



Neodymium isotope constraints on chemical weathering and past glacial activity in Svalbard

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ABSTRACT

Neodymium (Nd) isotopes in leached authigenic components of marine sediments have been increasingly used as a tracer of past ocean-water masses. Despite the general assumption that the Nd isotopic composition of solutes released during chemical weathering fingerprints the source rocks on continents, preferential dissolution of easily dissolvable phases may result in significant deviations in Nd isotopic composition between the solutes and the source rocks, with potential implications for the utility of Nd isotopes in paleoenvironmental studies. Here, we present the Nd isotopic compositions of leached and detrital fractions separated from bedrock and marine sediment samples from the Svalbard archipelago. Our goal is to further understand the behaviour of Nd isotopes during chemical weathering in glacial catchments and evaluate how glacier fluctuations and associated weathering congruency may have affected the export of dissolved Nd isotope signatures to seawater.

Our results confirm that terrestrial weathering on Svalbard causes considerable Nd isotopic decoupling between the leached and detrital fractions of fjord sediments ($\Delta\epsilon_{Nd}$), resulting from the preferential dissolution of marine precipitates in glaciated catchments dominated by sedimentary rocks. We also show that the degree of Nd isotopic decoupling has fluctuated in response to climate variability on Svalbard during the Holocene, which is also suggested by the occurrence of generally higher $\Delta\epsilon_{Nd}$ values during periods of glacier advances in sediment cores retrieved from two different fjords (Dicksonfjorden and Woodfjorden). We posit that the high $\Delta\epsilon_{Nd}$ values can be ascribed to incongruent chemical weathering of fresh rock flour produced by glacial abrasion. This finding suggests that the degree of Nd isotopic decoupling could be used as a new proxy for tracing glacial fluctuations and associated glacier-derived nutrient inputs to the marine realm.

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1. Introduction

Global warming is having a profound impact on marine and terrestrial environments, and its effects are particularly evident in polar regions, where glacier and sea-ice melting in combination with associated changes in surface albedo and ocean heat distribution are key mechanisms underlying polar amplification (Taylor

et al., 2013 and references therein). Glacier melting and accompanying glacial rock weathering have been recently identified as an important source of nanoparticulate nutrients available for primary production in the ocean (Hawkings et al., 2015; Wadham et al., 2019), and they potentially play an important role in marine biogeochemical cycles (review in Wadham et al., 2019). However, the impact of glacier melting on nutrient releases and marine productivity relies on sparse datasets (Hawkings et al., 2015). One important requirement for further evaluations of the future relationship among climate changes, glacial weathering (understood as chemical weathering in glacial catchments), glacier melting, and marine

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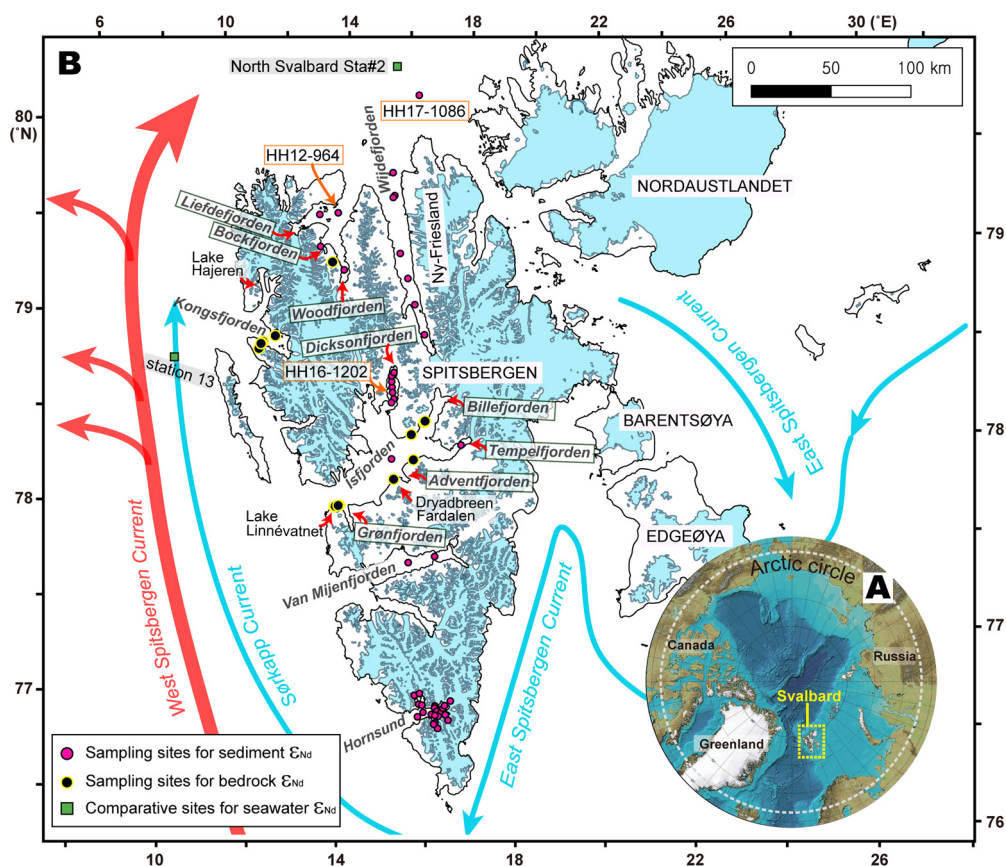


Fig. 1. (A) Map of the Arctic showing the location of the Svalbard archipelago. (B) Map of the Svalbard archipelago. Locations of bedrock and fjord-sediment samples as well as place names mentioned in the text are indicated. Sky blue land areas represent the glacier coverage at present. Red and sky blue arrows indicate the West Spitsbergen Current (WSC) and the Sørkapp Current (branch of the East Spitsbergen Current (ESC)), respectively. The comparative sites (North Svalbard Sta#2 and station 13) for the ϵ_{Nd} values of Atlantic Water and Arctic Water inflows carried by the WSC and the ESC, respectively, are also shown. See Table S1 for the coordinates of the sampling and comparative sites. (For interpretation of the colours in the figure(s), the reader is referred to the web version of this article.)

ecosystems is the reconstruction of past responses of glaciers to climate and subsequent changes in meltwater discharge.

Neodymium (Nd) isotopes (often expressed using the ϵ_{Nd} notation) are widely used for reconstructing past changes in ocean circulation and continental weathering fluxes (e.g., Bayon et al., 2002; Jang et al., 2017; Rutberg et al., 2000). A general consensus on the use of Nd isotopes in paleoenvironmental studies is that negligible decoupling occurs during continental weathering and sedimentary processes. However, in recent decades, multiple studies from river systems have shown that preferential dissolution of labile phases relative to more resistant minerals during weathering leads to markedly different Nd isotopic signatures in dissolved and solid loads (Goldstein and Jacobsen, 1987), especially in sub-Arctic regions and glacier-covered areas (Andersson et al., 2001; Hindshaw et al., 2018a; Süfke et al., 2019). This evidence for decoupling of Nd isotopes in the glacial environment suggests that past episodes of glacial retreats and advances could have possibly affected the ϵ_{Nd} signatures of the dissolved loads exported to the ocean over centennial to millennial time scales in high-latitude regions (Hindshaw et al., 2018a). For instance, an increased supply of fresh rock substrates by glacial abrasion could have led to enhanced incongruent weathering on continents (Vance et al., 2009), which would have resulted in greater Nd isotopic decoupling during past cold events because of the intrinsic Nd isotopic differences in rock-forming minerals. The importance of Nd isotopes in provenance studies and their utility for tracing past meltwater releases call for a better understanding of their behaviour during glacial weathering and further evaluation of how past glacial dynamics may have influenced the isotopic composition of dissolved Nd exported to the ocean.

In this study, we present an extensive set of Nd isotopic compositions measured in both labile and detrital fractions of a series of sedimentary and metamorphic rock samples and surface sediment samples from across the Svalbard archipelago. Our aim is to characterize the effect of incongruent silicate weathering on Nd isotopes. Two marine sediment cores from fjords covering the Holocene period were also analyzed to assess the impact of past glacial advances and retreats on Nd isotopic decoupling and the preservation of ϵ_{Nd} signatures by surrounding water masses. The Svalbard archipelago is largely covered by glaciers and has diverse bedrock compositions, including sedimentary, metamorphic and volcanic rocks (Dallmann and Elvevold, 2015). Thus, the area is well suited for investigating the behaviour of Nd isotopes during glacial weathering.

2. Background

2.1. Nd isotope behaviour during weathering

Easily dissolvable components in rocks and sediments, such as marine precipitates (e.g., bio-skeletal carbonates, phosphates and Fe-Mn oxides) and labile detrital minerals (e.g., volcanic components and accessory minerals) (Goldstein and Jacobsen, 1987; Hindshaw et al., 2018a), can be selectively extracted using chemical leaching methods (Bayon et al., 2002; Gutjahr et al., 2007). The comparison of radiogenic isotopic compositions between 'leachable' and residual silicate phases indicates whether isotopic decoupling may occur during continental weathering (e.g., Dausmann et al., 2019). A recent experimental investigation of Nd isotopes conducted on Svalbard (Dryadreen and Fardalen river basins; Fig. 1) clearly showed that bedrock leachates and river waters presented

more radiogenic Nd isotopic values (= higher ε_{Nd}) than the parent bedrocks (Hindshaw et al., 2018a), suggesting that the isotopic composition of dissolved Nd is controlled by the preferential dissolution of labile phases during weathering. While such decoupling is mostly observed in sedimentary catchments (Goldstein and Jacobson, 1987), where radiogenic labile components typically occur as marine precipitates (e.g., Hindshaw et al., 2018a), preferential dissolution of easily dissolvable accessory minerals such as apatite or titanite, can also result in decoupling in granitic catchments (e.g., Dausmann et al., 2019; Sűfke et al., 2019).

On continents, the extent or magnitude of incongruent weathering is considered to be climate-dependent (Vance et al., 2009). Enhanced incongruent weathering of glacially eroded rock substrates is generally thought to have occurred during past cold periods, and they most likely resulted in an intensified release of radiogenic Nd and Pb isotopes into dissolved river loads and the marine environment. This process has been inferred in part on the basis of pronounced radiogenic Pb isotope excursions in various late Quaternary sediment records of dispersed Fe-Mn oxyhydroxide phases from the North Atlantic (Gutjahr et al., 2009) and high Alpine lake sediments (Sűfke et al., 2019). Furthermore, Sűfke et al. (2019) documented a trend towards more radiogenic authigenic Nd isotopic values in an Alpine lake sediment record, suggesting that Nd isotopes could also record enhanced incongruent weathering of glacially derived material in the past. Similarly, long-term variations in Pb and Nd isotopes in North Atlantic ferromanganese crusts across the onset of Northern Hemisphere glaciations have also been attributed to changes from chemical- to physical-dominated weathering regimes on surrounding continents (Gutjahr et al., 2009; von Blanckenburg and Năgler, 2001).

2.2. Study area

This study is based on bedrock samples from multiple onshore locations as well as sediment samples from various fjords on Spitsbergen, the largest island of the Svalbard archipelago (Fig. 1). The Svalbard archipelago is located between approximately 74–81°N and 10–35°E and surrounded by the Barents Sea and the Greenland Sea (Fig. 1). Three main water masses contribute to the hydrography of Spitsbergen fjords: (1) relatively warm and saline Atlantic Water (AW; $T > 3.0$ and $S > 34.65$; Cottier et al., 2005 and references therein) carried by the West Spitsbergen Current (WSC; Fig. 1), (2) cold and relatively fresh Arctic water (ArW; $-1.5 < T < 1.0$ and $34.30 < S < 34.80$; Cottier et al., 2005 and references therein) by the Sørkapp Current, an extension of the East Spitsbergen Current (ESC; Fig. 1), and (3) glacial meltwater (Cottier et al., 2005). Their mixing varies depending on location within a fjord, water depth, and season of a year, resulting in complex hydrography containing various water masses such as Surface Water (SW), Local Water (LW), Intermediate Water (IW), Transformed Atlantic Water (TAW), and Winter Cooled Water (WCW) (e.g., Promińska et al., 2017; Zajăczkowski et al., 2010). A top-most layer in late spring and summer is occupied by fresh SW ($S < 34.00$), which is strongly affected by glacial meltwater. LW ($34.30 < S < 34.85$, $-0.5 < T < 1.0$) and WCW ($T < -0.5$, $34.4 < S < 35.00$) are formed by surface cooling during autumn and brine formation during winter, respectively, while TAW ($1.0 < T < 3.0$, $34.65 < S$) and IW ($34.00 < S < 34.65$, $T > 1.0$) are formed by mixing between AW and ArW, as well as various fjord masses (Cottier et al., 2005).

The bedrock geology of Svalbard in general as well as in the catchment areas of the studied fjords, is very diverse and ranges from Proterozoic metamorphic rocks to Phanerozoic sedimentary rocks (Dallmann and Elvevold, 2015) (Fig. 2). Hornsund is dominated by Proterozoic metamorphic rocks at the fjord mouth in the west to Palaeogene sedimentary rocks at the fjord head in the east. They display complicated structures, including folds, faults,

and overthrusts (Dallmann and Elvevold, 2015). Similarly, the Van Mijenfjorden and Isfjorden systems display the longitudinal (W-E) variability in bedrock geology from Proterozoic metamorphic rocks to Carboniferous to Palaeogene sedimentary rocks. The catchment area of Dicksonfjorden, a northern tributary of Isfjorden, is composed of sedimentary sequences, including Devonian Old Red Sandstones and siltstones and Carboniferous-Permian carbonate-siliciclastic strata. The catchment area of Woodfjorden primarily consists of the Devonian Old Red Sandstone with a minor Quaternary volcanic complex, and Mesoproterozoic metamorphic and igneous rocks located at the heads of its tributaries (Fig. 2). The catchment of Wijdefjorden is dominated by Proterozoic to lower Silurian metamorphic rocks along the eastern shore and Upper Silurian to Devonian sedimentary rocks in the west (Dallmann and Elvevold, 2015).

During the Holocene, Svalbard experienced several phases of glacier advances and retreats. The deglaciation of the inner fjords of west Spitsbergen terminated approximately 11.2 ka (Baeten et al., 2010; Forwick and Vorren, 2009), and a warming period in western Spitsbergen lasted until 9 to 8 ka (Forwick and Vorren, 2009; Rasmussen et al., 2012). Gradual cooling accentuated by several asynchronous cooling steps (e.g., Rasmussen et al., 2012) followed this period (Forwick and Vorren, 2009). In particular, the enhanced growth and/or reformation of glaciers (Fjeldskaar et al., 2018), such as Linnėbreen, Nordenskiöldbreen, Tunabreen and those in the Hajeren lake catchment occurred between 6 and 4 ka (Baeten et al., 2010; Forwick et al., 2010; Svendsen and Mangerud, 1997; van der Bilt et al., 2015). Most glaciers reached their maximum Holocene extents during the Little Ice Age (LIA) (~66 % with a total area of 38,871 km²; Martín-Moreno et al., 2017). Glaciers have been retreating since the termination of the LIA, and approximately 57% of Svalbard is currently covered with glaciers (total area of 33,775 km²; Martín-Moreno et al., 2017 and references therein).

The study focuses in particular on the analysis of two sediment cores from Dicksonfjorden and Woodfjorden. The sediment supply to Dicksonfjorden occurs mainly by glacio-fluvial runoff. On the other hand, the main sediment sources in Woodfjorden are glacio-fluvial rivers in inner Woodfjorden and Bockfjorden as well as tidewater glaciers in Liefdefjorden. At present, WCW forms by convection during the fall and winter seasons in both fjords. However, considering the known past climate and glacier variations in Svalbard during the Holocene (Baeten et al., 2010; Rasmussen et al., 2012; Svendsen and Mangerud, 1997), the oceanographic and sedimentary regime were different in the past. The relative sea level fell by up to 65 m on Spitsbergen during the last 12 ka (Forman et al., 2004). The most pronounced changes occurred at the end and shortly after the deglaciation, and only minor changes have occurred since the middle Holocene.

3. Materials and methods

3.1. Materials

3.1.1. Bedrock

A total of 18 fresh unweathered rock samples from representative bedrock units of Svalbard were collected (Isfjorden, $n = 10$; Kongsfjorden, $n = 7$; Woodfjorden, $n = 1$; Table 1 and Fig. 2). Metamorphic rocks (i.e., quartzite and phyllite) were collected from the upper Proterozoic Generalfjella Formation in the Kongsfjorden area, while sandstone samples were collected from Isfjorden, Billefjorden and Woodfjorden. The Woodfjorden sandstone corresponds to the Devonian Old Red Sandstone, which represents the main source of sediments in Dicksonfjorden and inner Woodfjorden. Carboniferous bedrock samples from the Kongsfjorden and Isfjorden

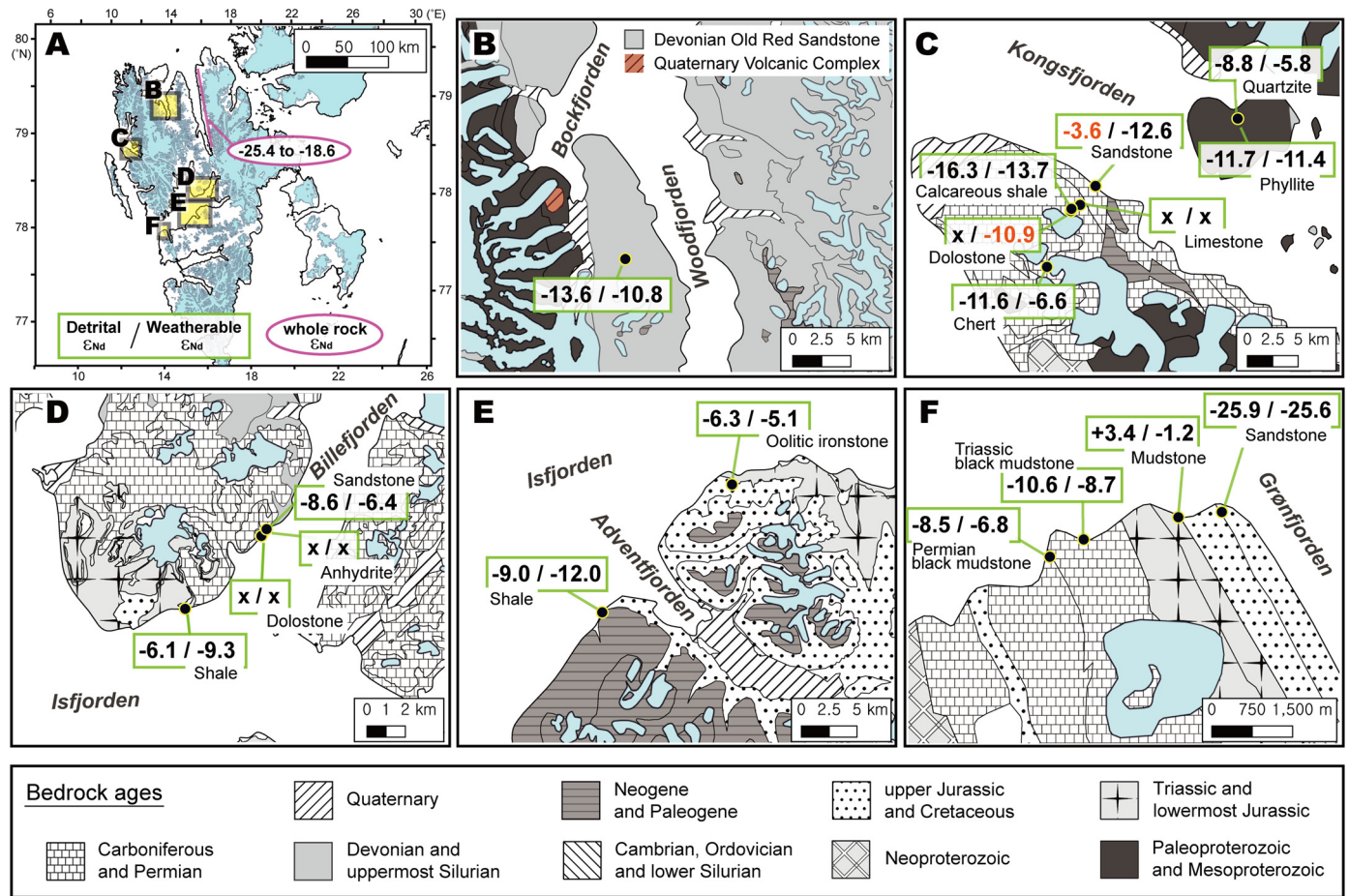


Fig. 2. (A) Map of Svalbard showing the sampling locations (B) to (F) for the bedrock. The ϵ_{Nd} values of detrital and weatherable components in the catchment areas of Woodfjorden (B), Kongsfjorden (C) and Isfjorden and its tributary Billefjorden (D, E, and F) are marked with green rectangles. Samples with too little Nd to obtain data are marked with an 'x'. ϵ_{Nd} values with high uncertainties (> 0.5 epsilon-unit) are in red numbers. The bedrock samples include sixteen sedimentary rocks and two metamorphic rocks. Various bedrock ages are marked with different symbols. For comparison, the ϵ_{Nd} value measured from whole rocks in Ny-Friesland (for location see Fig. 1) is also marked with pink ellipses in Fig. 2A (data from Johansson et al., 1995). The geologic map is modified after Dallmann and Elvevold (2015).

areas are mainly composed of calcareous sedimentary rocks (limestone and dolostone), while the Mesozoic bedrock samples from Isfjorden include siliciclastic mudstones, siltstones, and sandstones.

3.1.2. Surface sediments

A total of 45 surface (0–1 cm sediment depth) and sub-surface sediment samples (up to 5–6 cm sediment depth) (hereafter, surface sediments) from Bockfjorden ($n = 1$), Dicksonfjorden ($n = 8$), Hornsund ($n = 21$), Liefjefjorden ($n = 1$), Tempelfjorden ($n = 1$), Van Mijenfjorden ($n = 2$), Wijdefjorden ($n = 8$) and Woodfjorden ($n = 2$) (Fig. 3) were collected to characterize the modern distribution of sedimentary Nd isotopes in Spitsbergen fjords. The material was retrieved with a giant box corer ($50 \times 50 \times 50 \text{ cm}^3$) or a gravity corer during cruises of the *RV Helmer Hanssen* of UiT The Arctic University of Norway in 2012, 2015, 2016, and 2017 (Supplementary Table S1). The sampling sites are located at water depths ranging from 37 to 322 m, and they are in contact with the water masses mentioned earlier (section 2.2) depending on the fjord location, water depth and distance from the fjord mouth (e.g., Promińska et al., 2017; Zajączkowski et al., 2010).

3.1.3. Core sediments

Two gravity cores from Dicksonfjorden (HH16-1202-GC; 78.727°N , 15.310°E , ~ 87 m water depth) and Woodfjorden (HH12-964-GC; 79.651°N , 13.755°E , ~ 173 m water depth) were retrieved to reconstruct the evolution of sedimentary Nd isotopic compositions during the Holocene. Core HH16-1202-GC is ~ 274 cm long

and spans the last 8.2 ka (Joo et al., 2019), with an average sediment accumulation rate of ~ 33 cm/ka. The lithology is dominated by reddish-brown mud that includes varying amounts of sandy detrital clasts. Core HH12-964-GC is ~ 334 cm long and spans the last ~ 13.3 ka (Hansen, 2014). The average sediment accumulation rate is 25.1 cm/ka, although rates of up to 91 cm/ka were calculated for the lower parts of the core. The lithology of core HH12-964-GC is dominated by silt-sized sediment, with the occurrence of coarser intervals at ~ 110 cm and near the bottom of the core (Hansen, 2014). In each core, 1 cm thick sediment layers were collected at 10 cm intervals for the Nd isotope analyses ($n = 28$ for HH16-1202-GC and $n = 35$ for HH12-964-GC).

3.2. Analytical methods

3.2.1. Sample preparation

In this study, we define 'authigenic' and 'weatherable' components as the easily dissolvable fractions (e.g., carbonate, and Fe-Mn oxyhydroxide) extracted by chemical leaching from unwashed ground bulk sediments and from bedrock, respectively (Bayon et al., 2002; Gutjahr et al., 2007; Jang et al., 2017; Rutberg et al., 2000; Werner et al., 2014). The protocol used for chemical leaching is based on the addition of 0.02 M hydroxylamine hydrochloride in 25% acetic acid buffered to pH ~ 4 using NH_4OH (hereafter, HH solution) for less than 1 hour (Jang et al., 2017). The reagent-to-sample volume ratio was approximately 1 (Jang et al., 2017) to prevent the dissolution of labile detrital components (e.g., volcanic

Table 1Detrital and weatherable ϵ_{Nd} values of Svalbard bedrocks.

Sample ID	Coordinate	Area	Stratigraphic unit	Age*	Rock type	Detrital ϵ_{Nd}	Weatherable ϵ_{Nd}
13071104	78.987°N, 12.087°E	Blomstrandhalvøya, Kongsfjorden	Generalbjella Formation (Fm.), Krossfjorden Group	Upper Proterozoic	Quartzite	-8.8 ± 0.2	-5.8 ± 0.2
13071105	78.987°N, 12.087°E	Blomstrandhalvøya, Kongsfjorden	Generalbjella Fm., Krossfjorden Group	Upper Proterozoic	Phyllite	-11.7 ± 0.3	-11.4 ± 0.4
SK01	78.545°N, 16.183°E	Skansen, Isfjorden- Billefjorden	Wood Bay Fm., Dicksonfjorden Member	Devonian	Sandstone	-8.6 ± 0.3	-6.4 ± 0.3
080302	79.389°N, 13.622°E	Halvdanpiggen, Woodfjorden	Wood Bay Fm.	Devonian	Red sandstone	-13.6 ± 0.3	-10.8 ± 0.2
13071005	78.953°N, 11.758°E	Brøggerhalvøya, Kongsfjorden	Brøggertinden Fm.	Carboniferous	Red sandstone	-3.6 ± 1.3	-12.6 ± 0.2
SYB05	78.944°N, 11.724°E	Brøggerhalvøya, Kongsfjorden	Wordiekammen Fm.	Carboniferous	Limestone	–	–
SYB56	78.941°N, 11.703°E	Brøggerhalvøya, Kongsfjorden	Wordiekammen Fm.	Carboniferous	Dolostone	–	-10.9 ± 0.6
SK20	78.539°N, 16.159°E	Skansen, Isfjorden- Billefjorden	Wordiekammen Fm.	Carboniferous	Dolostone	–	–
SYB40.1	78.942°N, 11.704°E	Brøggerhalvøya, Kongsfjorden	Wordiekammen Fm.	Carboniferous- Permian	Calcareous shale	-16.3 ± 0.3	-13.7 ± 0.3
SF00.5	78.915°N, 11.654°E	Brøggerhalvøya, Kongsfjorden	Kapp Starotin Fm.	Permian	Chert	-11.6 ± 0.5	-6.6 ± 0.2
080109	78.091°N, 13.81°E	Festningen, Isfjorden- Grøn fjorden	Kapp Starotin Fm.	Permian	Black mudstone	-8.5 ± 0.3	-6.8 ± 0.2
Sval central	78.545°N, 16.183°E	Skansen, Isfjorden- Billefjorden	Kapp Starotin Fm.	Permian	Anhydrite	–	–
KIGAM #25	78.475°N, 15.812°E	Rotundafjellet, Isfjorden- Billefjorden	Sassendalen Group	Early – Mid. Triassic	Shale	-6.1 ± 0.4	-9.3 ± 0.2
080111	78.094°N, 13.837°E	Festningen, Isfjorden- Grøn fjorden	Verdebukta Fm.	Triassic	Black mudstone	-10.6 ± 0.3	-8.7 ± 0.2
20150901-02	78.341°N, 15.857°E	Janusfjellet, Isfjorden- Adventfjorden	Marhøgda Bed, Agardhfjellet Fm.	Jurassic	Oolitic ironstone	-6.3 ± 0.3	-5.1 ± 0.3
KIGAM #16	78.098°N, 13.913°E	Festningen, Isfjorden- Grøn fjorden	Janusfjellet Subgroup	Mid. Jurassic - Early Cretaceous	Shale	3.4 ± 0.4	-1.2 ± 0.3
KIGAM #5	78.239°N, 15.337°E	Platåberget, Isfjorden- Adventfjorden	Carolinefjellet Fm.	Early Cretaceous	Shale	-9.0 ± 0.2	-12.0 ± 0.2
080114	78.099°N, 13.948°E	Festningen, Isfjorden- Grøn fjorden	Helvetiafjellet Fm.	Cretaceous	Sandstone	-25.9 ± 0.2	-25.6 ± 0.2

* Dallmann and Elvevold (2015).

material) during the leaching process. Following the extraction of the authigenic and/or weatherable components, a second leaching step was carried out using the HH solution for ~24 hours to eliminate any remaining carbonate or Fe-Mn oxyhydroxide materials from the residual 'detrital' fraction (Jang et al., 2017).

After rinsing with Milli-Q water (Millipore, >18.2 MΩ cm), the detrital fractions were digested by alkaline fusion for Nd isotopic measurements (Bayon et al., 2009). Approximately 100 mg of dry ground detrital sediment was transferred into a glassy carbon beaker (SIGRADUR GAB 3, HTW) with 1.2 g of Na₂O₂ (>95%, small beads, ACS grade, Sigma-Aldrich) and 0.6 g of NaOH (>98.0%, pellet, ACS grade, Sigma-Aldrich) and placed in a furnace at ~700 °C for ~15 min. Neodymium and other reactive elements were then co-precipitated onto Fe oxide phases after the addition of ultra-pure Milli-Q water. After centrifugation, Fe oxide precipitates were rinsed in Milli-Q water and dissolved in 2 M HNO₃. Neodymium was purified by ion chromatography following a protocol modified from Pin and Zalduegui (1997) (Jang et al., 2017).

3.2.2. Elemental concentration analysis

The elemental abundances (Al, K, Ca, Mg, Fe, and rare earth elements (REEs)) of whole-rock samples and detrital fractions were determined by inductively coupled plasma optical emission spectrometry (ICP-OES, OPTIMA 8300, PerkinElmer) and mass spec-

trometry (ICP-MS, Thermo X-5, Thermo Scientific) at the Korea Basic Science Institute (KBSI). Prior to the elemental analyses, the samples were dissolved by acid digestion in a PTFA digestion vessel using a 4:4:1 mixture of HNO₃, HF, and HClO₄. Repeated analyses of USGS rock reference powders (BCR-2 and BHVO-2) yielded a reproducibility better than ±5% (1σ, standard deviation).

3.2.3. Nd isotope analyses

Neodymium isotopic measurements were performed by thermal ionization mass spectrometry (TIMS, Triton, Thermo Scientific) at the Korea Polar Research Institute. All measured ratios were corrected for mass fractionation using $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. Repeated analyses of JNdi-1 standard solutions yielded an average value of $^{143}\text{Nd}/^{144}\text{Nd} = 0.512100 \pm 0.000011$ (2σ, n = 30), which were consistent with the reference value (0.512115) of Tanaka et al. (2000). The validity of the analytical procedure was further evaluated by analyzing the USGS reference materials BCR-2 (0.512631 ± 0.000013, 2σ, n = 3) and BHVO-1 (0.512975 ± 0.000009, 2σ, n = 3), which were also consistent with values from the literature (e.g., Raczek et al., 2003). The total procedure blank was < 0.2% Nd for the detrital fractions and an order of magnitude less for the leached authigenic phases. In this study, Nd isotopic ratios are reported using the epsilon notation (ϵ_{Nd}), which represents the deviation of any measured $^{143}\text{Nd}/^{144}\text{Nd}$ ratio relative to the value

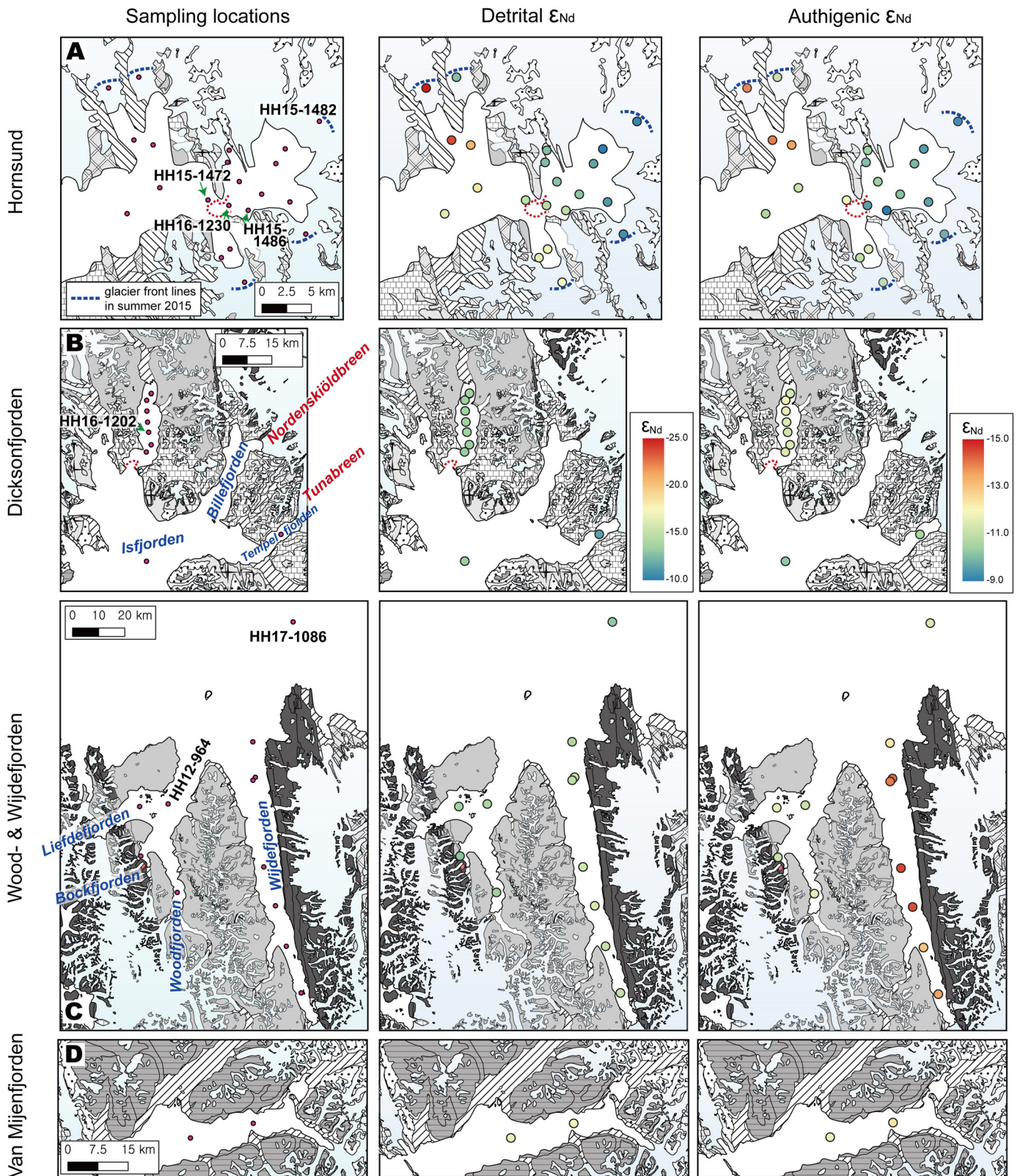


Fig. 3. Maps of the locations of fjord sediments (left column). ϵ_{Nd} of detrital (central column) and authigenic (right column) components within surface sediments collected from (A) Hornsund, (B) Dicksonfjorden, Isfjorden, Tempelfjorden, (C) Woodfjorden, Wijdefjorden, and (D) Van Mijenfjorden. The present glacier fronts in Hornsund (summer 2015) are indicated by blue dashed lines, and major sills in Hornsund and Dicksonfjorden are marked by red dotted lines.

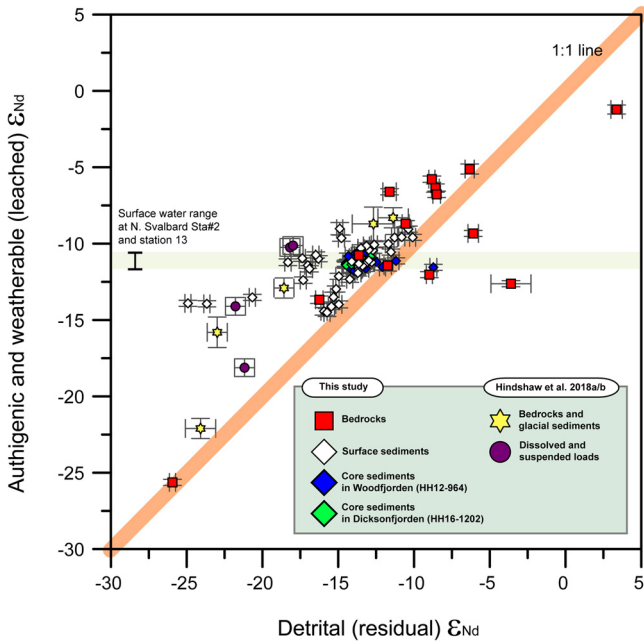


Fig. 4. ϵ_{Nd} data comparison between leached (authigenic and weatherable) and residual components (detrital) of the bedrock samples (red squares), surface sediments (white diamonds), core sediments at sites HH12-964 (blue diamonds) and HH16-1202 (green diamonds). For further comparison, the ϵ_{Nd} values of the corresponding components within bedrocks and glacial sediments (yellow stars) and riverine dissolved and suspended loads (purple circles) in Svalbard, as well as sea-water range at the Spitsbergen margin (green bar) are also shown (data from Andersson et al., 2008; Hindshaw et al., 2018a,b; Laukert et al., 2017).

for the modern chondritic uniform reservoir ($(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}} = 0.512638$; Jacobsen and Wasserburg, 1980).

4. Results

4.1. Svalbard bedrocks

Elemental and isotopic data for the detrital and weatherable fractions of the bedrock samples ($n = 18$) are reported in Supplementary Table S2, Table 1, and Fig. 2. The Nd concentrations in these samples vary from ~ 0.3 to ~ 43 mg/kg for the bulk samples and from ~ 0.03 to ~ 33 mg/kg for the detrital components. Limestone, dolostone, and anhydrite are characterized by much lower Nd concentrations (0.3 to 4.0 mg/kg for bulk samples) compared with those in the siliceous sedimentary rocks, such as the Devonian Old Red Sandstone and the Permian black mudstone (up to ~ 43 mg/kg for bulk and ~ 35 mg/kg for the detrital component).

The detrital and weatherable fractions of the bedrocks display wide ranges of ϵ_{Nd} values, from -25.9 to 3.4 and from -25.6 to -1.2 , respectively (Fig. 2). The average values of detrital and weatherable ϵ_{Nd} are -9.8 ± 6.6 (1σ , $n = 14$) and -9.8 ± 5.7 (1σ , $n = 15$), respectively, and the two isotopic compositions are strongly correlated with each other ($r = 0.82$, $n = 14$). The Cretaceous sandstone and the mid-Jurassic to early Cretaceous shale collected close to the mouth of Isfjorden presented the minimum ϵ_{Nd} value (-25.9 for detrital and -25.6 for weatherable) and maximum ϵ_{Nd} value ($+3.4$ for detrital and -1.2 for weatherable) measured in the bedrocks. Note that analyses of Nd-poor bedrocks, such as anhydrite (< 0.4 mg/kg for bulk), limestone (< 4 mg/kg for bulk) and two dolostone samples (< 1.3 mg/kg for bulk), were not possible due to insufficient intensity signals during the TIMS analysis. Except for the Cretaceous sandstone and the upper Proterozoic phyllite, the Nd isotopic compositions of the weatherable fractions are statistically different from the corresponding detrital fractions. Except for three Mesozoic shales, the weatherable ϵ_{Nd} signatures are

systematically more radiogenic (up to 5 epsilon-units higher) than the corresponding detrital fractions (Figs. 3 & 4).

4.2. Fjord surface sediments

The Nd concentrations of the bulk samples and their detrital components of the surface sediments show average values of 48 and 40 mg/kg, respectively. Thus, they are higher than the maximum concentrations reported for the bedrock samples (see section 4.1). The ϵ_{Nd} compositions of the detrital components range from -24.9 to -10.1 (with an average of -14.7 ± 3.0 ; 1σ , $n = 45$), while those of the authigenic components are more radiogenic, ranging from -14.5 to -9.0 (with an average of -11.5 ± 1.5 ; 1σ , $n = 45$) (Figs. 3 & 4). The correlation between the detrital and authigenic ϵ_{Nd} is strong ($r = 0.62$) but not as strong as that of the bedrock. Note that the sediments collected from Hornsund, with average detrital and authigenic ϵ_{Nd} values of -15.1 ± 3.0 and -10.8 ± 1.5 (1σ , $n = 21$), respectively, display a large spatial variability reflecting the presence of various bedrock types in the surrounding catchment area, including Proterozoic metamorphic rocks and Palaeozoic, Mesozoic and Palaeogene sedimentary sequences (Dallmann and Elvevold, 2015). The sediments recovered from Dicksonfjorden are comparatively uniform in terms of Nd isotopes, with average detrital and authigenic ϵ_{Nd} values of -13.6 ± 0.1 and -11.5 ± 0.2 , respectively, which are very similar to the composition of the Devonian Old Red Sandstone (-13.6 ± 0.3 and -10.8 ± 0.2 (1σ , $n = 8$), respectively). Similar detrital ϵ_{Nd} values were also obtained for sediments from Woodfjorden and its tributary fjords (-13.6 ± 0.5 , 1σ , $n = 4$). However, the elemental ratios (e.g., Fe/Ca and Ca/Al; Supplementary Table S3) and sediment colours are different from that in Dicksonfjorden. In Wijdefjorden, the detrital ϵ_{Nd} values of surface sediments are between those of the Devonian Old Red Sandstone (-13.6 ± 0.3) collected from the Woodfjorden catchments (Fig. 2B) and Paleoproterozoic meta-igneous rocks from Ny-Friesland ($\epsilon_{\text{Nd}} \sim -25.4$ to -18.6 from Johansson et al., 1995) (Fig. 2A). The sediments collected in Van Mijenfjorden are characterized by less radiogenic detrital ϵ_{Nd} values of -17.3 and -16.9 .

4.3. Core sediments

4.3.1. Dicksonfjorden

Sediments at site HH16-1202 in Dicksonfjorden are characterized by detrital and authigenic ϵ_{Nd} values ranging from -14.5 to -12.9 (average -13.5 ± 0.3 , 1σ , $n = 28$), and from -11.5 to -10.4 (average -11.2 ± 0.2), respectively (Fig. 5). Compared with the bedrock and core-top sediment samples, the detrital and authigenic ϵ_{Nd} values at site HH16-1202 display only a moderate correlation over the past 7.6 ka ($r = 0.43$, $n = 28$). Some ϵ_{Nd} excursions can be detected in the core for both detrital and authigenic fractions (e.g., the more radiogenic value at ~ 3.5 ka) as well as only for the detrital (less radiogenic ϵ_{Nd} shifts during the last millenia) or authigenic fractions (e.g., more radiogenic authigenic value at ~ 4.5 ka).

4.3.2. Woodfjorden

The detrital ϵ_{Nd} values at site HH12-964 in Woodfjorden fluctuated from -14.5 to -8.7 over the last 13.3 ka (Fig. 6), whereas the authigenic fractions displayed a much narrower range of ϵ_{Nd} signatures (from -11.8 to -10.6 ; average value -11.2 ± 0.3 , 1σ , $n = 35$) (Fig. 6). For the detrital fractions, the average ϵ_{Nd} value of -13.4 is consistent with that of the Devonian Old Red Sandstone. Prominent peaks in the detrital ϵ_{Nd} values towards radiogenic signatures occurred at ~ 7.2 ka and ~ 13.2 ka. The overall downcore trend of authigenic values ϵ_{Nd} does not correlate with that of the detrital ϵ_{Nd} values ($r = -0.07$), which is probably due to several outliers in the detrital ϵ_{Nd} trend.

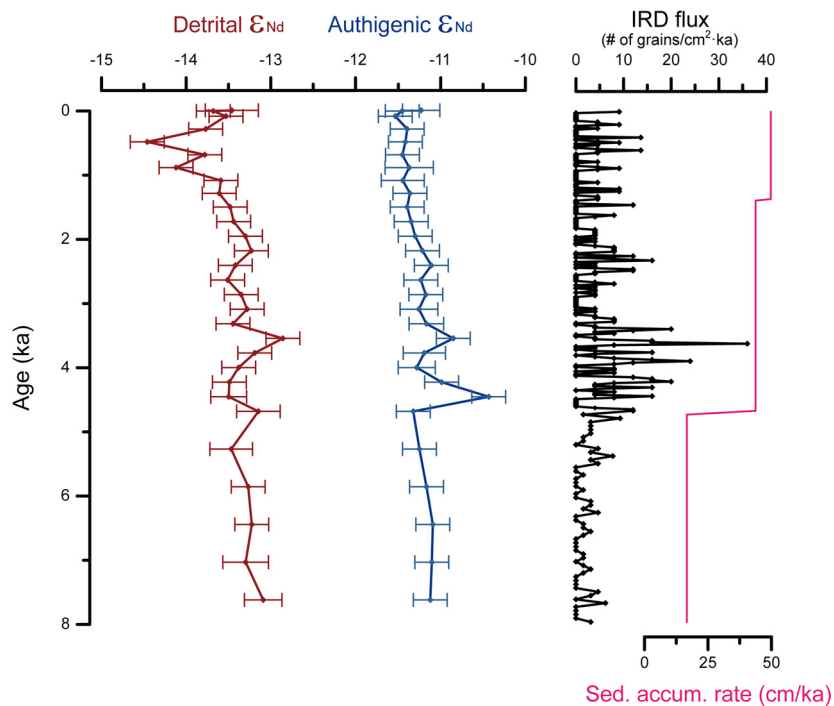


Fig. 5. Temporal variation in ϵ_{Nd} values of detrital (red) and authigenic (blue) components at site HH16-1202 in Dicksonfjorden. The ϵ_{Nd} data are compared to the IRD flux (black) and sediment accumulation rate (dark pink) (Joo et al., 2019). Error bars for ϵ_{Nd} represent $\pm 2\sigma$ uncertainties.

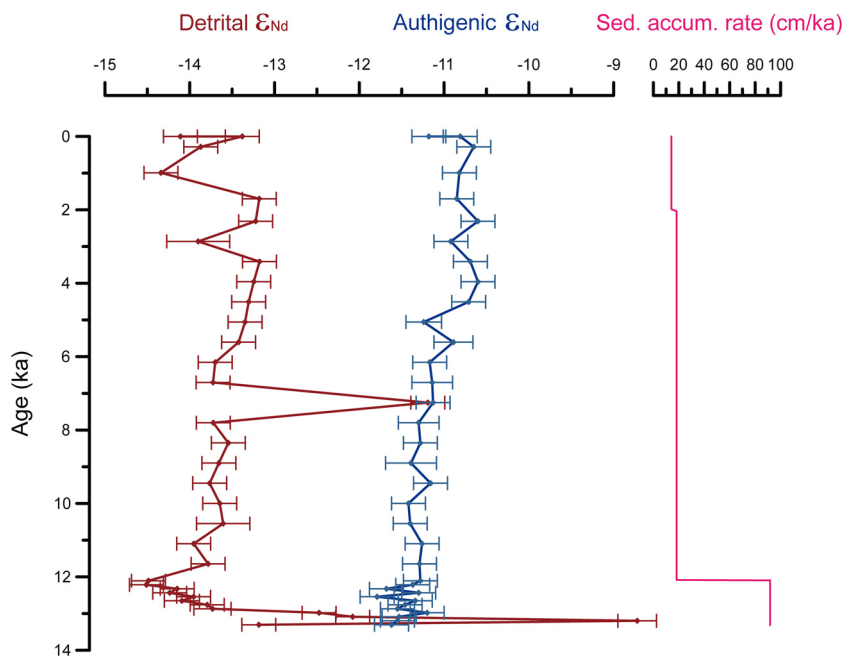


Fig. 6. Temporal variation in ϵ_{Nd} values of detrital (red) and authigenic (blue) components at site HH12-964 in Woodfjorden. The ϵ_{Nd} data are compared to the sediment accumulation rate (Hansen, 2014). Error bars for ϵ_{Nd} represent $\pm 2\sigma$ uncertainties.

5. Discussion

5.1. Nd isotopic decoupling during incongruent terrestrial weathering in Svalbard

The weatherable components extracted from the analyzed bedrocks usually display more radiogenic Nd isotopic compositions than the corresponding detrital silicate fractions (Figs. 3 & 4). Enriched radiogenic Nd isotope ratios of leachates could be found, such as in volcanogenic sediments (e.g., Elmore et al., 2011). However, we are confident that the observed radiogenic Nd isotopic

composition in the weatherable fraction does not result from the preferential dissolution of labile volcanic components due to four reasons (Supplementary Text 1). First, our mild leaching procedure would have prevented any significant detrital contamination (Jang et al., 2017). Second, if significant detrital contamination had occurred, then preferential leaching of the volcanogenic sediment in core HH12-964 should have occurred, which would have yielded pronounced radiogenic signatures in the leached authigenic fractions (which is not the case). Third, the authigenic ϵ_{Nd} signature at site HH17-1086 in Wijdefjorden (Fig. 1) is statistically consistent with the neighbouring seawater ϵ_{Nd} (see more detail in

section 5.2.1). Finally, although the sediments collected from two proximal sites in Hornsund display similar detrital ε_{Nd} and major element geochemistry, they present different authigenic ε_{Nd} values, which is consistent with their overlying bottom water masses showing contrasting water temperatures (see more detail in section 5.2.2).

As a consequence, the observed Nd isotopic decoupling between the weatherable and detrital components of Svalbard bedrocks is most likely best explained by the preferential dissolution of marine precipitates hosted by sedimentary rocks and/or labile accessory mineral phases in silicate rocks. The same hypothesis was recently proposed by Hindshaw et al. (2018a) to account for the observation that dissolved riverine loads in Svalbard were more radiogenic than corresponding suspended sediments, bedrocks, and glacial sediments (Fig. 4). In full agreement with this finding, our rock leaching experiments demonstrate that incongruent weathering in Svalbard can lead to significant Nd isotopic decoupling (Supplementary Text 2). Sedimentary rocks consist of various biogenic and authigenic components that are generally admixed with older recycled detrital minerals derived from the erosion of the basement and presumably characterized by less radiogenic ε_{Nd} compositions (e.g., Hindshaw et al., 2018b).

Previous studies have also reported pronounced Nd isotopic differences between dissolved loads and suspended sediments or bedrocks in catchments (Andersson et al., 2001; Goldstein and Jacobsen, 1987), particularly in sub-arctic river systems draining old metamorphic rocks (Andersson et al., 2001). Preferential alteration of easily dissolvable and radiogenic accessory minerals, such as apatite or titanite, is likely to occur during the weathering of igneous and metamorphic rocks (e.g., Andersson et al., 2001; von Blanckenburg and Nagler, 2001) as demonstrated by the rock leaching experiments (Dausmann et al., 2019).

In this study, significant Nd isotopic decoupling between detrital and weatherable fractions (> 1.2 epsilon-units) was observed in one of the two metamorphic rock samples and 11 of the 12 sedimentary rock samples, with the most pronounced isotopic decoupling (about 9 epsilon-units) occurring in the Carboniferous sandstone (Figs. 3 & 4). Interestingly, the ε_{Nd} differences between the weatherable and detrital fractions in sedimentary rocks were not always positive and presented negative values in four samples (Figs. 3 & 4). This finding contradicts the common assumption that incongruent weathering leads to the release of more radiogenic Nd fractions compared to residual detrital fractions (Andersson et al., 2001; Dausmann et al., 2019; Goldstein and Jacobsen, 1987; Hindshaw et al., 2018a; von Blanckenburg and Nagler, 2001). The negative ε_{Nd} offsets were found in the Carboniferous sandstone (with a Nd isotopic difference of -9 epsilon-units) and three Mesozoic shale samples (with ε_{Nd} differences between -3.0 and -4.6 epsilon-units); thus, in these samples, the detrital ε_{Nd} values are comparatively more radiogenic than the weatherable ε_{Nd} values. The presence of abundant radiogenic mineral phases, such as zircon or garnet (characterized by high Sm/Nd ratios; see Fig. 2 in Bayon et al., 2006), in silicate residual phases could account for the observed ε_{Nd} differences between the weatherable and detrital fractions. However, it is also likely that these negative ε_{Nd} differences reflect the fact that detrital fractions in sedimentary rocks may have been derived from the erosion of silicate bedrocks that are more radiogenic than associated weatherable marine precipitates.

5.2. Origin of the authigenic ε_{Nd} in Svalbard fjord sediments

5.2.1. Terrestrial signals

The strong Nd isotope correlation ($r = 0.62$) observed between authigenic and detrital fractions of Svalbard fjord sediments suggests that the ε_{Nd} signature of leached phases is mainly influenced

by terrestrial inputs (Fig. 4). As reported previously, the presence of preformed Fe oxides delivered together with detrital inputs could at least partially account for these 'terrestrial' ε_{Nd} signatures (Bayon et al., 2004; Werner et al., 2014). Such preformed terrestrial Fe oxides can be entrained in sea-ice (Werner et al., 2014) or delivered via riverine inputs (Bayon et al., 2004). However, preformed Fe oxides do not account for the characteristic features of authigenic ε_{Nd} described in section 5.2.2.

Alternatively, glacial meltwater runoff to the fjords through glacial rivers or direct outflows from tidewater glaciers is known to deliver substantial amounts of dissolved and nanoparticulate iron (Poulton and Raiswell, 2005; Wadham et al., 2019; Wehrmann et al., 2014), and this is also the case for Svalbard, where large inputs of dissolved iron and Fe oxides can occur as a result of the glacial abrasion and subsequent dissolution of silicate rocks or iron-bearing sedimentary rocks (Hindshaw et al., 2018a; Wehrmann et al., 2014). This mechanism can be further promoted in polar regions by the photoreductive dissolution of Fe oxides trapped in ice (Kim et al., 2010). In Svalbard, this process may be linked to markedly higher contents of highly reactive Fe oxides observed in fine-grained glacial meltwater sediments in Finsterwalderbreen compared to other glacial and riverine sediments in the world (Poulton and Raiswell, 2005). Iron oxide in such sediments is mainly hosted by clay-bound nanoparticulate Fe oxides (10–20 nm diameter) (Poulton and Raiswell, 2005), which can be easily dissolved in seawater (Moore et al., 1979), hence representing a potential source of dissolved iron (and presumably REEs) for subsequent authigenic Fe-oxyhydroxide co-precipitation in Svalbard fjords.

Vertical benthic fluxes related to the early diagenetic iron cycle in marine sediments represent another substantial source of dissolved iron in Svalbard fjords. The diagenetic reduction of glacially derived Fe oxides and the subsequent release of dissolved iron into pore waters may enhance the *in-situ* formation of authigenic minerals in near-surface sediments, e.g., monosulphides and siderite (Gorlich, 1986). This process is revealed by the high dissolved Fe concentrations (up to 800 μM) determined in porewaters in Kongsfjorden and Van Keulenfjorden (Wehrmann et al., 2014) and other evidence for intense early diagenetic Fe reductions in other Svalbard fjords (e.g., Hornsund, Tempelfjorden and Van Mijenfjorden; Vandieken et al., 2006 and references therein). The reduction of Fe oxide releases abundant REEs. Therefore, the high export fluxes of dissolved Fe to Svalbard fjords are also accompanied by the delivery of substantial amounts of dissolved REEs.

5.2.2. Seawater signals

One seawater ε_{Nd} data point is available from North Svalbard Sta#2 ($\varepsilon_{\text{Nd}} \sim -11.8 \pm 0.4$ from Andersson et al., 2008; 78.834°N, 9.330°E, ~ 199 m water depth) located only approximately 22 km north-north west from site HH17-1086 (top-core authigenic $\varepsilon_{\text{Nd}} \sim -11.4 \pm 0.3$), i.e., the northernmost station of Wijdefjorden (Fig. 1). The two sites lie at water depths of 150 m and 322 m in comparable water masses characterized by the same density. The ε_{Nd} values of seawater and authigenic sediment at both sites are consistent within uncertainty, suggesting the authigenic ε_{Nd} as an archive of seawater Nd isotopic signatures (not preformed Fe oxides).

The seawater signals recorded in the authigenic ε_{Nd} can also be evaluated by comparing the results from two nearby sites in Hornsund; HH15-1472 and HH16-1230 (for locations see Table S2 and Fig. 3). These two sites are located less than 2.2 km from each other but are separated by a sill and hence are affected by different water masses, namely, TAW (HH15-1472) and WCW (HH16-1230) (Promińska et al., 2017; Zajczkowski et al., 2010) (Fig. 3). The ambient bottom water masses at both sites can be clearly distinguished by different summer season temperatures, which are

approximately 1 °C in the outer fjord basin (TAW) and below -1 °C in the inner basin (WCW) (Zajączkowski et al., 2010). The surface sediments collected at the two sites show the same detrital ε_{Nd} values and almost identical major elemental geochemical composition, such as K/Al, Ca/Al, Fe/Ca and Mg/Al ratios (Supplementary Table S3). However, the samples display different authigenic ε_{Nd} values (-12.1 at site HH15-1472 and -9.6 at site HH16-1230). This observation further highlights that the authigenic ε_{Nd} signal preserved in the surface fjord sediments in Svalbard could be derived, at least to some extent, from local water masses.

The authigenic ε_{Nd} values of the surface sediments collected near glacier fronts in Hornsund (i.e., sampling sites HH16-1230, HH15-1482, and HH15-1486) are much more radiogenic than those of the regionally advected water mass within the Sørkapp Current, which is an extension of the ESC (-11.6 < ε_{Nd} < -10.6 from Laukert et al., 2017; station 13; 78.834°N, 9.330°E, ~199 m water depth) (Fig. 1), suggesting strong preservation of radiogenic terrestrial inputs near the sources. Strong terrestrial signals could be partly due to efficient flocculation and, thus, the capture of small particles (including Fe oxides) and their rapid deposition in the proximal environment (Markussen et al., 2016). However, limitations exist in the simple comparison of isotopic compositions. Due to the transformation of water masses carried by ESC and mixing with Atlantic water masses within WSC on the continental shelf of western Spitsbergen, the water masses entering Hornsund fjord are a mixture of various water masses (e.g., Promińska et al., 2017). Therefore, we cannot rule out the influence of seawater inflow on the radiogenic authigenic ε_{Nd} .

5.3. Variation in the degree of Nd isotopic decoupling linked to climate-driven glacial system changes

In Svalbard fjords, the authigenic ε_{Nd} values measured in surface sediments are systematically more radiogenic than the corresponding detrital ε_{Nd} signatures. Consistent with previous experimental investigations (see section 5.1), this observation further suggests that the preferential release of radiogenic Nd occurs during weathering in a glacial environment.

Similar observations can be drawn from the downcore records acquired from Dicksonfjorden (site HH16-1202; Supplementary Table S4) and Woodfjorden (site HH12-964; Supplementary Table S5), where the authigenic ε_{Nd} values are typically more radiogenic than the detrital ε_{Nd} values (Figs. 5 & 6). Interestingly, the differences between the authigenic and detrital values ($\Delta\varepsilon_{\text{Nd}}$) at these two sites display similar variability over time (Fig. 7). Neodymium isotopic decoupling was enhanced (i.e., leading to more positive $\Delta\varepsilon_{\text{Nd}}$ values) since the mid-Holocene, when new glaciers formed (Fjeldskaar et al., 2018; Svendsen and Mangerud, 1997; van der Bilt et al., 2015) (growth curve of valley glacier in Fig. 7) and/or existing glaciers on Svalbard readvanced ~4.5 ka (Baeten et al., 2010; Forwick et al., 2010) (growth curve of ice cap in Fig. 7). Decoupling is even more markedly enhanced during the transition period and main phase of LIA, when most glaciers on Svalbard reached their maximum extent (e.g., Martín-Moreno et al., 2017; Svendsen and Mangerud, 1997) (Fig. 7).

Based on the above discussion (section 5.2), the observed strong decoupling between authigenic and detrital signatures during the colder periods could indicate increasing contribution from (1) less radiogenic detrital materials, (2) inflow of radiogenic seawater and/or (3) more labile terrestrial sources in the glaciated catchments. The first hypothesis, although it could partly explain maximum $\Delta\varepsilon_{\text{Nd}}$ values at the onset of and during parts of the LIA when decreased ε_{Nd} of detrital fractions were documented (Figs. 5, 6, and 7), cannot account for the significant change in the $\Delta\varepsilon_{\text{Nd}}$ value during the period of glacier growth at ~4.5 ka. Unfortunately, the very high diversity of rock types on Svalbard combined with the

sparse dataset on the measured ε_{Nd} in rocks (Table 1) limits the ability to provide an exact explanation (sediment sources) for the observed temporal variability of ε_{Nd} in the studied sediment cores. The second hypothesis concerning the enhanced inflow of advected seawater likely cannot explain the observed changes, at least in Dicksonfjorden because of a shallow (~20 m) sill at the fjord mouth, which has become slightly shallower in response to an isostatic rebound during the mid-Holocene (Forman et al., 2004), thereby further restricting the inflow of external water masses. The third hypothesis concerning the increased input of labile terrestrial materials seems to provide a good explanation of the changes, particularly those recorded in Dicksonfjorden, where the increased sediment accumulation rate ~4.5 ka (Fig. 5) indicates enhanced terrestrial inputs related to the glacio-fluvial processes at that time. The glacial activity in Dicksonfjorden likely increased at approximately 4.5 ka. This timing is synchronous with the glacier reformation in the catchment areas of Linnévatnet (5 - 4 ka from Svendsen and Mangerud, 1997) and Hajeren lakes (~4.3 and ~3.3 ka from van der Bilt et al., 2015) (Fig. 7). It also corresponded to the periods when Nordenskiöldbreen in Billefjorden (5.5 - 3.2 ka from Baeten et al., 2010) and Tunabreen in Tempelfjorden (6 - 4 ka from Forwick et al., 2010) grew (Fig. 7). Based on the evidence above, we propose that the downcore fluctuations in $\Delta\varepsilon_{\text{Nd}}$ in Svalbard fjord sediments, mainly at ~4.5 ka when several catchments were likely re-occupied by glaciers, reflected changes in the degree of ε_{Nd} decoupling related to the presence/growth (or absence/retreat) of adjacent glaciers and associated weathering conditions. We hypothesize that the growth of glaciers provided freshly eroded rock substrates available for subglacial weathering, thereby leading to the preferential release of radiogenic Nd (e.g., Süfke et al., 2019) and ultimately more pronounced isotopic decoupling between authigenic and detrital ε_{Nd} values in the fjord sediments (Fig. 7). An increase in the $\Delta\varepsilon_{\text{Nd}}$ value at site HH12-964 in mid-Woodfjorden at approximately 4.5 ka may also be attributed to the growth of tidewater glaciers draining into the Woodfjorden system (Fig. 7), which is supported by an abrupt increase in the sand content at that time (from less than 2 to 4.4%; Hansen, 2014). We ruled out the potential influence of preformed Fe oxides based on the discussion in section 5.2. However, the presence of preformed phases within sediment that survived weak chemical weathering during the periods of glacier advances might partly intensify $\Delta\varepsilon_{\text{Nd}}$ by increasing the authigenic ε_{Nd} .

Our finding that $\Delta\varepsilon_{\text{Nd}}$ variability in Svalbard fjord sediments may reflect weathering-induced changes in Nd isotopic decoupling related to glacier cover has far-reaching implications. For instance, $\Delta\varepsilon_{\text{Nd}}$ could be used in future studies to reconstruct the impact of past glacial meltwater on marine productivity (e.g., Wadham et al., 2019) as a proxy to infer historical glacial releases of key nutrients (e.g., P and Fe) associated with the dissolution of accessory phosphate minerals and nanoparticulate iron oxides.

6. Conclusion

Our survey of Nd isotopes in leached and residual (detrital) components of Svalbard bedrock and fjord sediment samples confirms that weathering under glacial conditions is accompanied by incongruent weathering leading to significant Nd isotope decoupling. In Arctic environments, such as Svalbard, glacier melting provides substantial amounts of nanoparticulate Fe oxides and dissolved elements, resulting in abundant co-precipitation of hydrogenous Fe-oxyhydroxide phases in the nearby seawater/fjord environments and controlling, to a large extent, the distribution of leached authigenic ε_{Nd} values in fjord sediments. The difference between authigenic and detrital ε_{Nd} values (termed $\Delta\varepsilon_{\text{Nd}}$) in the Holocene sedimentary records from the two fjords, Dicksonfjorden and Woodfjorden show trends that mimic the evolution of glacier

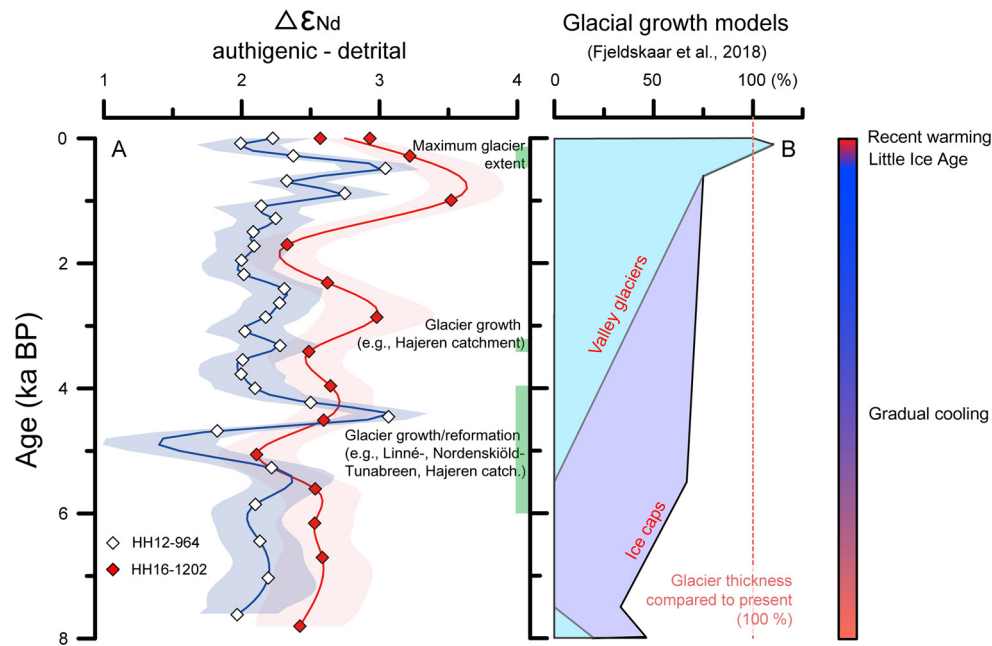


Fig. 7. Comparison of (A) the differences between the authigenic and detrital ϵ_{Nd} values ($\Delta\epsilon_{\text{Nd}}$) at sites HH12-964 (red) and HH16-1202 (blue) and (B) the glacier thickness reconstructed by the best-fit model after Fjeldskaar et al. (2018) over the last 8 ka. Higher $\Delta\epsilon_{\text{Nd}}$ values typically occur during the periods of glacier advances (green bars). Error ranges for $\Delta\epsilon_{\text{Nd}}$ were calculated based on a smoothing spline Monte-Carlo simulation (2σ , resampled at every 0.1 ka, $n = 100,000$).

activity over the last millennia, related to climate variability. The $\Delta\epsilon_{\text{Nd}}$ values and inferred Nd isotopic decoupling were higher during the periods of glacier advances, e.g., at ~ 4.5 ka and during the LIA, suggesting that increased glacier cover and concomitant glacial abrasion of fresh rock substrates resulted in intensified incongruent chemical weathering. This novel Nd isotope approach ($\Delta\epsilon_{\text{Nd}}$) may be used in future studies as a new proxy for past glacial changes and associated glacial-derived nutrient inputs to the marine realm.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

No competing interests.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.epsl.2020.116319>.

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