheric CO₂ rise.

The North Sea is a continental shelf sea in northwestern Europe that acts as a highly efficient sink for organic carbon of the North Atlantic Ocean. More than 90% of the CO₂ absorbed from the Ocean is transferred to the Atlantic Ocean, with an uptake rate of 1.38 mol C m⁻² yr⁻¹ in the North Sea (Thomas et al., 2005). The organic carbon accumulation rates in the upper 0.1 m of North Sea sediments amounts to about 231 Tg C, representing 1.7 % of the global carbon stock on continental shelves (Diesing et al., 2021). However, since industrial times the marine carbon cycle in the North Sea and its POC accumulation have been heavily affected by human interventions (ICES, 2020; OSPAR, 2023). This includes bottom trawling activities (Rijnsdorp et al., 1998; Pitcher et al., 2000; Bellman et al., 2005; Murawski et al., 2005; Eigaard et al., 2017), coastal protection means (Hoeksema, 2007; Hofstede, 2008), land reclamation (Flemming and Nyandwi, 1994), sand mining (De Groot, 1986), dredging and ditching of rivers (Ericson et al., 2006; Goelz, 2008; Rovira et al., 2014), and eutrophication (Pätsch et al., 2010; Skogen et al., 2014; Axe et al., 2017). Bottom trawling, for example, penetrates the seafloor to sediment depths of up to 35 cm (Eigaard et al., 2017), resuspending significant amounts of fine-grained sediments across the North Sea. This human activity impacts approximately 63% of the North Sea seafloor (Eigaard et al., 2017). Furthermore, the construction of dams and dredging activities along major mid-European rivers such as the Elbe, Weser, Ems, and Rhine have significantly reduced riverine sediment inputs into the North Sea (Puls et al., 1997; Syvitski et al., 2005; Ericson et al., 2006; Goelz, 2008; Rovira et al., 2014). These interventions have resulted in the disposal of substantial amounts of fine-grained sediments in particular areas of the North Sea (OSPAR, 2017). This large-scale impact on both sediment dynamics and carbon cycling has led to a decreased fixation of particulate organic carbon in the sediments, thereby enhancing CO₂ release to the atmosphere (De Borger et al., 2020; Bradshaw et al., 2021; Atwood et al., 2024, Zhang et al., 2024). The APOC project aimed to quantify these anthropogenic impact factors and the contribution of POC burial in the North Sea. By generating this knowledge, effective protection strategies can be developed to preserve vulnerable regions in the North Sea that serve as effective long-term carbon storage.

The overarching goal of GEOMAR, AWI, and Hereon encompassed investigating POC dynamics by reconstructing changes in mass and POC accumulation rates at the key depocenters of the North Sea, such as the Skagerrak and Helgoland Mud Area (HMA), alongside a detailed characterization of the sedimentary processes in these depositional areas.

At Hereon state-of the-art sedimentation models across the coastal and shelf seas have been developed and applied, which resolve POC cycle, including the sources and sinks in benthic and pelagic compartments. Additionally, at Hereon and UHH POC cycling on different time-scales has been simulated including the identification of human impacts influencing turnover and burial of POC in the entire North Sea region, which helps to assess the temporal evolution and variability of POC deposition and distribution in the North Sea region. Within these tasks, the BUND acted as a link between scientists and policymakers, translating scientific findings into practical guidance and integrating them into social discourse as a foundation for climate policy action at national, regional and EU level.

1.3 Motivation and objectives of this thesis

Sedimentary processes play a crucial role in the burial and long-term storage of POC by directly controlling its accumulation in marine sediments and its removal from the carbon cycle. Thereby the POC accumulation is predominantly influenced by sediment transport, erosion and depositional processes (Tissot et al., 1978). In the North Sea, these sediment dynamics have been significantly shaped by both anthropogenic and climate-driven changes over the past century, influencing the sediment budget of the region (Kuijpers et al., 1993; Flettweis et al., 2010). It is crucial to understand the main sediment transport pathways and sedimentation processes in the North Sea to evaluate how this system has changed over the last century. Sediments deposited in natural sink areas serve as valuable archives for reconstructing sedimentary processes in continental shelf seas like the North Sea. Terrigenous fine-grained sediments and autochthonous marine particles are transported and redistributed across the shelf by the tidal and wind driven current regime and ultimately settle in depocenters where low bottom-current velocities allow their accumulation (Rodhe, 1996; De Haas and Van Weering, 1997; De Haas et al., 2002). These deposits serve as crucial indicators of historical sedimentary and carbon cycling processes, highlighting the interplay between environmental forces and carbon sequestration. The suspended particulate matter (SPM) transported through the entire North Sea region originates from various sources, including the northern Atlantic Ocean, European rivers, coastal and seafloor erosion, the Baltic Sea, and primary production (Eisma, 1981). This material is ultimately deposited in a number of depositional environments, such as river estuaries, tidal flats, the Helgoland Mud Area (HMA), the Skagerrak, and the Norwegian Trench (Eisma, 1981; Eisma and Kalf, 1987; Van Weering et al., 1987; Van Weering et al., 1993; Pohlmann and Puls, 1994; Oost et al., 2021). The total sediment supply to the North Sea

was estimated to range from 44 – 78 Mt yr⁻¹ (Oost et al., 2021), with approximately 28 – 46 Mt yr⁻¹ accumulating in the Skagerrak basin (Van Weering, 1987; De Haas and Van Weering, 1997; Spiegel et al., 2024). This highlights that the Skagerrak represents the largest depocenter for fine-grained sediments within the entire North Sea region. The origin and transport pathways of the sediments deposited in the Skagerrak basin are reflected in their geochemical composition, which can provide detailed insights into the sediment dynamics of the North Sea. Previous studies have investigated the sediment contributions to the Skagerrak based on clay mineral compositions and heavy metal distributions (Zöllmer and Irion, 1993; Bengtsson and Stevens, 1998; Irion and Zöllmer, 1999; Lepland et al., 2000). These studies demonstrated that the clay mineral distribution reflects the transport pathways which are controlled by the regional circulation pattern and indicates a predominant sediment supply from the southern North Sea to the Skagerrak region (Kuijpers et al., 1993; Pederstad et al., 1993). However, the primary sediment contributors to the Skagerrak and their exact proportions remain unclear and have yet to be fully quantified. Furthermore, the extent to which anthropogenic and climatic forcing factors have influenced transport processes and the relative contributions of key sediment sources to the Skagerrak basin remains uncertain. Alongside anthropogenic impacts, climate variability has also played a significant role in shaping sediment transport regimes and distribution in the North Sea over the past century (Kuijpers et al., 1993). Intensified storminess is assumed to have resulted in more energetic hydrodynamic conditions, resulting in increased SPM concentrations in the region (Capuzzo et al., 2015; Wilson and Heath, 2019). This interplay of natural and anthropogenic factors complicates efforts to disentangle their respective impacts.

The geochemical characterization of North Sea and Skagerrak sediments using radiogenic isotope signatures of clay-sized particles, combined with clay mineral compositions, serves as a robust approach to trace sediment provenance and reconstruct transport pathways within the shelf sea. The investigation of the temporal variability in Skagerrak sediment records provides valuable insights into the key factors driving sediment transport and will help to evaluate whether, and to what extent, the sedimentary system in the North Sea has changed over the past 150 years. To achieve the specific objectives of the APOC project, this study contributes by addressing the following research questions:

1. What are the primary sediment sources of detrital clays deposited in the Skagerrak basin, and what are their relative contributions to this region?

2. What are the main transport mechanisms in the North Sea, and what key factors drive these processes?
3. How have the contributions from different sediment sources to the Skagerrak basin changed since the onset of industrialization?
4. To what extent have climatic changes and anthropogenic activities influenced the transport, deposition, and provenance of fine-grained sediments in the North Sea and Skagerrak basin?

1.4 Radiogenic isotopes as a provenance tracer

Radiogenic isotope compositions of the detrital sediment fraction are a powerful tool for tracing the provenance of sediments in marine and terrestrial environments, widely used in many provenance studies (Kessarkar et al., 2003; Ehlert et al., 2011; Rodrigo-Gámi et al., 2015; Liu et al., 2016; Yu et al., 2019; Bretschneider et al., 2021; Guinoiseau et al., 2022). The radiogenic isotope ratios $^{87}\text{Sr}/^{86}\text{Sr}$, ϵNd , and ϵHf provide individual fingerprints that reflect the geochemical composition of the source rocks.

Radiogenic isotopes are formed by the decay of parent isotopes, resulting in distinct isotope signatures in rocks, which arise from variations in the relative abundances of the parent and daughter isotopes controlled by the decay processes (Dickin, 2005). Parent and daughter isotopes are fractionated based on their compatibility during crustal differentiation from the mantle as well as the formation of silicate rocks from partial melts. This refers to their ability to replace major elements in the mineral lattice. Consequently, the degree of compatibility controls the distribution of distinct isotopes in the resulting minerals. This fractionation processes result in different radiogenic isotope ratios for mantle and continental crust rocks, the compositions of which are a function of their age and rock type (Dasch, 1969; Goldstein and Jacobsen, 1987; Grousset et al., 1988; Revel et al., 1996; Colin et al., 1999).

The radiogenic isotope ^{87}Sr , which results from the radioactive decay of the parent isotope ^{87}Rb , is typically normalized to the primordial ^{86}Sr (Blum and Erel, 2003; Steiger and Jäger, 1977). Rb is preferably incorporated into magmatic melts over remaining in the mantle, which lead to higher Rb/Sr ratios in the mantle, while the continental crust exhibits higher ratios. Consequently, the preferential partitioning results in higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of old crust-derived rocks and lower ratios in young mantle-derived rocks. The radiogenic Sr isotope composition is also influenced by the degree of isotopic fractionation during chemical weathering, as

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different mineral phases display variations in their $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. During weathering, minerals with distinct isotope compositions are released from the bulk rock. This can result in large differences between the Sr isotope signature of the bulk source rock and the transported sediment (Eisenhauer et al., 1999, Blum and Erel, 2003).

The radiogenic ^{143}Nd isotope, which is produced by the decay of the parent isotope ^{147}Sm , is typically normalized to the primordial ^{144}Nd (Lugmair and Marti, 1978). Sm and Nd rare earth elements with similar chemical behaviour but Sm is slightly more compatible than Nd, resulting in its preferential retention in the residual mantle. This contrasts with the behaviour observed in the Rb-Sr isotope system. As a result, mantle rocks display high Sm/Nd ratios and correspondingly elevated $^{143}\text{Nd}/^{144}\text{Nd}$ ratios compared to older continental crust (DePaolo and Wasserburg, 1976). Due to the small differences in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios, Nd isotope compositions are commonly expressed as ϵ_{Nd} values, calculated using the following equation (Jacobsen and Wasserburg, 1980):

$$\epsilon_{\text{Nd}} = \frac{\left(\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ sample}\right) - \left(\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ CHUR}\right)}{\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ CHUR}} * 10000$$

where CHUR refers to the chondritic uniform reservoir and has a present day $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.512638. Since the parent and daughter isotopes in the Sm-Nd isotope system behave chemically similar, the differences in Sm/Nd ratios between the bulk source rock and the weathered sediment are minimal. Consequently, the chemical weathering of Nd isotopes is generally considered congruent.

The radiogenic ^{176}Hf isotope, produced by the β -decay of ^{176}Lu , is commonly normalized to primordial ^{177}Hf (Nir-El and Lavi, 1998; Scherer et al., 2001). Similar to the Sm-Nd isotope system, the compatible parent isotope Lu prefers to stay in the mantle, leading to higher $^{176}\text{Hf}/^{177}\text{Hf}$ ratios in the mantle than in the continental crust. Hf isotope compositions are expressed as ϵ_{Hf} values, analogous to ϵ_{Nd} values, using a present-day chondritic $^{176}\text{Hf}/^{177}\text{Hf}$ reference value of 0.282785 (Bouvier et al., 2008):

$$\epsilon_{\text{Hf}} = \frac{\left(\frac{^{176}\text{Hf}}{^{177}\text{Hf}} \text{ sample}\right) - \left(\frac{^{176}\text{Hf}}{^{177}\text{Hf}} \text{ CHUR}\right)}{\frac{^{176}\text{Hf}}{^{177}\text{Hf}} \text{ CHUR}} * 10000$$

The Hf isotope signatures of the bulk source rocks are also influenced by incongruent weathering such as the zircon effect (Patchett et al., 1984; Bayon et al., 2009). Zircons, which

are highly resistant to chemical weathering, contain significant amounts of Hf and are preferentially incorporated into coarser-grained sediments. This process results in less radiogenic Hf isotope signatures and in the decoupling of Hf and Nd isotopes (Patchett et al., 1984).

The similar behaviour of the radiogenic Nd and Hf isotope compositions in terrestrial rocks leads to a positive correlation commonly referred as the “terrestrial array” (Vervoort et al., 1999). Old crustal rocks of the continental crust reveal unradiogenic Nd and Hf as well as radiogenic Sr isotope signatures, while young volcanic rocks with mantle origin are characterized by radiogenic Nd and Hf and unradiogenic Sr isotope compositions. During transport and weathering processes, the zircon effect results in marine sediments having more radiogenic Hf isotope compositions relative to their Nd isotope signatures when compared to igneous rocks. This shift defines the characteristic "seawater array" observed in marine sediments (Albarède et al., 1998). Detrital clays tend to exhibit less radiogenic Hf isotope for a given Nd isotope signature compared to coarser sediments. The "clay array", derived from a combination of Chinese and Mongolian loess and dust records (Chen et al., 2013; Zhao et al., 2014) along with global river clay data, represents sedimentary rocks primarily composed of weathering products (Bayon et al., 2016). Positioned between the terrestrial array and the seawater array, the clay array reflects the effects of incongruent weathering of bulk silicate rocks. This makes it a valuable tool for tracing sediment sources and assessing terrestrial weathering intensity (Bayon et al., 2016).

This systematic variability in radiogenic isotope composition of different rocks enables to use them as a powerful tool for identifying the source rocks of the sediments deposited in the Skagerrak region. Additionally, it allows for the reconstruction of mixing relationships between different source materials, providing valuable insights into sediment transport pathway within the North Sea region. Detrital sediments derived from old Precambrian Scandinavian bedrock exhibit low $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$ ratios, accompanied by high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. In contrast, sediments originating from central Europe, which represent a diverse geology dominated by younger and Quaternary formations (Pawlewicz et al., 1997), are rather characterized by high $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$ ratios and low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios.

1.5 Study site and research cruise AL561

The North Sea represents a marginal sea located on the north-west European continental shelf that is divided into a shallow southern and deeper northern part (Fig. 1.1 A). Sediment transport and distribution in this area are primarily controlled by tidally and wind-driven currents (De Haas et al., 2002), which are forced by the inflow of the Atlantic water entering the North Sea from the south-west through the English Channel and from the north via the passage between the British and Norwegian coast. Large mud deposits are found in the deeper parts of the North Sea, where current velocities diminish, such as in the Skagerrak, the Norwegian Channel, the Helgoland Mud Area (HMA), river estuaries, tidal flats, and the Fladen Ground (Johnson and Elkins, 1979; Eisma, 1981; Bockelmann et al., 2018). Amongst these the Skagerrak represents the largest depocenter for fine-grained sediments in the North Sea.

To identify source areas of the fine grained-sediments transported across the North Sea and deposited in the Skagerrak basin, surface sediment samples were collected from the North Sea, Scandinavian fjords, the Baltic Sea, and the Skagerrak basin (Fig. 1.1 A). This enables an evaluation of the spatial distribution of radiogenic isotope signatures of clay-sized sediments and clay minerals within the North Sea region. To investigate the temporal variability of Skagerrak sedimentation and to assess changes in contributions from different sediment sources, research cruise AL561 with RV Alkor was conducted from August 2 to 13, 2021 (Schmidt, 2021).

During the cruise, undisturbed multiple cores (MUCs) approximately 50 cm in length were obtained from the central part of Skagerrak basin, near the Norwegian coast, and within the Kattegat near the Swedish coast (Fig. 1.1 B). Sedimentation rates in the inner Skagerrak are estimated to reach up to 1 cm yr⁻¹ (Ståhl et al., 2004). To receive sedimentary records covering the past century, additional gravity cores exceeding 2 meters in length were also collected.

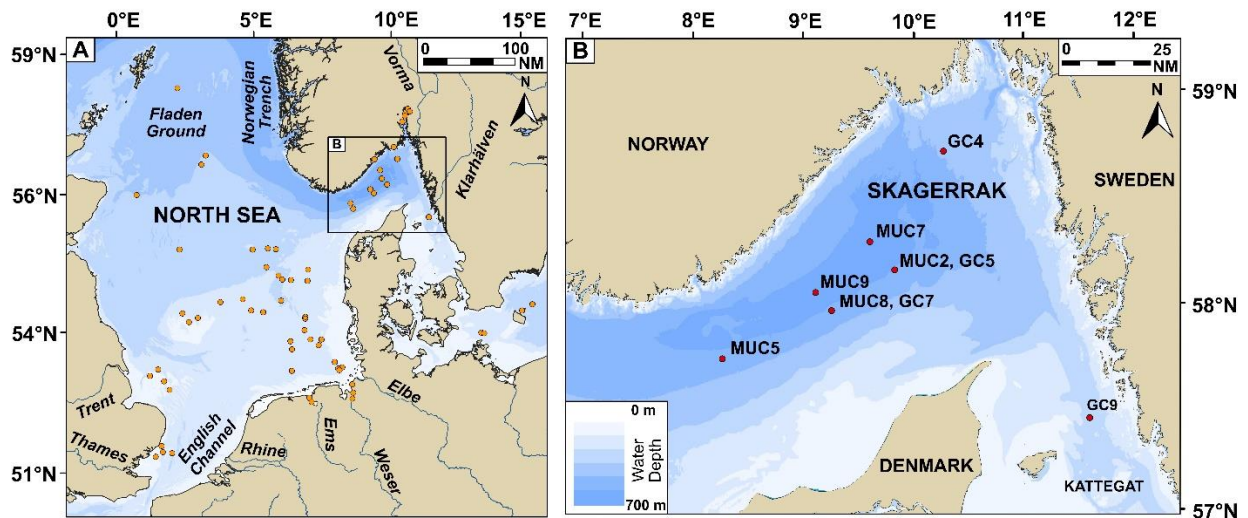


Fig. 1.1: (A) Map of the North Sea with surface sediment samples (orange dots) collected during various research cruises. (B) Skagerrak with core locations (red dots) of multiple and gravity cores collected during research cruise AL561

1.6 Outline of the thesis and declaration of contribution

The thesis is structured into three scientific chapters (I – III). Each one addresses specific research questions developed within the framework of the APOC project ("Anthropogenic impacts on particulate organic carbon cycling in the North Sea"), funded by the Federal Ministry of Education and Research, Germany (BMBF). Each scientific chapter represents an individual manuscript intended for publication in scientific journals. Chapter I has been published in *Frontiers in Marine Sciences*, chapter II has been submitted to *Continental Shelf Research*, and chapter III is currently in preparation for submission to a peer-reviewed journal.

Scientific chapter I investigates the sediment provenance of the Skagerrak deposits using radiogenic Sr, Nd, and Hf isotope signatures combined with clay mineral compositions of the detrital clay fraction. Surface sediment samples were collected from across the North Sea, Scandinavian fjords, the Baltic Sea, and the Skagerrak basin, enabling an evaluation of the spatial distribution of radiogenic isotopes and clay minerals within the North Sea region. This approach allows to distinguish between the main sediment sources, including the northern and southern North Sea as well as Scandinavia. The results provide valuable insights into the main sedimentary process in the North Sea and Skagerrak region, highlighting the important role of coastal and seabed erosion.

Contribution to chapter I: Martin Frank and Klaus Wallmann proposed this study. I coordinated the collection of surface sediment samples from various research institutes, which were

obtained during previous North Sea and Baltic Sea cruises. In the laboratory, I processed all samples, including sieving, leaching, alkaline fusion, and column chemistry, after receiving training from Ed Hathorne. I conducted isotope measurements on the mass spectrometer at GEOMAR with guidance and support from Christopher Siebert and Ed Hathorne. For clay mineral analysis, I was introduced to quantitative clay mineral preparation by Stefan Dreibrodt and subsequently carried out the laboratory preparations independently. The clay mineral samples were then measured by Brendan Ledwig at the University of Kiel (CAU). I interpreted the datasets, created all figures, and wrote the manuscript, while all co-authors provided valuable comments and feedback throughout the entire review process.

Scientific chapter II presents a down-core record of radiogenic Sr, Nd, and Hf isotope signatures as well as clay mineral composition of detrital clay-sized sediments obtained from Skagerrak sediment cores for investigating temporal variations in sediment supply to the Skagerrak basin over the past 150 years. The results provide insights into shifts in the relative contributions of different sediment sources over time. The findings highlight a reduction in sediment input from the southern North Sea over this period of time. In addition, grain-size analyses of the sediment cores were conducted to examine changes in hydrodynamic conditions within the Skagerrak basin. An upward coarsening trend in the cores suggests a shift toward increasingly energetic hydrodynamic conditions over time.

Contribution to chapter II: Martin Frank and Klaus Wallmann proposed this study. The multi-cores for this study were collected during the AL561 cruise to the Skagerrak by the scientific crew and myself. I carried out the laboratory work, including sieving, leaching, alkaline fusion, and column chemistry. I conducted isotope measurements of the prepared samples at GEOMAR with support from Christopher Siebert. For clay mineral analysis, I prepared the samples in the laboratory and Brendan Ledwig conducted the measurements at the University of Kiel (CAU). Grain size measurements were carried out by Sebastian Lindhorst at the University of Hamburg. Timo Spiegel developed the age-depth model used for this study, and I assisted in sample preparation for ^{210}Pb and ^{137}Cs measurements. I analyzed and interpreted the datasets, created all figures, and wrote the manuscript, while co-authors contributed constructive comments and feedback throughout the review process.

Scientific chapter III presents XRF core scanning records of Skagerrak sediments to reconstruct the impact factors influencing the Skagerrak sedimentation in response to climatic variability. It provides new insights into how these factors have shaped sediment provenance during colder and warmer climatic periods. The results demonstrate that during colder and

windier periods, more energetic hydrodynamic conditions have led to enhanced sediment supply from the Scandinavian coast and the northern North Sea.

Contribution to chapter III: Nina Lenz, Martin Frank and Klaus Wallmann proposed this study. The gravity cores for this study were retrieved during the AL561 cruise to the Skagerrak by the scientific crew and myself. XRF core scanning was performed by Sascha Plewe and Helge Arz at the Leibniz Institute for Baltic Sea Research Warnemünde (IOW). Grain-size measurements were conducted by Sebastian Lindhorst at the University of Hamburg. The age-depth model used for this study was established by Timo Spiegel. I helped with the sample preparation for ^{210}Pb and ^{137}Cs measurements. Matthias Moros measured the Hg concentrations at IOW. I interpreted the data, created all figures and wrote the original draft with the constructive feedback of all co-author

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2. Methods

This chapter provides detailed information on the analytical procedures used to extract Sr, Nd, and Hf from the detrital clay fraction, including the alkaline fusion method and ion-exchange chromatographic purification. The sample collection, leaching procedure, and acquisition of radiogenic isotope ratios are already outlined in the methods section of the scientific chapters (3.2.2, 4.3.3). Additionally, the specific methodologies for sample preparation (3.2.1, 4.3.1, 5.3.1), clay mineral analysis (3.2.3, 4.3.3), grain size analysis (4.3.4, 5.3.2), and XRF core scanning (5.3.4) can be found in the corresponding method sections of the respective scientific chapters (3.2, 4.3, 5.3).

2.1 Alkaline fusion method

For precise determination of isotope composition, a total sample digestion was conducted using the alkaline fusion method developed at GEOMAR to avoid HF-acid digestion (Bayon et al., 2009). 80 ng of Tm were added to crucible beakers and evaporated on a hotplate at 80°C. Subsequently, 100 mg of the detrital clay fraction together with 1.2 g of Na₂O₂ and 0.6 g of NaOH pellets were placed in the beaker. For samples with limited clay material, half of these amounts were used. These crucibles were put into a muffle furnace and heated above 15 650°C for 15 min. After cooling, 10 ml of Milli-Q water was added to dissolve the melt and precipitate the iron hydroxides, with the solution kept on a hotplate at 80°C. The solution was then transferred to 30 ml teflon vials after rinsing with Milli-Q water and heated at 130°C for two hours. Once cooled, the solution was centrifuged in pre-cleaned centrifuge tubes for 3 minutes at 3000 rpm. The supernatant was decanted, and the tube was refilled with 15 ml of Milli-Q water, stirred, and centrifuged again. This washing process was repeated twice. Following the cleaning of Fe-(Ti) hydroxides, 4 ml of 6 M HCl was added to dissolve the solids. The resulting solution was divided into three aliquots: 2 ml for Nd-Hf analysis, 1 ml for Sr analysis, and 1 ml reserved for storage.

2.2 Ion exchange chromatographic purification of Sr, Nd, and Hf

For separation and purification of Sr, Nd, and Hf, standard ion chromatographic procedures were applied (Cohen et al., 1988; Galer and O'Nions, 1989; Pin and Zalduegui, 1997). Before column chromatography, all samples were centrifuged to prevent undissolved particles from being introduced into the ion exchange columns. Sr was separated from matrix elements using

Methods

50 µl Eichrom Sr-Spec resin (50-100 µm mesh) following the modified protocol of Horwitz et al. (1992; Tab. 2.1). Nd and Hf were initially separated from other rare earth elements (REE) and matrix elements using a cation exchange resin (Bio-Rad AG 50W-X8, 200-400 µm mesh) according to the procedure outlined in Table 2.2. Nd was further separated from the other REEs using 2 ml Eichrom Ln-Spec resin (50-100 µm mesh), following the modified version of the method by Pin and Zalduegui (1997; Tab. 2.3). Hf was purified using 2 ml of Bio-Rad AG1-X8 resin (200-400 µm mesh), following the modified procedure of Sahoo et al. (2006; Tab. 2.4). Finally, to remove any residual traces and organic compounds, the Sr, Nd, and Hf cuts were treated with 100 µl of concentrated HNO₃ as well as 100 µl of 30 wt.% suprapur H₂O₂.

Step	Volume	Reagent
clean column	1 res.	0.1M H ₂ SO ₄
clean column	2 x 1 res.	Milli-Q
load resin	50 µl	Resin
clean resin	1 ml	0.1M H ₂ SO ₄
clean resin	2 x 1 res.	Milli-Q
condition resin	2 x 50 µl	3M HNO ₃
condition resin	2 x 75 µl	3M HNO ₃
load sample	50 µl	3M HNO₃ (sample)
elute sample	2 x 50 µl	3M HNO ₃
elute sample	300 µl	3M HNO ₃
collect Sr	500 µl	Milli-Q
salvage used again	backwash	Milli-Q

Tab. 2.1: Eichrom® Sr-Spec (50 µl, 50-100 µm) column chemistry for Sr purification (modified from Horwitz et al., 1992)

Step	Volume	Reagent
change the acid	2*1 ml	Milli-Q
pre-clean	10 ml	6M HCl/0.05M HF
pre-clean	0.5 ml	1M HCl/0.05M HF
pre-condition	1 ml	1M HCl/0.05 HF
load and directly collect Hf	2 ml	1MHCl/0.05 HF
collect Hf	2 ml	1MHCl/0.05 HF
elute Fe	5 ml	3M HCl
change the acid	2*1 ml	Milli-Q
elute Ba	10 ml	2M HNO ₃
collect Nd/REE	6.5 ml	6M HNO₃
clean	10 ml	6M HNO ₃ /0.5M HF
pass/store	2+2 ml	Milli-Q

Tab. 2.2: Cation AG50W-X8 (1.4 ml, 200-400 μ m) column chemistry to separate Hf and REE

Step	Volume	Reagent
pre-clean	8 ml	6M HCl
pre-condition	0.5 ml	0.1M HCl
pre-condition	1 ml	0.1M HCl
load sample	0.5 ml	0.1M HCl
wash-in	0.5 ml	0.1M HCl
elute LREE	8.5 ml	0.25M HCl
collect Nd	5 ml	0.25M HCl
clean	8 ml	6M HCl
pass and store	1+1 ml	0.3M HCl

Tab. 2.3: Eichrom® Ln-Spec (2 ml, 50-100 μ m) column chemistry for Nd purification (modified from Pin and Zaldugui, 1997)

Step	Volume	Reagent
clean	10 ml	1M HCl/1mM DTPA
clean	2 + 2 + 2 ml	Milli-Q
pre-condition	2 + 2 ml	0.5M HCl/0.5M HF
load sample	2.5 ml	0.5M HCl/0.5M HF
wash Ti repeat 3 times	10 ml	3.6M Acetic/8mM HNO ₃ /1% H ₂ O ₂
washout H ₂ O ₂	3 ml	9M Acetic
collect Hf (Zr)	5 ml	6M HCl/0.06M HF
clean	10 ml	6M HNO ₃ /0.2M HF 1% H ₂ O ₂
pass/store	2 + 2 ml	Milli-Q

Tab. 2.4: cation AG1-X8 resin (2ml, 200-400 µm) column chemistry for Hf purification (modified from Sahoo et al. (2006))

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3. Scientific Chapter I

Provenance of clay-sized detrital sediments in the North Sea and the Skagerrak region based on radiogenic Nd-Sr-Hf isotopes and clay mineral compositions: Assessing the impact of coastal and seabed erosion

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Abstract

The Skagerrak basin represents the main sink area for fine-grained sediment in the North Sea region and constitutes a natural deposition centre for sediments that are supplied from the Atlantic, the Baltic Sea and the surrounding continental margins and coasts. However, the exact sources and their proportional contributions to the North Sea sediments and to the Skagerrak deposits are not well understood. To trace the predominant sources of the sediment and to gain a better understanding of the sedimentary processes in the North Sea and the Skagerrak basin, radiogenic Sr, Nd, and Hf isotope signatures and clay mineral compositions of the detrital clay fraction of surface sediment samples from the North Sea, the Scandinavian margins and the Baltic Sea were measured. The results indicate that the major source for Skagerrak clay-size sediments is the northern North Sea but Scandinavia as well as the southern North Sea including the southern England coast also contribute material. Seabed and coastal erosion in the northern North Sea are enhanced by the inflowing Atlantic Currents, which provide the Skagerrak with high amounts of clay size sediments. In contrast, the southern North Sea, the Baltic Sea and mid-European rivers such as Weser, Elbe and Ems are only minor contributors. As Skagerrak deposits are dominated by clay sized material (up to 60%), the reconstructed sediment processes related to this study deviate from findings in previous sediment budget studies, which were based on both clay and silt fraction and indicated predominant influences from the southern

North Sea. These results highlight that coastal and seabed erosion in the North Sea is a previously underestimated source of fine-grained sediments for depocenters in the entire North Sea. With regard to climate change, the global sea-level rise will likely enhance erosional processes and can therefore significantly influence the sediment budget of the entire North Sea.

3.1 Introduction

3.1.1 The sedimentary system of the North Sea

The Skagerrak basin represents a major natural deposition centre for fine-grained sediments supplied from various regions of the North Sea. More than half of the sediments initially deposited in the entire North Sea finally end up in the Skagerrak region following current-driven transport (Eisma and Irion, 1988; Van Weering, 1981; De Haas and Van Weering, 1997; Spiegel et al., 2024).

At the same time, the sediment dynamics of the North Sea are heavily affected by anthropogenic processes, i.e., bottom trawling, land reclamation and sand mining, which may lead to enhanced bottom current velocities and sediment erosion (O'Neill and Ivanović, 2016). Riverine sediment input into the North Sea has been drastically reduced due to constructions of dams and dredging activities in particular affecting the large European rivers Rhine and Elbe (Puls et al., 1997; Rovira et al., 2014). About one billion tons of dredged material is dumped in different areas of the North Sea annually (OSPAR, 2017) which significantly influences the sediment balance and dynamics of the North Sea.

Moreover, climate forcing factors such as increasing storm intensity and wave activity have particularly changed sediment dynamics in the shallow areas of the North Sea. These anthropogenic and climate-driven effects have changed the sediment budget in the North Sea (Fettweis et al., 2010), which can be observed in large depocenters where the particles settle down.

The distribution of fine-grained sediments, their main sediment transport pathways, and potential sources in the North Sea have been the subject of various studies (Eisma and Kalf, 1987; Van Weering, 1981; Irion and Zöllmer, 1999). The terrigenous mud is transported and redistributed across the shelf by the water masses of the North Sea and finally accumulates in depocenters where low bottom current velocities are prevailing (De Haas et al., 2002). The suspended particulate matter (SPM) in the North Sea originates from the Atlantic Ocean,

European rivers, coastal erosion, the Baltic Sea as well as seafloor erosion. Sediment deposition predominantly occurs in the Skagerrak, the Norwegian Channel, the Helgoland Mud Area (HMA), tidal flats and river estuaries (Eisma, 1981; Eisma and Kalf, 1987; Van Weering et al., 1987; Van Weering et al., 1993; Pohlmann and Puls, 1994; Oost et al., 2021).

Several provenance studies have been published for the North Sea, which have mainly been based on clay mineral assemblages and heavy metal distributions in surface sediments (Zöllmer and Irion, 1993; Bengtsson and Stevens, 1998; Irion and Zöllmer, 1999; Lepland et al., 2000). These studies showed that the clay mineral distribution reflects the regional current patterns and indicates a predominant supply of sediments originating from the southern North Sea and the Atlantic, and to a minor extent from the Baltic Sea (Kuijpers et al., 1993; Pederstad et al., 1993; Bengtsson and Stevens, 1996). However, the main sediment contributors and their precise proportions in the Skagerrak are still not fully understood and has not been quantified yet. Furthermore, it is still unclear how anthropogenic impacts affect the sediment transport processes and sediment budget in the North Sea at present. As the main depocenter, the Skagerrak area receives sedimentary inputs from the entire North Sea. Given that multiple impact factors have influenced the North Sea sedimentation in the last century, it is crucial to understand the main present-day transport processes as well as the predominant sources of Skagerrak sediments. This knowledge, will allow the interpretation of down-core data of Skagerrak sediments in studies to investigate temporal changes of the sediment provenance and their driving factors over the last century or further back in time. This will help to evaluate whether and to what extent the sedimentary system in the North Sea has changed over time due to enhanced human activities and climate changes and may help to distinguish between these two impact factors. Consequently, it is crucial to determine and quantify the end-members contributing today to the main depocenter of the North Sea.

A wide range of methods exist that can be applied to identify sediment sources and quantify their contributions. They include the quantitative distribution of clay minerals (Bengtsson and Stevens 1998; Gingele et al., 2001; Bout-Roumazeilles et al., 2013), the analyses of heavy mineral content (Luepke and Clifton, 1983; Bengtsson and Stevens, 1996; Lang and Stevens, 1999), sedimentological methods including grain-size analyses of the bulk composition (Garzanti et al., 2009), and geochemical analyses based on radiogenic isotope compositions (Kessarkar et al., 2003; Ehlert et al., 2011; Bretschneider et al., 2021).

Hence, radiogenic isotope signatures of clay size sediments and clay mineral compositions represent two promising sediment provenance tracers and their combination provides a tool to

gain detailed insight into characterizing the source rock and the sediment transport pathways. This enables the reconstruction of precise proportions of the different sediment contributors applying end-member modelling.

The radiogenic Nd and Sr isotope systems are primarily controlled by the age and type of the continental rocks (Dasch, 1969; Goldstein and Jacobsen, 1987; Grousset et al., 1988; Revel et al., 1996; Colin et al., 1999). The $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ compositions of detrital sediments have thus been shown to be a powerful tracer to identify source areas and their mixing relationships and have been widely used to trace sediment provenance (Ehlert et al., 2011; Liu et al., 2016; Yu et al., 2019; Bretschneider et al., 2021).

In addition to the bulk sediment provenance, the radiogenic Sr isotope signatures are controlled by the degree of isotopic fractionation during chemical weathering due to different mineral phases of rocks exhibiting variations in the $^{87}\text{Sr}/^{86}\text{Sr}$ compositions. This can lead to large variations in radiogenic Sr isotope compositions between different mineral phases and grain sizes and consequently result in differences between the Sr isotope signatures of the bulk source rocks and the transported sediment (Eisenhauer et al., 1999, Blum and Erel, 2003). Many studies have also applied radiogenic $^{176}\text{Hf}/^{177}\text{Hf}$ signatures to reconstruct sediment transport pathways taking into account that the Hf isotope signature of the bulk source rock is also modified during sedimentation and is influenced by incongruent weathering and the zircon effect (Patchett et al., 1984; Bayon et al., 2009). Large amounts of Hf are contained in zircons, which are resistant to chemical weathering processes and are preferentially incorporated into coarser-grained sediments leading to less radiogenic Hf isotope signatures (Patchett et al., 1984). Investigations of the grain-size fractions revealed that radiogenic isotopes are useful source tracers, in particular when the same grain size fractions, e.g. clays, are considered in provenance studies (Walter et al., 2000).

In this study, we present radiogenic Nd, Sr and Hf isotope as well as clay mineral compositions of clay-sized surface sediments from the North Sea region to identify the sources of the detrital sediments and their mixing. We concentrate on the clay fraction ($< 2 \mu\text{m}$) since it can be transported by ocean currents over long distances and in contrast to bulk sediment, it is not biased by grain-size sorting effects. We then evaluate the dominant sediment transport pathways in the entire North Sea and quantify the sources of sediments deposited in the main depocenter, the Skagerrak.

3.1.2 Oceanographic and geographic setting

The North Sea is a marginal sea on the continental shelf of north-west Europe that can be divided into a deep northern and shallow southern part (Otto et al., 1990). The deeper northern part has a mean depth of 60 m and is geographically constrained by the Fladen Ground and the Norwegian Channel. The shallow southern part is dominated by the glacially formed Dogger Bank and Oyster Grounds as well as tidally generated sand banks (e.g. Norfolk Banks, Flemish Banks). The water depth in this area varies between 20 and 60 m and is on average 40 m. The sea floor basement mainly consists of Quaternary deposits with overlying reworked Holocene sediments (Stabell and Thiede, 1985; von Haugwitz and Wong, 1993). High mud contents are located in the deeper parts of the North Sea where current velocities decrease and bed shear stress is low, e.g. the Norwegian Channel, the Skagerrak, the Helgoland Mud Area (HMA), river estuaries, tidal flats, and the area southwest of the Doggerbank (Eisma and Kalf, 1987). Large areas of mud deposition are also located in the Fladen Ground and Oyster Ground where recent sedimentation of fine-grained sediments is restricted and fine-grained reworked early Holocene deposits prevail (Johnson and Elkins, 1979; Eisma, 1981; Bockelmann et al., 2018).

The Skagerrak basin represents the main depocenter for fine-grained sediments with annual mass accumulation rates of 28 - 46 Mt (Van Weering, 1987; De Haas and Van Weering, 1997; Spiegel et al., 2024a). It is located in the northeastern part of the North Sea between Denmark, Norway and Sweden with a mean water depth of 210 m and a maximum depth of 700 m in its central part and forms a connection with the shallow North Sea and the Baltic Sea (Fig. 3.1). The Skagerrak sediments are characterized by fine-grained sediments comprising the silt and clay fractions with small amounts of sandy material near the coastline (Stevens et al., 1996). In the deeper part of the Skagerrak basin the sediments contain clay contents above 50 %, which correlate positively with depth (Pederstad et al., 1993; Bengtsson and Stevens, et al. 1998).

3.1.3 Circulation pattern

The water circulation of the North Sea is primarily controlled by the North Atlantic Ocean and tidal forcing but also wind-driven currents play a role (Böhnecke, 1922; Furnes, 1980). The Atlantic water masses enter the North Sea from the north between the British and Norwegian coast and from the south-west through the English Channel (Fig. 3.1). The Atlantic inflow causes an anti-clockwise circulation in the North Sea which dominates the transport processes of suspended matter along the North Sea coast. The North Atlantic inflow, which generally

represents bottom waters, can be separated into three distinct flow paths: the Orkneys-Shetland region, the Shetland shelf area and the western part of the Norwegian Trench (Fig. 3.1). Parts of the Atlantic water masses flow south along the east coast of England via the Southern Trench Current (STC) and continue eastward through the central North Sea into the Skagerrak (Rohde, 1998). In contrast, the Atlantic surface water masses flowing westward in the Norwegian Trench as the Tampen Bank Current (TBC) is advected directly into the Skagerrak (Rohde, 1998).

The southern North Sea is mainly influenced by the inflow of Atlantic water masses through the English Channel that follows the southern coastline where the mouths of large rivers draining central Europe (e.g. Rhine and Elbe) are located. The SPM supplied by those rivers and the Atlantic Ocean in the southern North Sea is transported northward along the Danish coast by the Jutland Current (JC) to the Skagerrak (Eisma and Kalf, 1987). When the waters from the Atlantic and the Jutland Current reach the inner part of the Skagerrak, they encounter the water masses of the Baltic Current (BC) which flows northward within the Kattegat between Denmark and Sweden. This results in a counter-clockwise circulation in the Skagerrak flowing northwards along the Swedish and Norwegian coastline as the Norwegian Coastal Current (NCC) and entering the Atlantic via the Norwegian Channel (Longva and Thorsnes, 1997).

3.1.4 Geological setting

The geological structure of the hinterland of the North Sea is highly diverse (Fig. 3.1). The Fennoscandian shield forms the northern part of Europe (Scandinavia) and mainly consists of Precambrian bedrock representing the oldest rocks of the European continent (Kärki et al., 1993; Huhma et al., 2004). It includes metamorphic rocks with several generations of granitoids as well as metasedimentary and metavolcanic rocks. The oldest rocks are Archean gneisses and greenstone belts that crop out in the Kola peninsula and north-eastern Finland, where also some Paleoproterozoic rocks prevail (Gaál et al., 1987). The bedrock of Great Britain largely consists of metamorphic Precambrian and older Palaeozoic rocks whereby Scotland and Scandinavia show similar geological settings that were separated by the Mesozoic rifting of Great Britain to the west (Hallam, 1971). The southern part of England consists of younger Jurassic, Cretaceous and Tertiary rocks representing parts of a former sedimentary basin (Fig. 3.1). Compared to northern Europe, the geology of central Europe, in particular the sedimentary sequences of the coastal hinterland of the southern North Sea are much younger and of Quaternary origin (Pawlewicz et al., 1997). Furthermore, in parts of central Europe, Cenozoic volcanic rocks and

Mesozoic sedimentary rocks crop out, which are drained by the Mid-European rivers that finally enter the coastal areas of the North Sea (Bernet et al., 2004).

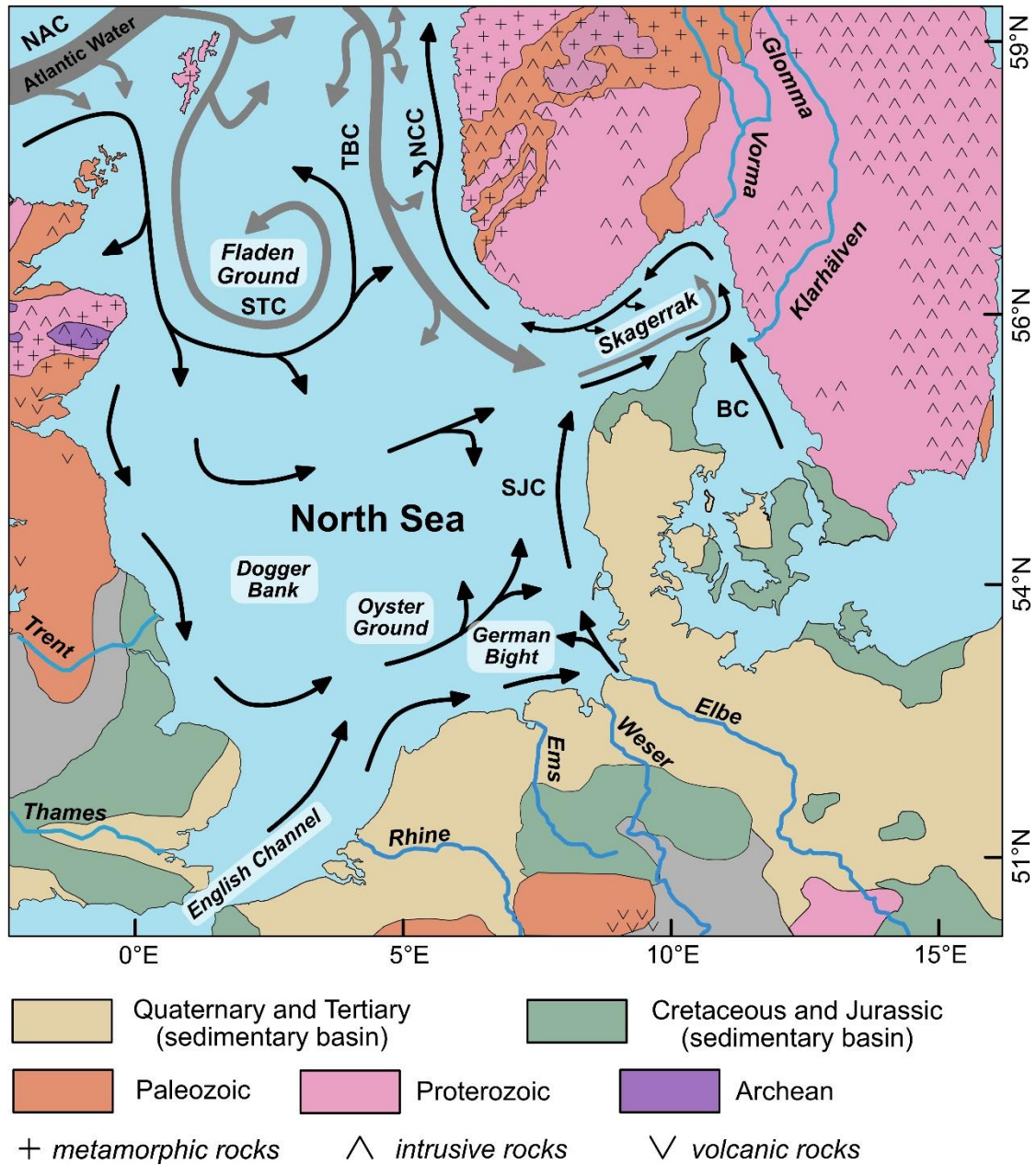


Fig. 3.1: Clay mineral distribution of the clay sized surface sediments in the North Sea in percentage: a) chlorite, (b) smectite, (c) kaolinite, (d) illite. Black dots indicate the sample locations. The maps were generated by using ODV software, version 5.6.3 (Schlitzer, Reiner, Ocean Data View, <https://odv.awi.de>, 2021).

3.2 Methods

3.2.1 Sample preparation

In total, 71 surface sediment samples (Fig. S3.1) in most cases the uppermost 0-5 cm subsampled from multi- and box corers were analysed for radiogenic Sr, Nd, and Hf isotope compositions of the clay-size fraction, as well as 32 surface samples for semiquantitative clay mineral compositions. The sample locations cover the entire North Sea and the western part of the Baltic Sea and thus all potential sediment sources.

3.2.2 Radiogenic isotopes

The sediment samples were freeze-dried and sieved through a 63 μm sieve to collect the fine-fraction in plastic bags. This was leached with a 0.05 M hydroxylamine hydrochloride (HH) – 15% acetic acid – 0.03 M Na-EDTA leaching solution to remove authigenic phases. Authigenic Fe-Mn oxyhydroxide phases were removed in a separate step following the modified leaching procedure described by Gutjahr et al. (2007). The sediments were first washed twice to remove the easily soluble fractions. Every rinsing step included the suspension of the sediment in deionized water and centrifugation to decant the supernatant. For decarbonization, the samples were treated with 40% acetic acid. This step was repeated until no reaction was visible anymore. Afterwards, the samples were treated with H_2O_2 to destroy the organic matter. The samples were rinsed three times and then the clay fraction ($< 2 \mu\text{m}$) was separated using a centrifuge-based Atterberg-method. The clay fraction was split into two aliquots for radiogenic isotope and clay mineral analyses. For radiogenic isotope analyses, the samples were leached with the HH leaching solution overnight to remove the remaining Fe-Mn oxyhydroxides and washed three times with deionized water.

After the leaching procedure, the samples were dried at 40°C in an oven. The clay fraction was completely digested using the alkaline fusion method described by Bayon et al. (2009). For separation and purification of Sr, Nd, and Hf, standard ion chromatographic procedures were applied (Cohen et al., 1988; Galer and O'Nions, 1989; Pin and Zalduegui, 1997). Sr was separated from matrix elements using 50 μl Eichrom Sr-Spec resin (50-100 μm mesh; Horwitz et al., 1992). Nd and Hf were separated from the other rare earth elements (REE) and other matrix elements using a cation exchange resin (Bio-Rad AG 50W-X8, 200-400 μm mesh). Then Nd was separated from the other REEs using 2 ml Eichrom Ln-Spec resin (50-100 μm mesh;

Pin and Zalduegui, 1997). Hf was separated using 1.6 ml of Bio-Rad AG1-X8 resin (200-400 μm mesh; Sahoo et al., 2006).

The radiogenic isotope measurements of Sr, Nd, and Hf were performed on a Nu Plasma high-resolution multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) located at GEOMAR Helmholtz Centre for Ocean Research Kiel in Germany. Before measuring the isotope compositions, the concentrations of every sample solution were determined on an aliquot of the sample solution collected after the purification procedure. Prior to the measurements, the samples were diluted to the same concentrations (50 ppb for high concentration samples and 10 ppb for lower concentration samples) as the standards, which were measured repeatedly between samples.

The instrumental mass bias was corrected by using $^{88}\text{Sr}/^{86}\text{Sr}$ of 0.1194 (Steiger and Jäger, 1977) and normalized to the NIST SRM 987 $^{87}\text{Sr}/^{86}\text{Sr}$ value 0.710245. Procedural blanks contributed between 0.6 and 18.9 % of the total Sr signal, whereby most of the samples show blank contributions below 8 %. This high blank contribution originates from the sodium hydroxide (NaOH) and sodium peroxide (NaO_2) used for the alkaline fusion. For the blank correction, the Sr concentrations and isotopic compositions of the blanks were measured. The calculated blank contribution was then removed from the total Sr isotope signal of the sample by mass balance calculation following Bretschneider et al. (2021a,b). Repeated measurements of the processed USGS rock reference material STM-1 ($n = 5$) gave a mean $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.703859 (± 0.00013), which is in good agreement with the average value of 0.703701 (± 0.000006) of the GeoReM database (Weis et al., 2006).

The measured Nd isotope ratios were mass bias corrected using a $^{146}\text{Nd}/^{144}\text{Nd}$ of 0.7219 (Vance and Thirlwall, 2002) and normalized to the JNdi-1 standard value 0.512115 (Tanaka et al., 2000). The Nd isotope ratios are reported as ϵNd values due to the small differences in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios between samples, which are calculated using the following equation (Jacobsen and Wasserburg, 1980):

$$\epsilon_{\text{Nd}} = \frac{\left(\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ sample}\right) - \left(\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ CHUR}\right)}{\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ CHUR}} * 10000$$

where CHUR is the present day chondritic $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.512638. Procedural blanks for Nd were not higher than 0.8 % of the total amount of Nd in the samples. Repeated measurements of the USGS rock reference material STM-1 ($n = 5$) show a mean ϵNd value of

5.15 (+/- 0.48), which agrees well with the mean value of 5.36 (+/- 0.18) provided by Weis et al. (2006).

The Hf isotope ratios were corrected for instrumental mass bias using a $^{179}\text{Hf}/^{177}\text{Hf}$ of 0.7325 and then normalized to a JMC 475 value of 0.282163 (Blichert-Toft and Albarède, 1997; Blichert-Toft, 2008). The Hf isotope compositions are reported as ϵHf values analogous to the ϵNd values using a present day chondritic $^{176}\text{Hf}/^{177}\text{Hf}$ value of 0.282785 (Bouvier et al., 2008). Procedural blanks for Hf were not higher than 2.9 % of the total amount of Hf in the samples. Repeated measurements of the USGS rock reference material STM-1 ($n = 5$) give a mean ϵHf value of 8.44 (+/- 0.68), which agrees well with the mean value of 8.36 (+/-0.79) given in the GeoReM database (Jochum et al., 2005).

3.2.3 Clay minerals

For clay mineral analyses, subsamples were taken from the clay-size fraction obtained from the Atterberg separation. Pedogenic iron oxides of the clays were removed using 0.3 M sodium citrate, 0.2 M sodium hydrocarbonate (NaHCO_3) and sodium dithionite. The dissolved iron compounds were washed out four times with 2M sodium chloride. 1/3 of the clay suspension was saturated and flocculated with an excess of K^+ -ions and 2/3 of the clay suspension/solution with an excess of Mg^{++} -ions. The ion excess of both saturated clay suspensions was removed via dialysis tubes with deionised water to electrical conductivity values below 20 μS . The K- and Mg- saturated samples were freeze-dried and 50 mg of the respective clay fraction was dispersed in 4 ml deionised water in an ultrasonic bath for 10 min. A subsample of 2 ml of the suspension was placed on a ceramic slide/plate and was dried for 48 hours in preparation of the measurement. Clay mineral measurements were performed at the Institute of Geosciences of the University of Kiel (CAU) by x-ray diffractometry (XRD) using a D8 Discover (Bruker AXS) with a Cu-cube as well as θ - θ -geometry. Samples were measured with 2θ at 0.037° steps with a scanning speed of 1.5 s at each step. The measurement was repeated after the oriented Mg-saturated sample slide was treated with ethylene glycol saturation and the K-saturated sample was heated to 550°C for two hours.

The analyses of the diffractograms were performed via the open-source software Profex using the BMBG and COD database for identifying the mineral phases. Four main groups of clay mineral phases, illite, kaolinite, chlorite and smectite were differentiated. The semi-quantitative analyses of the clay mineral content were carried out by determining the peak areas of the

respective mineral phases and were calculated following Whitton and Churchman (1987) and by using the weighting factors recommended by Laves and Jähn (1972).

3.3 Results

3.3.1 Radiogenic isotope characterization of the clay-size surface sediments

The $^{87}\text{Sr}/^{86}\text{Sr}$, ϵNd and ϵHf signatures of the detrital clay fraction ($< 2 \mu\text{m}$) of the surface samples across the entire North Sea are reported in Tab. S3.1 and Figure 3.2, respectively. The Sr isotope signatures range from 0.726905 to 0.775977 and show the most radiogenic values in the Scandinavian fjords and the Baltic Sea. Lower radiogenic Sr isotope ratios are found in the southern North Sea, especially in the samples originating from European estuaries (Weser, Ems, Elbe).

The ϵNd values range between -17.7 and -10.6 and show the least radiogenic values in the Baltic Sea and Scandinavian rivers and most radiogenic values in the southern North Sea and German river estuaries. The ϵHf signatures vary between -15.2 and -4.8 with the least radiogenic values in the central North Sea and the highest values in the southern North Sea. However, some sediment samples from the Weser and Ems estuaries also show low ϵHf signatures between -6.7 and -5.7.

The isotopic signatures of the surface samples collected from the sediment basin of the Skagerrak range between intermediate ϵNd values of -14.6 to -13.4, between 0.73875 and 0.74260 for the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic compositions and are comparable to the average isotopic values of the surface sediment samples. Similarly, the ϵHf signatures of the Skagerrak surface sediments are close to the average North Sea values ranging between -8.4 and -7.0. The surface samples from the northern North Sea reflecting the Atlantic inflow into the northern North Sea are also characterized by intermediate ϵNd values of around -14 and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios around 0.74.

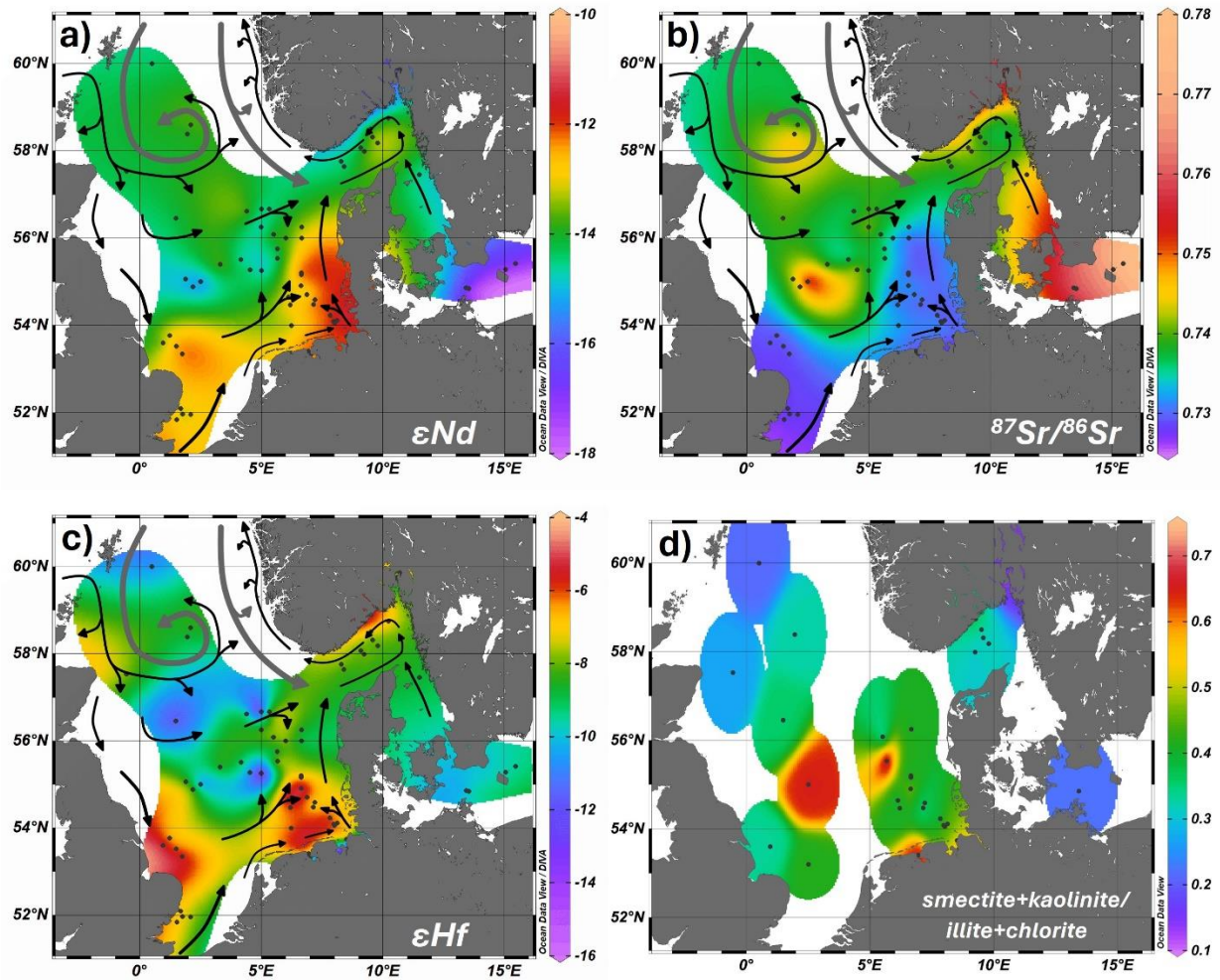


Fig. 3.2: Distribution of radiogenic isotopes of the clay-sized surface samples in the North Sea with circulation patterns: (a) ϵNd (b) $^{87}Sr/^{86}Sr$ (c) ϵHf (d) Clay mineral distribution of the secondary and primary clay minerals (smectite+kaolinite /illite+chlorite). Black dots indicate the sample locations. Black arrows indicate the subsurface currents and grey arrows mark the Atlantic deep water inflow. The maps were generated by using ODV software, version 5.6.3 (Schlitzer, Reiner, Ocean Data View, <https://odv.awi.de>, 2021).

3.3.2 Clay mineralogical composition of the surface samples

The smectite distribution in the North Sea shows a clear pattern with highest contents in the sediments of the southern North Sea and the Central-European rivers with a maximum value of 32.3 % in the Ems estuary and a decreasing trend along the west coast of Denmark with lowest values in the Skagerrak between 4.48 and 8.37 % (average of 13.7; Tab. S3.2; Fig. 3.3). The surface samples in the northern North Sea which likely reflect the Atlantic input reveal contents similar to the Skagerrak. Compared to the smectite distribution the kaolinite content in the North Sea reveals smaller variations between 7.2% and 22.1 % (average of 14.3 %). The

dominant clay mineral contents of illite and chlorite exhibit values ranging from 38.8 to 67.9 % (average of 53.1 %) and 3.7 and 38.18 % (average of 18.9 %), respectively.

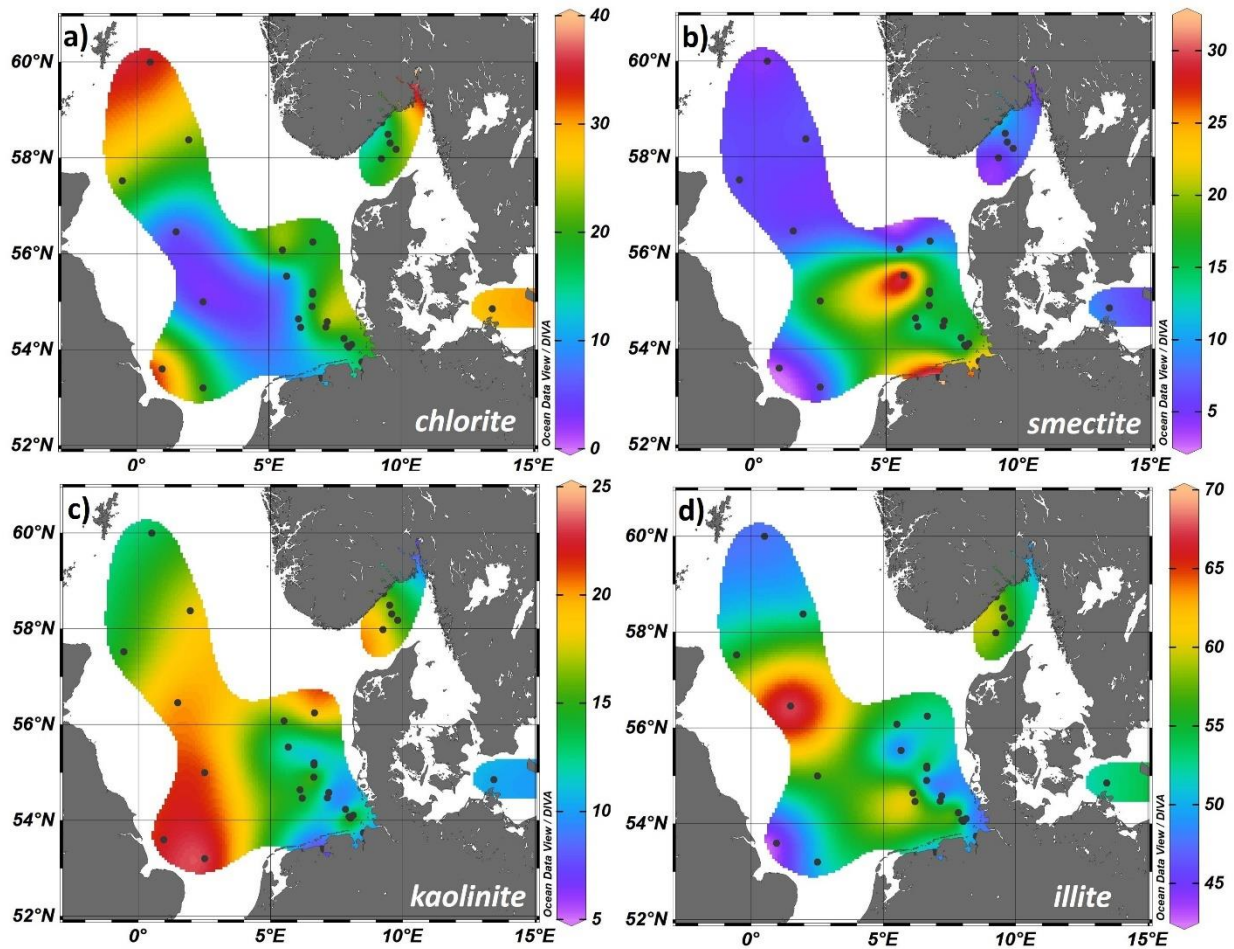


Fig. 3.3: Clay mineral distribution of the clay sized surface sediments in the North Sea in percentage: a) chlorite, (b) smectite, (c) kaolinite, (d) illite. Black dots indicate the sample locations. The maps were generated by using ODV software, version 5.6.3 (Schlitzer, Reiner, Ocean Data View, <https://odv.awi.de>, 2021).

3.4 Discussion

3.4.1 The main sediment contributors to the Skagerrak region

To disentangle the sediment contributions from different sources that accumulate in the depocenter of the Skagerrak, the potential source areas need to be defined and characterized. The isotopic signatures reveal a sharp boundary between the northern and southern North Sea reflecting the contribution from different source areas (Fig. 3.2). The clays originating from the southern North Sea and in particular of the estuaries of the middle European rivers Elbe, Weser and Ems are characterized by more radiogenic ϵNd and less radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ isotope values

than clays originating from the Baltic Sea and the Scandinavian coast (Fig. 3.2 a, b), which reflects the distinct source rocks of the hinterland. During partial melting the mother isotope of the radiogenic ^{143}Nd behaves more compatible in the mantle than the primordial ^{144}Nd resulting in lower $^{143}\text{Nd}/^{144}\text{Nd}$ ratios in continental crust compared to the mantle (DePaolo and Wasserburg, 1976). In contrast, the mother isotope of the radiogenic ^{87}Sr is incorporated into the magmatic melt rather than staying in the mantle leading to higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in old continental rocks compared to young mantle-derived rocks (Wilson, 1989). The sediments from Scandinavia typically originate from old Paleozoic-Proterozoic rocks (Fig. 3.1) and consequently detrital sediments from Scandinavian fjords have unradiogenic Nd and radiogenic Sr isotope ratios. Surface sediments from the Baltic Sea show even less radiogenic ϵNd and more radiogenic Sr isotope signatures than the surface sediments of the Norwegian fjords. This is consistent with the estuarine systems of the Baltic Sea predominantly supplying older crustal Proterozoic-Archean sediments into the Baltic Sea (Andersson et al., 1992). Furthermore, most fine-grained sediments in the Baltic Sea are provided by coastal and seabed erosion, particularly of glacial till (Wallmann et al., 2022) originating from Scandinavia, particularly from Finland characterized by unradiogenic ϵNd and radiogenic Sr isotope values. Moreover, the measured isotope signatures of the Baltic Sea are consistent with ϵNd signatures of Scandinavian-river sediments (Kymijoki, Ume, Lule) ranging between -17.6 and -20.4 previously determined by Bayon et al. (2015).

Clay-size sediments in the southern North Sea, including the English Channel, German Bight and large parts of the Danish North Sea sector, reveal more radiogenic ϵNd and less radiogenic Sr isotope signatures with peak values observed for the clay fractions of the central European-river estuaries (Fig. 3.2 a, b). These sediment signatures reflect the hinterland geology of central Europe which is dominated by Quaternary and Tertiary sediments (Fig. 3.1; Pawlewicz et al. 2002). Highly radiogenic Nd and isotope signatures of -9.10 and -11.5 were also found for sediments of Rhine and Seine by Bayon et al. (2015).

The Skagerrak clay sediments are a mixture of these two source regions given that the radiogenic isotope signatures are intermediate between Mid-European-like and Scandinavian-like isotope ratios. That implies that the surrounding Scandinavian hinterland is not the main contributor to Skagerrak sediments. This is supported by previous studies who reported that large amounts of particles (50 - 90%) are transported laterally into the Skagerrak, whereas only a small proportion of Skagerrak deposits originate from local sources including local riverine and coastal erosion inputs (Van Weering et al., 1993; De Haas and Van Weering, 1997; Spiegel

et al., 2024). Strong differences between the isotope signatures of the Skagerrak and the Baltic Sea surface sediments (Fig. 3.2) suggest that the Baltic Sea is an insignificant source for Skagerrak clays. This is in good agreement with further studies which have shown that the SPM supply entering the North Sea through the Skagerrak-Kattegat transition is comparatively low (Eisma and Irion, 1988; Pohlmann and Puls, 1994). Conversely, the clays in the northern part of the North Sea reveal isotopic ratios similar to the Skagerrak sediments (Fig. 3.2). These clays partly reflect the Atlantic input following the circulation pattern that clearly shows that Atlantic water masses entering the North Sea from the north between the British and Norwegian coasts flow directly into the Skagerrak region (Fig. 3.1). In fact, Bengtsson and Stevens (1997) as well as Pederstad et al. (1993) already mentioned that the North Atlantic deep water transports muds into the Skagerrak, but only in small proportions compared to the Jutland Current coming from the southern North Sea. However, when following the Jutland Current from the southern North Sea to the Skagerrak, the isotopic compositions of the clays clearly show a decreasing ϵ_{Nd} and increasing Sr isotope signature (Fig. 3.2 a, b), which makes the southern North Sea and the Mid-European rivers unlikely main contributors to the Skagerrak. This is also supported by the clay mineral distribution which reveals a profound decrease in smectite and increase in kaolinite content from the German Bight along the west coast of Denmark to the Skagerrak (Fig. 3 b, c).

Combining mineralogical and isotopic data, the Skagerrak and the northern North Sea surface sediments display similar characteristics in similar ranges of the data (Fig. 3.4) which implies that the northern North Sea is the predominant supply area of fine-grained sediments in the Skagerrak.

These observations based on Nd and Sr isotopic data as well as clay mineral data allow the definition of three main source end members including the northern North Sea, southern North Sea, and Scandinavia, whereby the northern North Sea appears to be the dominant source of the Skagerrak clays, which cannot be explained by a two-endmember mixture of the southern North Sea and Scandinavia only (Fig. 3.5). This contrasts with the conclusions of previous provenance studies of Skagerrak sediments, which rather considered the southern North Sea to be the dominant contributor (Zöllmer and Irion, 1993; Bengtsson and Stevens, 1998; Irion and Zöllmer, 1999; Lepland et al., 2000).

Figure 3.5 shows that the Skagerrak clays are plotting close within the field defining samples from the northern North Sea, suggesting high supplies from this region, although some Skagerrak samples seem to be rather a mixture between the southern North Sea and Scandinavian region. However, the spatial distributions of the radiogenic isotopes and the clay

minerals (Fig. 3.2, 3.3) clearly indicate that the northern North Sea and the Skagerrak sediments have similar geochemical characteristics, which differ from the southern North Sea and Scandinavian sediments. This shows that the northern North Sea is the predominant sediment contributor to the Skagerrak basin.

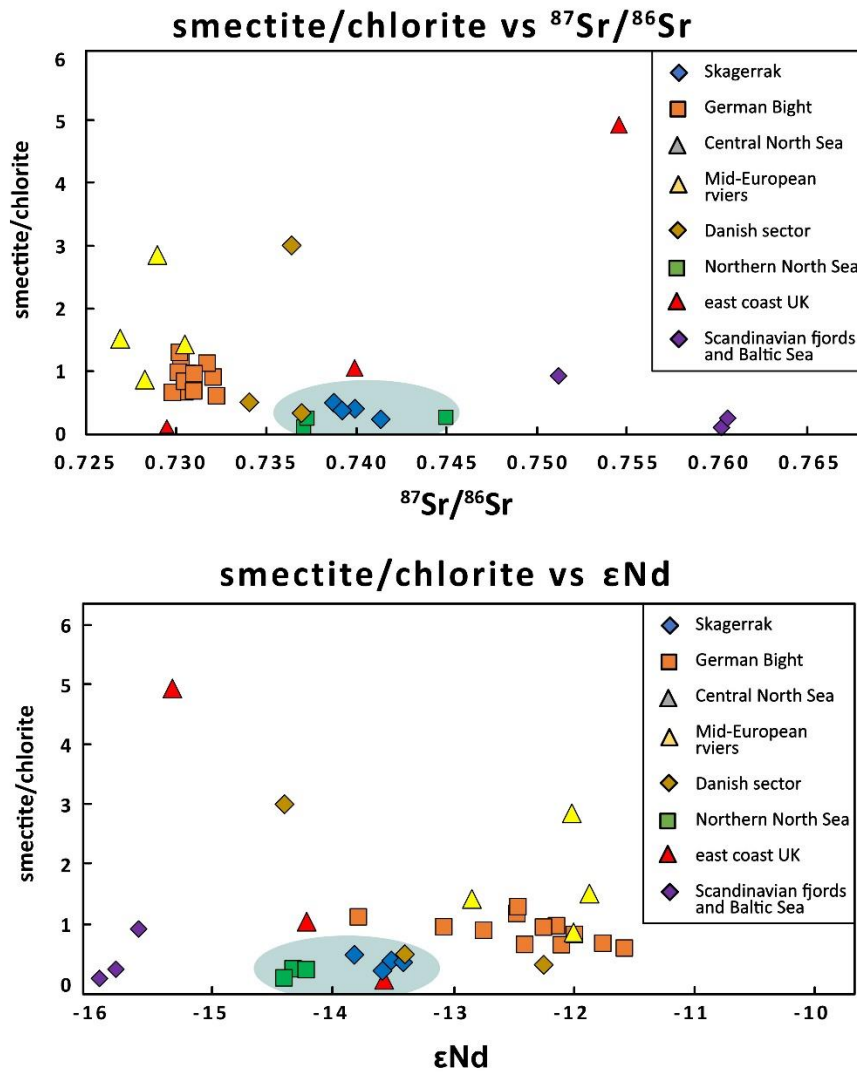


Fig. 3.4: Clay minerals (smectite/chlorite) and radiogenic isotopes ($^{87}\text{Sr}/^{86}\text{Sr}$, ϵNd) in surface samples of the North Sea. The light blue colored area marks the northern North Sea and Skagerrak surface sediments with similar isotopic and clay mineral characteristics.

Our results demonstrated that the radiogenic isotope signatures not only provide information about geochemical characteristics of the parent bedrock of the respective source, but also about the transport pathways of clays and the circulation pattern which significantly influences the distribution of fine-grained-sediments in the North Sea. This can be seen in the northern North Sea where major transport pathways from the North Atlantic such as the Tampen Bank and Central North Sea Current transport particles directly to the Skagerrak resulting in similar

isotopic characteristics in both regions. Most previous studies mentioned that the SPM dynamics of the North Sea are dominated by the input from the Atlantic, although the sediment supply across Dover Strait is higher than that of the northern pathway (Eisma and Irion, 1988; Sündermann, 1993; Puls et al. 1997). This results in higher SPM concentrations in the southern North Sea compared to the northern North Sea (Eisma, 1981). This evidence is now compared to the clay mineralogical and radiogenic hafnium isotope distribution in the study area.

3.4.2 Clay mineral distribution in the North Sea

In general, the clay mineral distribution shows a domination of the primary clay minerals illite and chlorite together representing 72% of the fine-grained North Sea sediments with the highest abundances in the northern North Sea, as well as the Skagerrak (Fig. 3.2d). These abundances reflect limited influence of clay minerals newly formed during weathering processes and confirm dominant physical erosion under cold climatic conditions, which is consistent with the environmental conditions prevailing in the North Sea hinterland where reworking of immature glacial deposits currently occurs. During cold periods of the Pleistocene the ice sheets transported illite and chlorite-rich till from the Fennoscandian Shield into the North Sea basin, resulting in high illite and chlorite contents in the northern part of the basin (Irion and Zöllmer, 1999; Fig. 3a, d). Griffin et al. (1968) and Chamley (1975) also found high illite and chlorite contents in marine sediments from the North Atlantic, which would be consistent with an Atlantic input to the northern North Sea and which complicates distinguishing these source areas. However, the low smectite content in the Fladen Ground (Fig. 3.3), which represents a hotspot of mud deposition, differ from clay mineral contents determined by Johnson and Elkins (1979) in sediments north of the North Sea shelf, which makes the Atlantic an unlikely dominant source for northern North Sea sediments. Furthermore, the SPM concentrations in the North Atlantic at the northern part of the North Sea shelf are much lower than observed in the Fladen Ground area (Pohlmann and Puls, 1994; Gayer et al., 2006). This supports the conclusions of Eisma (1973) and Eisma and Kalf (1987) that Fladen Ground sediments are reworked early Holocene deposits and that the North Atlantic is only a minor contributor to North Sea sediments. From these results it can be concluded that the sediments entering the Skagerrak via the Atlantic Currents preferentially originate from seabed erosion of old relict deposits in the northern North Sea rather than from the Atlantic inflow north of the Shetland Islands. This is also consistent with the previously mentioned observation that Skagerrak sediments display similar mineralogical and isotopic characterizations as the northern North

Sea sediments and supports the conclusion that the northern North Sea is a predominant source for clays deposited in the Skagerrak.

The kaolinite content in the North Sea shows strikingly high values at the (southern) east coast of England (Fig. 3.3c). Kaolinite is primarily formed by continental chemical weathering processes and mostly originates from Weichselian tillites with Tertiary origin or old eroded rocks transported seawards by fluvial or coastal processes (Exley 1976; Chamley 1989). Furthermore, it is the dominant clay mineral in the Upper Triassic and Lower Jurassic rocks (Mørk et al. 2003) which dominate the south-eastern coast of England (Fig. 3.1). Consequently, the kaolinite-rich sediments in the south-western North Sea imply an England origin by riverine input or coastal erosion. McCave (1987), as well as McManus and Prandle (1997), found that the supply from intensive cliff erosion at the Holderness and Norfolk cliffs, are the predominant source of SPM in this region, whereas fluvial supply is negligible. This demonstrates the importance of coastal erosion in the sediment budget of the sediments in the south-eastern North Sea. Furthermore, most clays from the east coast of UK plot outside of the Nd-Sr mixing triangle (Fig. 3.5) suggesting a fourth end member in addition to the previously defined endmembers for the Skagerrak (southern North Sea, northern North Sea, Scandinavia). With the strong influences of cliff erosion in these regions in mind, it is likely that coastal erosion is a dominant sediment source for suspended matter at the east coast of England. It is conspicuous that certain parts of the Skagerrak show relatively high kaolinite contents (Fig. 3.3c) which are also observed in the clay mineral data of Irion and Zöllmer (1999) from East Anglia to the Skagerrak region. This implies a significant input from the south-eastern coast of England to the Skagerrak sediments.

The southern North Sea is characterized by higher secondary clay mineral contents compared to the northern part of the North Sea (Fig. 3.2 d). This is the result of relatively high amounts of smectite in the south-eastern North Sea with the highest contents in the German river estuaries (Fig. 3.3) which may be supplied by altered volcanic rocks from central Europe. Therefore, the profoundly lower smectite contents in the German Bight sediments indicate only a relatively small fluvial sediment supply from Mid-European rivers. Puls et al. (1997) established a SPM budget for the German Bight that clearly indicated that German rivers only supply negligible amounts of SPM to the southern North Sea. Further investigations of sediment dynamics in the southern North Sea have shown that the highest concentrations of suspended matter prevail in the English Channel where they are entrained by currents from the south into the southern North Sea and the German Bight (Eisma, 1981; Pohlmann and Puls, 1994). In fact,

the southwestern North Sea, including the English Channel and the southwest coast of the UK, show isotope signatures similar to the German Bight. Hence, given that the German rivers show much higher smectite contents than the open southern North Sea also including the English Channel, it is likely that the radiogenic isotope distribution of the clay fraction in this area reflects the circulation pattern of the southern North Sea.

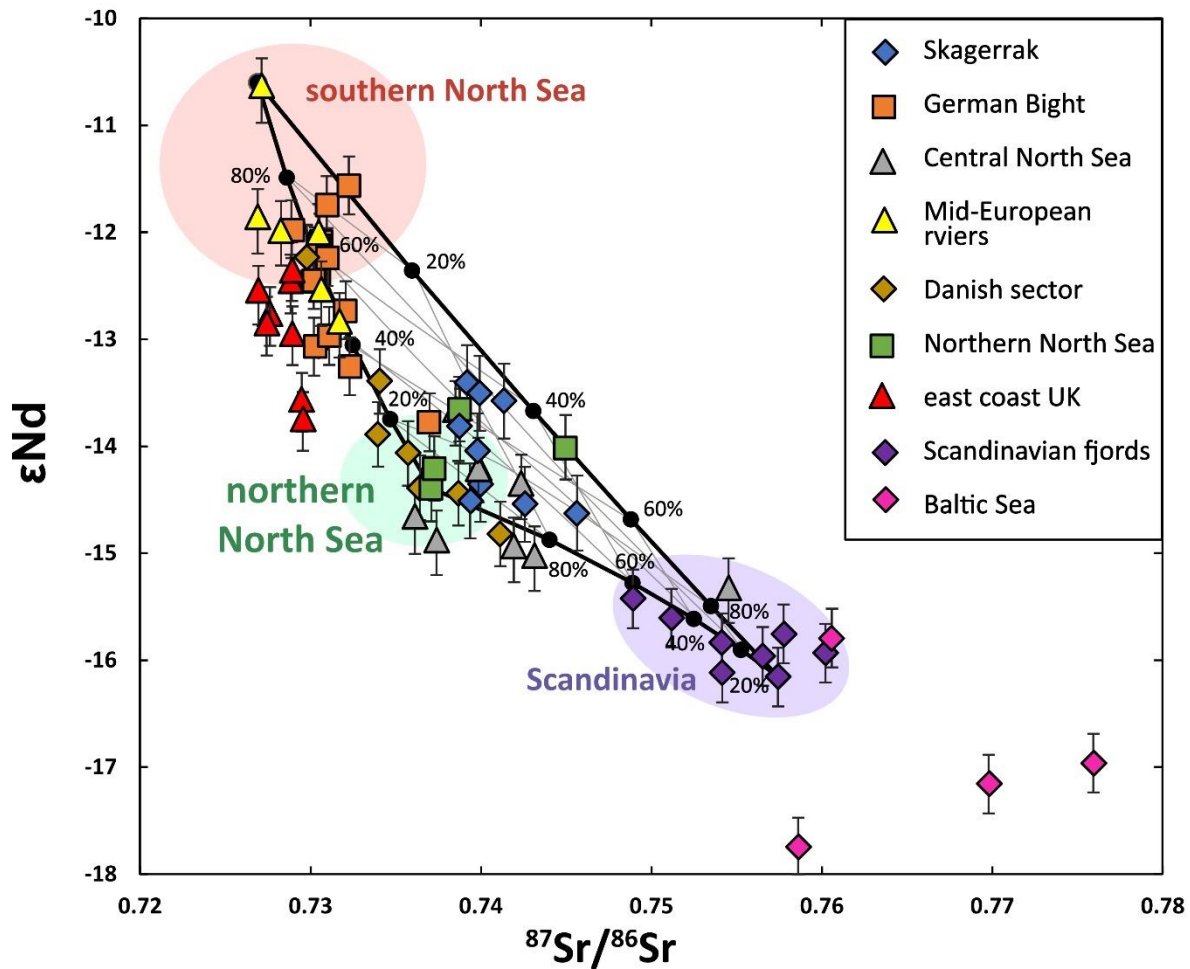


Fig. 3.5: Nd-Sr isotope composition of the clay size fraction of the surface sediments in the North Sea with the defined end members of the southern North Sea, northern North Sea and Scandinavia and the resulting mixing field. The 2SD error bars for Sr isotopes are smaller than the symbol size. The grid was calculated according to a 10% increment of the mixture composition. The isotopic compositions are based on measured data of the respective source area. The concentration data for the calculated endmembers is based on literature data of the respective source area (Blanchet et al., 2019).

Irion and Zöllmer (1999) presented an extensive study of the clay mineral distribution of the entire North Sea that is similar to our study. Their clay mineral calculation follows Biscaye (1965) and therefore resembles the calculation method used in this study. However, our

smectite contents in the Skagerrak and parts of the German Bight reveal much lower values and resemble the smectite contents measured by Bengtsson and Stevens (1997) in the Skagerrak area. Irion and Zöllmer (1999) mainly analysed surface samples from the upper 20 cm which represent modern sedimentary conditions of the North Sea, but rather within the Holocene period. In contrast, surface sediments from our study and those from Bengtsson and Stevens (1997) only include the upper 0-5 cm of the sediments which is more representative for the current century in the Skagerrak. Therefore, these differences in smectite contents may suggest a decline of smectite input to the southern North Sea and the Skagerrak. Since the Mid-European rivers constitute a predominant source of smectite, this likely indicates a decreasing riverine input to the North Sea over time. A possible explanation could be river damming, which has progressively increased for major European rivers such as the Elbe, Weser, and Ems (Hübner and Schwandt, 2018) and has led to a decrease in the SPM concentration in the rivers (Ericson et al., 2006; Rovira et al., 2014). Consequently, river damming may have diminished the riverine sediment supply to the ocean and resulted in a reduced contribution to the German Bight and Skagerrak sediments.

3.4.3 Radiogenic Hf isotopes tracing sedimentary processes

Despite that the radiogenic Hf isotope composition can be strongly influenced by fractionation processes during weathering, the ϵHf distribution in the North Sea reveals patterns generally similar to the Nd and Sr radiogenic isotope systems, but also shows distinct differences in specific regions (Fig. 3.2). These imply a decoupling of Nd and Hf isotopic composition in the respective source areas due to incongruent weathering and sorting processes. Comparison of ϵHf and ϵNd data exhibits that the clays are isotopically similar to global river clays (Bayon et al. 2016; Fig. 3.6a) suggesting that the clay fraction experienced a loss of Hf-bearing zircons and only a moderate degree of chemical weathering, which is consistent with expectations for relatively cold climate conditions. However, some samples, in particular most of the Weser and Ems surface samples, plot near the terrestrial array (Vervoort et al., 1999) with unexpectedly unradiogenic ϵHf signatures. Even though zircons are thought to only occur in negligible amounts in the clay fraction, Marchandise et al. (2014) concluded that an influence of zircons remains in clay-sized sediments given that they can also occur in very fine grains. Prior to grain-size separation for the clay analyses, the river sediments revealed high sand proportions. It is likely that the clay-sized sediments we measured are partly experienced a high degree of physical weathering during long transport pathways through rivers causing the disintegration

and abrasion of sand-sized sediments which tends to contain high amounts of Hf-bearing zircons. It is noteworthy that the ϵ_{Hf} signatures in the German Bight are much more radiogenic than the river sediments of Weser and Ems (Fig. 3.2 c), which supports the observation that German rivers are minor contributors to the sediment budget of the North Sea and that major amounts of SPM are supplied to the southern North Sea via the English Channel.

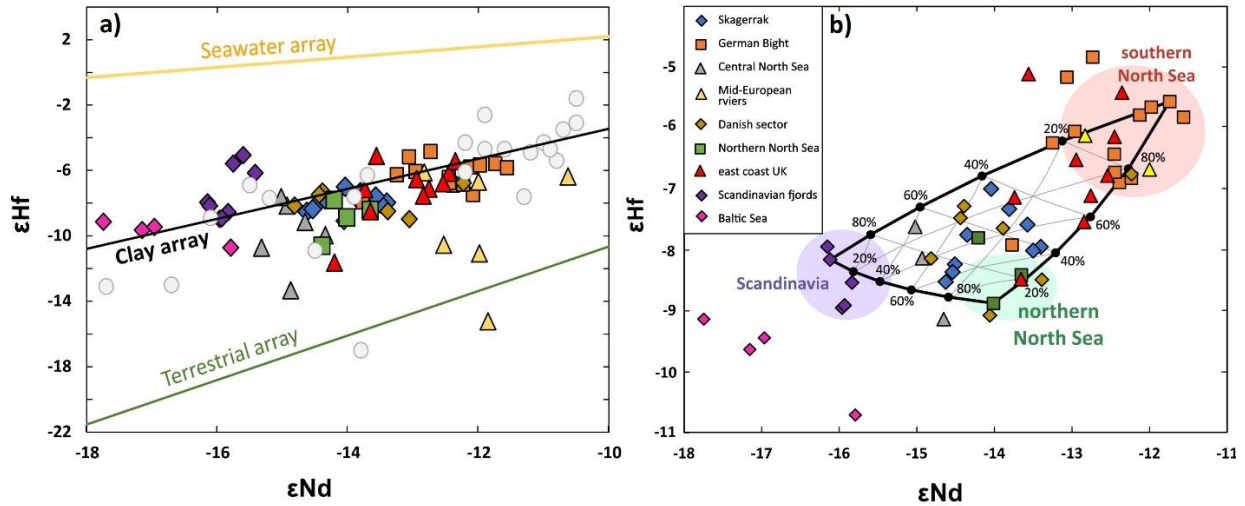


Fig. 3.6: Combination of ϵ_{Hf} and ϵ_{Nd} signatures of surface samples of the North Sea: a) clay surface samples with Seawater Array (Albarede et al., 1998), Clay Array (Bayon et al., 2015), and Terrestrial Array (Vervoort et al., 1999), b) Hf-Nd isotope with the defined end members of the southern North Sea, northern North Sea and Scandinavia and the resulting mixing field. The grid was calculated according to a 10% increment of the mixture composition. The isotopic compositions are based on measured data of the respective source area. The concentration data for the calculated endmembers is based on literature data of the respective source area (Blanchet et al., 2019).

The $\Delta\epsilon_{\text{Hf}}_{\text{clay}}$ signatures, which describe the deviation of the measured Hf isotopic compositions from the Clay Array, are linked to climatic parameters and correlate positively with the degree of weathering (Bayon et al. 2016). Since some samples show relatively high positive or negative $\Delta\epsilon_{\text{Hf}}_{\text{clay}}$ signatures (Fig. S3.2) probably due to strong fractionation processes, e.g. incongruent weathering, they were excluded in the end-member mixing field of ϵ_{Hf} and ϵ_{Nd} (Fig. 3.6b). Compared to the Nd-Sr isotope mixing field where most clays from the east coast of UK show less radiogenic Sr values and therefore plotting outside the mixing field (Fig. 3.5), which were previously interpreted to be influenced by coastal erosion, plot near the southern North Sea end member in the Hf-Nd isotope mixing relationship (Fig. 3.6b). Sediments transported via large rivers are often physically and chemically weathered by intensive erosion and transport processes (Bouchez et al., 2012). Therefore, river sediments mostly have a higher degree of

alteration than sediments originating from rock coast erosion which supplies newly exposed rocks from cliffs (Costa et al., 2015; Prémaillon et al., 2018). The east coast of England, as Holderness and Norflok, is composed mainly of Quaternary sediments with glacial till (McCave, 1987; Pye and Blott, 2015), but also large parts at the south-eastern coast of England consist of upper Triassic and Lower Jurassic rocks where chalk cliffs are present (Mørk et al. 2003) which exposed sediments are likely less altered than river sediments. Consequently, the slight shift of some UK clays toward less radiogenic values can possibly be explained by a lower chemical weathering intensity compared to other samples originating from the southern North Sea when it is assumed that these sediments are mainly contributed by cliff erosion. This would also support the finding that UK sediments are mainly contributed by coastal erosion.

When calculating the contributions of the northern North Sea, the southern North Sea and Scandinavia to the sediments deposited in the Skagerrak based on the three radiogenic systems of Nd, Sr and Hf, it results in 43 % originating from the north, 26 % from the south and 31 % from Scandinavia. These calculated contributions of the respective endmembers are in good agreement with the previous findings and confirms that the seabed erosion in the northern North Sea plays a major role in contributing sediments to the Skagerrak compared to the southern North Sea. This also demonstrates that the combination of all three isotope systems is a sensitive and powerful tool to trace and quantify sources of erosional material.

3.4.4 Reconstruction of the sedimentary systems in the North Sea and the role of coastal and seabed erosion

Previous studies on the sediment budget of the North Sea have shown that the main source of SPM in the North Sea is the North Atlantic which supplies material between Scotland and Norway and across the Strait of Dover (McCave, 1973; Eisma and Kalf, 1987; Puls et al., 1997). As strong currents prevail in the English Channel, the SPM does not settle in this region but is transported further into the southern North Sea. These strong currents lead to intensive erosion processes due to high bed shear stress velocities in the eastern Channel where sediment is also supplied by coastal and seabed erosion, which results in much higher suspended matter concentrations compared to the Atlantic entry (Eisma, 1981; Puls and Sündermann, 1990). This is also supported by much higher suspended matter concentrations in the Dover Strait than observed in the adjacent Atlantic (UKMMAS, 2010). In addition, coastal cliff erosion at the west coast of the North Sea, especially at the East Anglia Coast with the highest SPM concentrations in the North Sea (Stanev et al., 2009), could lead to high suspended matter

concentration in this region. These observations are also supported by similar isotopic signatures in the English Channel, the southern east coast of England and the German Bight indicating that the coastal erosion at the English coast and within the English Channel supplies the southern North Sea with SPM. Furthermore, the rather radiogenic ϵNd values suggest a source rock of Mid-European origin which can be found at the coast surrounding the English Channel and the southern North Sea (Fig. 3.1, 3.2). Since we already showed that Mid-European rivers only provide negligible amounts of suspended matter to the ocean, it is likely that large proportions of fine-grained sediments which are transported and partly deposited in the inner German Bight originate from erosion processes within the English Channel.

The northern North Sea, which we proposed to be the predominant source for Skagerrak sediments, is dominated by the Fladen Ground where fine-grained sediments can be found (Eleftheriou and Basford, 1989; Bailey et al., 1993). Eisma (1981) suggested that seafloor erosion in the northern North Sea represents a source for fine-grained sediment, which is primarily caused by the inflow of Atlantic Waters supplying the Skagerrak with sediment (Eisma and Kalf, 1987; Weering and Kalf, 1987). This suggests, that Skagerrak sediments mainly originate from erosion processes occurring in the northern North Sea by the Atlantic Currents, as our study already indicated by the geochemical characterization of the surface samples. However, most studies about the provenance of the Skagerrak and sediment budget of the North Sea indicated a predominant influence from the southern North Sea in the Skagerrak sediments (Eisma and Irion, 1988; Zöllmer and Irion, 1993; Bengtsson and Stevens, 1998; Lepland et al., 2000). It is noted that most of these studies focused on the mud fraction, which comprises both clay and silt sized sediments. However, the sediment depocenter of the Skagerrak mainly consists of clay deposits up to 60% (Lepland and Stevens, 1996). Bockelmann et al. (2018) predicted the spatial distribution of the median grain size in the North Sea including the clay size and clearly showed that especially the Fladen Ground and the Skagerrak are clay dominated in comparison to other depocenters in the North Sea. This strengthens the assumption that the clays deposited in the Skagerrak predominantly originate from erosion processes in the Fladen Ground with some Atlantic input. Since our study only considers the clay fraction, this would explain the apparently discrepant findings between the older studies and ours. In addition, Oost et al. (2021) suggested that the Wadden Sea region acts as a major sink of sediments transported along the southern pathway of the North Sea, which would explain why large proportions of suspended matter in the southern North Sea do not reach the Skagerrak, which is in a good agreement with our radiogenic isotopic distributions.

The above-mentioned findings emphasize the importance of coastal and seabed erosion for the sediment budget of the entire North Sea region. Puls et al. (1997) and Gerritsen et al. (2000) already mentioned that coastal erosion provides large amounts of suspended matter, which is transported by currents and deposited in deeper parts of the North Sea. However, these studies suggested smaller contributions of coastal erosion than Atlantic supplies. This contrasts with recent studies about the Baltic Sea (Wallmann et al. 2022) that showed that the coastal and seabed erosion is the main sediment contributor in this region surrounded by abundant coastal cliffs. This may indicate that the estimated seabed erosion of 6 -12 Mt per year calculated by van Alphen (1990) as well as Eisma and Irion (1988) for the entire North Sea is an underestimated source for fine-grained sediments and thus also highlighted the crucial role of erosion processes in the sediment budget. Furthermore, in the context of climate change, a rising sea-level enhances erosional processes especially at coastal areas (Bird, 1996; Leatherman et al., 2000) and may lead to an increasing SPM budget and consequently to a higher sediment supply to the depocenters of the North Sea.

In order to gain a better understanding of whether and to what extent these climate forcing factors change the sedimentary processes in the North Sea, a future work is necessary to resolve the temporal variability of the contribution of different endmembers. Additionally, it is still unclear how anthropogenic impacts such as bottom trawling as well as the dumping from dredging activities influence the sediment supply to the depocenters in the North Sea. A temporal resolution of the endmembers may also help to identify and assess these anthropogenic sediment sources.

3.5 Conclusions

The application of the radiogenic isotopic systems of Nd, Sr and Hf to clay-sized sediments in combination with clay mineralogy provides new insights into the sedimentary processes in the North Sea and allows to distinguish between three main sources of clay-sized sediments, which are the northern North Sea, Scandinavian fjords and Mid-European rivers. The isotopic and mineralogical data clearly show that the Skagerrak clay sediments predominantly originate from the northern North Sea. Coastal and seabed erosion, especially in the Fladen Ground, are shown to contribute high amounts of clays to the Skagerrak. In contrast, Scandinavian fjords and central European rivers are only minor contributors of clays. Our study also highlights that it is important to differentiate between different grain sizes of clay and silt when investigating the source of Skagerrak sediments, given that this region represents the main depocenter for

clays. In addition, we demonstrate that suspended matter transported along the southern North Sea mainly originates from coastal erosion in the English Channel and along the south-eastern coast of England. Conversely, the central European rivers only provide negligible amounts to the SPM budget in the southern North Sea. Furthermore, the material originating from coastal erosion in the south-western North Sea is shown to be a minor contributor to Skagerrak sediments, consistent with the hypothesis that large proportions of SPM are deposited in the inner German Bight or trapped in the Wadden Sea region before reaching the Skagerrak.

3.6 Acknowledgements

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3.7 Supplement

3.7.1 Supplementary figures

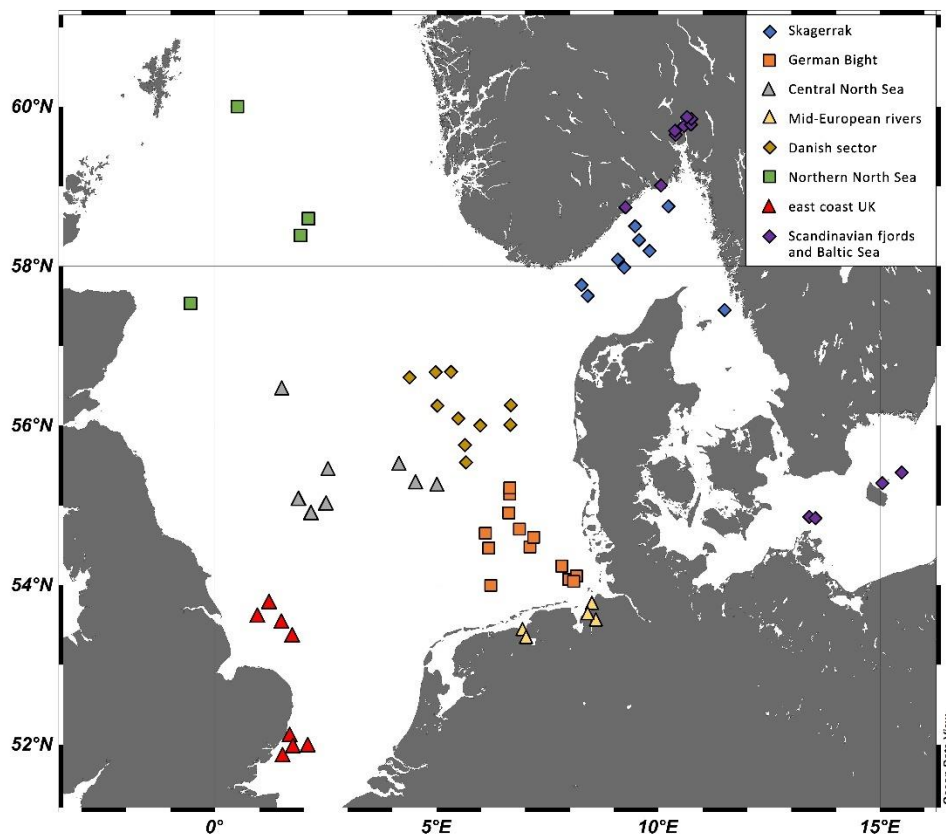


Fig. S3.1: Map of the North Sea with surface sediment samples from defined areas.

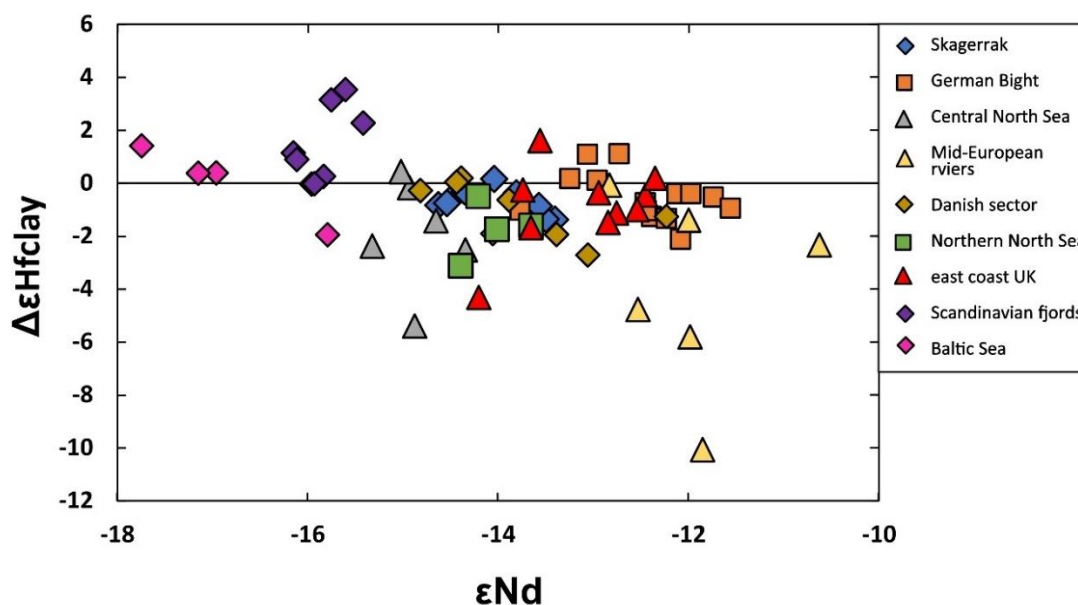


Fig. S3.2: $\Delta\epsilon_{\text{Hfclay}}$ and ϵ_{Nd} signatures, whereby the $\Delta\epsilon_{\text{Hfclay}}$ represents the deviation of Hf isotopic compositions from the Clay Array (Bayon et al. 2015).

3.7.2 Supplementary tables

Tab. S3.1: Radiogenic Sr, Nd, and Hf isotope data from surface sediment samples.

Sample ID	Latitude	Longitude	$^{87}\text{Sr}/^{86}\text{Sr}$	2 σ	ϵNd	2 σ	ϵHf	2 σ
AL561_MUC2	58.18141	9.79359	0.739205	2.15E-05	-13.4	0.39	-7.95	0.58
AL561_MUC7	58.31295	9.57211	0.739924	2.15E-05	-13.5	0.39	-8.02	0.58
AL561_MUC8	57.98803	9.23826	0.741358	2.15E-05	-13.6	0.39	-7.59	0.58
AL557_65	58.49988	9.499967	0.738754	7.27E-05	-13.8	0.39	-7.33	0.58
AL561_MUC6	57.63488	8.39954	0.739411	9.10E-05	-14.5	0.39	-8.24	0.58
AL561_MUC5	57.75328	8.28623	0.739997	9.10E-05	-14.4	0.39	-7.76	0.58
AL561_MUC3	58.74799	10.22404	0.739817	9.10E-05	-14.0	0.39	-7.00	0.58
AL561_MUC1	57.45044	11.50793	0.745643	9.10E-05	-14.6	0.39	-8.53	0.58
AL561_MUC9	58.07261	9.09593	0.742600	9.10E-05	-14.5	0.39	-8.37	0.58
HE400_38B	54.90583	6.63217	0.732047	1.03E-04	-12.7	0.27	-4.84	0.9
HE416_4B	55.2015	6.63983	0.730636	1.03E-04	-12.1	0.27	-7.47	0.9
HE416_44B	54.64267	6.1165	0.730271	1.03E-04	-13.1	0.27	-5.16	0.9
HE436_4B	55.1625	6.63983	0.730141	1.03E-04	-12.5	0.27	-6.43	0.9
HE416_49B	54.23367	7.82583	0.730485	1.03E-04	-12.1	0.27	-5.79	0.9
HE435_58B	53.4109	6.9354	0.728971	1.03E-04	-12.0	0.27	-5.66	0.9
HE447/108	56.0823	5.50035	0.736960	1.03E-04	-13.8	0.27	-7.92	0.9
HE416_11B	54.58733	7.20167	0.730496	7.27E-05	-12.4	0.27	-6.90	0.9
MSM99/2_25-1	54.11281	8.12506	0.731001	7.27E-05	-12.2	0.27	-6.83	0.9
MSM99/1_51-1	54.08345	7.9673	0.730976	7.27E-05	-11.7	0.27	-5.57	0.9
MSM99/2	54.05151	8.0101	0.732244	7.27E-05	-11.6	0.27	-5.82	0.9
HE400_109B	54.70717	6.87917	0.731106	7.27E-05	-13.0	0.27	-6.06	0.9
AL557_15	53.98953	6.229433	0.732327	7.27E-05	-13.3	0.27	-6.25	0.9
AL557_3-2	54.06108	8.0153	0.730202	7.27E-05	-12.4	0.27	-6.73	0.9
HE468_19	55.27	4.53	0.742371	7.27E-05	-14.3	0.27	-9.96	0.9
HE468_23	55.25	5	0.737419	7.27E-05	-14.9	0.27	-13.32	0.9
AL557_47	55.50282	4.164817	0.736144	7.27E-05	-14.7	0.27	-9.15	0.9
JC_145VC	55.05938	1.88805	0.741945	9.10E-05	-14.9	0.3	-8.15	0.9
AL557_43	55.00003	2.500633	0.754558	9.10E-05	-15.3	0.3	-10.72	0.9
JC123_144C	54.88359	2.17869	0.743157	9.10E-05	-15.0	0.3	-7.63	0.9
TS_277	53.3202	7.021	0.730638	9.10E-05	-12.5	0.3	-10.54	0.9
TS_276	53.7415	8.5084	0.730492	9.10E-05	-12.0	0.3	-6.68	0.9
SS5.1_U5	54.4663	6.194	0.731718	9.10E-05	-12.8	0.3	-6.13	0.9
BH4-3_U5	53.5391	8.576	0.726905	9.10E-05	-11.9	0.3	-15.22	0.9

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BH2.1_U5	53.5391	8.576	0.728275	9.10E-05	-12.0	0.3	-11.08	0.9
MSM99/2_25-2	54.11283	8.12494	0.727139	9.10E-05	-10.6	0.3	-6.37	0.9
HE532_20	54.47567	7.14183	0.729797	9.10E-05	-12.2	0.3	-6.76	0.9
HE497_9	55.53182	5.664664	0.736416	9.10E-05	-14.4	0.3	-7.28	0.9
HE507_10	56.2501	6.6593	0.734072	9.10E-05	-13.4	0.3	-8.49	0.9
HE497_15	55.74894	5.65707	0.735731	9.10E-05	-14.1	0.3	-9.08	0.9
HE507_5	55.99975	6.001083	0.733951	9.10E-05	-13.9	0.3	-7.65	0.9
HE507_16	56.24978	4.999183	0.741138	9.10E-05	-14.8	0.3	-8.15	0.9
HE507_18	56.66263	5.326633	0.738694	9.10E-05	-14.4	0.3	-7.48	0.9
HE507_9	56.00013	6.659633	0.730489	9.10E-05	-13.1	0.3	-8.98	0.9
AL557_38	59.9999	0.50095	0.737073	9.10E-05	-14.4	0.3	-10.60	0.9
JC123_108VC	57.52952	-0.55072	0.737267	9.10E-05	-14.2	0.3	-7.80	0.9
JC077_VC01	58.59472	2.08823	0.738746	9.10E-05	-13.7	0.3	-8.41	0.9
JC077_VC-20	58.38378	1.95385	0.744952	9.10E-05	-14.0	0.3	-8.88	0.9
JC123_127VC	56.45793	1.47912	0.739905	9.10E-05	-14.2	0.3	-11.63	0.9
JC123_165VC	53.59911	0.95253	0.729491	9.10E-05	-13.6	0.27	-5.11	0.9
THA_VC2	51.95816	1.7535	0.729582	9.10E-05	-13.7	0.27	-7.14	0.9
JC123_153VC	55.3949	3.30848	0.738521	9.10E-05	-13.7	0.27	-8.49	0.9
JC123_170VC	53.76385	1.22485	0.728946	9.10E-05	-12.9	0.27	-6.52	0.9
JC123_169VC	53.51873	1.49698	0.728865	9.10E-05	-12.5	0.27	-6.15	0.9
JC123_168VC	53.34442	1.74007	0.728946	9.10E-05	-12.4	0.27	-5.42	0.9
THA_VC15	51.83883	1.52081	0.726946	9.10E-05	-12.5	0.27	-6.78	0.9
THAMES_VC18	52.08853	1.68717	0.727632	9.10E-05	-12.8	0.27	-7.11	0.9
THA_VC28	51.95803	2.08857	0.727447	9.10E-05	-12.8	0.27	-7.55	0.9
M86-1/18-2	54.852	13.43283	0.760584	9.10E-05	-15.8	0.27	-10.72	0.9
M86-1/25-2	55.41117	15.46817	0.775977	9.10E-05	-17.0	0.27	-9.45	0.9
M86-1/19-5	54.838	13.53367	0.758642	9.10E-05	-17.7	0.27	-9.14	0.9
M86-1/22-2	55.28213	15.0469	0.769846	9.10E-05	-17.2	0.27	-9.64	0.9
A	59.68793	10.37124	0.757456	9.10E-05	-16.2	0.27	-7.95	0.9
C	59.7511	10.57382	0.756552	9.10E-05	-16.0	0.27	-8.95	0.9
D	59.78977	10.71923	0.754146	9.10E-05	-15.8	0.27	-8.54	0.9
F	59.85783	10.69517	0.754156	9.10E-05	-16.1	0.27	-8.17	0.9
B	59.6477	10.4134	0.760231	9.10E-05	-15.9	0.27	-8.92	0.9
E	59.84405	10.72595	0.757793	9.10E-05	-15.8	0.27	-5.58	0.9
Larvik	59.0075	10.07122	0.748944	9.10E-05	-15.4	0.27	-6.15	0.9
Oster	58.7409	9.254733	0.751209	9.10E-05	-15.6	0.27	-5.06	0.9
HE606_13	56.66725	5.0016	0.739502	9.10E-05	-14.6	0.27	-13.68	0.9

JC123_115VC 56.61597 4.39717 0.736842 9.10E-05 -13.7 0.27 -8.95 0.9

Tab. S3.2: Clay mineral compositions of surface sediment samples

Sample ID	Chlorite [%]	Smectite [%]	Kaolinite [%]	Illite [%]
HE436-4B	12.02	14.45	13.85	59.69
HE416-11B	24.62	16.98	10.18	48.22
HE416-44B	17.93	17.62	10.09	54.36
HE400_38B	18.32	16.98	16.44	48.26
HE416-49B	16.42	16.46	12.68	54.36
HE447-108	12.34	14.15	14.65	58.86
HE416-4B	21.55	14.67	11.05	52.74
HE435-58B	17.81	15.22	9.1	57.88
AL557_St_3-2	17.03	22.46	14.88	45.63
MSM99_2_24_2	15.65	9.77	15.65	58.93
MSM99_2_25_2	18.33	17.92	14.45	48.3
MSM99_2_51-1	16.97	11.95	15.53	55.54
TS-276	11.22	32.27	7.21	49.3
SS5_1-45	16.67	24.14	9.23	49.96
BH4_3-U5	16.04	24.66	10.42	48.88
BH2.1-U5	24.61	21.8	14.78	38.81
JC077_VC20	23.58	6.6	18	51.82
AL557_St 38	34.56	4.38	13.31	47.75
108VC	23.57	6.31	14.91	55.21
AL557 St 43	3.66	18.16	21.25	56.93
127VC	5.8	6.2	20.14	67.86
165VC	31.52	3	22.12	43.35
AL557 St 65	16.32	8.37	17.83	57.48
MUC7_1	18.05	7.55	15.09	59.3
MUC2_1	20.54	8.03	15.82	55.61
MUC8_1	18.14	4.48	18.87	58.51
HE532-20	22.96	7.98	15.57	53.49
HE507-10	18.59	9.66	19.45	52.29
HE497-9	10	30.31	11.78	47.91
B	38.18	4.46	7.79	49.57
oster fjord	13.56	12.78	15.1	58.56
M86-1_18-2	29.16	7.83	10.28	52.73

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4. Scientific Chapter II

Variability of Skagerrak sediment provenance over the past 150 years

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Abstract

The Skagerrak basin acts as a major sink for fine-grained sediments of the entire North Sea. Skagerrak sediments therefore provide valuable insights into the sedimentary dynamics of the North Sea, which is subjected to extensive human-induced disturbances since the onset of industrialisation and exposed to recent climate change. However, there is little knowledge on the impact of these changes on the sediment dynamics in the North Sea in the most recent past. To reconstruct temporal variations in sediment supply from different source areas, time series of radiogenic Nd, Sr and Hf isotope as well as clay mineral compositions of detrital clay-sized sediments from Skagerrak sediment cores were investigated. The results reveal more radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ signatures and lower radiogenic ϵNd and ϵHf values since the end of the 20th century indicating decreased sediment input from the southern North Sea. This was likely caused by enhanced sediment trapping efficiency of the Wadden Sea promoted by increasing storminess and the ongoing sea-level rise. In addition, the radiogenic isotope signatures also suggest an increase in sediment supply from the northern North Sea as a result of intensified bottom trawling in the 1980s and 1990s. Conversely, the deepening of Mid-European rivers has apparently not affected the sediment contributions to the Skagerrak. A coarsening upward trend in the sediment cores suggests more energetic hydrodynamic conditions since around the 1960s in the Skagerrak region, likely due to increased storminess during multi-decadal periods. This

may imply that the clay and fine silt fractions have been hampered from settling in the inner part of the Skagerrak basin, which resulted in decreasing sediment deposition rates in the second part of the last century. These findings emphasize the direct influence of climate change on the sedimentation in the Skagerrak and clarify that these driving factors will likely sustain imbalances of the North Sea sedimentary system in the future.

4.1 Introduction

The North Sea is a continental shelf sea where strong tidal currents and intensive wave activity prevail. Both enhance the redistribution of suspended particulate matter (SPM), especially in the shallow regions, while at the same time sediment accumulation takes place in few sedimentary depocenters.

Since the onset of industrialisation, sediment dynamics in the North Sea have increasingly been impacted by human activity (Halpern et al., 2008). Bottom trawling, for example, penetrates the seafloor up to 35 cm depth (Eigaard et al., 2017) and causes resuspension of large amounts of fine-grained material in the entire North Sea. About 63 % of North Sea sediments are affected by trawling (Rijnsdorp et al., 1998; Pitcher et al., 2000; Bellman et al., 2005; Murawski et al., 2005; Eigaard et al., 2017); mainly in the sediment depositional areas, i.e. the Skagerrak, the Norwegian Trench and the Fladen Ground (ICES, 2019). This emphasizes the large-scale impact on both sediment dynamics and carbon cycling and results in a decreased fixation of particulate organic carbon in the sediments and consequently contribute to an enhanced release of CO₂ to the atmosphere (De Borger et al., 2020; Bradshaw et al., 2021; Atwood et al., 2024, Zhang et al., 2024). In addition, the construction of dams and dredging activities along larger European rivers such as Elbe, Weser, Ems and Rhine led to substantially reduced riverine inputs into the North Sea (Puls et al., 1997; Syvitski et al., 2005; Ericson et al., 2006; Goelz, 2008; Rovira et al., 2014), dumping of large amounts of fine-grained sediments in different areas of the North Sea (OSPAR, 2017) or in deposition areas on land (Netzband et al., 2002). These anthropogenic activities can affect the sediment balance but the dimensions of these impacts and their drivers are still not fully understood. By generating this knowledge, concrete protection measures can be developed to preserve regions in the North Sea that are effective areas for long-term carbon storage. Furthermore, the North Sea ecosystem and its sediment dynamics are also affected by climate change, which is accompanied by long-term variability in storminess and wave activity (Fettweis et al., 2010).

A number of sedimentological studies concluded that sediment dynamics of the North Sea have undergone changes over the last century (Dellwig et al., 2000; Cappuzzo et al., 2015; Emeis et al., 2015; Benninghof and Winter, 2019; Wilson and Heath, 2019; Oost et al., 2021). Cappuzzo et al. (2015) as well as Wilson and Heath (2019) discovered that the concentration of SPM in the water column of the North Sea generally increased by over 20%, in particular in its southern part, during the second half of the last century due to a combination of increased wave activity, amplified tidal range and bottom trawling activities which enhanced resuspension of seabed sediments.

Sedimentary depocenters of the North Sea provide a high-resolution record for the reconstruction of the sedimentary history. One of these, the Skagerrak basin, represents a natural sediment trap and forms the largest depocenter of fine-grained sediments derived from the northern and southern North Sea, as well as from Scandinavia and the Baltic Sea, suggesting that significant variability in sediment provenance is to be expected here. Between 45 – 80% of the suspended particulate matter that is transported across the North Sea ends up in the Skagerrak (Oost et al. 2021; Spiegel et al., 2024). However, the mass accumulation rates decreased during the second half of the last century, the driving factors of which are not clear (Spiegel et al., in review). A detailed look into further down-core records of Skagerrak sediments is necessary to assess temporal changes and their reasons further back in time. The reconstruction of changes in the provenance of the sediments that are supplied to the Skagerrak helps to identify multi-decadal variations in the current system of the North Sea or changes within the various areas of the predominant sediment suppliers.

Radiogenic isotope signatures of clay-size sediments in combination with clay mineral compositions are established tools for characterizing sediment sources and reconstructing sediment transport pathways (Kessarkar et al., 2003; Ehlert et al., 2011; Bretschneider et al., 2021a; Lenz et al., 2024). Radiogenic Nd, Sr and Hf signatures of detrital material are predominantly controlled by the age and type of the rocks in the hinterland, and enable tracing of sediment sources (Dasch, 1969; Goldstein and Jacobsen, 1987; Grousset et al., 1988; Revel et al., 1996; Colin et al., 1999). The isotopic composition of a rock depends on the fractionation between the parent and daughter isotope and the timing of melt extraction. When rocks undergo weathering processes, sediment particles with certain radiogenic isotope compositions are released. Consequently, these isotopic compositions serve as valuable tools for reconstructing the source of the eroded material.

In addition, composition and abundance of clay minerals is controlled by the geology of their source areas and environmental conditions during erosion and transport (He et al., 2013). They therefore represent ideal complements of sediment provenance studies based on radiogenic isotopes.

Lenz et al. (2024) established the present-day provenance of clay-sized sediments deposited in the North Sea and the Skagerrak, which provides the basis for investigating the temporal variability of the main sediment contributors. This study showed that the northern North Sea, Scandinavia, and the southern North Sea are the three main sources for clay-sized sediments deposited in the Skagerrak, whereby the northern North Sea is the predominant source area of sediments derived from coastal as well as seabed erosion. Conversely, Mid-European rivers are only minor contributors of fine-grained sediments to the southern North Sea and consequently to the Skagerrak. The temporal variability of the contribution of the different source area, however, remains unknown so far.

This study presents the first time series of clay-size radiogenic Nd, Sr and Hf isotope signatures and clay mineral compositions originating from the North Sea region that have been delivered to the Skagerrak basin during industrialization. This approach allows the investigation whether and to what extent the sediment sources for the Skagerrak have changed during that period of time to assess which driving factors were mainly controlling the North Sea sedimentation.

4.2 Study site – Oceanographic setting and circulation pattern

The Skagerrak is a semi-enclosed basin located in the northeastern part of the North Sea with a maximum water depth of 700 m. It connects the North Sea with the Kattegat as part of the Baltic Sea and is bordered by Norway, Sweden and the Jutland peninsula of Denmark (Fig. 4.1). The North Sea is a shallow (average depth 90 m) marginal sea of the Atlantic Ocean with a circulation pattern primarily forced by the North Atlantic Current (Svansson, 1975; Rodhe, 1987; Otto et al., 1990). Atlantic water masses enter the North Sea from the south-west through the English Channel and from the north between Scotland and Norway resulting in an anti-clockwise circulation (Fig. 4.1A). Parts of the Atlantic water masses originating from the northwestern entrance flow along the British coast via the Southern Trench Current, carry on eastwards through the central North Sea, and continue further into the Skagerrak (Rohde, 1998). By contrast, the Atlantic inflow west of Norway flows directly into the Skagerrak via the Tampen Bank Current (Rohde, 1998). In the southern North Sea, the South Jutland Current

from the English Channel flows along the Danish west coast and continues to the Skagerrak (Fig. 4.1A). As a result, terrigenous mud carried by the Jutland Current merges with material carried by Atlantic water masses from the northern entrance. When these water masses reach the inner part of the Skagerrak, they mix with fresher and colder waters of the Baltic Current (BC) originating from the Kattegat between Sweden and Denmark further south (Fig. 4.1B). This leads to an anti-clockwise current in the Skagerrak, which exits the basin as the Norwegian Coastal Current (NCC). These water masses reach depths of up to 500 m within the basin with essentially stagnant water masses underneath (Rohde, 1996). With increasing water depth and decreasing flow velocities, fine grained-sediments can settle and is deposited in the central and northeastern part of the Skagerrak (Van Weering, 1982; Rodhe and Holt, 1996; Bøe et al., 1998).

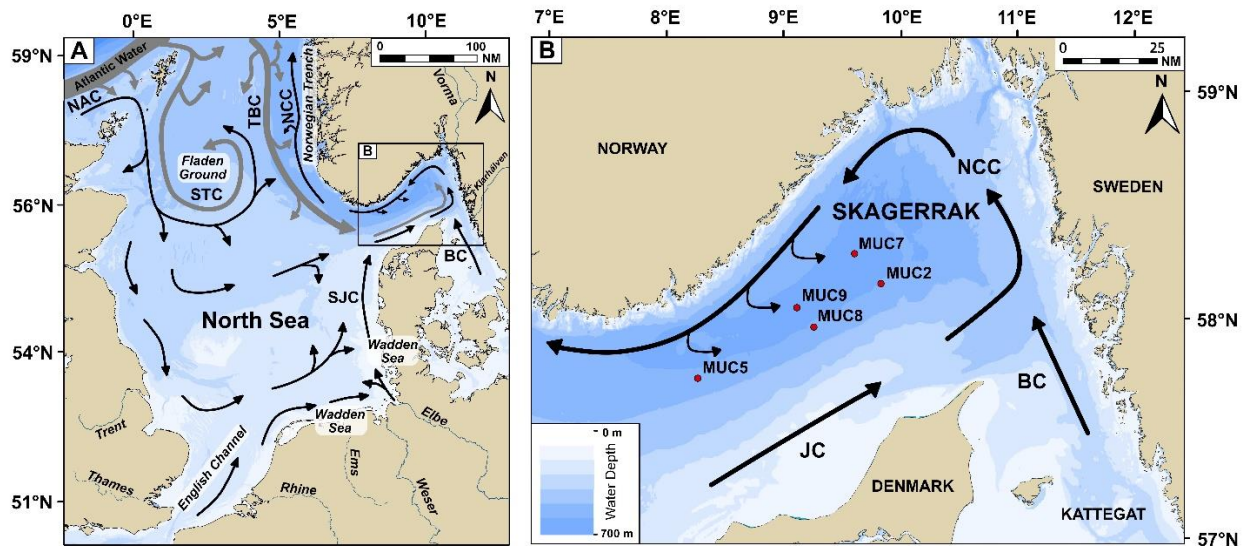


Fig. 4.1: (A) Water depth and circulation pattern of the North Sea with major river systems modified after Turrell (1992) including the main currents: NAC = North Atlantic Current, STC = Southern Trench Current, TBC = Tampen Bank Current, NCC = Norwegian Continental Current, NJC = North Jutland Current, SJC = South Jutland Current, and BC = Baltic Current. (B) Skagerrak basin with water depth and circulation pattern modified after Longva and Thorsnes (1997) including main currents: JC = Jutland Current, BC = Baltic Current, and NCC = Norwegian Coastal Current. Red dots indicate core locations.

The Skagerrak basin forms the largest sink for fine-grained sediments in the North Sea with sedimentation rates reaching 1 cm yr^{-1} in its central part (Van Weering, 1982; Bøe et al., 1998) and mass accumulation rates of $28 - 46 \text{ Mt yr}^{-1}$ (Van Weering, 1987; De Haas and Van Weering, 1997; Spiegel et al., 2024). Skagerrak sediments are characterized by high amounts of silt and

clay with only small amounts of sand near the Scandinavian coastline (Pederstad et al., 1993; Stevens et al., 1996; Spiegel et al., 2023).

4.3 Methods

4.3.1 Sample preparation

This study is based on down-core sediment samples of five multi-cores (MUC) which were collected during the AL561 cruise in the Skagerrak in August 2021 (Schmidt et al., 2021; Fig. 4.1B). Cores originate from the central part of the Skagerrak from water depths between 434 and 677 m and recovered undisturbed sediment successions (Fig. 4.1, Tab. 4.1). The cores recovered sediment down to depths of 30 - 45 cm below the seafloor. The clay-sized detrital sediments were analysed for radiogenic Sr, Nd, and Hf isotope compositions as well as for clay mineral compositions of the sediments from cores MUC2, MUC7 and MUC8.

Station	Latitude N	Longitude E	Length [cm]	Water depth [m]	MAR before 1963 [g cm ⁻² yr ⁻¹]	MAR after 1963 [g cm ⁻² yr ⁻¹]
MUC2	58° 10.884'	09° 47.624'	41	500	0.22	0.1
MUC5	57° 45.191'	08° 17.173'	45	434	0.18	0.08
MUC7	58° 18.785'	09° 34.335'	44	677	0.11	0.08
MUC8	57° 59.286'	09° 14.305'	40	490	0.3	0.14
MUC9	58° 04.352'	09° 05.736'	37	604	0.12	0.06

Tab. 4.1: Cores used for this study. Mass-accumulation rates (MAR) are from Spiegel et al. (in review)

4.3.2 Radiogenic isotopes

Sediment samples were sieved through a 63 µm sieve. To remove the authigenic Fe-Mn oxyhydroxide fraction, the fine-fraction collected was leached with a 0.05 M hydroxylamine hydrochloride (HH) – 15% acetic acid – 0.03 M Na-EDTA leaching solution following the procedure described by Gutjahr et al. (2007). The remaining sediment was rinsed with deionized water to remove the soluble fraction, was fully decarbonated with 40% acetic acid and then treated with H₂O₂ to destroy organic matter. Afterwards the clay-size sediment (< 2 µm) was separated using a centrifuge-based Atterberg method. The clay sediments were divided into two aliquots for radiogenic isotope and clay mineral analyses. For radiogenic isotope analyses, the samples were again leached with the HH leaching solution buffered to pH 4 to

remove the remaining Fe-Mn oxyhydroxides. After washing the samples three times with deionized water, the samples were dried at low temperatures (40°C) in an oven. 100 mg of the clays were fully digested using the alkaline fusion method modified of Bayon et al. (2009). For separation and purification of Sr, Nd, and Hf, standard ion chromatographic procedures were applied (Cohen et al., 1988; Galer and O'Nions, 1989; Pin and Zalduegui, 1997).

For Sr separation from the matrix elements 50 µl Eichrom Sr-Spec resin were used (50-100 µm mesh; Horwitz et al., 1992) and for separation of Nd and Hf from the other rare earth elements (REE) and other matrix elements a cation exchange resin was used (Bio-Rad AG 50W-X8, 200-400 µm mesh). Then Nd was separated from the other REEs using 2 ml Eichrom Ln-Spec resin (50-100 µm mesh; Pin and Zalduegui, 1997). Hf was separated using 1.6 ml of Bio-Rad AG1-X8 resin (200-400 µm mesh; Sahoo et al., 2006).

The radiogenic isotope measurements were performed on a Nu Plasma high-resolution multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at GEOMAR Helmholtz Centre for Ocean Research Kiel in Germany.

Sr isotope ratios were corrected for instrumental mass bias by using $^{88}\text{Sr}/^{86}\text{Sr}$ of 0.1194 (Steiger and Jäger, 1977) and normalized to the NIST SRM 987 $^{87}\text{Sr}/^{86}\text{Sr}$ value 0.710245. Procedural blanks contributed between 1.33 and 13.25 % of the total Sr signal. This high blank contribution originates from using sodium hydroxide (NaOH) and sodium peroxide (NaO₂) for the alkaline fusion. To calculate the blank contribution, the Sr concentration and isotopic composition of the blanks were measured. This blank contribution was then deducted from the total Sr isotope signal of the sample by mass balance calculation as described by Bretschneider et al. (2021a, b). The digested and repeatedly measured USGS rock reference material STM-1 (n = 3) gave a mean $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.703835 (+/- 0.00016), which is in good agreement with the average value of 0.703701 (+/- 0.000006) of the GeoReM database (Weis et al., 2006).

Nd isotope ratios were corrected for instrumental mass bias using a $^{146}\text{Nd}/^{144}\text{Nd}$ of 0.7219 (Vance and Thirlwall, 2002) and normalized to the JNdi-1 standard value 0.512115 (Tanaka et al., 2000). The Nd isotope ratios are reported as ϵ_{Nd} values calculated by using the following equation (Jacobsen and Wasserburg, 1980):

$$\epsilon_{\text{Nd}} = \frac{\left(\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ sample}\right) - \left(\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ CHUR}\right)}{\frac{^{143}\text{Nd}}{^{144}\text{Nd}} \text{ CHUR}} * 10000$$

where CHUR is the present day chondritic $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.512638. The blank contribution of Nd was below 0.29 % of the total amount of Nd in the samples. Measurements of the USGS rock reference material STM-1 ($n = 3$) revealed a mean ϵNd value of 5.49 (± 0.56), which agrees well with the mean value of 5.36 (± 0.18) provided by Weis et al. (2006).

Hf isotope ratios were corrected for mass bias using a $^{179}\text{Hf}/^{177}\text{Hf}$ of 0.7325 and then normalized to a JMC 475 value of 0.282163 (Blichert-Toft and Albarède, 1997; Blichert-Toft, 2008). The Hf isotope ratios are reported as ϵHf values similar to the ϵNd values using a present day chondritic $^{176}\text{Hf}/^{177}\text{Hf}$ value of 0.282785 (Bouvier et al., 2008). Blank contributions for Hf were below 2.9 % of the total amount of Hf in the samples. Measurements of the USGS rock reference material STM-1 ($n = 3$) revealed a mean ϵHf value of 8.44 (± 0.68), which agrees well with the mean value of 8.36 (± 0.79) given in the GeoReM database (Jochum et al., 2005).

4.3.3 Clay minerals

The leached clay fraction obtained by Atterberg-separation were used for clay mineral analyses. To remove pedogenic iron oxides from the clay fraction, 0.3 M sodium citrate, 0.2 M sodium hydrocarbonate (NaHCO_3) and sodium dithionite were used and the dissolved iron compounds were subsequently washed out four times with 2M sodium chloride. The clay suspension was then saturated and flocculated for 1/3 by an excess of K^+ -ions with KCl and 2/3 by an excess of Mg^{++} -ions with MgCl. The ion excess which is not bound to clay mineral surfaces was removed with dialysis tubes filled with deionised water to electrical conductivity values below 20 μS . Afterwards, the saturated clay samples were freeze-dried and 50 mg of the material was dispersed in 4 ml deionised water in an ultrasonic bath for 10 min. 2 ml of the clay suspension was placed on a ceramic slide and dried for 48 hours. These oriented clay mineral samples were measured with x-ray diffractometry (XRD) using a D8 Discover (Bruker AXS) at the Institute of Geosciences of the University of Kiel (CAU). The samples were measured with a Cu-cube as well as θ - θ -geometry of 2θ at 0.037° steps with a scanning speed of 1.5 s at each step. The measurement of the oriented Mg-saturated sample slide was repeated after the clays were treated with ethylene glycol saturation and the K-saturated sample was heated to 550 $^\circ\text{C}$ for two hours.

The XRD analyses of the diffractograms were performed via the open-source software Profex using the BMBG and COD database for identifying the different mineral phases. The main clay mineral phases illite, kaolinite, chlorite and smectite were differentiated. The semi-quantitative

clay mineral analyses were performed by determining the peak areas of the respective mineral phases and were calculated following Whitton and Churchman (1987). Based on Moore and Reynolds (1989) the standard deviations for these clay mineral groups amounts to +/- 5%.

4.3.4 Grain-size analysis

Sediment cores were sampled equidistantly (1.5 cm³ every 1 cm) for determination of grain-size distributions. Samples were treated with 10 % to 30 % hydrogen peroxide to dissolve organic matter and then decarbonated with 60 % acetic acid. Afterwards, samples were suspended in water with tetra-sodium diphosphate decahydrate as dispersing agent. Grain-size measurements were conducted at the Institute for Geology, University of Hamburg, with a laser-diffraction particle-sizer (Sympatec HELOS/KF Magic; range 0.5/18 to 3200 3500 µm). An in-house grain-size standard was measured regularly to ensure accuracy of measurements (standard deviation for mean grain size and D50 over the analysis period was < 1.1 µm). Statistical analysis of grain-size data is based on graphical method (Folk and Ward, 1957), calculated using Gradistat (Blott and Pye, 2001).

4.3.5 Age-depth model

The depth distribution of the radiogenic isotopes, clay minerals and grain sizes were converted to temporal distributions using age-depth models that have been applied to the same sediment cores in a previous study (Spiegel et al., in review). In brief, age-depth relationships were determined based on the known decay rate of lead-210 (²¹⁰Pb) and a time marker triplet consisting of caesium-137 (¹³⁷Cs), fraction modern carbon-14 (¹⁴C) and mercury (Hg). The sediment cores were virtually split into two sections separated by the depth of the time marker maxima representing the year 1963. Age-depth relationships in the upper section were determined using a reaction transport model that simulated sediment burial, mixing, compaction and ²¹⁰Pb decay. In the sediment section below the time marker peak, the constant flux and constant sedimentation (CFCS) model (Sanchez-Cabeza and Ruiz-Fernández, 2012) was applied to ²¹⁰Pb data to derive age-depth relationships.

4.4 Results

4.4.1 Radiogenic isotopes and clay minerals

A summary of the $^{87}\text{Sr}/^{86}\text{Sr}$, ϵNd and ϵHf signatures of the clay-sized detrital down-core samples with the respective ages is shown in Figure 4.2 and also reported in the supplementary Table S4.1. Based on ^{210}Pb data of the same cores, the bioturbation depth ranges between the upper 5-15 cm in the respective cores (Spiegel et al., in review). The bioturbation depths in the upper sediment sections were assigned to the time interval younger than approximately 1980 in all cores (Fig. 4.2). As within the bioturbation depth the resolution of the radiogenic isotope record influences the accuracy, variations observed during this period represent integrated changes after 1980.

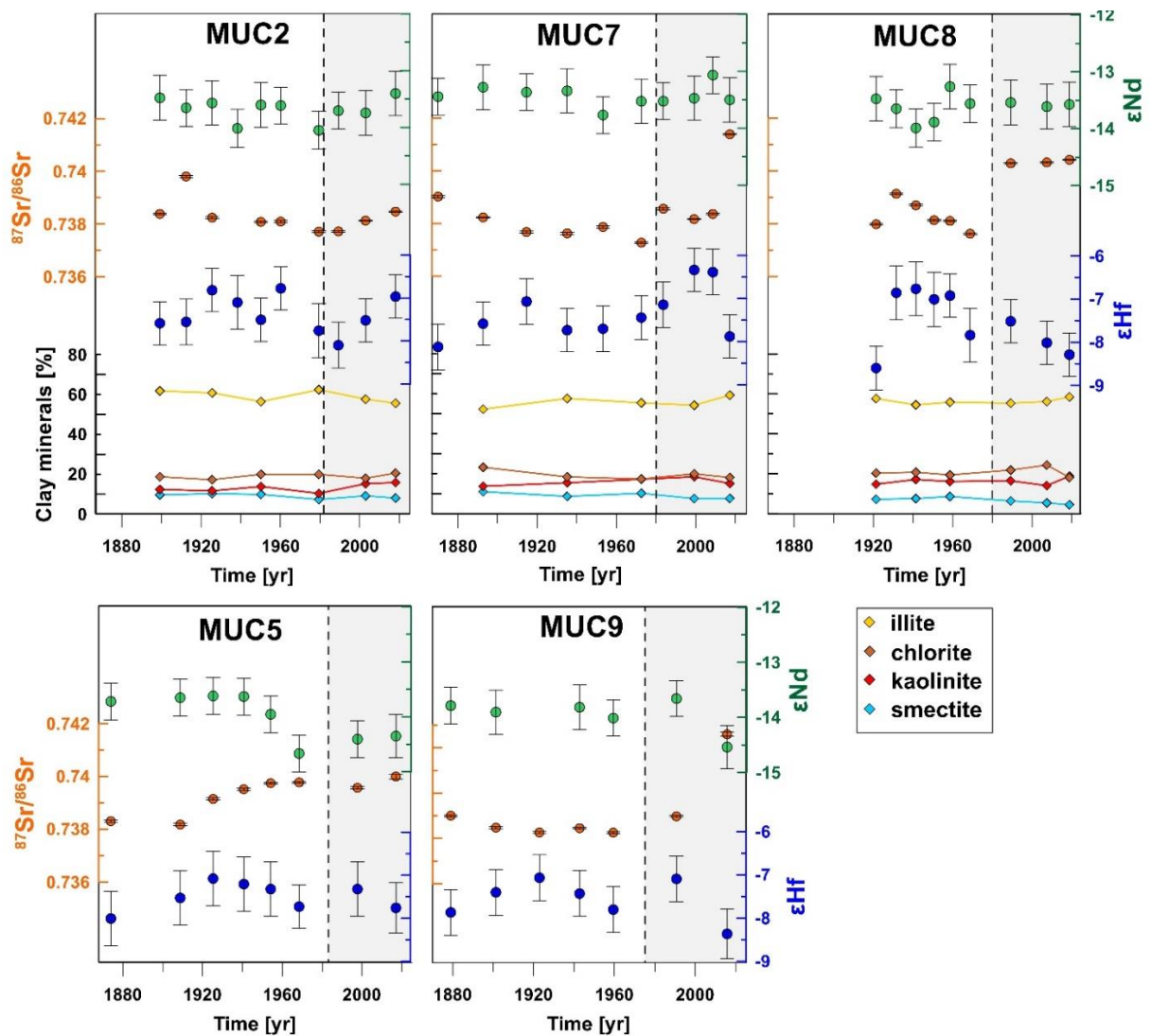


Fig. 4.2: Down-core radiogenic Sr, Nd, and Hf isotope compositions and clay mineral compositions. The dashed line and the light grey shaded area mark the bioturbation depth.

The ϵNd signatures range from -14.7 to -13.1 and within the standard deviations (2SD) the values represent constant ϵNd values over time in each core. The only exceptions are the slightly lower ϵNd signatures since around the 1970s in core MUC5 and the lower surface value in core MUC9 (Fig. 4.1B). In contrast to ϵNd , $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are more sensitive to variations in the isotopic composition due to the large range of the signatures and comparatively small analytical uncertainties which results in significant variability in all cores over time. The radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios vary between 0.737274 and 0.742600 and all MUC cores generally show a trend to higher, more radiogenic Sr isotope ratios at the end of the 20th century starting approximately in the 1980s. The ϵHf signatures of the sediment cores vary between -8.6 and -6.3 and overall show only minor variability outside analytical uncertainty. MUC8 reveals the highest sedimentation rates (Tab. 4.1) and shows a decreasing trend of ϵHf values since the 1980s. A slight declining trend can also be observed in MUC7 and MUC9 in the time interval after 2000. Overall, the radiogenic isotope signatures in all cores remained stable until approximately 1980. The only exceptions are slightly higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios around the 1920s observed in MUC2 and MUC8 which are accompanied by lower ϵHf signatures in MUC8.

The clay mineral compositions of the core samples are shown in Figure 4.2 and reported in the supplementary Table S4.2. The main components include illite, chlorite, kaolinite, and smectite with average contents of 56.9 %, 19.8 %, 15.3 %, and 8.0 %, respectively. Overall, the proportions of the four clay mineral phases in the down-core samples of MUC2, MUC7, and MUC8 remained constant within error ranges over time and were essentially the same at all coring sites located in the inner part of the Skagerrak.

4.4.2 Grain-size data

MUC cores reveal a succession of sediments in the silt to clay size range with an average mud content (particles < 63 μm) of 92.8 to 99.3 % (Fig. S4.1). Only MUC2 and MUC8 show larger portions of sediment in the sand size (up to 9.6 %; Fig. S4.1), with an increase of the very fine sand fraction (63 – 125 μm) since the mid-20th century (Fig. 4.3). The amount of fine silt and clay ($\leq 15.6 \mu\text{m}$) declines in the same time period. By contrast to this, the proportion of the coarse silt fraction (31 – 63 μm) increases simultaneously in MUC2, MUC7, and MUC9, whereas it decreases in MUC8 since around the 1970s.

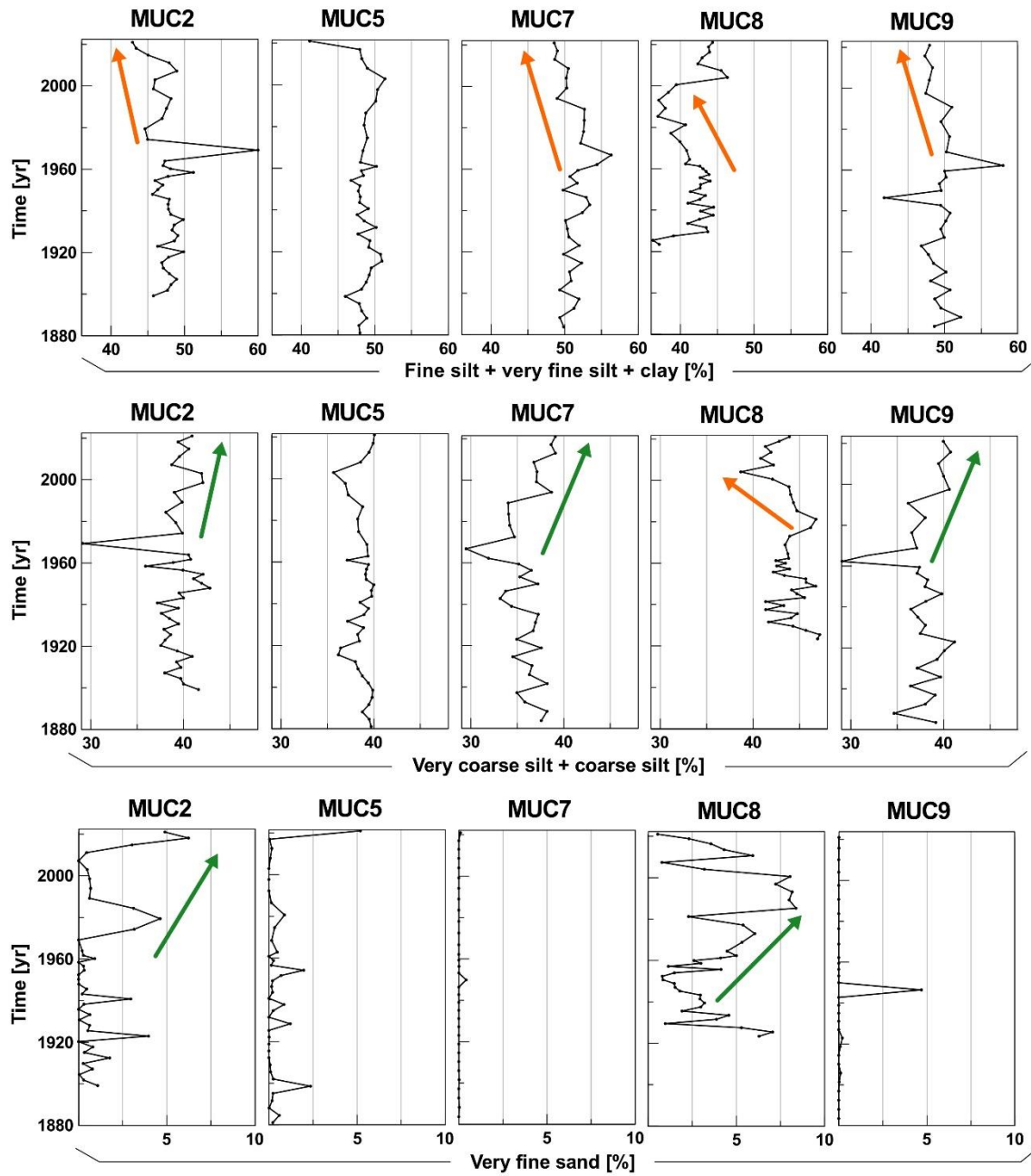


Fig. 4.3: Grain-size data of the MUCs shown as the percentage of fine silt fraction ($\leq 15.6 \mu\text{m}$), coarse silt fraction ($31 - 63 \mu\text{m}$), and very fine sand fraction ($63 - 125 \mu\text{m}$). The green arrows indicate an increase and the orange arrows a decrease of the respective sediment fraction.

4.5 Discussion

4.5.1 Evolution of the provenance of the Skagerrak sediments over the last century

Despite the large range of isotopic signatures of the source rocks of the sediments in the study area (Fig. 4) the down-core radiogenic Nd and Hf isotope data of the Skagerrak sediments (Fig. 4.2) remained largely stable throughout the 20th century, documenting the absence of large shifts in sediment supply of the main sources. This finding is consistent with the evolution of the clay mineral compositions, which reveal no significant variability over the same time period. The radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ signatures, however, which are more sensitive for variations in source provenance compared to ϵNd and ϵHf , reveal a trend to higher radiogenic values since the end of the 20th century (Fig. 4.2). This trend is also reflected by some less radiogenic ϵNd values in MUC5 and less radiogenic ϵHf values in MUC8 during the same time interval, which is consistent with the general radiogenic isotope geochemical signatures of rocks. The shift in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios imply a shift in the relative sediment proportions received from the different sources (Fig. 4). Since the Skagerrak represent the largest sedimentary sink of the entire North Sea system, this observation suggests that the sedimentary dynamics remained stable throughout the past century until approximately the 1980s and then experienced changes in the sediment provenance afterwards.

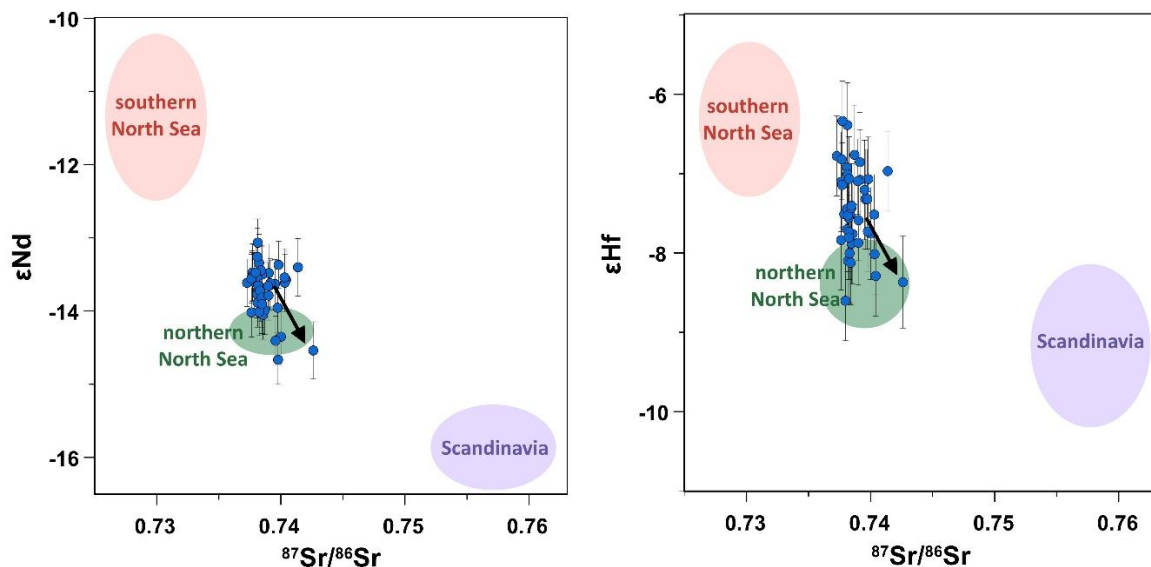


Fig. 4.4: Radiogenic Nd-Sr and Hf-Sr isotope compositions of Skagerrak sediments (this study) marked by the blue dots with defined end members of the North Sea (Lenz et al., 2024). The 2SD error bars for Sr isotopes are smaller than the symbol size. Black arrows indicate shifts of radiogenic isotope signatures since around the 1980s.

Transport and deposition of fine-grained sediments in the entire North Sea are mainly controlled by the circulation pattern (Fig. 4.1A). This also applies to the Skagerrak basin which is predominantly supplied by the Atlantic inflow coming from the northern North Sea where erosional processes lead to an enhanced remobilization of fine-grained material (Lenz et al., 2024). Given that the circulation of the North Sea persisted since around 600-700 AD (Hass, 1994), it can be assumed that the principal sediment transport pathways were stable at least through the last Millennium. In fact, the rather small range in the radiogenic isotope fluctuations imply no fundamental changes in the source contributions of the Skagerrak clays. During the last century, however, climatic and anthropogenic factors have obviously altered the sedimentary processes in the North Sea; most obvious since around the 1980s by shifts in the radiogenic isotope signatures (Fig. 4.2).

The observed fluctuations in sediment composition also emphasize the sensitivity and the importance of radiogenic isotope signatures of clays to detect even small temporal provenance variations whereas the clay mineral records are apparently not sufficiently sensitive to reflect such changes. Sediments collected in different parts of the Skagerrak region (Fig. 4.1B) show no significant spatial variations in the radiogenic isotope signatures and even minor variations in the $^{87}\text{Sr}/^{86}\text{Sr}$ composition can be traced in all cores. In contrast to our results, Hass (1993) found that granulometric signatures of neighbouring cores in the Skagerrak are difficult to correlate, due to rapid lateral changes in the depositional environment of the Skagerrak basin. The similarity of radiogenic isotope signatures observed in the various Skagerrak cores show that the core locations in the western and eastern part of the Skagerrak are supplied by similar sediment sources and that the variable but local SPM discharge of the adjacent Scandinavian rivers (Pederstad et al., 1993) do not affect the sediment provenance in different parts of the inner Skagerrak basin.

4.5.2 Changes in the hydrodynamic conditions of the Skagerrak

Grain-size of Skagerrak sediments is dominated by high mud ($< 63 \mu\text{m}$) contents. Sediments cored originate from parts of the Skagerrak where weak bottom currents prevail (maximum velocities between $5 - 10 \text{ cm s}^{-1}$; Rodhe, 1987) and grains in the clay and fine silt range are preferably deposited. The upward-increase in the amount of coarse silt in MUC2, MUC7 and MUC9, as well as fine sand in MUC2 and MUC8, (Fig. 4.3), suggests an increase of bottom current intensity since the second half of the 20th century. This is also corroborated by a general decline of the fine silt and clay fraction since around the 1970s (Fig. 4.3). It has been suggested

that accelerated bottom-current velocities increased the transport of bedload sediments like coarse silt and sand from coastal areas to the central part of the basin, while finer grains preferably remained in suspension (Engelhardt, 1977) or are winnowed. For the last decades, Sandén and Håkansson (1996) as well as Frigstad et al. (2023) reported an increase in suspended particle concentrations in Skagerrak waters. This is partly connected to the variability in the hydrography of the Skagerrak (Frigstad et al., 2023), which is forced by wind strength and has been closely correlated with NAO (North Atlantic Oscillation; Sætre, 2007). During periods of a positive NAO, strong westerly winds increase the inflow of Atlantic water and intensify the currents within the Skagerrak region, likely leading to the resuspension of particles in coastal waters. With this regard, Hass (1993, 1996) inferred that intensification of bottom-current circulation occurs during periods of more frequent westerly storms, resulting in generally coarser deposits in the Skagerrak. It is therefore likely that the upward-coarsening of Skagerrak sediments since around the 1970s reflects intensified NAO-related storminess, as this has been particularly observed in the 1980s and 1990s. This is in line with findings of De Haas (1996) who concluded that sedimentation pattern and rate in the Norwegian Channel is in particular influenced by the hydrological regime. As a consequence, the suspension load only settles in the deeper parts of the Norwegian Channel, where current velocities are less pronounced, which favours a permanent deposition resulting in higher sedimentation rates especially in the northern part of the Norwegian Channel. Increasing currents also favour the inflow of the Baltic Current into the Skagerrak during positive NAO phases and storm surges, as strong westerly winds push surface waters from the central North Sea to the Skagerrak intensifying the inflow of water masses from the Baltic Sea (Winther and Johannessen, 2006). This may explain, the observed lower radiogenic ϵ_{Hf} signatures in recent sediment layers of MUC7 and MUC8 (Fig. 4.2), potentially indicating enhanced sediment supply from the Baltic Sea due to higher storm intensity over the last decades. Recent findings by Lenz et al. (2024) also show that the Skagerrak region experienced a reduced sediment supply from the southern North Sea since the end of the 20th century, which is consistent with decreasing sedimentation rates in the region since 1963 as suggested by Spiegel et al. (in review; Tab. 4.1). However, the decline in average mass accumulation rates (MAR) from 0.17 to 0.09 g cm⁻² yr⁻¹ over this short time period appears too large to be explained solely by the detected change in the sediment provenance. In addition, the reduced deposition of sediment in the fine silt range may have resulted in declining sedimentation rates as observed past the 1960s. With this regard, it is suspicious that MUC8 which exhibits the most pronounced decline in MARs among all the stations (Tab. 4.1) also show a significant decrease in particles in both the fine as well as coarse silt fraction, while the

sand content increases considerably during the second half of the 20th century (Fig. 4.3). This indicates that, compared to the other core locations, station MUC8 experienced the most intense hydrodynamic conditions.

Our findings demonstrate that the sedimentation in the Skagerrak is controlled by both sediment transport processes in the North Sea and the local hydrodynamic effects in the Skagerrak region, predominantly forced by storminess. Sedimentation rates in the Skagerrak, a sedimentary system that is mainly supplied by fine-grained sediments, decrease with increasing hydrodynamic activity. This implies that an ongoing increase in storm intensity will further decline sedimentation rates within the Skagerrak basin and that fluctuations in the accumulation rates are rather controlled by short-term climatic changes than by human activities. It remains unclear whether the fine fraction, which is prevented from settling in the inner Skagerrak, is subsequently transported and deposited in the area of the Norwegian Trench. To clarify where this fine-grained material settles, it remains essential to determine the temporal evolution of MARs along a northward transect of the Norwegian Trench.

4.5.3 Changes in the sedimentary system of the North Sea over the last century

4.5.3.1 Climatic impact

A change in the sediment provenance is indicated by the fluctuations in the radiogenic isotopes since around the 1980s (Fig. 4.2). Previous studies show that clay size sediments of the southern North Sea are characterized by more radiogenic Sr isotope and less radiogenic Nd as well as Hf isotope signatures (Fig. 4.4; Lenz et al., 2024). Consequently, the shift observed in the most recent sediments may indicate a reduced supply of material originating from the southern North Sea. Changes in the sediment transport paths of the southern North Sea were also observed in the Wadden Sea area, which plays a crucial role in the sediment balance of the southeastern North Sea being a natural sink for fine-grained sediments. Changes of the sedimentary framework conditions, like sea-level rise and increased storminess have led to a larger tidal range leading to lateral extension and vertical growth of tidal flats (Diekmann et al., 1987; Pye and Blott, 2014; Benninghof and Winter, 2019). The capability of tidal flats to keep pace with sea-level rise (Andersen et al., 2010; Friedrichs, 2011) depends on the sediment availability and requires a continuous sediment supply from external sources. Postma (1981) and Oost et al. (2021) suggested that the fine-grained sediments originate from outside the tidal flats such as

the English Channel and local rivers. This suggests that the Wadden Sea acts as a natural trap for sediments that are transported along the southern North Sea coast, the efficiency of which increased in the past century due to climate change. The latter is also observed in various salt marshes along the German and Danish coasts which shown accelerating accretion rates, especially in the 1980s and 1990s due to accumulated storm-surge events (Pedersen and Bartholdy, 2006; Schürch et al., 2011; Lenz et al., 2023). These trends are mirrored by the shift of the radiogenic isotopes in the Skagerrak (this study) past around 1980, which indicate that higher amounts of SPM were trapped in the Wadden Sea area before reaching the Skagerrak region. In view of recent sea-level rise and the predicted increase in storminess in the near future, it is assumed that increasing trapping efficiency of tidal flats will further deteriorate the supply of sediments from the southern North Sea to the Skagerrak.

The isotope signatures of the uppermost sediment layers assigned to the time interval past 1980 closely resemble those from the Fladen Ground, the predominant source of Skagerrak clays in the northern North Sea (Fig. 4.4; Lenz et al., 2024). This finding may indicate an enhanced influence of the Atlantic Current to the Skagerrak region. The inflow of Atlantic water into the northern North Sea varies seasonally (Robertson 1909; Schott, 1966) and can be intensified by westerly winds. The Atlantic inflow can also be subject of rapid fluctuations in its geographical extension in the northern North Sea (Dooley, 1974). Furthermore, Winther and Johannessen (2006) pointed out that inter-annual to multi-decadal variations in the circulation pattern of the North Sea are linked to the NAO, with positive NAO phases being characterized by strong westerly winds in northern Europe (Hurrell, 1995; Carretero et al., 1998; Alexandersson et al., 2000; Hurrell et al., 2003; Dangendorf et al., 2012). This in mind, it appears logical, that an enhanced Atlantic inflow to the Skagerrak basin is the result of strong westerly winds observed during the 1980s and 1990s, triggered by a pronouncedly positive NAO index. Similar atmospheric conditions prevailed during the 1920s, where radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in cores MUC2 and MUC8 reflect higher values (Fig. 4.2). This climatic influence could also account for the lower radiogenic ϵHf signatures observed in MUC8 during this period, which contrast with the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the same core.

To summarise, the radiogenic isotope data suggest that climatic changes significantly impacted the sedimentary regime of the North Sea and the Skagerrak, especially since the second half of the 20th century. Climatic key factors include increased storminess, particularly bound to the NAO, which may intensify erosional processes in the northern North Sea and amplify sedimentation in Wadden Sea depocenters of the southern North Sea.

4.5.3.2 Human impact on the North Sea sedimentary system

In addition to natural forcing, sedimentary changes in the northern North Sea may also result from increased bottom-trawling activity in the Fladen Ground (northern North Sea, Fig. 4.1). Here, fishing activity was intensified in the second half of the 20th century due to technological advances (Jenning et al., 1999). It is likely that the trawling activities increased the amounts of SPM in the North Sea during the second half of the 20th century. Couce et al. (2020) showed that the trawling activity in the Fladen Ground was peaked in the 1980s and 1990s. As this region acts as the main source for Skagerrak sediments, an increased supply of Fladen Ground sediments has caused the shift in radiogenic isotope signatures of Skagerrak sediments deposited after 1980, when the signatures closely resemble those from the Fladen Ground (northern North Sea; Fig. 4.4; Lenz et al., 2024). This would explain the observed relative decrease in sediment supplied from the southern North Sea.

The deepening of river courses and ongoing dredging activities in large European rivers discharging to the North Sea, like Elbe, Weser, and Ems, has also been related to sedimentary changes in the southern North Sea (Kerner, 2007). So, has dredging of the river Elbe drastically increased since 1990 (Dücker et al., 2006), accompanied by a decline in riverine discharge of SPM into the North Sea (Goelz, 2008; Rovira et al., 2014). However, the overall invariant ϵ_{Hf} signatures observed in the Skagerrak cores as well as the respective lower radiogenic values in recent sediment layers (Fig. 4.2) do not mirror a reduced Mid-European riverine sediment contribution of the Skagerrak over time as the sediments supplied by these rivers are generally characterized by low radiogenic ϵ_{Hf} signatures. This makes human intervention less likely to impact the sediment budget of the North Sea during the last century.

Our findings imply that the observed changes in Skagerrak sediment composition over the last century result from both, climate related environmental changes (regional wind field, storminess, sea level) and anthropogenic measures (mainly fishery) affecting the sediment dynamics of the North Sea. Results also emphasize the difficulty to distinguish between natural and anthropogenic factors and question if it will be possible quantify their respective influences. However, our geochemical data underline that environmental changes in most recent decades have altered provenance of sediments in the Skagerrak depocenter. In times of rapid ongoing climate change, observed trends may accelerate potentially resulting in an imbalance of the North Sea sedimentary system.

4.6 Conclusions

Radiogenic Nd, Sr and Hf isotopes of clay-sized sediments from the Skagerrak provide new insights into changes of sediment provenance and the sedimentary dynamics of the North Sea during the last century. Variability of the radiogenic isotopes since the second half of the 20th century imply a decline in sediment supply from the southern North Sea as a result of the enhanced sediment trapping efficiency of the Wadden Sea promoted by increased storm intensity and sea-level rise. By contrast, intensified bottom trawling in the 1980s and 1990s in the northern North Sea led to enhanced seabed erosion in the Fladen Ground region, the predominant source of Skagerrak clays. This resulted in an increased transport of SPM from the northern North Sea to the Skagerrak. Enhanced sediment supply may be promoted by strong westerly winds, which intensify the Atlantic inflow. Radiogenic isotopes also indicate that intensified dredging and deepening of large European rivers has not significantly affected the sediment supply to the Skagerrak depocenter. In contrast to the clay minerals, which revealed no significant variability over time, fluctuations of the radiogenic isotope signatures demonstrate the sensitivity of these proxies and their importance in identifying temporal changes in sediment provenance and supply. We show that the proposed decline in accumulation rates of the Skagerrak sediments can be explained not only by changes in the sediment supply but also by enhanced hydrodynamics in the Skagerrak region. In the course of amplified storm and wave intensity, fine-grained sediments are hampered from settling in the inner part of the Skagerrak resulting in lower sedimentation rates. Finally, our findings highlight that changes in the sedimentary system of the Skagerrak basin over the last century have been a result of both climate and anthropogenic forcing

4.7 Acknowledgements

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4.8 Supplement

4.8.1 Supplementary figures

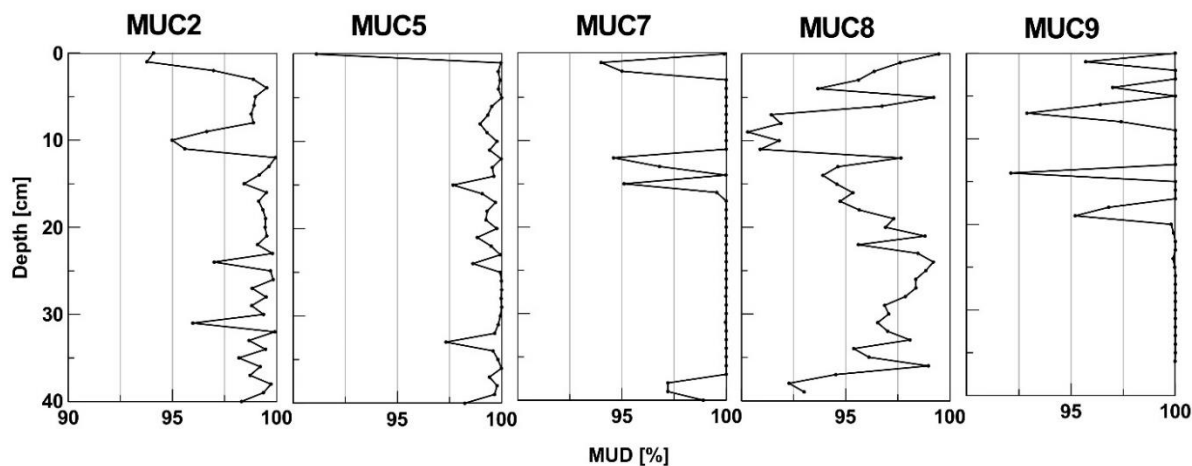


Fig. S4.1: Mud content of the MUCs.

4.8.2 Supplementary tables

Tab. S4.1: Radiogenic isotope compositions of down-core samples with analytical uncertainties ($2\sigma_{SD}$) of the respective mass spectrometry sessions.

core ID	depth [cm]	time [yr]	eNd	sigma2	$^{87}\text{Sr}/^{86}\text{Sr}$	sigma2	eHf	sigma2
MUC2_1	1	2018	-13.40	0.39	0.741379	0.000021	-6.97	0.50
MUC2_5	5	2003	-13.75	0.39	0.738363	0.000038	-7.52	0.5
MUC2_8	8	1989	-13.71	0.32	0.738167	0.000021	-8.10	0.53
MUC2_10	10	1979	-14.05	0.33	0.738559	0.000038	-7.76	0.63
MUC2_15	15	1960	-13.61	0.32	0.737274	0.000021	-6.78	0.50
MUC2_20	20	1950	-13.61	0.39	0.737869	0.000038	-7.51	0.50
MUC2_25	25	1938	-14.02	0.33	0.737626	0.000038	-7.10	0.63
MUC2_30	30	1926	-13.57	0.39	0.737676	0.000038	-6.82	0.50
MUC2_35	35	1912	-13.66	0.32	0.738230	0.000021	-7.56	0.53
MUC2_40	40	1899	-13.48	0.39	0.739020	0.000038	-7.59	0.50
MUC7_1	1	2017	-13.50	0.39	0.738474	0.000021	-7.88	0.50
MUC7_3	3	2009	-13.07	0.32	0.738131	0.000021	-6.39	0.53
MUC7_5	5	1999	-13.47	0.39	0.737721	0.000038	-6.34	0.50

MUC7_8	8	1984	-13.53	0.32	0.737714	0.000038	-7.14	0.53
MUC7_10	10	1973	-13.53	0.39	0.738095	0.000038	-7.44	0.50
MUC7_15	15	1953	-13.77	0.32	0.738083	0.000021	-7.70	0.53
MUC7_20	20	1935	-13.34	0.39	0.738244	0.000038	-7.73	0.50
MUC7_25	25	1915	-13.37	0.32	0.739805	0.000038	-7.07	0.53
MUC7_30	30	1893	-13.28	0.39	0.738530	0.000021	-7.58	0.50
MUC7_35	35	1870	-13.44	0.32	0.738385	0.000021	-8.12	0.53
MUC8_1	1	2019	-13.57	0.39	0.740417	0.000021	-8.29	0.50
MUC8_5	5	2007	-13.61	0.39	0.740319	0.000021	-8.01	0.50
MUC8_10	10	1989	-13.54	0.39	0.740287	0.000021	-7.52	0.50
MUC8_15	15	1969	-13.56	0.33	0.737618	0.000038	-7.84	0.63
MUC8_20	20	1954	-13.26	0.39	0.738103	0.000021	-6.92	0.50
MUC8_25	25	1951	-13.89	0.33	0.738128	0.000038	-7.01	0.63
MUC8_30	30	1941	-13.99	0.33	0.738699	0.000038	-6.76	0.63
MUC8_35	35	1932	-13.65	0.33	0.739129	0.000038	-6.85	0.63
MUC8_40	40	1921	-13.47	0.39	0.737976	0.000021	-8.60	0.50
MUC5_1	1	2017	-14.4	0.39	0.739997	0.000091	-7.76	0.58
MUC5_5	5	1998	-14.41	0.33	0.739569	0.000038	-7.32	0.63
MUC5_10	10	1968	-14.67	0.33	0.739773	0.000038	-7.73	0.50
MUC5_15	15	1954	-13.96	0.33	0.739743	0.000038	-7.32	0.63
MUC5_20	20	1941	-13.63	0.33	0.739516	0.000038	-7.20	0.63
MUC5_25	25	1925	-13.62	0.33	0.739147	0.000038	-7.08	0.63
MUC5_30	30	1909	-13.65	0.33	0.738175	0.000038	-7.53	0.63
MUC5_40	40	1874	-13.72	0.33	0.738305	0.000038	-8.00	0.63
MUC9_1	1	2016	-14.5	0.39	0.742600	0.000091	-8.37	0.58
MUC9_5	5	1991	-13.66	0.32	0.738972	0.000038	-7.09	0.53
MUC9_10	10	1959	-14.01	0.32	0.738255	0.000038	-7.80	0.53
MUC9_15	15	1943	-13.82	0.40	0.738449	0.000038	-7.43	0.53
MUC9_25	25	1901	-13.91	0.40	0.738471	0.000038	-7.40	0.53
MUC9_30	30	1879	-13.79	0.33	0.739000	0.000038	-7.87	0.53

Tab. S4.2: Clay mineral distribution of down-core samples.

core ID	Depth [cm]	Kaolinit [%]	Illite [%]	Chlorit [%]	Smektit [%]
MUC7_1	1	15.1	59.3	18.1	7.6
MUC7_5	5	18.4	54.2	19.9	7.5

MUC7_10	10	17.1	55.4	17.3	10.1
MUC7_20	20	15.4	57.6	18.4	8.6
MUC7_30	30	13.7	52.3	23.2	10.9
MUC2_1	1	15.8	55.6	20.5	8.0
MUC2_5	5	15.2	57.6	18.0	9.1
MUC2_10	10	10.4	62.4	19.9	7.3
MUC2_20	20	13.8	56.4	19.9	9.9
MUC2_30	30	11.7	60.8	17.3	10.2
MUC8_1	1	18.9	58.5	18.1	4.5
MUC8_5	5	14.1	56.2	24.4	5.4
MUC8_20	20	16.1	55.9	19.4	8.6
MUC8_30	30	17.2	54.5	20.8	7.6
MUC8_40	40	14.8	57.7	20.3	7.2

Tab. S4.3: Grain size fractions of MUC2.

Depth [cm]	mean [μm]	v fine sand [%]	v coarse silt [%]	coarse silt [%]	fine silt [%]	v fine silt [%]	clay [%]
0	8.49	4.9%	15.2%	25.7%	10.8%	10.8%	21.3%
1	8.38	6.2%	14.3%	25.1%	10.9%	10.9%	21.5%
2	7.69	3.0%	14.2%	26.4%	11.4%	11.4%	22.3%
3	6.81	0.5%	12.3%	27.3%	12.1%	12.1%	23.7%
4	6.56	0.0%	11.4%	27.3%	12.3%	12.3%	24.3%
5	7.32	0.5%	14.5%	27.4%	11.6%	11.6%	22.8%
6	7.39	0.6%	14.7%	27.4%	11.5%	11.5%	22.7%
7	6.74	0.7%	11.8%	27.2%	12.1%	12.1%	23.9%
8	6.90	0.6%	12.5%	27.3%	12.0%	12.0%	23.6%
9	7.12	3.1%	11.9%	26.2%	11.8%	11.8%	23.3%
10	7.89	4.6%	13.4%	25.7%	11.3%	11.3%	22.1%
11	7.72	3.2%	13.6%	26.3%	11.3%	11.3%	22.3%
12	3.05	0.0%	0.0%	4.8%	19.2%	19.2%	37.7%
13	6.95	0.2%	13.0%	27.6%	11.9%	11.9%	23.5%
14	7.02	0.3%	13.4%	27.4%	11.9%	11.9%	23.3%
15	6.77	0.9%	11.9%	27.0%	12.1%	12.1%	23.8%
16	6.08	0.0%	8.9%	27.0%	12.9%	12.9%	25.4%
17	6.85	0.3%	12.6%	27.3%	12.0%	12.0%	23.7%
18	7.31	0.3%	14.5%	27.6%	11.6%	11.6%	22.8%
19	7.01	0.0%	13.2%	27.9%	11.9%	11.9%	23.3%
20	7.19	0.0%	14.0%	27.9%	11.7%	11.7%	23.0%
21	7.40	0.0%	14.9%	27.9%	11.5%	11.5%	22.6%
22	6.80	0.5%	12.2%	27.3%	12.1%	12.1%	23.8%
23	6.83	0.2%	12.5%	27.5%	12.0%	12.0%	23.7%
24	6.89	3.0%	11.2%	26.0%	12.1%	12.1%	23.7%
25	6.75	0.3%	12.1%	27.4%	12.1%	12.1%	23.9%

26	6.37	0.0%	10.5%	27.1%	12.6%	12.6%	24.7%
27	6.64	0.6%	11.3%	27.1%	12.3%	12.3%	24.1%
28	6.71	0.0%	11.9%	27.5%	12.2%	12.2%	23.9%
29	6.52	0.6%	10.7%	27.1%	12.4%	12.4%	24.3%
30	6.64	0.5%	11.4%	27.2%	12.3%	12.3%	24.1%
31	7.34	4.0%	12.3%	25.7%	11.7%	11.7%	23.0%
32	6.36	0.0%	10.3%	27.2%	12.6%	12.6%	24.7%
33	6.83	0.8%	12.1%	27.2%	12.1%	12.1%	23.7%
34	7.05	0.3%	13.4%	27.6%	11.8%	11.8%	23.3%
35	7.04	1.8%	12.4%	26.8%	11.9%	11.9%	23.4%
36	6.79	0.3%	12.3%	27.4%	12.1%	12.1%	23.8%
37	6.57	0.8%	10.9%	27.1%	12.3%	12.3%	24.2%
38	6.76	0.0%	12.4%	27.3%	12.1%	12.1%	23.9%
39	6.86	0.3%	12.5%	27.4%	12.0%	12.0%	23.6%
40	7.40	1.1%	14.4%	27.2%	11.5%	11.5%	22.7%

Tab. S4.4: Grain size fractions of MUC5.

Depth [cm]	mean [μm]	v fine sand [%]	v coarse silt [%]	coarse silt [%]	fine silt [%]	v fine silt [%]	clay [%]
0	9.37	5.2%	15.1%	25.0%	10.4%	10.4%	20.4%
1	6.78	0.0%	12.3%	27.6%	12.1%	12.1%	23.8%
2	6.72	0.2%	11.9%	27.5%	12.2%	12.2%	23.9%
3	6.53	0.1%	11.1%	27.5%	12.4%	12.4%	24.3%
4	6.04	0.0%	8.7%	27.0%	13.0%	13.0%	25.5%
5	6.25	0.0%	9.8%	27.1%	12.7%	12.7%	25.0%
6	6.29	0.0%	10.0%	27.2%	12.6%	12.6%	24.8%
7	6.59	0.1%	11.4%	27.4%	12.3%	12.3%	24.2%
8	6.66	0.9%	11.2%	27.1%	12.2%	12.2%	24.1%
9	6.54	0.3%	11.1%	27.3%	12.4%	12.4%	24.3%
10	6.68	0.2%	11.8%	27.4%	12.2%	12.2%	24.0%
11	6.77	0.5%	12.0%	27.4%	12.1%	12.1%	23.8%
12	6.28	0.0%	10.0%	27.1%	12.7%	12.7%	24.9%
13	6.73	0.3%	11.9%	27.5%	12.1%	12.1%	23.9%
14	6.67	0.1%	11.7%	27.5%	12.2%	12.2%	24.0%
15	7.14	2.0%	12.4%	26.7%	11.8%	11.8%	23.2%
16	6.79	0.7%	11.9%	27.3%	12.1%	12.1%	23.8%
17	6.84	0.2%	12.5%	27.5%	12.0%	12.0%	23.7%
18	6.78	0.2%	12.3%	27.5%	12.1%	12.1%	23.8%
19	6.79	0.2%	12.3%	27.4%	12.1%	12.1%	23.8%
20	6.53	0.0%	11.3%	27.2%	12.4%	12.4%	24.3%
21	6.89	0.9%	12.2%	27.2%	12.0%	12.0%	23.6%
22	6.64	0.2%	11.5%	27.4%	12.2%	12.2%	24.1%
23	6.29	0.0%	10.1%	27.1%	12.7%	12.7%	24.9%
24	6.87	1.2%	11.9%	27.0%	12.0%	12.0%	23.6%
25	6.46	0.0%	10.9%	27.4%	12.4%	12.4%	24.4%
26	6.50	0.0%	11.0%	27.4%	12.4%	12.4%	24.4%
27	6.17	0.0%	9.4%	27.1%	12.8%	12.8%	25.2%
28	6.13	0.0%	9.0%	27.2%	12.9%	12.9%	25.3%

29	6.43	0.0%	10.7%	27.3%	12.5%	12.5%	24.5%
30	6.48	0.1%	10.9%	27.4%	12.4%	12.4%	24.4%
31	6.58	0.1%	11.4%	27.3%	12.3%	12.3%	24.2%
32	6.73	0.2%	12.0%	27.4%	12.2%	12.2%	23.9%
33	7.38	2.4%	13.3%	26.6%	11.6%	11.6%	22.8%
34	6.81	0.2%	12.4%	27.4%	12.1%	12.1%	23.7%
35	6.72	0.2%	11.9%	27.6%	12.2%	12.2%	23.9%
36	6.57	0.0%	11.5%	27.3%	12.3%	12.3%	24.2%
37	6.83	0.6%	12.2%	27.3%	12.1%	12.1%	23.7%
38	6.78	0.2%	12.3%	27.4%	12.1%	12.1%	23.8%
39	6.92	0.3%	12.8%	27.4%	12.0%	12.0%	23.5%
40	7.68	1.3%	15.0%	26.9%	11.3%	11.3%	22.3%
41	7.17	0.7%	13.7%	27.3%	11.7%	11.7%	23.1%
42	7.07	1.0%	13.0%	27.2%	11.8%	11.8%	23.3%
43	7.14	0.3%	13.8%	27.4%	11.8%	11.8%	23.1%
44	7.03	0.3%	13.3%	27.5%	11.9%	11.9%	23.3%

Tab. S4.5: Grain size fractions of MUC7.

Depth [cm]	mean [μm]	v fine sand [%]	v coarse silt [%]	coarse silt [%]	fine silt [%]	v fine silt [%]	clay [%]
0	6.64	0.1%	11.7%	27.4%	12.2%	12.2%	24.1%
1	6.53	0.0%	11.1%	27.5%	12.4%	12.4%	24.3%
2	6.62	0.0%	11.5%	27.6%	12.3%	12.3%	24.1%
3	6.22	0.0%	9.6%	27.2%	12.7%	12.7%	25.0%
4	6.27	0.0%	9.9%	27.3%	12.7%	12.7%	24.9%
5	6.27	0.0%	9.9%	27.2%	12.7%	12.7%	24.9%
6	6.54	0.0%	11.1%	27.5%	12.3%	12.3%	24.3%
7	5.80	0.0%	7.4%	26.6%	13.3%	13.3%	26.1%
8	5.80	0.0%	7.3%	26.7%	13.3%	13.3%	26.1%
9	5.82	0.0%	7.4%	26.7%	13.3%	13.3%	26.1%
10	5.89	0.0%	7.8%	26.9%	13.2%	13.2%	25.9%
11	5.21	0.0%	4.2%	25.3%	14.2%	14.2%	27.9%
12	5.52	0.0%	5.9%	26.0%	13.7%	13.7%	27.0%
13	5.96	0.0%	8.1%	27.0%	13.1%	13.1%	25.7%
14	6.16	0.0%	9.3%	27.2%	12.8%	12.8%	25.1%
15	5.97	0.0%	8.2%	27.0%	13.0%	13.0%	25.6%
16	6.36	0.4%	9.9%	27.3%	12.6%	12.6%	24.7%
17	5.75	0.0%	7.1%	26.6%	13.3%	13.3%	26.2%
18	5.68	0.0%	6.7%	26.4%	13.5%	13.5%	26.5%
19	5.85	0.0%	7.6%	26.8%	13.2%	13.2%	26.0%
20	6.30	0.0%	10.0%	27.2%	12.6%	12.6%	24.8%
21	6.24	0.0%	9.7%	27.3%	12.7%	12.7%	25.0%
22	6.20	0.0%	9.4%	27.3%	12.7%	12.7%	25.1%
23	5.93	0.0%	8.0%	27.0%	13.1%	13.1%	25.8%
24	6.35	0.0%	10.2%	27.3%	12.6%	12.6%	24.7%
25	5.86	0.0%	7.7%	26.8%	13.2%	13.2%	25.9%
26	6.18	0.0%	9.3%	27.2%	12.8%	12.8%	25.1%
27	6.13	0.0%	9.1%	27.2%	12.8%	12.8%	25.2%

28	6.45	0.0%	10.7%	27.5%	12.4%	12.4%	24.4%
29	5.93	0.0%	8.0%	26.9%	13.1%	13.1%	25.7%
30	6.05	0.0%	8.6%	27.2%	12.9%	12.9%	25.4%
31	6.45	0.0%	10.7%	27.5%	12.4%	12.4%	24.5%
32	6.36	0.0%	10.3%	27.3%	12.6%	12.6%	24.7%
33	6.24	0.0%	9.6%	27.2%	12.7%	12.7%	25.0%
34	6.22	0.0%	9.6%	27.2%	12.7%	12.7%	25.0%
35	6.14	0.0%	9.1%	27.4%	12.8%	12.8%	25.1%
36	5.85	0.0%	7.6%	26.8%	13.2%	13.2%	26.0%
37	6.04	0.0%	8.5%	27.1%	13.0%	13.0%	25.5%
38	6.33	0.0%	10.2%	27.3%	12.6%	12.6%	24.7%
39	6.17	0.0%	9.3%	27.2%	12.8%	12.8%	25.1%
40	6.33	0.0%	10.1%	27.4%	12.6%	12.6%	24.7%
41	5.92	0.0%	8.0%	26.9%	13.1%	13.1%	25.8%
42	6.25	0.0%	9.8%	27.2%	12.7%	12.7%	25.0%
43	6.36	0.0%	10.2%	27.5%	12.5%	12.5%	24.7%

Tab. S4.6: Grain size fractions of MUC8.

Depth [cm]	mean [μm]	v fine sand [%]	v coarse silt [%]	coarse silt [%]	fine silt [%]	v fine silt [%]	clay [%]
0	7.85	0.5%	16.8%	27.1%	11.2%	11.2%	22.0%
1	8.08	2.3%	16.2%	26.6%	11.0%	11.0%	21.7%
2	8.08	3.6%	15.2%	26.1%	11.1%	11.1%	21.8%
3	8.48	4.3%	16.2%	25.8%	10.8%	10.8%	21.3%
4	8.81	5.9%	15.8%	25.1%	10.7%	10.7%	21.0%
5	7.47	0.8%	15.0%	27.2%	11.5%	11.5%	22.6%
6	7.31	3.2%	12.7%	26.0%	11.7%	11.7%	23.0%
7	10.30	8.0%	17.8%	24.3%	9.9%	9.9%	19.5%
8	10.75	7.2%	19.1%	24.7%	9.7%	9.7%	19.0%
9	11.34	8.2%	19.8%	24.2%	9.3%	9.3%	18.4%
10	10.94	8.0%	19.9%	24.4%	9.6%	9.6%	18.8%
11	11.33	8.4%	20.4%	24.3%	9.3%	9.3%	18.3%
12	9.30	2.3%	20.5%	26.3%	10.3%	10.3%	20.2%
13	10.42	5.4%	20.9%	25.3%	9.8%	9.8%	19.2%
14	9.84	6.0%	18.7%	25.2%	10.1%	10.1%	19.8%
15	9.41	5.3%	18.1%	25.4%	10.3%	10.3%	20.2%
16	9.16	4.5%	17.9%	25.8%	10.4%	10.4%	20.4%
17	9.43	5.0%	18.2%	25.6%	10.3%	10.3%	20.2%
18	8.58	4.1%	16.6%	25.8%	10.8%	10.8%	21.1%
19	8.36	2.6%	17.0%	26.5%	10.9%	10.9%	21.3%
20	8.22	3.0%	16.3%	26.3%	11.0%	11.0%	21.6%
21	8.01	1.1%	16.8%	27.2%	11.1%	11.1%	21.7%
22	8.56	4.1%	16.3%	25.9%	10.8%	10.8%	21.1%
23	7.97	1.5%	16.3%	27.0%	11.1%	11.1%	21.8%
24	8.38	0.8%	18.3%	27.4%	10.8%	10.8%	21.2%
25	8.39	0.8%	18.3%	27.4%	10.8%	10.8%	21.2%
26	8.93	1.5%	19.5%	27.2%	10.4%	10.4%	20.5%
27	8.20	1.5%	17.3%	26.9%	10.9%	10.9%	21.5%

28	8.46	1.8%	17.7%	27.0%	10.7%	10.7%	21.1%
29	9.14	3.0%	19.0%	26.6%	10.3%	10.3%	20.3%
30	7.93	2.9%	15.8%	25.6%	11.2%	11.2%	22.1%
31	8.49	3.2%	16.9%	26.4%	10.8%	10.8%	21.2%
32	7.99	3.0%	16.3%	25.0%	11.2%	11.2%	22.0%
33	8.50	1.9%	18.0%	26.8%	10.7%	10.7%	21.1%
34	9.26	4.6%	18.3%	25.8%	10.3%	10.3%	20.3%
35	8.27	3.9%	15.9%	25.8%	11.0%	11.0%	21.6%
36	8.07	1.0%	17.3%	27.0%	11.0%	11.0%	21.7%
37	10.21	5.3%	20.1%	25.6%	9.8%	9.8%	19.4%
38	11.35	7.1%	22.2%	25.0%	9.1%	9.1%	18.0%
39	11.00	6.3%	21.7%	25.3%	9.4%	9.4%	18.4%

Tab. S4.7: Grain size fractions of MUC9.

Depth [cm]	mean [µm]	v fine sand [%]	v coarse silt [%]	coarse silt [%]	fine silt [%]	v fine silt [%]	clay [%]
0	6.79	0.0%	12.3%	27.7%	12.1%	12.1%	23.8%
1	6.95	0.0%	13.1%	27.7%	11.9%	11.9%	23.4%
2	6.70	0.0%	12.0%	27.5%	12.2%	12.2%	24.0%
3	6.80	0.0%	12.4%	27.6%	12.1%	12.1%	23.8%
4	6.92	0.0%	13.0%	27.6%	12.0%	12.0%	23.5%
5	6.12	0.0%	8.9%	27.3%	12.9%	12.9%	25.3%
6	6.44	0.0%	10.5%	27.5%	12.5%	12.5%	24.5%
7	6.16	0.0%	8.1%	28.4%	12.8%	12.8%	25.1%
8	6.26	0.0%	8.9%	28.2%	12.7%	12.7%	24.9%
9	4.98	0.0%	3.0%	24.4%	14.6%	14.6%	28.7%
10	6.32	0.0%	9.5%	27.9%	12.6%	12.6%	24.8%
11	6.27	0.0%	9.3%	27.9%	12.7%	12.7%	24.9%
12	6.47	0.0%	10.2%	28.1%	12.4%	12.4%	24.4%
13	6.42	0.0%	10.1%	27.9%	12.5%	12.5%	24.5%
14	9.02	4.7%	14.5%	25.3%	10.5%	10.5%	20.7%
15	6.44	0.0%	10.3%	27.8%	12.5%	12.5%	24.5%
16	6.16	0.0%	8.5%	28.0%	12.8%	12.8%	25.2%
17	6.29	0.0%	9.4%	27.8%	12.6%	12.6%	24.9%
18	6.44	0.0%	10.3%	27.7%	12.5%	12.5%	24.5%
19	6.33	0.0%	9.5%	28.0%	12.6%	12.6%	24.8%
20	7.07	0.2%	13.4%	27.8%	11.8%	11.8%	23.2%
21	6.83	0.1%	12.5%	27.6%	12.0%	12.0%	23.7%
22	6.66	0.0%	11.8%	27.5%	12.2%	12.2%	24.0%
23	6.28	0.0%	9.8%	27.3%	12.7%	12.7%	24.9%
24	6.75	0.1%	12.1%	27.6%	12.1%	12.1%	23.8%
25	6.14	0.0%	9.1%	27.3%	12.8%	12.8%	25.2%
26	6.62	0.0%	11.5%	27.5%	12.3%	12.3%	24.1%
27	6.43	0.0%	10.7%	27.3%	12.5%	12.5%	24.5%
28	5.89	0.0%	7.8%	26.8%	13.2%	13.2%	25.9%
29	6.64	0.0%	11.7%	27.4%	12.3%	12.3%	24.1%
30	6.68	0.0%	11.9%	27.5%	12.2%	12.2%	24.0%
31	6.82	0.0%	12.5%	27.6%	12.1%	12.1%	23.7%

32	6.83	0.0%	12.5%	27.6%	12.1%	12.1%	23.7%
33	6.72	0.0%	12.0%	27.5%	12.2%	12.2%	23.9%
34	6.39	0.0%	10.2%	27.6%	12.5%	12.5%	24.6%
35	6.70	0.0%	11.9%	27.6%	12.2%	12.2%	24.0%
36	7.04	0.0%	13.4%	27.8%	11.8%	11.8%	23.3%

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5. Scientific Chapter III

Influence of climate variability on sediment dynamics in the Skagerrak basin over the past 150 years

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Abstract

The North Sea represents a highly dynamic sedimentary system where sediment transport and deposition are primarily controlled by tidal and wind-driven currents. The Skagerrak basin acts as the main depositional sink for fine-grained sediments transported across the North Sea region. However, little is known about how climate variability has influenced sediment dynamics at this depocenter, particularly concerning the contributions from various sediment sources. To investigate how sedimentation has responded to climate variability, XRF core scanning records from Skagerrak sediment cores were analysed, covering the transition from colder, stormier conditions of the Little Ice Age to the warmer Modern Climate Optimum. This analysis also provides the opportunity to assess sediment dynamics under largely natural conditions prior to intense human activities since the beginning of the industrialization. The results indicate that during colder periods, stronger bottom currents and enhanced Atlantic inflow into the northern North Sea and Skagerrak, controlled by intense westerly storms, led to coarser sediment deposition, as reflected by higher Zr/Rb ratios. As a result, intensified seabed erosion in the Fladen Ground and coastal erosion along the Scandinavian coastline caused an increased supply of sediments from these regions into the central part of the Skagerrak. This is

indicated by higher K/Rb and lower Fe/Ti ratios, since potassium-enriched minerals are more abundant in material sourced from Scandinavia, while iron-rich sediments are more prevalent in those from central Europe. In contrast, during calmer hydrodynamic conditions, the Skagerrak received higher sediment contributions from the southern North Sea. The climate-driven variability observed in the sedimentation pattern appears to be linked to the NAO (North Atlantic Oscillation), with a positive NAO index correlating with enhanced hydrodynamic conditions and increased sediment input from Scandinavian sources. These findings highlight the important role of climate change in shaping the sedimentary system and document that climatic forcing factors have had a stronger influence on sedimentation than anthropogenic activities.

5.1 Introduction

In recent times, climate variability has emerged to play an important role in controlling the sedimentary system of the North Sea. As a semi-enclosed basin of the Atlantic Ocean located in northern Europe, it represents a highly dynamic system receiving sediment input from surrounding European landmasses via rivers, the Atlantic Ocean, Baltic Sea, seabed and coastal erosion (Eisma, 1981; Eisma and Kalf, 1987). The sedimentation is controlled by permanent and tidal currents, which regulate the sediment transport across the shelf sea (Eisma, 1988; De Haas et al., 2002). A shift in meteorologically forced processes such as increased storminess led to stronger hydrodynamic conditions that changed the sediment transport regime (Kuijpers et al., 1993). Studies have shown that shifts in climate have influenced the amount of transported material leading to increased suspended particulate matter (SPM) concentrations in the North Sea (Capuzzo et al., 2015, Wilson and Heath, 2019) and its depocenters such as the Skagerrak basin (Sandén and Håkansson, 1996; Frigstad et al., 2023).

However, since the beginning industrial times, the influence of climate-forcing factors on sedimentary processes has been increasingly compounded by human activities (Halpern et al., 2008), i.e., bottom trawling (Bellman et al., 2005; Eigaard et al., 2017) and dredging of major European rivers (Rovira et al., 2014). This makes it challenging to disentangle the main impacts of natural and anthropogenic factors. Understanding the historical variability of climate and its interaction with the sedimentary system is essential to establish a baseline for distinguishing between the key drivers and also to reliably predict future sedimentation scenarios.

The transition from the Little Ice Age (LIA) to the recent warm period (Modern Climate Optimum) provides a valuable opportunity to investigate changes in sediment dynamics under predominantly natural conditions, prior to human impact that has prevailed since industrialisation. This period also marks a transition from colder, stormier conditions to the onset of the present-day warm period, offering insights into how varying climatic conditions have altered the energy dynamics and have shaped the sedimentary regime in the North Sea. The LIA was the coldest period within the last 8000 years, which lasted approximately between 1250 to 1860 AD (Grove, 1988; Mayewski et al., 1993, 1994). The on average colder temperatures can be attributed to a higher frequency of cold winters, which increased the hydrodynamics and enhanced erosional processes in the North Sea region (Nordberg, 1991). Over the last centuries, European climate has been closely linked to the intensity of the North Atlantic Oscillation (NAO), which has been interpreted to be a key driver of climate variability in this region (Hurrell, 1995; Wanner et al., 2001, Trouet et al., 2009), whereby a positive NAO index leads to increased storm intensity and frequency in northern Europe (Dangendorf et al., 2012; Hurrell, 1995; Hurrell et al., 2003).

Sediments continuously deposited in a depocenter provide an archive for reconstructing climate variability and its impact on sedimentary processes. The Skagerrak basin acts as the largest depocenter for fine-grained material transported from the southern and northern North Sea as well as Scandinavian rivers (Eisma and Kalf, 1987; Van Weering, 1982; Lenz et al., 2024). The northern North Sea area represents the predominant source for sediments, whereby seabed erosion is amplified by the inflow of the Atlantic currents providing the Skagerrak with fine-grained sediments (Lenz et al. 2024). The deposited sediments reach accumulation rates of up to 1 cm yr⁻¹ in the central part of the Skagerrak (Van Weering, 1982; Bøe et al., 1998). By analysing sedimentary and paleoenvironmental proxies of a sediment record from the Skagerrak, which includes the transition zone from the LIA to recent times, it is possible to assess how sediment transport and deposition has been influenced by climate variability in the Skagerrak and the North Sea region. Moreover, this also allows a better understanding how significant shifts in energy conditions affect changes in the provenance of Skagerrak sediments driving factors of which are predominantly the current system and erosional processes of the North Sea (Lenz et al. 2024).

X-ray fluorescence (XRF) core scanning is a highly effective tool for studying the sedimentary history of a depositional environment like the Skagerrak by analysing records of element variability. Physical factors such as variations in water content, grain size and organic matter

impact scanned XRF counts of single elements (Löwemark et al., 2011; Thomson et al., 2006). To address these limitations and improve data accuracy, element ratios are widely applied as proxies (Weltje and Tjallingii, 2008). Numerous studies have demonstrated the utility of XRF-derived elemental ratios in identifying changes in sediment provenance and energy conditions, often linked to climatic variability (e.g., Rothwell et al., 2006; Diekmann et al., 2008; Siani et al., 2010; Nieto-Moreno et al., 2011). Zirconium (Zr) predominantly resides in coarser grains, while rubidium (Rb) is mainly concentrated in clay-sized particles. Consequently, the Zr/Rb record in sediment cores serves as a reliable high-resolution proxy for grain-size variability. The approach of using Zr/Rb ratios as a proxy for tracking depositional grain-size changes and hydrodynamic variability has been proven valid in previous studies (e.g., Chen et al., 2006; Wang et al., 2011; Wu et al., 2020). Similarly, potassium (K), which is typically enriched in terrestrial siliciclastic e.g. illite clays and potassium feldspar (Croudace and Rothwell, 2015), commonly found in Scandinavian rocks, serves as a robust provenance proxy. It has been widely used in numerous studies to distinguish between different sediment sources, particularly where potassium-bearing rocks play a prominent role in the sediment composition (e.g. Richter et al., 2006; Diekmann et al., 2008; Hodell et al., 2010). The Fe/Ti also functions as a reliable proxy for determining sediment provenance, particularly in identifying contributions from Fe-rich materials like hematite and magnetite (Marsh et al., 2007), which are abundant in central Europe (Peine et al., 2000). Together, these geochemical proxies provide a comprehensive tool for reconstructing the sediment dynamics and differentiating between sources from northern and central Europe.

This study presents the first XRF core scanning record of Skagerrak sediments and aims to achieve two main objectives: (1) to reconstruct the factors influencing Skagerrak sedimentation in response to climatic variability, and (2) to provide new insights into how these factors have shaped sediment provenance during colder and warmer climatic periods. We present records of selected elemental ratios of cores collected from the central and outer part of the Skagerrak as well as the adjacent Kattegat, which represents the transitional zone between the Baltic Sea and the North Sea. The ratios were carefully selected to act as proxies for both hydrodynamic processes and sediment provenance. By comparing the elemental ratios across these locations, this study aims to identify patterns and variations that reflect the interaction between the currents, transport dynamics, and the sources of sediment input.

5.2 Study site

The Skagerrak strait is a shallow semi-enclosed sedimentary basin that connects the North Sea with the Baltic Sea (Fig. 5.1). It is located in the northeastern part of the North Sea which is a shallow marginal sea of the Atlantic Ocean in northwestern Europe. The Skagerrak represents the deepest part of the entire North Sea area where water depths reach up to 700 m in its central part. The circulation pattern of the North Sea is characterized by a counter-clockwise current system that is mainly forced by the Atlantic water masses that enter the marginal sea across its northern entrance between Scotland and Norway and to a lesser extent from the south-west across the English Channel (Svansson, 1975; Rodhe, 1987; Otto et al., 1990; Fig. 5.1A).

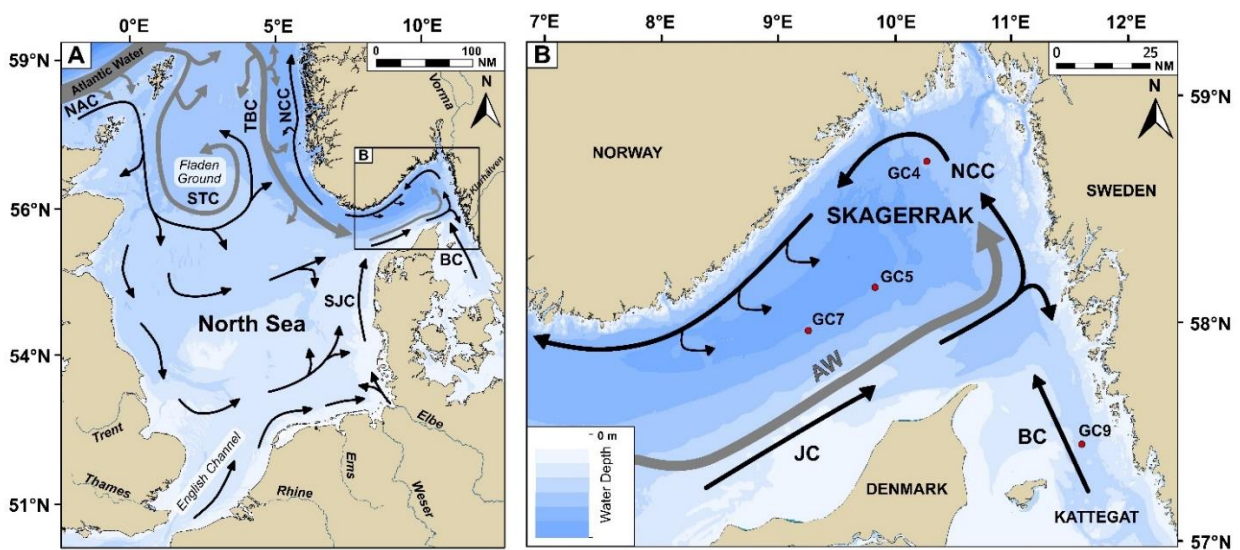


Fig. 5.1: (A) Water depth and circulation pattern of the North Sea modified after Turrel (1992) containing the main currents: NAC = North Atlantic Current, STC = Southern Trench Current, TBC = Tampen Bank Current, NCC = Norwegian Continental Current, SJC = South Jutland Current, and BC = Baltic Current **(B)** Skagerrak basin with collected gravity core sites and bathymetry. Black arrows mark the surface water circulation modified after Longva and Thorsnes (1997) including main currents: JC = Jutland Current, BC = Baltic Current, and NCC = Norwegian Coastal Current. The grey arrow indicates the Atlantic water inflow.

Water masses of the North Atlantic Current originating from the northward flow along the British coast partly continue eastward to the central part of the North Sea and end up in the Skagerrak. East of Norway, the Atlantic inflow carries on directly into the Skagerrak via the Tampen Bank Current. The southern North Sea is dominated by the South Jutland Current which carries mixed water masses from the English Channel as well as the southern North Sea and flows north along the Danish west coast further into the Skagerrak basin (Fig. 5.1A). As a result, the North Atlantic Current and Jutland Current, both climate-sensitive current systems

that are highly sensitive to wind forcing (Fonselius, 1990), provide the Skagerrak basin with fine-grained sediments stemming from the North Sea. As these currents enter the inner Skagerrak, they blend with the fresher, colder waters of the Baltic Current (BC), which originates from the Kattegat and leads to an anti-clockwise circulation pattern in the Skagerrak region (Svansson, 1975; Fig. 5.1B). The mixed water masses decelerate again further east and subsequently exit the Skagerrak as the Norwegian Coastal Current. During the cyclonic rotation, increasing water depths and decreasing flow velocities facilitate the rapid deposition of fine-grained sediment in the central and northeastern Skagerrak (Van Weering, 1982a; Rodhe and Holt, 1996; Bøe et al., 1996). The sedimentation in the current-controlled sedimentary system is dominated by mud and clay deposition in the inner part of the Skagerrak (Pederstad et al., 1993; Stevens et al., 1996; Spiegel et al., 2023) and forms a major sink area for fine-grained particles transported across the entire North Sea region (Van Weering, 1982; Bøe et al., 1998). The shallow areas near the Scandinavian coastline, where stronger hydrodynamic conditions prevail due to higher current and wave energy (Fonselius, 1990), consist of coarser sand and sediment coarse silt deposits.

5.3 Methods

5.3.1 Sample preparation

The data of this study are based on four sediment gravity cores (GC) collected during the AL561 research cruise with RV Alkor in August 2021 (Schmidt, 2021). The cores exhibit an undisturbed sediment sequence recovered from the central and northeastern part of the Skagerrak basin and the Kattegat (Fig. 5.1, Tab. 5.1). At each station, cores of 200 cm length below seafloor were recovered using a gravity corer. The uppermost 10 cm of GC4 exhibit a sedimentation gap and are therefore excluded from analyses.

Station	Latitude N	Longitude E	Water depth [m]	Porosity [%]	mean grain-size [μm]
GC5	58° 10.891'	09° 47.566'	492	0.75	7.58
GC7	57° 59.275'	09° 14.362'	490	0.73	10.03
GC4	58° 44.878'	10° 13.447'	214	0.72	8.06
GC9	57° 27.021'	11° 30.492'	66	0.70	11.99

Tab. 5.1: Gravity core locations with general information. Porosity and mean grain size represent mean values of the entire cores.

5.3.2 Grain-size analysis

The gravity cores were sampled equidistantly every 5 cm (volume per sample of 1.5 cm³) for the determination of grain-size distribution. Sediment samples were treated with 10% to 30% hydrogen peroxide and 60% acetic acid to dissolve organic matter and carbonate, respectively. Subsequently, sample were dispersed in water with tetra-sodium diphosphate decahydrate to bring the particles in suspension. Grain-size measurements were carried out at the CEN, University of Hamburg with a laser-diffraction particle-sizer (Sympatec HELOS/KF Magic; range 0.5/18 to 3200 3500 µm). To ensure accuracy of the measurements and to exclude effects of long-term instrumental drift an in-house grain-size standard was measured regularly (standard deviation for mean grain size and D50 over the analysis period was < 1.1 µm). The statistical assessment of grain-size data is based on graphical method (Folk and Ward, 1957), calculated using Gradistat (Blott and Pye, 2001).

5.3.3 Age model

Analyses of the radionuclides ²¹⁰Pb and ¹³⁷Cs were carried out to establish an age depth model for the gravity cores. Sediment samples were taken equidistantly every 10 cm. The porosity of the sediment samples was calculated from the loss of water after freeze-drying by assuming a density of 2.5 g cm⁻³ for the dry solids. For that wet sediment samples were weighed before and after freeze-drying to assess the wet and dry sediment mass. For the radionuclide analyses, the dry sediments were grinded and subsequently embedded in tubes which were sealed with a two-component epoxy resin. To ensure a steady-state equilibrium between ²²⁶Ra and ²¹⁴Bi the samples were stored for two weeks. The ²¹⁰Pb and ¹³⁷Cs activities were measured at GEOMAR Helmholtz Centre for Ocean Research Kiel using gamma spectrometry on n-type planar or coaxial Ge-detectors for GC5 and GC7. The natural background decay of ²²⁶Ra (295 keV) was also measured and subtracted from total ²¹⁰Pb activity to calculate the unsupported ²¹⁰Pb, which represents the excess of ²¹⁰Pb (²¹⁰Pb_{ex}). Mercury (Hg) content was measured equidistantly every 10 cm using a DMA-80 Analyser from the company MLS, following the methodology described by Leipe et al. (2013). Before measurements, samples were dried and weighed (ranging between 20 and 100 mg). Calibration was conducted with certified reference material CRM (BCR) 142R and soil standard SRM 2709.

The profiles of excess ²¹⁰Pb, ¹³⁴Cs activity, and Hg content in the gravity cores were compared with corresponding radionuclide and Hg profiles from cores collected at the same locations

using a multi-corer (MUC). Age-depth models for these MUCs have previously been developed by Spiegel et al. (in review). These models were constructed based on the decay of ^{210}Pb and a set of independent time markers, including ^{137}Cs peaks, fraction of modern ^{14}C , and Hg concentrations. The sediment cores were divided into two sections at the depth corresponding to the independent 1963-time marker. For the upper section, age-depth relationships were established using a reaction-transport model that accounted for sediment burial, mixing, compaction, and ^{210}Pb decay. In the lower section, a constant flux and constant sedimentation (CFCS) model (Sanchez-Cabeza and Ruiz-Fernández, 2012) was applied to the ^{210}Pb data to calculate the age-depth relationships. This age-depth model was applied to the gravity cores by aligning the radionuclide and Hg depth profiles of the respective cores.

5.3.4 XRF core scanning and radiographs

The X-ray Fluorescence (XRF) core scanning provides a non-destructive analytical approach to obtain a semi-quantitative, high-resolution down-core record of element concentration variations. Scanning was conducted at the Leibniz Institute for Baltic Sea Research Warnemünde (IOW) using an ITRAX XRF core scanner with a high-power chromium-XRF source. Before scanning, the uppermost sediment layer was removed to ensure an undisturbed sediment surface. All core sections were scanned with a resolution of 1 mm, 30 kV source voltage, a current of 55 mA and 5 s measurement time per step. Element intensities are reported as counts per second (cps) and cover elements from aluminium to barium. The data for the sediment surface of core GC7, which exhibits exceptionally high element ratios in the uppermost 3 cm and were excluded from further interpretations. Calibration was not needed due to the homogeneity of the physical properties of the cores such as porosity and textural variations including grain size (Löwemark et al., 2011; Bönning et al., 2007).

Radiographic images were obtained by using 60 kV, a current of 30 mA and 1,300 ms measuring time. The radiographs are represented as grey-scale images, the changes of which reflect variations in density and composition of the sediment. Lighter colour mark higher X-ray intensities indicating more compact material (i.e. high clay content), whereas darker intervals reflect a higher attenuation of X-rays with a lower density (i.e. higher silt or sand content).

5.4 Results

5.4.1 Sedimentological data

All gravity cores are composed of fine-grained sediments dominated by high mud ($< 63 \mu\text{m}$) content (Fig. S1) with average mean grain-sizes ranging between 7.58 and 11.99 μm (Tab. 5.1). GC7 and GC9 display slightly coarser grain-sizes than GC4 and GC5 (Tab. 5.1, Fig. 5.2, 5.3). The maximum sand contents for GC7 and GC9 are 15.1% and 13.4%, respectively, whereas GC4 and GC5 show significantly lower sand abundances of 4.1% and 4.6% (Fig. S1). The grain-size distribution of GC7 shows the highest variations between values of 7.29 and 15.19 μm (Fig. 5.2). Overall, the gravity core data show a fining-upward trend, starting at depths between 70 and 90 cm (Fig. 5.2, 5.3).

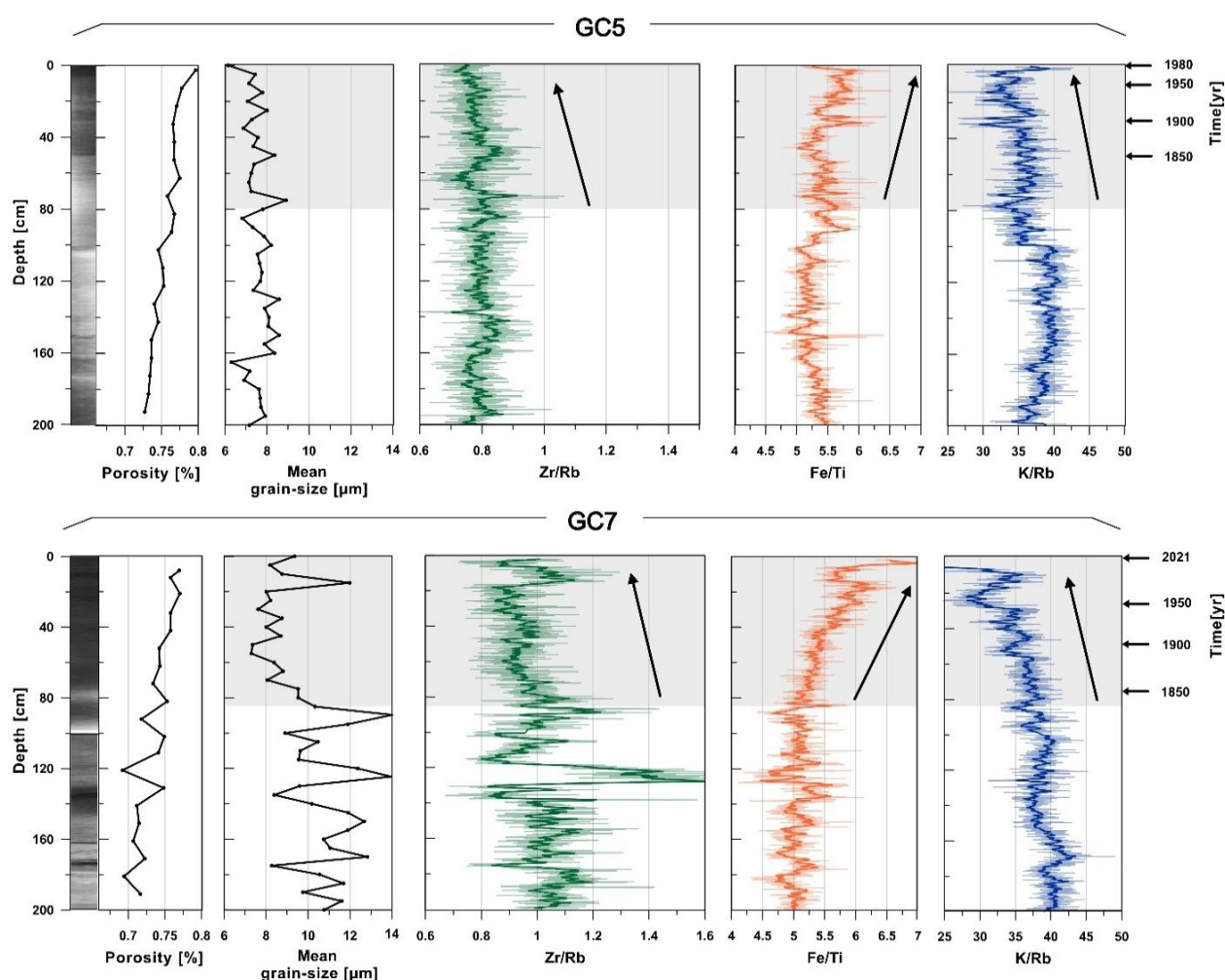


Fig. 5.2: X-ray radiographs, porosity, grain sizes and selected element ratios (Zr/Rb, Fe/Ti, K/Rb) of cores GC5 and GC7 located in the central part of the Skagerrak. Element ratios were calculated from XRF core scanning data. The bold lines of XRF ratios mark the 11-year running mean. Sediment ages are calculated based on data of Spiegel et al. (in review).

The radiographs reveal subtle changes in sediment composition with a darker grayscale intensity at core depths with finer grain-sizes. In contrast, sediment intervals with coarser sediments appear lighter on the X-ray radiographs. The uppermost part of Cores GC5, GC7 and GC9 is composed of radiographically more clayey material (darker). Porosity exhibits a consistent decrease with depth, accompanied by relatively minor variations throughout the sediment successions (Fig. 5.2, 5.3).

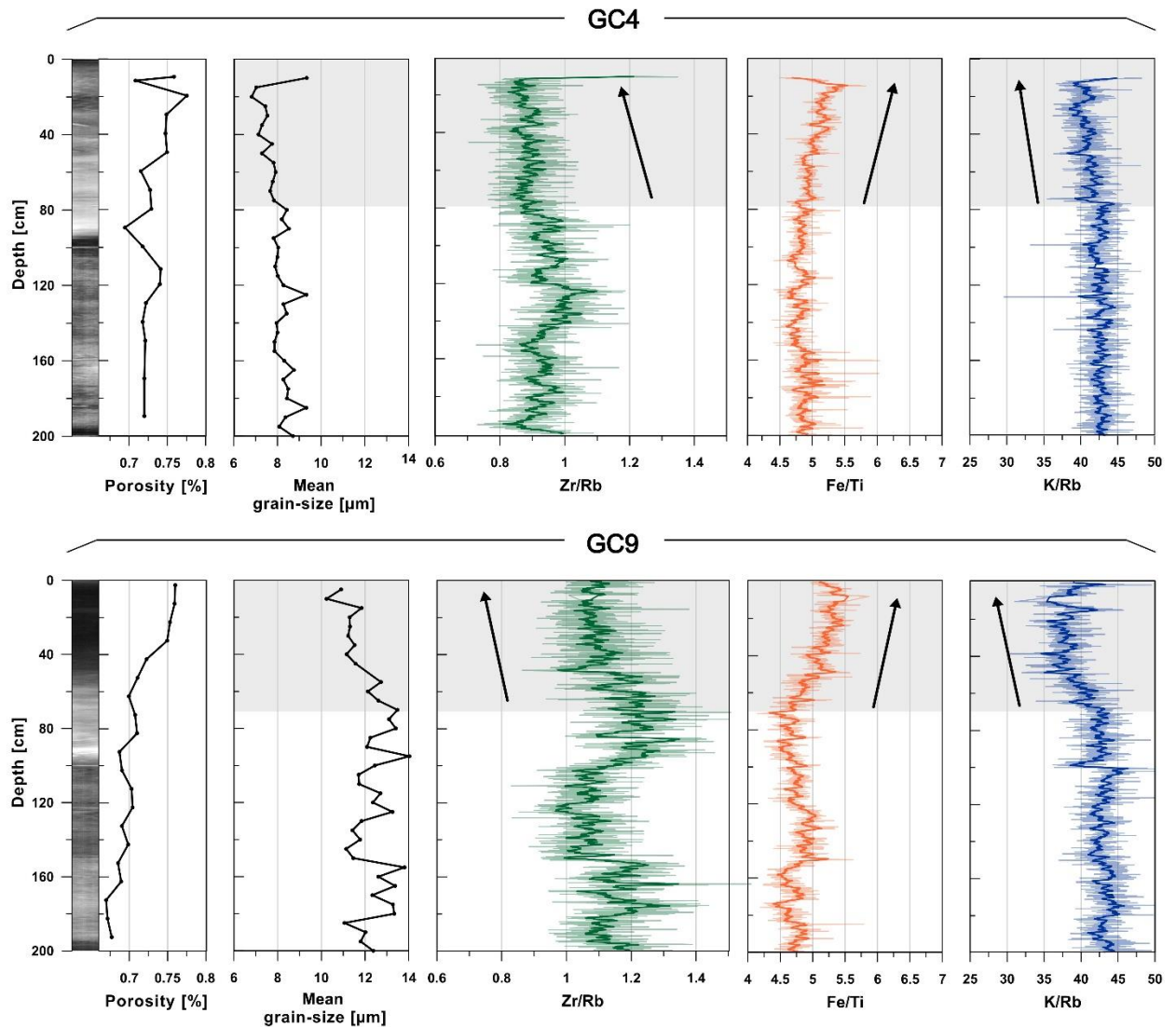


Fig. 5.3: X-ray radiographs, porosity, grain-sizes and selected element ratios (Zr/Rb, Fe/Ti, K/Rb) of cores GC4 and GC9 located in northeastern part of the Skagerrak (GC4) and the Kattegat (GC9). Element ratios were calculated from XRF core scanning data. Bold lines of XRF ratios mark the 11-year running mean.

5.4.2 Radionuclide and Hg data

The activity and concentration profiles of $^{210}\text{Pb}_{\text{ex}}$, along with the time markers ^{134}Cs and Hg of gravity core GC7, coincide with those of MUC8 when the topmost 3 cm of the gravity core are adjusted upwards (Fig. 5.4). Similarly, the profiles of GC5 correspond to those of MUC2 when shifted 10 cm downwards (Fig. 5.4). Since the gravity cores reveal the same $^{210}\text{Pb}_{\text{ex}}$ and ^{134}Cs activities as well as Hg concentrations as the multi cores collected at same core locations, the age models for MUC2 and MUC8, developed by Spiegel et al. (in review), were applied to GC5 and GC7 with the appropriate depth corrections. In contrast, GC4 and GC9 display evidence of sediment disturbances as indicated by fluctuations in the $^{210}\text{Pb}_{\text{ex}}$ down-core activities (Fig. S5.2), and do not allow the construction of a precise age-depth model.

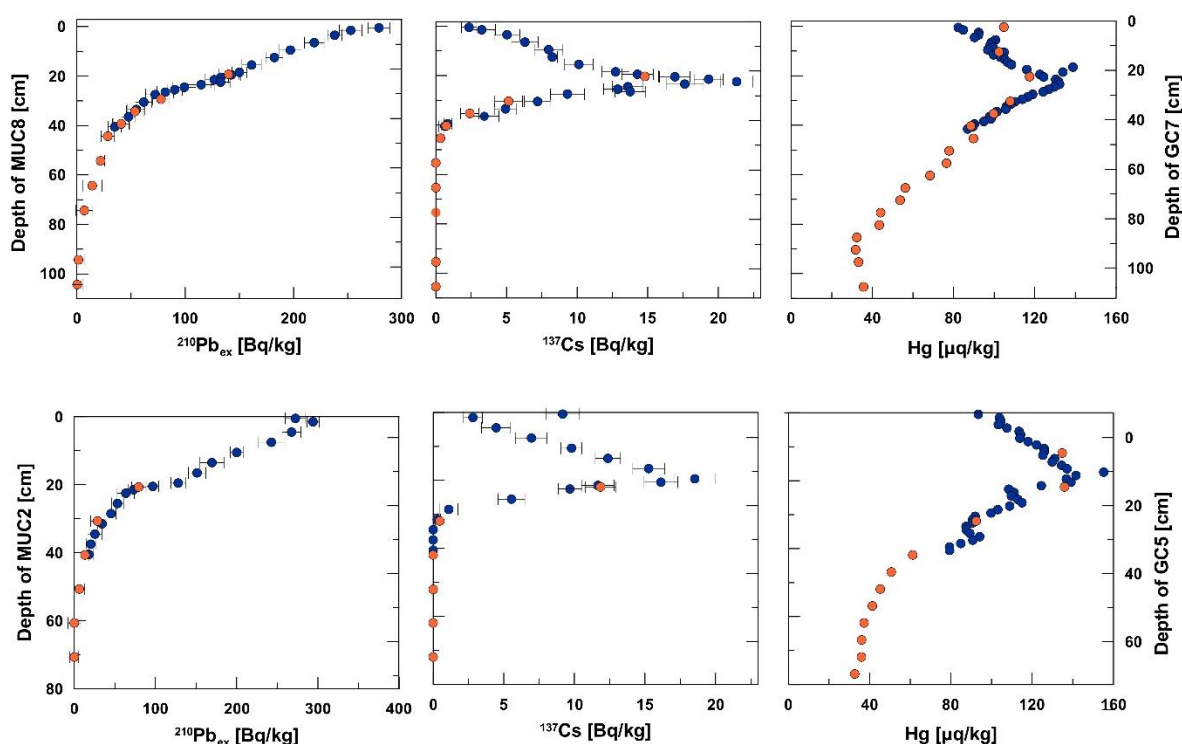


Fig. 5.4: Depth profiles of $^{210}\text{Pb}_{\text{ex}}$, ^{134}Cs and Hg. Blue dots indicate the profiles from MUC cores, while red dots represent those from GC cores. Radionuclide activity and Hg concentrations of the MUCs are from Spiegel et al. (in review).

5.4.3 XRF data

5.4.3.1 Depth-based data

Specific XRF ratio profiles were selected to investigate variations in hydrodynamic conditions and sediment provenance. The chemical compositions of the cores are highly variable at the

millimeter scale (Fig. 5.2, 5.3). The Zr/Rb ratios of the gravity cores visually correlate with the mean grain-size exhibiting a positive statistical relationship (GC5: $r = 0.74$; $p < 0.05$; GC7: $r = 0.78$; $p < 0.05$; GC4: $r = 0.74$; $p < 0.05$; GC9: $r = 0.61$; $p < 0.05$). The Zr/Rb ratio in the upper sections of all cores exhibits a long-term trend of upwards decreasing values, starting from a sediment depth of approximately 70 to 90 cm in the respective cores and continuing towards the surface sediments (Fig. 5.2, 5.3). Overall, these ratios are notably higher in GC7 and GC9 than in the other cores. Peaking Zr/Rb values are primarily observed in the lower portions of the cores. The K/Rb ratio shows similar long-term trends with depth as shown by Zr/Rb (Fig. 5.2, 5.3). In contrast, the Fe/Ti ratio behaves exactly opposite to those ratios (Fig. 5.2, 5.3). In general, the Fe/Ti values are consistently higher throughout the entire cores GC5 and GC7 than the other cores. In contrast, K/Rb values are higher in GC4 and GC9 than in GC5 and GC7.

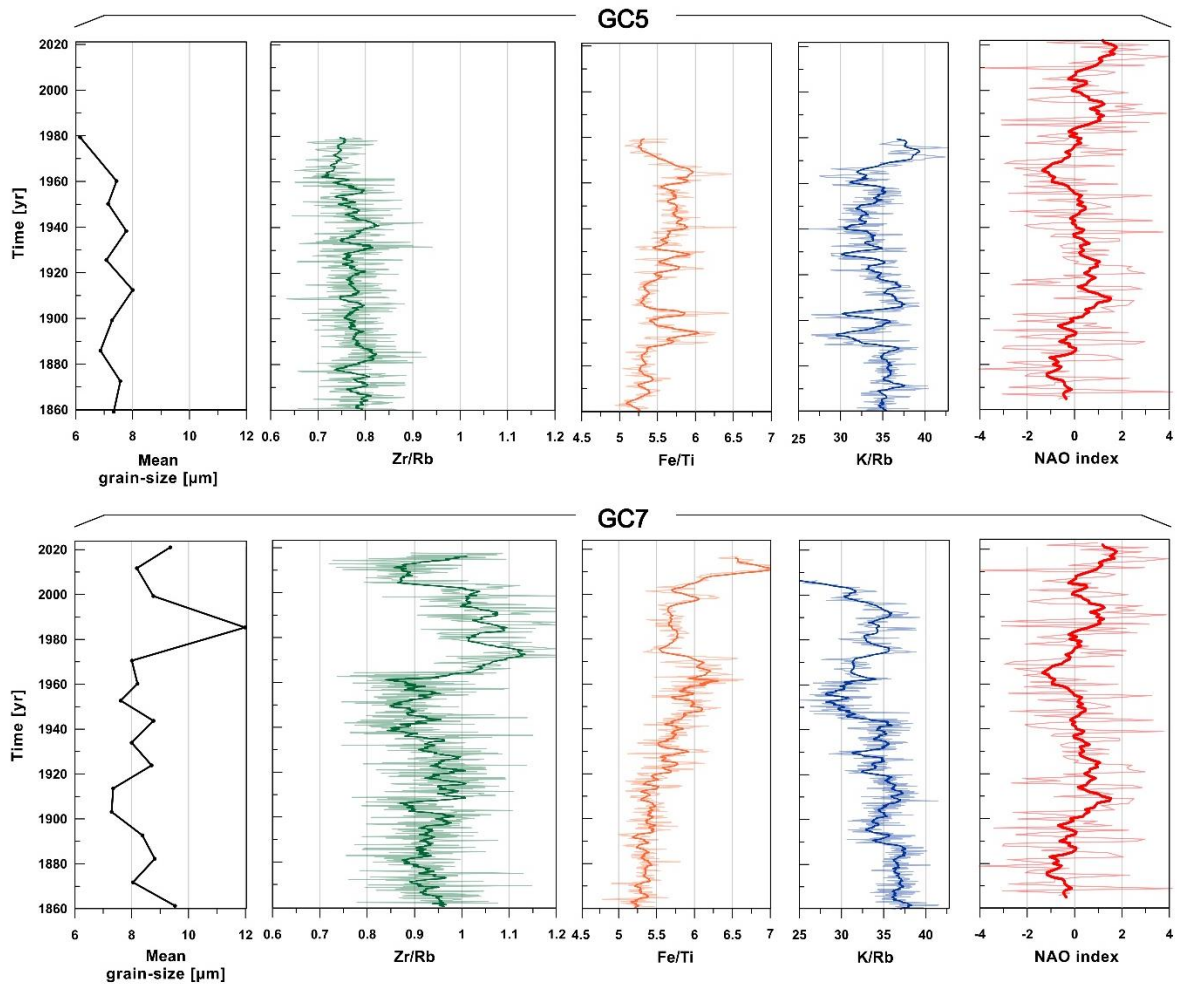


Fig. 5.5: Grain-size data and XRF ratios (Zr/Rb, Fe/Ti, K/Rb) compared with NAO index (data are from Hurrell et al., 2023). The bold lines of XRF ratios and NAO index mark the 11-year running mean. Sediment ages are calculated based on the data of Spiegel et al. (in review).

5.4.3.2 Age-based data

Figure 5 show the temporal variations in the respective XRF ratios for cores GC5 and GC7 over the past 160 years, with the age model for GC5 being constrained to data up to 1980. From 1860 to the 1960s, the Zr/Rb and K/Rb ratios display a consistent decreasing trend, while Fe/Ti values show a gradual increase over the same time period. Around the 1960s, an abrupt change in the chemical composition is observed. Zr/Rb (only notable in core GC7) and K/Rb ratios increase sharply, while Fe/Ti values decline. In GC7, this transition is particularly pronounced, with Zr/Rb and K/Rb ratios reaching their highest values and Fe/Ti their lowest in the 1980s during the second half of the 20th century. By approximately 2000, the geochemical ratios return to levels comparable to those observed in the 1960s. Additionally, there are notable significant correlations between the XRF ratios and the NAO index. The Zr/Rb and K/Rb ratios demonstrate a positive correlation with the NAO index, whereas the Fe/Ti ratios exhibit an inverse relationship.

5.5 Discussion

5.5.1 Shift in hydrodynamic conditions from colder to warmer periods

The strong correlation between the mean-grain size distribution and the Zr/Rb ratio observed in the gravity cores (Fig. 5.2, 5.3), demonstrates that this ratio serves as a reliable high-resolution proxy for grain-size variability. The granulometric characteristics of sediment deposits are primarily determined by hydrodynamic energy, which controls the erosion, transport, and deposition processes of particles. Higher current velocities enable the mobilization and deposition of coarser bedload (coarse silt and sand fraction), while lower energy conditions favour the sedimentation of fine-grained particles (Hjulstrom, 1935; Engelhardt, 1977). The Skagerrak sediment deposits are predominantly composed of silt (Fig. S5.1), reflecting the generally low-energy conditions and weak bottom currents within the basin (Rodhe, 1987), where suspension load preferably settles down. The significant upward-decline in the Zr/Rb ratios observed in the upper halves of the cores (Fig. 5.2, 5.3) indicates a decrease in bottom current intensity in the Skagerrak region since around 1850 AD, as seen in cores GC5 and GC7 (Fig. 5.2). Bottom current variability within the basin is strongly influenced by meteorological factors, such as wind strength and direction (Davies, 1981; Aure and Sætre, 1981). Intense storms with dominant westerly winds enhance the counter-clockwise circulation in the Skagerrak by driving the inflow of Atlantic Water and the Jutland Current into the basin,

resulting in increased bottom current velocities (Hognestad, 1987; Fonselius, 1989). These dynamics are reflected in the coarser sediments found in the lower sections of the cores, indicative of enhanced storm activity during these periods. Hass (1993, 1996) corroborated this relationship by demonstrating that coarser sediments in the Skagerrak primarily represent winter deposits, coinciding with intensified bottom currents. The LIA, which lasted approximately between 1250 and 1860 AD, was characterized by frequent cold winters across northern Europe and an increase in both the frequency and intensity of westerly storms (Lamb, 1979, 1984). As a result, the coarser sediments in the lower core sections reflect the more hydrodynamic winter conditions of this period, with the fining-upward trend marking the transition to the warmer and calmer climate of the Modern Climate Optimum.

The comparatively higher grain-sizes and elevated Zr/Rb ratios observed in cores GC7 and GC9 (Fig. 5.2, 5.3) suggest slightly more dynamic conditions with stronger bottom currents at the southern slope of the Skagerrak and in the central part of the Kattegat (Fig. 5.1), which is also supported by the higher sand contents recorded in these cores (Fig. S5.1). These cores also exhibit pronounced fluctuations in the Zr/Rb ratios, with peak values preceding the transition from the stormier conditions of the LIA to calmer modern conditions. One of the most turbulent periods of the LIA occurred between 1703 and 1860 AD (Lamb, 1991) and likely corresponds to these peaks reflecting maximum storm activity. This suggests that sedimentation in areas with stronger bottom currents is more sensitive to climatic fluctuations than in locations with weaker bottom currents.

These findings highlight the influence of climate variability on the bottom current circulation system in the Skagerrak region. Colder phases with frequent westerly storms promote the deposition of coarser sediments, whereas warmer phases with calmer conditions favour the accumulation of fine-grained particles. Similar hydrographic changes in the Skagerrak likely occurred across the broader North Sea, may also affecting the transport processes in the region and influencing the relative contributions of various sources of the sediments deposited in the Skagerrak.

5.5.2 Shift in sediment provenance from colder to warmer periods

The geochemical shifts observed in the Fe/Ti and K/Rb ratio in the transition zone from the LIA to modern times (Fig. 5.2, 5.3) indicate changes in the chemical composition of the sediments deposited in the Skagerrak. In contrast to the high variability of geology of central

Europe, the Scandinavian bedrock of the Fennoscandian Shield is predominantly composed of crystalline rocks such as granites and gneisses, which contain high amounts of potassium-bearing minerals like potassium feldspar and mica (Gaál and Gorbatshev, 1987; Gorbatshev and Bogdanova, 1993). Additionally, sediments originating from Scandinavia exhibit high contents of the clay mineral illite (Irion and Zöllmer, 1999), with deposits in the Skagerrak providing evidence of contributions from Scandinavian fjords (Lenz et al., 2024). This is reflected in the higher K/Rb ratios observed in Skagerrak cores GC4 and GC9, which are located near the Scandinavian coastline (Fig. 5.1). These cores are likely influenced by higher sediment input from Scandinavian rivers, which discharge potassium-enriched materials into coastal areas. Based on these findings, the general declining K/Rb trend in the Skagerrak cores over time (Fig. 5.2, 5.3) suggests a reduction in the contribution of potassium-rich sediments from Scandinavia. This trend may be linked to accelerated current velocities during colder climate periods, which enhanced coastal erosion and the transport of coarser silt and sand fractions to the central part of the basin. Concurrently, the increasing Fe/Ti ratio (Fig. 5.2, 5.3) indicates a growing fraction of sediment transport from central Europe. Unlike Scandinavia, the sedimentary geology of central Europe contains significant amounts of iron-rich sediments, especially derived from volcanic rocks abundant in hematite, goethite, and magnetite (Peine et al., 2000).

Zuther et al. (2000) demonstrated that K/Rb ratios increase from nearshore areas towards the open sea in the southern North Sea, attributed to higher concentrations of illite and K-feldspar in reworked marine sediments. These sediments are primarily derived from glacial erosion of the Fennoscandian Shield (Schwedhelm, 1984; Irion et al., 1987; Zöllmer and Irion, 1993), whereas the deposited nearshore sediments are rather supplied by Mid-European rivers. It is suspicious that the Fladen Ground sediments, which represent the predominant source for fine-grained Skagerrak deposits, also display reworked Pleistocene deposits (Jansen et al., 1979). Consequently, the elevated K/Rb ratios in the lower Skagerrak core sections, corresponding to colder and stormier LIA conditions, likely indicate increased sediment input not only from Scandinavia but also from the northern North Sea. The inflow of Atlantic Water into the Skagerrak from the northern North Sea has been most frequent during winter conditions, when strong westerly winds prevail (Ljøen, 1981). During the LIA, it is likely that the winter climate intensified seabed erosion in the northern North Sea, providing the Skagerrak with higher proportions of Fladen Ground sediments.

These findings imply that climate variability significantly influences the sediment provenance in the Skagerrak by controlling the current velocity of the North Sea and Skagerrak circulation systems. During colder climatic periods, such as those experienced during the LIA, intensified bottom currents have amplified sediment transport from the northern North Sea and have accelerated coastal erosion along the Scandinavian coastline. This process increases the proportion of northern-derived sediments, including materials eroded from the Scandinavian coast and from Pleistocene deposits in the northern North Sea, within the Skagerrak basin. Conversely, during periods of calmer climatic conditions, reduced current velocities hampered the transport of these sediments, resulting in higher contributions of sediment from the southern North Sea, primarily sourced from central Europe. These climate-driven shifts in sediment provenance underscore the profound influence of climate on the sediment dynamics across the entire North Sea region and highlight the crucial role of seabed and coastal erosion in this sedimentary system.

5.5.3 Impacts of climate variability on Skagerrak sedimentation during the last century

The shifts in sedimentary Zr/Rb and Fe/Ti, as well as K/Rb ratios during the 20th century recorded decadal changes in the hydrodynamic conditions and the sediment provenance in the Skagerrak (Fig. 5.5). In the second half of the 20th century, starting around the 1960s, the Skagerrak core locations have experienced more energetic hydrodynamic conditions, as indicated by peak Zr/Rb values. These conditions likely facilitated the deposition of coarser sediments originating from Scandinavian fjords, along with fine-grained material, potentially derived from intensified erosion of fine-grained deposits in the northern North Sea. Furthermore, the decline in the Fe/Ti ratio indicates a reduction of sediment supply from the southern North Sea. This observation aligns with findings by Lenz et al. (in review), who demonstrated that during this period of time, the increased trapping efficiency of the Wadden Sea, caused by elevated storminess, led to diminished sediment input from the southern North Sea, while intensified bottom trawling activity and stronger Atlantic inflow of currents at the Fladen Ground contributed to an enhanced sediment supply from this region to the Skagerrak. The key driving mechanisms of the decadal climate variability in the 20th century in northern Europe have been linked to the NAO, which has been shown to influence variations in the circulation pattern of the North Sea (Winter and Johannessen, 2006). Strong westerly winds are predominantly associated with positive NAO phases (Hurrell, 1995; Alexandersson et al., 2000;

Dangendorf et al., 2012). This is evident in the high positive NAO values during the 1980s and 1990s, coinciding with the deposition of coarser-grained sediments and an increased contribution from the northern North Sea and Scandinavian fjords (Fig. 5.5). This emphasizes that the sedimentary processes in the North Sea are impacted by a natural regional variability on multidecadal time scales as a response to forcing mechanisms originating from the North Atlantic. Over the last century, significant human interventions, including river management and increased fishing efforts, have expanded, coinciding with climatic effects (Murawski et al., 2005; Halpern et al., 2008; Rovira et al., 2014). For instance, bottom trawling activity and strengthened Atlantic currents in the Fladen Ground region led to similar outcomes, such as intensified seabed erosion, both peaking in the 1980s and 1990s (Winther and Johannessen, 2006; Couce et al., 2020). These superimposed influences of natural and anthropogenic activities create complex patterns of spatial and temporal variability of various processes of the North Sea system (Cappuzo et al., 2015; Emeis et al., 2015) and complicate precise assessments of individual contributions to sediment dynamics. The visual correlation between the NAO index and hydrodynamic and provenance proxies, such as Zr/Rb and K/Rb ratios, along with the negative correlation with Fe/Ti ratios in cores GC5 and GC7 during the 20th century (Fig. 5.5), suggests that climate variability, rather than anthropogenic impacts such as bottom trawling, predominantly controls energy conditions and sediment provenance. This conclusion is further supported by evidence from the LIA, when sedimentation patterns reflected past climate-driven hydrodynamics in essential absence of human activities.

Rising storminess in recent decades (Beniston et al., 2007) will intensify seabed and coastal erosion, causing intense redistribution of sediments across the North Sea. Future detailed analysis of sediment cores collected at different locations of the North Sea, such as the Wadden Sea, Norwegian Trench, and Fladen Ground, will provide a more comprehensive understanding of how the local and regional sedimentation is affected in different energetic settings.

5.6 Conclusions

The analysis of XRF core scanning data from Skagerrak sediment cores provides valuable insights into the impact of climate variability on its sediment dynamics and depositional history. Climate fluctuations have affected the provenance of Skagerrak sediments by controlling the current velocity and the dynamics of the circulation system. During colder periods like the LIA, frequent and intense westerly storms strengthened the Atlantic inflow into the northern North Sea and the Skagerrak basin, resulting in more energetic hydrodynamic conditions. Increased

bottom current velocities, reflected in coarser Skagerrak sediments with elevated Zr/Rb ratios, lead to intensified coastal erosion along the Scandinavian coastline and increased transport of sediments of Scandinavian origin into the central part of the Skagerrak. Furthermore, stronger Atlantic inflows into the northern North Sea amplified seabed erosion of Fladen Ground sediments, predominantly composed of reworked Pleistocene material derived from glacial erosion of Scandinavian sources, which resulted in elevated supply of material from that area to the Skagerrak region. The increased contributions of Scandinavian and Fladen Ground sediments are evident in higher K/Rb ratios in core sections representing colder climatic phases. These sediments are enriched in potassium-bearing minerals, which are characteristic of Scandinavian rocks. In contrast, calmer, similar to modern conditions favour increased sediment contributions from the southern North Sea due to lower energy conditions. We also show that the sediment dynamics are more strongly influenced by climate variability than by anthropogenic activities given that the NAO index exhibits a clear correlation with fluctuations in environmental conditions and changes in the sources of sediment contributions. These mechanisms illustrate the interplay between climatic conditions, hydrodynamic processes, and sediment provenance and highlight the important role of seabed and coastal erosion in the sediment contributions to the Skagerrak.

5.7 Acknowledgements

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5.8 Supplement

5.8.1 Supplementary Figures

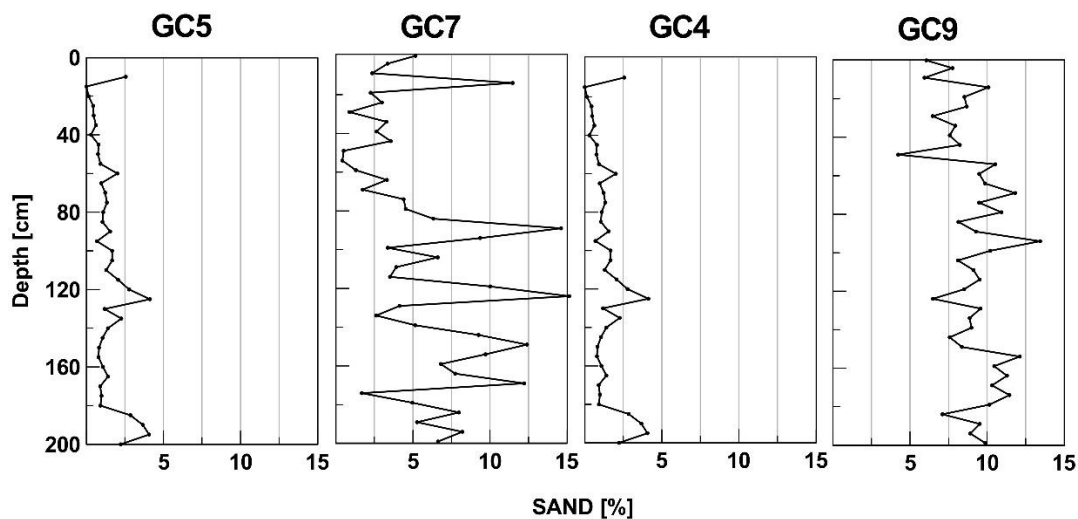


Fig. S5.1: Sand content of gravity cores.

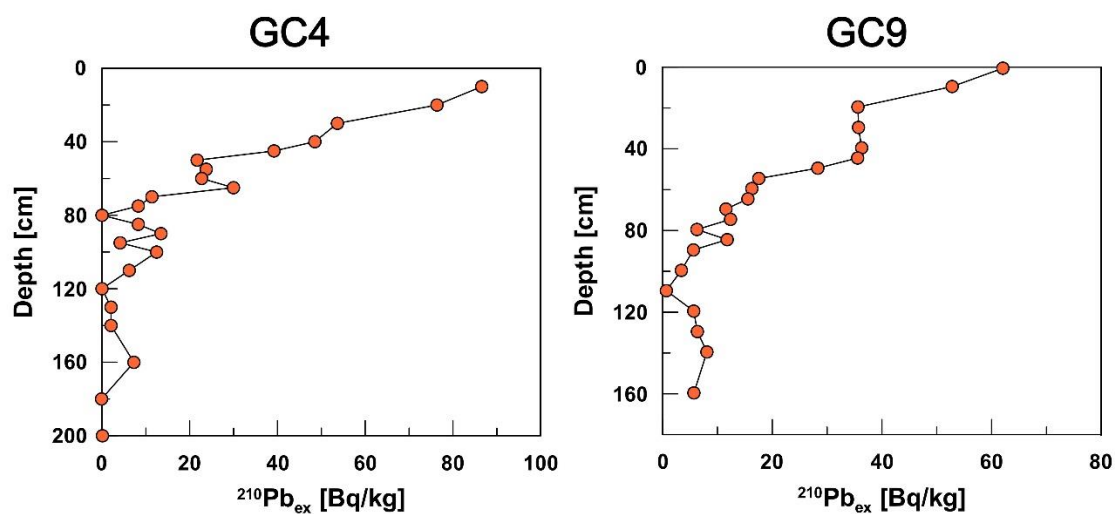


Fig. S5.2: Depth profiles of $^{210}\text{Pb}_{\text{ex}}$ of the gravity cores GC4 and GC9.

5.8.2 Supplementary Tables

Tab. S5.1: Grain size parameters of GC4 and GC5

GC4				GC5			
Deph [cm]	mean [μm]	sand [%]	mud [%]	Deph [cm]	mean [μm]	sand [%]	mud [%]
10	9.34	2.6%	97.4%	0	6.15	0.1%	99.9%
15	7.03	0.0%	100.0%	4	7.44	2.8%	97.1%

20	6.83	0.1%	99.8%	9	7.14	0.9%	99.1%
25	7.45	0.4%	99.5%	14	7.78	1.7%	98.1%
30	7.54	0.5%	99.5%	19	7.07	0.4%	99.5%
35	7.29	0.6%	99.4%	24	8.00	1.9%	97.8%
40	7.14	0.3%	99.7%	29	7.28	0.4%	99.5%
45	7.75	0.8%	99.2%	34	6.87	0.2%	99.8%
50	7.29	0.8%	99.2%	39	7.57	0.4%	99.6%
55	7.83	0.9%	99.1%	44	7.34	0.1%	99.9%
60	7.92	2.0%	98.0%	49	8.36	2.0%	98.0%
65	7.79	1.0%	99.0%	54	7.38	0.8%	99.2%
70	7.67	1.2%	98.8%	59	7.25	0.8%	99.1%
75	7.84	1.3%	98.7%	64	7.13	0.4%	99.6%
80	8.43	1.1%	98.9%	69	7.24	0.4%	99.6%
85	8.20	1.0%	98.9%	74	8.91	1.7%	98.3%
90	8.53	1.5%	98.5%	79	7.79	0.7%	99.3%
95	7.83	0.7%	99.3%	84	6.81	0.2%	99.7%
100	8.04	1.7%	98.3%	89	7.32	0.3%	99.7%
105	8.00	1.7%	98.2%	94	7.86	0.6%	99.3%
110	7.90	1.3%	98.7%	99	8.20	1.3%	98.6%
115	8.02	2.1%	97.9%	104	7.54	0.7%	99.1%
120	8.28	2.8%	97.2%	109	7.65	0.7%	99.2%
125	9.33	4.1%	95.9%	114	7.76	0.9%	99.1%
130	8.28	1.2%	98.8%	119	7.68	0.9%	98.9%
135	8.43	2.3%	97.7%	124	7.35	0.6%	99.3%
140	7.96	1.4%	98.6%	129	8.58	4.6%	95.4%
145	8.01	1.1%	98.9%	134	7.88	1.8%	98.2%
150	7.87	0.8%	99.1%	139	8.09	2.0%	97.9%
155	7.87	0.8%	99.2%	144	8.06	1.1%	98.7%
160	8.31	1.1%	98.8%	149	8.58	1.2%	98.8%
165	8.75	1.4%	98.4%	154	7.87	0.7%	99.3%
170	8.27	0.9%	98.8%	159	8.36	1.1%	98.9%
175	8.49	1.0%	99.0%	164	6.29	0.1%	99.9%
180	8.44	0.9%	99.1%	169	7.17	0.3%	99.7%
185	9.32	2.9%	97.1%	174	6.91	0.0%	99.9%
190	8.37	3.7%	96.3%	179	7.61	0.4%	99.3%
195	8.08	4.1%	95.9%	184	7.67	0.4%	99.6%
200	8.72	2.2%	97.8%	189	7.71	0.5%	99.3%
				194	7.91	0.6%	99.2%
				199	7.15	0.2%	99.7%

Tab. S5.2: Grain size parameters of GC7 and GC9

GC7				GC9			
Depth [cm]	mean [μm]	sand [%]	mud [%]	Depth [cm]	mean [μm]	sand [%]	mud [%]
0	9.35	5.2%	94.5%	0	9.37	6.1%	93.9%
4	8.18	3.4%	96.6%	4	10.91	7.8%	92.2%
9	8.76	2.4%	97.6%	9	10.24	5.9%	94.1%

14	11.98	11.5%	88.5%	14	11.84	10.1%	89.9%
19	8.01	2.3%	97.7%	19	11.29	8.5%	91.5%
24	8.21	3.0%	97.0%	24	11.31	8.7%	91.3%
29	7.62	0.9%	99.1%	29	11.23	6.5%	93.5%
34	8.76	3.3%	96.7%	34	11.52	7.9%	92.1%
39	8.00	2.6%	97.4%	39	11.15	7.6%	92.4%
44	8.70	3.6%	96.4%	44	11.56	8.2%	91.8%
49	7.35	0.5%	99.4%	49	9.22	4.2%	95.8%
54	7.29	0.4%	99.6%	54	12.73	10.5%	89.5%
59	8.37	1.3%	98.6%	59	12.13	9.5%	90.5%
64	8.81	3.3%	96.4%	64	12.61	9.9%	90.1%
69	8.05	1.7%	98.3%	69	13.48	11.8%	88.2%
74	9.53	4.4%	95.4%	74	13.09	9.5%	90.4%
79	9.53	4.5%	95.2%	79	13.40	10.9%	89.0%
84	10.33	6.3%	93.6%	84	12.24	8.1%	91.9%
89	15.19	14.6%	85.2%	89	12.09	9.3%	90.7%
94	11.90	9.4%	90.5%	94	14.03	13.4%	86.6%
99	8.91	3.4%	96.5%	99	12.45	10.2%	89.8%
104	10.47	6.6%	93.3%	104	11.71	8.1%	91.9%
109	9.63	3.9%	95.6%	109	11.73	9.1%	90.8%
114	9.56	3.5%	96.5%	114	12.70	9.5%	90.5%
119	12.37	10.0%	90.0%	119	12.36	8.5%	91.5%
124	14.39	15.1%	84.9%	124	13.25	6.5%	86.3%
129	9.59	4.1%	95.9%	129	11.84	9.6%	90.4%
134	8.39	2.6%	97.4%	134	11.42	8.9%	91.1%
139	10.18	5.2%	94.7%	139	11.78	9.0%	91.0%
144	11.91	9.3%	90.5%	144	11.14	7.6%	92.4%
149	12.68	12.4%	87.5%	149	11.46	8.4%	91.6%
154	11.89	9.7%	90.2%	154	13.80	12.1%	87.9%
159	10.74	6.8%	93.2%	159	12.61	10.5%	89.5%
164	11.06	7.7%	92.1%	164	13.37	11.3%	88.7%
169	12.83	12.2%	87.7%	169	12.33	10.3%	89.7%
174	8.26	1.7%	98.3%	174	13.27	11.4%	88.6%
179	10.55	5.0%	95.0%	179	13.33	10.1%	89.9%
184	11.68	8.0%	92.0%	184	11.05	7.1%	92.9%
189	9.74	5.3%	94.7%	189	12.01	9.5%	90.5%
194	11.61	8.2%	91.6%	194	11.80	8.9%	91.1%
199	10.76	6.6%	93.2%	199	12.37	9.9%	90.1%

5.8.2.1 Digital supplement for XRF data

Tab. S5.3: XRF data for gravity core GC4 (File: GC4_XRF_data.xlsx)

Tab. S5.4: XRF data for gravity core GC5 (File: GC5_XRF_data.xlsx)

Tab. S5.5: XRF data for gravity core GC7 (File: GC7_XRF_data.xlsx)

Tab. S5.6: XRF data for gravity core GC9 (File: GC9_XRF_data.xlsx)

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6. Summary, Conclusions and Outlook

6.1 Summary and Conclusions

This thesis provides new insights into the sediment dynamics of the Skagerrak basin and the entire North Sea region by establishing the provenance of the Skagerrak sediments using radiogenic Sr, Nd, and Hf isotope compositions of detrital clays in combination with clay mineralogical analyses. In addition, time series of the radiogenic isotope signatures and XRF core scanning data from Skagerrak sediment cores reveal changes in North Sea sediment transport, provenance and the depositional history of Skagerrak sediments over the last century. These findings enable the identification of the key driving factors, including climate and human impacts, that have shaped transport dynamics and have influenced the contributions of various sediment sources.

The radiogenic isotope composition of clay-sized surface sediment samples collected in the entire North Sea region allows to distinguish between three main sediment contributors, which are the northern North Sea, the southern North Sea, and the Scandinavian fjords. The geochemical data indicate that seabed erosion at the the Fladen Ground predominantly provides the Skagerrak with clay-sized sediments and is controlled by the inflow of the Atlantic current into the northern North Sea. Conversely, local and mid-European rivers, as well as the Baltic Sea, only contribute minor proportions to Skagerrak deposits. Coastal erosion along the southern coast of England and seabed erosion within the English Channel are assumed to be the primary sources of suspended matter in the southern North Sea. Large amounts of this material are mainly deposited in the depocenters of the German Bight and partially trapped in the Wadden Sea region before reaching the Skagerrak via the Jutland Current. Mid-European rivers are shown to contribute only negligible amounts to the SPM budget of the southern North Sea and, consequently, to the Skagerrak. These sedimentary processes have mainly been influenced by climate variability and, to a lesser extent, by intensified human activities, resulting in changes of the proportions of the contributors to the Skagerrak basin over the last century. Radiogenic isotope signatures and XRF data reveal a decline in sediment supply from the southern North Sea during the second half of the 20th century driven by increased storm intensity and frequency in the 1980s and 1990s. This was associated with a positive NAO index, which enhanced hydrodynamic conditions in the North Sea. Strong westerly winds during this period intensified the Atlantic inflow and therefore enhanced the sediment supply from the northern North Sea to the Skagerrak. Additionally, elevated bottom current velocities within

the Skagerrak basin, reflected in the deposition of coarser sediments, intensified coastal erosion along the Scandinavian coastline thereby increasing the transport of Scandinavian-derived sediments into the central part of the Skagerrak. These stronger currents likely hampered the deposition of fine-grained sediments such as clay and fine silt in the central depocenter, potentially explaining the observed decline in sediment accumulation rates over the past 60 years. Energetic conditions in the North Sea also likely increased the trapping efficiency of the Wadden Sea, which resulted in a reduction of the sediment input from the southern North Sea to the Skagerrak. Similarly, intensified bottom trawling activities in the northern North Sea during the 1980s and 1990s enhanced seabed erosion in the Fladen Ground and further promoted sediment contributions from this region to the Skagerrak. Conversely, intensified dredging and deepening of large European rivers has not significantly affected the sediment supply to the Skagerrak depocenter, as indicated by the radiogenic isotope data.

These findings demonstrate that climate variability has predominantly controlled the provenance of Skagerrak sediments and the transport and deposition of fine-grained sediments in various parts of the North Sea via changes in current velocity and circulation dynamics. Coastal and seabed erosion have played a crucial role in the sediment budget of the entire North Sea. In contrast, human interventions did not strongly impact the sediment contributions from different source areas to the Skagerrak. Ultimately, this thesis helps to understand not only the factors controlling Skagerrak sedimentation but also the processes shaping North Sea sedimentary dynamics. These insights directly address the research questions outlined within the APOC project.

6.2 Outlook

This thesis provides detailed insights into the provenance of Skagerrak sediments and the main factors controlling its variability, which allow a better understanding of the main transport processes in the North Sea region. At the same time, significant unresolved research questions emerged that need to be answered and new challenges are introduced that require future work.

The reconstruction of the provenance of Skagerrak sediments in this study focuses exclusively on the detrital clay-sized fraction. This approach revealed that the Fladen Ground, an area characterized by clay-rich sediments within the northern North Sea, supplies clay material to the Skagerrak, where deposits are also clay-dominated (up to 60%). This explains discrepant findings of previous sediment budget studies, which were based on the fine fraction smaller

than 63 μm , encompassing both silt and clay fraction, and were interpreted in terms of predominant supply from the southern North Sea (Eisma and Irion, 1988; Zöllmer and Irion, 1993; Bengtsson and Stevens, 1998; Lepland et al., 2000). This discrepancy clearly indicates that clay and silt behave completely different during transport and depositional processes. Clay particles are often transported and deposited as flocs, with flocculation influenced by factors such as salinity, organic matter and hydrodynamic conditions (Tan et al., 2014). In contrast, silt particles are typically transported as individual grains and their deposition is affected by bottom current velocities. This distinction highlights the importance of separately analysing these sediment fractions to fully understand the transport dynamics of fine-grained sediments in the North Sea. Future studies should analyse radiogenic Nd, Sr, and Hf isotope signatures of the silt-sized fraction to evaluate its contributions from different sources to the Skagerrak under varying climatic conditions. Furthermore, it is essential to differentiate between fine silt (2 – 15.6 μm), transported as suspension load, and coarse silt (31 – 63 μm), likely transported as bedload via saltation (Moss, 1972). This approach would provide a comprehensive understanding of the transport and depositional processes of fine-grained sediments in the region.

This study revealed the temporal variability in sediment contributions to the Skagerrak from the three main sources: the northern North Sea, the southern North Sea, and Scandinavia. A reduction in sediment supply from the southern North Sea accompanied by increased inputs from the northern North Sea to the Skagerrak have been observed during the second half of the 20th century. This shift is attributed to enhanced trapping efficiency in the Wadden Sea and intensified erosional processes in the Fladen Ground region. However, the relative contributions of these processes to the observed changes in the Skagerrak sediment provenance remain unclear. Addressing this uncertainty requires collecting sediment cores from both the Wadden Sea and the Fladen Ground to establish comprehensive time series of geochemical and sedimentological data of these regions, which would allow to investigate the effects of regional changes. In addition, a granulometric down-core record would reveal changes in hydrodynamic conditions in response to climate variability in both regions. In the Fladen Ground, coarser sediments would for example suggest increased bottom currents due to enhanced Atlantic inflow promoted by strong westerly winds. Characterizing the deposits in this region could also help to differentiate the effects of natural versus anthropogenic erosional processes, as bottom trawling typically results in mixed sediment layers (De Borger et al., 2021). Such data would provide insights into the main factors driving seabed erosion in this region. New sediment records from Wadden Sea cores from various locations along the southern North Sea coast

would enable the reconstruction of accumulation rates, thereby helping to identify key depocenters where southern North Sea sediments are trapped before transported further into the Skagerrak region. Additional down-core analyses of radiogenic isotopes will allow distinguishing between sediments from the southern North Sea, originating from erosional processes within the English Channel, and sediments discharged by mid-European rivers. Postma (1981) and Oost et al. (2021) suggested that the fine-grained sediments originate from outside the tidal flats such as the English Channel and local rivers. This thesis provides findings how the radiogenic Hf compositions can be used to differentiate between these two sediment sources. Consequently, additional sediment records of the Wadden Sea would provide valuable insights into the trapping efficiency under changing climate.

In this thesis it is assumed that the observed decline in accumulation rates during the second half of the 20th century is caused by more hydrodynamic conditions, which hamper fine-grained sediments to settle down in the central part of the Skagerrak basin. However, it remains unclear whether the fine fraction, prevented from settling in the inner Skagerrak, is subsequently transported and deposited in the area of the Norwegian Trench. To clarify this and to determine where this fine-grained material ultimately accumulates, it will be crucial to investigate the temporal evolution of MARs along a transect across the Norwegian Trench.

The approach of reconstructing sediment provenance and transport pathways through radiogenic isotope compositions combined with clay mineral assemblages, as also demonstrated by Ehlert et al. (2011) in the eastern Indian Ocean for promising paleoreconstructions of sedimentary processes, can be effectively extended to other shelf seas with large sediment depocenters. These regions, characterized by significant fine-grained sediment depocenters that serve as efficient carbon sinks, provide valuable opportunities to evaluate whether climatic changes over the past century have influenced their sediment dynamics to a similar extent as observed in the North Sea. For instance, the Bering Sea, one of the largest marginal seas, is characterized by a dynamic sedimentary system influenced by strong bottom currents and frequent coastal and offshore storm surges (Mason et al., 1996). This region acts as a significant carbon sink, with a net annual sequestration of approximately 7.9 TgC yr⁻¹ (Zhang et al., 2023). Investigating temporal variations in sediment contributions to its depocenters, such as Bristol Bay, can provide valuable insights into climate variability and its influence on sediment dynamics over the past century. Such studies also enhance understanding of erosional processes and their impact on the reduced sequestration of particulate organic carbon in sediments.

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