

Impact of Holocene climate change on silicon cycling in Lake 850, Northern Sweden

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The Holocene
1–11

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DOI: 10.1177/09596836211025973

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Abstract

Diatom-rich sediment in a small subarctic lake (Lake 850) was investigated in a 9400 cal. yr BP sediment record in order to explore the impact of Holocene climate evolution on silicon cycling. Diatom stable silicon isotopes ($\delta^{30}\text{Si}_{\text{BSi}}$) and biogenic silica (BSi) indicate that high BSi concentrations in sediment throughout the Holocene are associated with a lighter Si isotope source of dissolved silica (DSi), such as groundwater or freshly weathered primary minerals. Furthermore, higher BSi concentrations were favoured during the mid-Holocene by low detrital inputs and possibly a longer ice-free period allowing for more diatom production to occur. The diatom $\delta^{30}\text{Si}_{\text{BSi}}$ signature shows a link to changes in regional climate and is influenced by length of diatom growth period and hydrological fluctuations. Lighter Si isotopic values occur during the mid-Holocene, when climate is inferred to be more continental and drier, with pronounced seasonality. In contrast, a heavier Si isotopic signature is observed in the early and late Holocene, when oceanic influences are thought to be stronger and the climate wetter. The $\delta^{30}\text{Si}_{\text{BSi}}$ values have generally lighter signatures as compared with other studies, which supports a light DSi source.

Keywords

diatom, Holocene, isotope, lake, sediment, silicon

Received 14 January 2021; revised manuscript accepted 11 May 2021

Introduction

In natural waters silicon is primarily present in the dissolved form as silicic acid H_4SiO_4 , also called dissolved Si (DSi), which originates from the weathering of primary minerals. Rivers and lakes act as a sink for DSi in the global silicon cycle, where lakes store the DSi primarily as diatoms (Frings et al., 2014b), which are unicellular siliceous algae. The DSi delivered to a lake is a product of processes in the lake catchment area, such as weathering and erosion, and can be modified by vegetation and soils. Subsequently, diatom production and dissolution are partly controlled by the sensitivity of diatoms to DSi concentration (Hamm et al., 2003; Yool and Tyrrell, 2003), and thus both processes – production and dissolution – influence changes in lake DSi (Panizzo et al., 2017). Fossil diatoms in lake sediment are used as an archive of environmental history and can be used for unravelling changes in silicon cycling as a result of their high preservation potential. Biogenic silica (BSi) concentrations have been used previously in Canadian Arctic lakes as an indicator of aquatic palaeoproductivity (Fortin and Gajewski, 2009). In Lake Baikal the sedimentary BSi concentration recorded diatom responses to changes in summer temperatures associated with variation in summer insolation (Khursevich et al., 2001). However, BSi concentration in lake sediments is also affected by diatom preservation (Panizzo et al., 2016; Ryves et al., 2003). Additionally, low detrital input can result in high sediment BSi concentration (Conger, 1942; Zahajská et al., 2021b).

Diatoms preferentially incorporate the lighter Si isotope, ^{28}Si , to form the diatom frustule when sufficient DSi is present (De La Rocha et al., 1997). The stable silicon isotopes from diatoms

($\delta^{30}\text{Si}_{\text{BSi}}$) can be used to record DSi utilization controlled by diatom productivity associated with changes in climate (De La Rocha et al., 1998; Hendry and Brzezinski, 2014; Opfergelt and Delmelle, 2012), diatom production and dissolution (Chen et al., 2012; Street-Perrott et al., 2008), vegetation impacts on the Si cycle (Frings et al., 2014a, 2016; Leng et al., 2009; Sun et al., 2011), rates of nutrient supply (Swann et al., 2010) or DSi sources (Nantke et al., 2019; Vandevenne et al., 2015). Additionally, changes in $\delta^{30}\text{Si}$ in the water column can result from seasonal diatom DSi uptake and clay mineral formation in the sediments (Ehlert et al., 2016; Frings et al., 2014c; Geilert et al., 2020; Zhang et al., 2020). Moreover, studies on Lake Baikal suggest that long-term changes in isotopic composition are complex and involve several simultaneous processes, for example, changes in weathering processes, land use or climate within the lake

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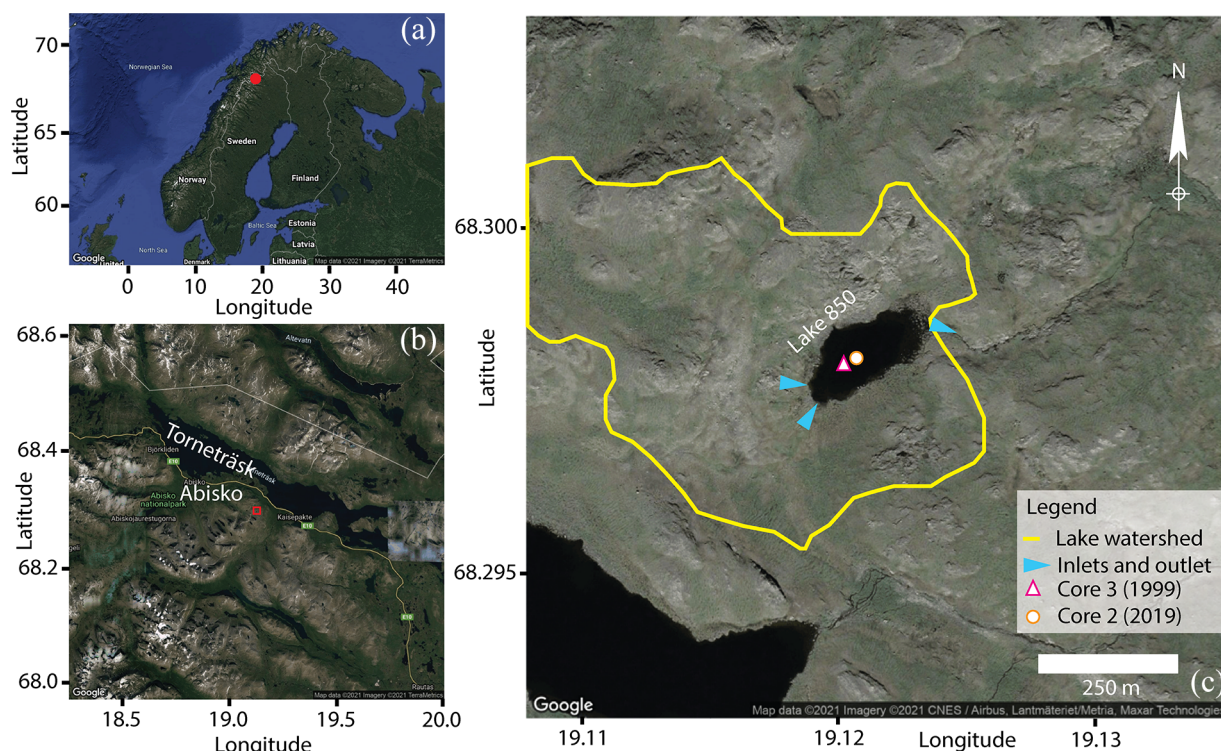


Figure 1. Location of Lake 850 (red circle) in northern Sweden on inset (a), the location of the lake in Abisko area (red square) on inset (b) and detailed map of the lake watershed (yellow line), inlets, outlet (blue arrows) and coring locations of two cores (from 1999 and 2019, triangle and circle, respectively) on inset (c). Plotted in R using package ggmaps (Kahle and Wickham, 2013) and modified.

catchment, as well as changes in lake mixing regimes, which affect the isotopic signature (Panizzo et al., 2017).

Changes in climate, recorded in lake sediments in northern Sweden spanning the last 9400 years, have been reconstructed using oxygen isotopes, changes in diatom communities (Bigler and Hall, 2003; Rosén et al., 2001; Shemesh et al., 2001), pollen-based vegetation reconstruction (Barnekow, 1999; Berglund et al., 1996; Seppä and Hammarlund, 2000) and chironomids (Rosén et al., 2001). In northern Sweden, post-glacial climate and vegetation development is commonly subdivided into three periods (Barnekow, 1999; Hammarlund et al., 2002; Seppä and Hammarlund, 2000). Early Holocene climate was characterized as humid oceanic, with warm summer temperatures, rising treeline and a shift in vegetation from subarctic shrub and birch tundra to boreal pine-birch forest (Seppä and Hammarlund, 2000). Mid-Holocene climate was more stable and continental compared with the early-Holocene, with warm and dry summers and strong seasonality (Berglund et al., 1996; Rosén et al., 2001). Gradual cooling with some short-term fluctuations is suggested for the late Holocene (Barnekow, 1999; Bigler et al., 2003; Rosén et al., 2001).

We examine here the impact of changes in climate on silicon cycling and BSi accumulation in Lake 850 within the framework of existing knowledge on Holocene climate changes in the Abisko area, northern Sweden (Berglund et al., 1996; Bigler and Hall, 2003; Hammarlund et al., 2002; Rosén et al., 2001; Seppä and Hammarlund, 2000). Pollen records in the Lake 850 area suggest that the lake has been above the pine treeline throughout the last 9400 cal. yr BP. We hypothesize that changes in DSi sources, derived from hydrological changes connected to changes in climate, are responsible for BSi concentration and $\delta^{30}\text{Si}_{\text{BSi}}$ variation. The long-term evolution of lake BSi accumulation is hypothesized to be dependent on climate-driven sedimentation rates, detrital input and diatom production. Sediment lithology, diatom BSi concentration and diatom stable silicon isotopes are used to constrain these processes. We hypothesize that BSi concentrations will be higher during warmer and drier periods due to lower allochthonous input and a prolonged

diatom growing season. We test this hypothesis by examining the lithology, BSi concentrations, proxies for detrital input (titanium and magnetic susceptibility) and comparison with diatom species composition data from Shemesh et al. (2001). Further, we hypothesize that changes in $\delta^{30}\text{Si}_{\text{BSi}}$ are connected to changes in DSi sources. Thus, isotopically heavier riverine DSi will influence the diatom $\delta^{30}\text{Si}_{\text{BSi}}$ in periods with increased precipitation, and isotopically lighter groundwater DSi will be more pronounced with decreased precipitation (Zahajská et al., 2021b).

Study area

Lake 850 is situated in northern Sweden, 14 km south-east from Abisko Research Station (388 m a.s.l.). Mean temperatures during summer and winter in the Abisko region are 9.8°C and −10.1°C, respectively, and the mean annual temperature (from 1913 to 2019) was −0.4°C (Abisko Scientific Research Station, 2019). The diatom growing season is from June to August (Shemesh et al., 2001). The lake, at an elevation of 850 m a.s.l. (68°15'N, 19°7'E), lies above treeline, which is currently at 600 m a.s.l. (Figure 1). The catchment area is 0.35 km² (Rubensdotter and Rosqvist, 2003), and the lake surface area is 0.02 km². Approximately 48% of the lake area lies within the deep basin, with a maximum depth of 8 m, and 52% of the lake surface area is shallow, with a depth of <4 m. The vegetation in the catchment area is comprised of Arctic species of mosses, grasses and shrubs, and the bedrock is composed of granites and syenites overlain by a thin layer of till (Shemesh et al., 2001). There are two ephemeral 1–2 cm deep inlets in the eastern part of the lake and one outlet in the western part (Figure 1). From mid-October until late May–early June, the lake is ice-covered, and its catchment area is snow-covered from mid-September to mid-June. In August the lake is well-mixed with no thermal stratification and has a pH of 6.8. The lake is classified as oligotrophic with a dissolved organic carbon concentration of 2.3 mg l^{−1}, and total phosphorus concentration of 6.5 µg l^{−1} (Bigler et al., 2002; Shemesh et al., 2001).

Table 1. Samples from Lake 850 sediment core from 2019 dated by ^{14}C AMS method.

^{14}C dated piston core					
Depth (cm)	Dated material	Weight (mg C)	Lab no	Radiocarbon age (BP)	Calibrated age ^a (cal. yr BP, 2 σ range)
28–29	<i>Betula nana</i> leaf	1.0	LuS15652	3405 $\pm$ 45	3562–3732
40–41	<i>Betula nana</i> leaf	0.8	LuS15655	3965 $\pm$ 45	4287–4528
54–55	<i>Betula nana</i> leaf	1.1	LuS15658	4525 $\pm$ 40	5047–5202
70–71	<i>Vaccinium</i> sp. leaf	1.4	LuS15661	6300 $\pm$ 50	7156–7366
73–74	<i>Vaccinium</i> sp. leaf	1.0	LuS15663	6370 $\pm$ 40	7247–7419

^a ^{14}C dates were calibrated using the IntCal20 radiocarbon calibration dataset by Reimer et al. (2020).

Methods

Core collection

In April 1999, a 125-cm sediment core was cored from the centre of the ice-covered Lake 850 using a modified Livingstone piston corer (Shemesh et al., 2001) at a water depth of 6.9 m. The core was subsampled into 62 2-cm sections, but only the top 56 samples contained sufficient biogenic silica (Shemesh et al., 2001) and are used here for stable Si isotope analysis.

To investigate BSi accumulation, a 74-cm-long sediment core was taken using a modified Livingston piston corer in March 2019 from the ice in the deep basin (68°17'53.2"N, 19°7'17.2"E) at a depth of 7.0 m. The piston core was scanned for density, magnetic susceptibility and X-ray fluorescence (XRF) with an ITRAX CS37 at the GLOBE Institute, Copenhagen University, Denmark. The core was correlated with the previously collected piston core (Core 3) from 1999 (Rubensdotter and Rosqvist, 2003; Shemesh et al., 2001) using age-depth models and total organic carbon (TOC) (see section Chronology).

Sediment characterization

The 2019 core was halved, and one half was continuously subsampled in 1-cm sections and placed into cubic boxes with known volume. The other half of the piston core was used for XRF and magnetic susceptibility scanning and archived. All sediments were weighed before and after freeze drying for water content, to obtain porosity and wet and dry bulk densities.

Total organic carbon (TOC) and total nitrogen (TN) analyses were carried out on freeze dried samples ($n = 28$), where 5–10 mg of dry sediment was packed into tin capsules. Six samples throughout the core were tested for carbonate by acidifying with HCl and heating to 60°C before total carbon (TC) measurements were conducted (Brodie et al., 2011). The measurements were done on a COSTECH ECS4010 elemental analyser at the Department of Geology, Lund University, with the mean analytical uncertainty for TOC of 0.3 wt% based on duplicate analysis ($n = 17$). The carbonate content was calculated as a difference in TC between de-calcified and bulk sample. Because these analyses indicated that the sediments do not contain carbonate, the TC measurements are used as a measure of TOC. The published LOI data from the 1999 core (Rubensdotter and Rosqvist, 2003) show values above 20%, thus an experimental conversion factor from LOI to TOC of 2 was used to recalculate LOI into TOC (Bojko and Kabała, 2016). Analysis for stable carbon isotopes analysis was performed on de-calcified bulk sediment from 2019 core (250 mg in 50 ml of 5% HCl for 24 h (Brodie et al., 2011)) and analysed with Elemental Analyser connected to a MAT-252 mass spectrometer at Weizmann Institute of Science, Rehovot.

The BSi concentration was analysed at a resolution of 1 cm by sequential alkaline extraction (Conley and Schelske, 2002). Freeze-dried homogenized samples were digested in 0.1 M Na_2CO_3 in a shaking bath at 85°C. Subsamples were taken at 3, 4 and 5 h and neutralized with HCl to examine for the dissolution of

minerals. There were no changes in the amount of total amorphous SiO_2 extracted during the time course of the dissolution, therefore mean values were used to estimate BSi concentration with no mineral correction applied (Conley, 1988).

Chronology

Five terrestrial macrofossil samples from the piston core were dated by ^{14}C using accelerator-mass-spectrometer at the Radiocarbon Dating Laboratory, Department of Geology, Lund University (Table 1). Radiometric dates were calibrated with IntCal20 radiocarbon calibration dataset (Reimer et al., 2020). The age-depth model (Figure 2) was established based on ^{14}C dates using the software package Bacon with five age controls (Blaauw, 2010). A priori assignment of mean sediment accumulation rate, based on the sediment accumulation rate from a previously dated core from 1999 (Shemesh et al., 2001), was set to 100 yr cm^{-1} , as suggested by Bacon. Thickness for spline calculation was set at 15.5 cm, above which the model diverged greatly from the age controls provided. The age-depth model had 100% of the dates overlapping within the mean 95% confidence ranges.

An updated age-depth model for the core from 1999 (Supplemental Figure S1) was established based on six original ^{14}C dates from the published age-depth model (Shemesh et al., 2001) using the same approach as described above. A priori assignment of mean sediment accumulation rate was set to 100 yr cm^{-1} , and thickness for spline calculation was set at 24.5 cm. Only 86% of the dates overlapped with the age-depth model's mean 95% confidence ranges.

Both cores were aligned based on age-depth models. A secondary control of the alignment was done through comparison of TOC calculated from loss on ignition (LOI) on the core from 1999 and a proxy for organic carbon (incoherent/coherent scanning ratio) measured on the new piston core (Figure 3), which showed good fit. The proxy for organic content is based on the rhodium (Rh) scatter peak areas, expressed as $\text{Rh}_{\text{inc}}/\text{Rh}_{\text{coh}}$, where the Rh incoherent (Rh_{inc}) responds to valence of electrons and Rh coherent (Rh_{coh}) scatter peak (K-alpha) responds to bulk electrons in sample material (Burnett et al., 2001). When the number of valence electrons is large compared to the number of bulk electrons, the density of the sample is lower, which indicates high organic content. All data are plotted with age, as the ages of both cores do not correspond to similar depths.

Stable Si isotopes analysis

Clean bulk diatom material (several diatom species with a minor contribution from chrysophytes, Supplemental Figure S2) from Lake 850 with 2-cm resolution was obtained from a 125-cm-long core used in a previous study (Shemesh et al., 2001) and processed for stable silicon isotopes. Sample purity was evaluated using a scanning electron microscope Tescan Mira3 at the Department of Geology, Lund University. Cleaned diatom samples ($\sim 0.8 \text{ mg}$) were digested with 0.5–1 ml of 0.4 M NaOH (analytical purity) at 50°C for at least 48 h. When all diatoms were dissolved, samples were diluted with Milli-Q[®] water to prevent precipitation and

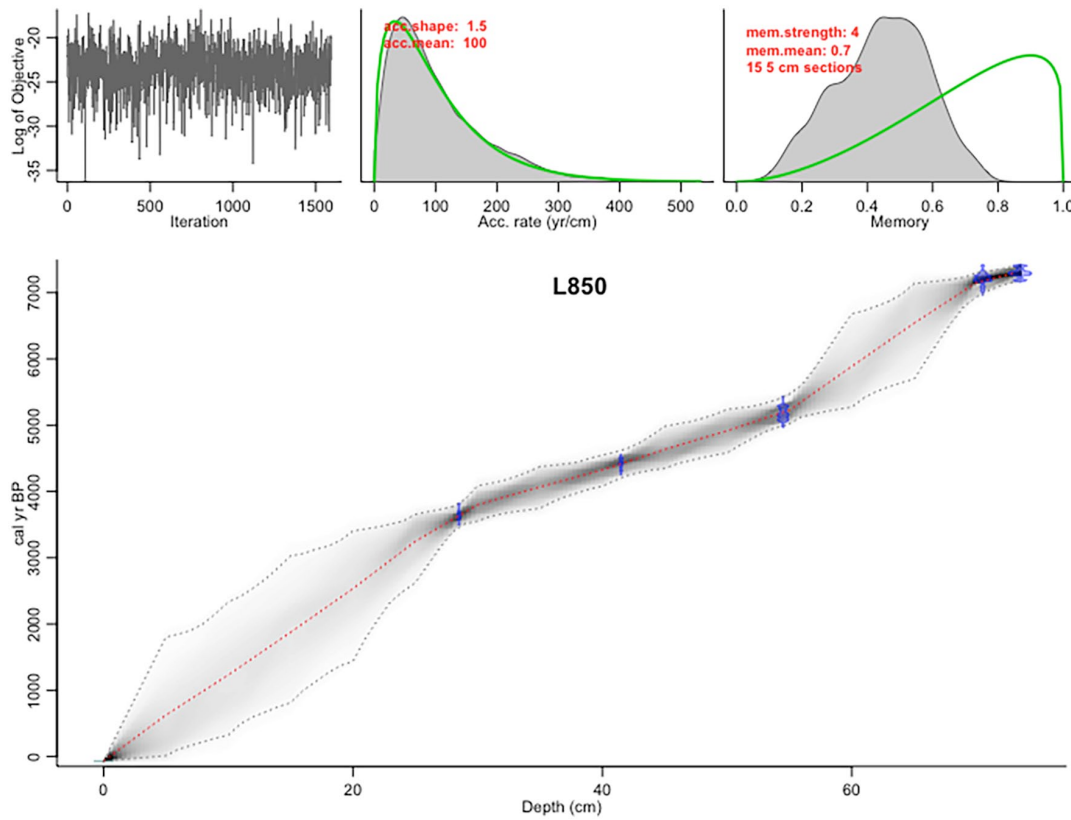


Figure 2. The age-depth model of piston core from 2019 from Lake 850 based on ^{14}C data (see Table 1) using the software package Bacon (Blaauw, 2010). The red line is the median probability age from all age-depth iterations, representing the best point estimate of age for any given depth. Grey shading represents the age model probability and contains the 95% confidence interval (dashed lines). Iteration history (left inset), prior and posterior densities of the mean accumulation rate (middle inset), and prior and posterior of the memory (right inset).

fractionation of amorphous silica, then neutralized by 0.5–1 ml of 0.4 M suprapur[®] HCl. The solutions were measured for their silicon concentration to obtain Si recovery, which was between 90% and 100% of the calculated concentration based on initially weighed and dissolved diatom SiO_2 . Samples solutions, Si standards RM-8546 and Diatomite prepared by NaOH fusion (Georg et al., 2006) were purified for silicon analysis by ion-chromatographic separation using 1.5 ml cation-exchange DOWEX[®] 50W-X8 (200–400 mesh) resin following the method by Georg et al. (2006).

Stable Si isotopes were measured on a NuPlasma (II) HR multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS, Nu Instruments[™]) with an Apex HF desolvation nebulizer at the Vegacenter, Swedish Natural History Museum, Stockholm. The ^{28}Si signal of full procedural blanks was determined to be $<0.35\%$ of the total signal, thus sample contamination was not observed. All samples were diluted to $2\text{--}3\text{ mg l}^{-1}$ of Si and matrix matched with standards in 0.12 M SeaStar HCl. Further, all samples and standards were doped to contain 3 mg l^{-1} of Li (IPC-MS standard) to match the matrix of Vegacenter standards prepared by LiBO_2 fusion (Sun et al., 2010).

Silicon isotopes data are reported as deviations of $\frac{^{30}\text{Si}}{^{28}\text{Si}}$ and $\frac{^{29}\text{Si}}{^{28}\text{Si}}$ from the RM-8546 (former NBS-28) in ‰, denoted $\delta^{30}\text{Si}$ and $\delta^{29}\text{Si}$ as follows:

$$\delta^{30}\text{Si} = \left(\frac{\left(\frac{^{30}\text{Si}}{^{28}\text{Si}} \right)_{\text{sample}}}{\left(\frac{^{30}\text{Si}}{^{28}\text{Si}} \right)_{\text{NBS28}}} - 1 \right) \cdot 1000. \quad (1)$$

Each sample was measured three to six times with bracketing of NBS-28 in between. Full chemical replicates were measured on 68% of all samples ($n = 56$). A three-isotope plot $\delta^{30}\text{Si}$ vs $\delta^{29}\text{Si}$ is used to ensure that there are no polyatomic interferences present during mass spectrometry measurements. All measured samples should fall on the expected mass-dependent fractionation line with a slope of 0.5092 (Supplemental Figure S3, Reynolds et al., 2007).

Long-term (ca. 3 years) variance from the secondary reference materials is as follows: Diatomite $\delta^{30}\text{Si} = 1.24 \pm 0.20\text{‰}$ ($2\text{SD}_{\text{repeated}}$, $n = 285$), Big-Batch $\delta^{30}\text{Si} = -10.63 \pm 0.34\text{‰}$ ($2\text{SD}_{\text{repeated}}$, $n = 109$) and IRMM $\delta^{30}\text{Si} = -1.77 \pm 0.22\text{‰}$ ($2\text{SD}_{\text{repeated}}$, $n = 195$). All secondary reference materials values were in good agreement with values from a previous interlaboratory comparison (Reynolds et al., 2007). The reproducibility of all samples was $<0.2\text{‰}$. At the Vegacenter laboratory, the long-term precision is 0.15‰ (expressed as 2SD).

Statistical analyses

For testing the statistical significance of proxy correlations, a Pearson correlation test was run on data in R software. A statistically significant correlation was considered to have a confidence interval of 95% and thus, the p -value < 0.05 . The correlation tests were run on the entire core record, as well as on three chosen zones. The $\delta^{30}\text{Si}_{\text{BSi}}$, BSi, LOI, TOC or OC proxy, Ti, Fe and MS were tested for correlation to identify processes connected to the $\delta^{30}\text{Si}_{\text{BSi}}$ signature and variation of BSi. The correlation results are heavily dependent on the alignment of two cores, thus a significant correlation between LOI and the OC proxy, which supports the robustness of the core alignment, was tested before any other correlation tests.

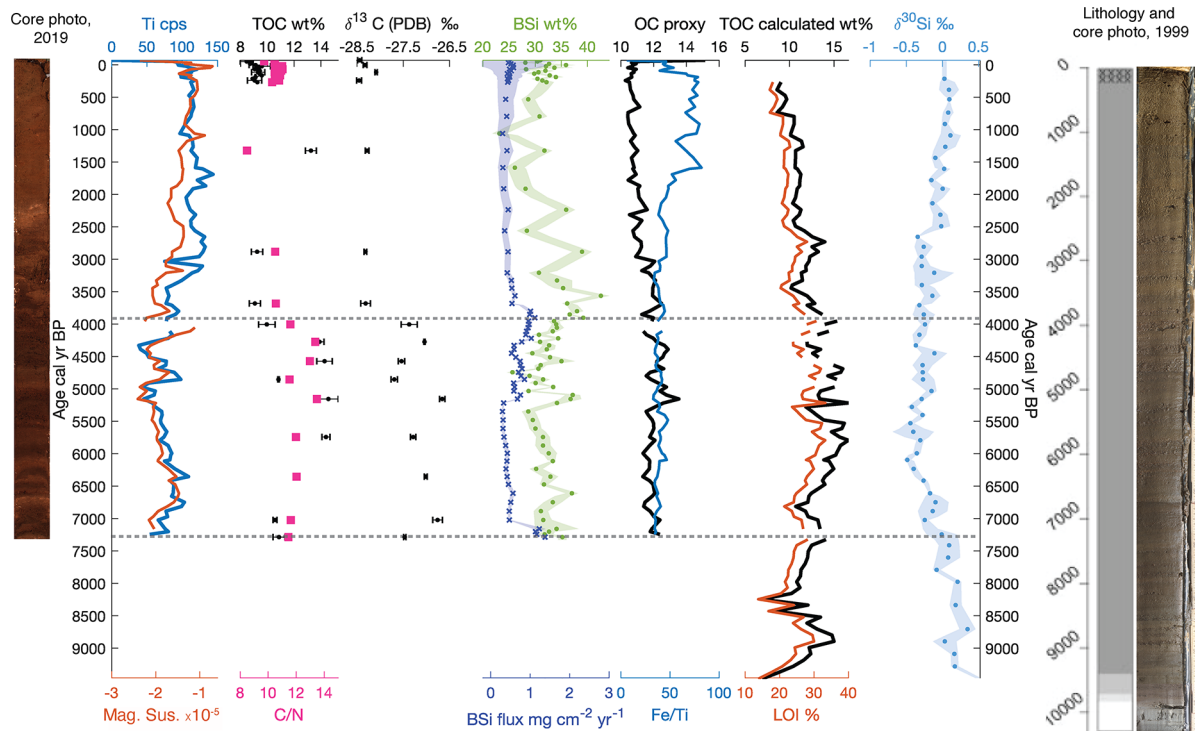


Figure 3. On the left, data for the 2019 piston core: core photo with lithology, titanium (Ti in cps) as a detrital input proxy, magnetic susceptibility (MS) as the grain size changes proxy, total organic carbon (TOC wt%), C/N ratios and $\delta^{13}\text{C}$ of bulk sediment, biogenic silica (BSi) as SiO_2 weight percentage of dry sediment, BSi fluxes ($\text{mg cm}^{-2} \text{yr}^{-1}$), the ($R_{\text{inc}}/R_{\text{coh}}$) as an additional OC proxy and Fe/Ti as an additional detrital input proxy. On the right, data for the core from 1999: TOC (wt%) calculated from LOI and LOI% (Rubensdotter and Rosqvist, 2003) are used for correlation of the two cores, stable silicon isotope record ($\delta^{30}\text{Si}$ in ‰) of cleaned bulk diatoms with shading of 1 SD from replicates, lithology and photo of the 1999 core modified from Rubensdotter and Rosqvist (2003), where dark-grey colour represents gyttja, lighter grey layer is clay-gyttja, the lightest grey is gyttja-clay and the white section is clay/silt. Horizontal dashed lines show early, mid- and late Holocene boundaries based on similar lakes in the Scandes mountain range (Bigler and Hall, 2003; Rosén et al., 2001).

Results

Age-depth model and core alignment

The base of the 2019 piston core was dated at approximately 7400 cal. yr BP (Table 1). The mean sedimentation rate over the entire core was estimated to be 0.012 cm yr^{-1} or 83.3 yr cm^{-1} (Figure 2), which is in the same range as a previously published sedimentation rate from Lake 850 of 0.013 cm yr^{-1} (Shemesh et al., 2001). Based on this sedimentation rate, the mass accumulation rate was calculated to be $1.8 \text{ mg cm}^{-2} \text{yr}^{-1}$. However, the age-depth model is not linear, especially in the top 28 cm. The increased sedimentation rate in the top of the core has a high uncertainty, as no age constraint was obtained between the surface and 28 cm. The top of the core is considered to have the age of the coring year. However, the sediment-water interface was disturbed during coring and thus, the age-depth model can suffer from the incompleteness of the record. The age-depth model of a short core from Lake 850 also shows non-linear changes in sedimentation rate in the surface of the core (Zahajská et al., 2021b).

A new age-depth model of the published core from 1999 (Supplemental Figure S1) indicates that the base of the core is approximately 9400 cal. yr BP and shows linear changes in sedimentation rate through time, which is in agreement with the previous Holocene age-depth model (Rubensdotter and Rosqvist, 2003; Shemesh et al., 2001).

Lithology

The 74-cm 2019 sediment core was examined for its lithology, elemental composition using XRF, total organic carbon (TOC), total nitrogen (TN), biogenic silica (BSi) and magnetic susceptibility. Low and negative values of magnetic susceptibility show

that the whole core is diamagnetic, which indicates that the core is composed of clay and/or pure organic. This result corresponds well with the sediment composition of the previously published 125-cm sediment core (Shemesh et al., 2001), which lithology is predominantly carbonate-free clay gyttja. The sediment porosity (data not shown) is generally high, averaging 86%, with maximum values of 90%. The wet bulk density (data not shown) is 1.15 g cm^{-3} , and the dry bulk density is 0.16 g cm^{-3} .

The XRF measurements generally showed low counts, that is, below 1000 counts per second, for the majority of elements, which may indicate low allochthonous input. Therefore, the magnetic susceptibility (MS) and the titanium (Ti) and iron normalized by Ti (Fe/Ti) XRF data are used as proxies for changes in grain-size and detrital/terrigenous input into the sediment, respectively (Figure 3). The Fe/Ti ratio was considered to represent detrital input as iron XRF counts were 10–100 times higher than all other identified elements (Figure 3), thus all trends found in redox proxies, such as Fe/Mn or Fe/Ti, are strongly influenced by the Fe counts, which show trends similar to MS. Titanium, iron and magnetic susceptibility show a generally increasing trend from the base to the top of the core. An increase in all three proxies is observed, especially in the last 3000 cal. yr BP. In contrast, the TOC and C/N data show elevated values from 7400 to 3500 cal. yr BP and are generally stable during the last 3500 cal. yr BP. Total organic carbon and BSi share some similarities in their trends, with increased values from 7400 to 3500 cal. yr BP, followed by lower values in last 3500 cal. yr BP. However, BSi varies greatly, whereas TOC and C/N display smaller variations. BSi concentrations and TOC together constitute from 38 wt% to 51 wt% of the sediment. The remainder of the sediment is considered to be minerogenic.

The non-minerogenic component of the sediment is used as a proxy for changes in productivity or terrigenous input. TOC varies greatly throughout the core, from $8.4 \pm 0.5 \text{ wt} \%$ to $14.6 \pm 0.7 \text{ wt} \%$ with a mean of $11 \pm 2.2 \text{ wt} \%$. One standard deviation is used as uncertainty when data are presented. Typical C/N values for autochthonous aquatic production range from 4 to 10, whereas terrestrial plants have $\text{C/N} > 20$ (Meyers, 1994). Thus, our values, which range from 8.5 to 13.5, represent the dominance of autochthonous sources. Stable carbon isotopes measured on bulk sediments support an autochthonous origin of carbon, with values typical for algal production (Meyers, 1994; Meyers and Lallier-Vergés, 1999) of $\delta^{13}\text{C}$ ranging between -28.5‰ and -27.7‰ , with mean at -27.7‰ .

The BSi shows large variability, with the BSi minimum of $23.2 \pm 1.3 \text{ wt} \%$ SiO_2 at 2280 cal. yr BP, maximum of $39.2 \pm 0.7 \text{ wt} \%$ at 3900 cal. yr BP and the core mean of BSi is $32.3 \pm 3.0 \text{ wt} \%$. A distinct local minimum in BSi concentration occurs at 4750 cal. yr BP, with BSi of $25.7 \pm 1.0\%$. Elevated BSi concentrations above 30 wt% are observed ~ 2800 cal. yr BP, 3800 cal. yr BP, 5100 cal. yr BP and 6600 cal. yr BP. The minerogenic fraction resulting from the BSi and TOC measurements varies between 48 wt% and 67 wt%.

Biogenic silica fluxes

The Holocene BSi flux into the sediment was determined from the piston core BSi and the mass accumulation rate of $1.85 \text{ mg cm}^{-2} \text{ yr}^{-1}$. The mean BSi flux into the sediment from 7300 cal. yr BP to present is $0.60 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$ (Figure 3). At the bottom of the core, the BSi flux is higher, with values up to $1.38 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$, and with a mean of $1.23 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$ from the period 7300 to 7100 cal. yr BP. From approximately 7100 cal. yr BP to 5200 cal. yr BP, the BSi flux stabilizes at a mean of $0.40 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$. An increasing trend in the BSi accumulation rate during younger ages is observed from 5200 cal. yr BP to 3700 cal. yr BP, with values varying from $0.53 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$ up to $1.12 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$, with a mean of $0.75 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$. From 3700 cal. yr BP to present, the BSi flux is stable at $0.46 \text{ mg SiO}_2 \text{ cm}^{-2} \text{ yr}^{-1}$, with some minor variation (Figure 3). Large uncertainties in the top part of the core are propagated from the age-depth model, where ages are extrapolated between the youngest ^{14}C date and the top of the core, which is considered to be recent.

Stable Si isotope record

The stable silicon isotopes measured on cleaned bulk diatoms (Supplemental Figure S2) from the 125-cm-long core from 1999 show generally light $\delta^{30}\text{Si}_{\text{BSi}}$ of preserved diatoms with subtle variations. A minor presence of chrysophytes does not affect the $\delta^{30}\text{Si}_{\text{BSi}}$ values as it was shown that chrysophytes likely fractionate silicon comparably to diatoms because of the similarity between silicon transporter SIT genes (Marron et al., 2013). Three Holocene periods with different trends (Figure 3) are distinguished based on records from similar lakes in the Scandes mountain range: Sjuodjijäure (Rosén et al., 2001) and Njulla (Bigler et al., 2003). During the early Holocene ($\sim 9400 - 7300$ cal. yr BP), there is a decreasing trend from heavier $\delta^{30}\text{Si}_{\text{BSi}}$ to lighter $\delta^{30}\text{Si}_{\text{BSi}}$ from the base upward, with the heaviest $\delta^{30}\text{Si}_{\text{BSi}} = 0.51 \pm 0.03\text{‰}$ (1SD, $n = 2$) at the bottom of the core, a mean $\delta^{30}\text{Si}_{\text{BSi}}$ of $0.17 \pm 0.16\text{‰}$ (1SD, $n = 10$) and shifts between two subsequent samples $< 0.34\text{‰}$ (Figure 3). Uncertainties are expressed as one standard deviation from replicates or total numbers of samples in the case of means. Analytical errors for individual $\delta^{30}\text{Si}_{\text{BSi}}$ measurements, expressed as 2SD, are shown in Supplemental Figure S3 and can be found in the source data Zahajská et al. (2021a). The mid-Holocene (7300 – 3900 cal. yr BP) has a light $\delta^{30}\text{Si}_{\text{BSi}}$ signal that oscillates very little ($\pm 0.25\text{‰}$), including the

lightest signal in the entire core ($\delta^{30}\text{Si}_{\text{BSi}} = -0.49 \pm 0.31\text{‰}$, 1SD, $n = 2$) and mean values of $-0.27 \pm 0.12\text{‰}$ (1SD, $n = 24$). In the late Holocene $\delta^{30}\text{Si}_{\text{BSi}}$ values are lighter and stable between 3900 cal. yr BP and 2700 cal. yr BP, ranging between -0.34‰ and -0.11‰ with a small shift between two subsequent samples of 0.18‰ . The topmost part of the core from 2700 cal. yr BP to present shows a gradual shift towards heavier $\delta^{30}\text{Si}_{\text{BSi}}$, with values ranging from -0.15‰ to 0.11‰ and varying by 0.18‰ . The core $\delta^{30}\text{Si}_{\text{BSi}}$ mean is $-0.19 \pm 0.16\text{‰}$ (1SD, $n = 56$).

Statistical analysis

Pearson correlation tests conducted on the entire core record of XRF, LOI, OC, TOC, BSi and $\delta^{30}\text{Si}_{\text{BSi}}$ data showed weak or no correlation between $\delta^{30}\text{Si}_{\text{BSi}}$, BSi and the proxy for production (OC, LOI, TOC) or the proxy for detrital input (MS, Ti). All data show a normal distribution, and our linear regression models were tested for residuals and variance.

Magnetic susceptibility, reflecting changes in grain-size, is positively correlated with the detrital input proxy – Ti ($R = 0.74$, $p\text{-value} < 2.2 \cdot 10^{-16}$, residual SD = 0.26, degrees of freedom (df) = 97), except at a few depths, where it may be more affected by organic matter content or sediment of autochthonous origin (Figure 3). Similarly, Fe is positively correlated with MS ($R = 0.76$, $p\text{-value} < 2.2 \cdot 10^{-16}$, residual SD = 0.24, df = 97), which suggests that MS, Fe and Ti similarly record detrital input. Mineral input is generally low, based on the low XRF counts of detrital elements and on the sedimentation rate. Further, the negative correlation between OC and density ($R = -0.52$, $p\text{-value} = 5.18 \cdot 10^{-6}$, residual SD = 0.73, df = 69) suggests that OC matter content may be responsible for the changes in the sediment density. Thus, 26% of the variation in sediment density can be attributed to organic carbon variation and 25% to detrital input represented by Ti ($R^2 = 0.49$, $p\text{-value} = 8.8 \cdot 10^{-6}$, residual SD = 21.3, df = 69). A few pulses of detrital input, identified by elevated MS, Ti and Fe, occur during the late and mid-Holocene periods around 4700 cal. yr BP, 3000 cal. yr BP and 1700 cal. yr BP.

A strong positive correlations between Fe, MS and Ti together with the negative correlation between BSi and Fe ($R = -0.26$, $p\text{-value} = 0.03$, residual SD = 3.2, df = 69) suggests that iron can be used as a proxy of detrital material which dilutes the diatom BSi in the sediment. Lower iron content occurs during the mid-Holocene period, together with higher BSi accumulation.

Discussion

Factors influencing the diatom accumulation and $\delta^{30}\text{Si}_{\text{BSi}}$

To explain sedimentary $\delta^{30}\text{Si}_{\text{BSi}}$, possible factors influencing the source of DSi to the system in which the diatoms grew must be considered. In DSi-limited systems, $\delta^{30}\text{Si}_{\text{BSi}}$ reflects diatom production limited by processes in the watershed and relative changes in DSi sources (Nantke et al., 2019; Phillips, 2020; Zhang et al., 2020). Factors influencing the $\delta^{30}\text{Si}_{\text{BSi}}$ in a lake include riverine diatom production, vegetation, the soil Si pool, groundwater input and secondary mineral formation (Frings et al., 2016; Opfergelt and Delmelle, 2012; Sutton et al., 2018; Zahajská et al., 2021b). Lake 850 is situated above the pine and birch treeline and is surrounded by bedrock with very sparse pockets of poorly developed soils. The primary water input to the lake is from groundwater with additional contributions from small ephemeral inlets (Zahajská et al., 2021b). Thus, none of those factors are likely to have had an influence on the $\delta^{30}\text{Si}$ of the source DSi, although these factors are important in environments below the treeline (Fontorbe et al., 2013; Frings et al., 2016; Nantke et al., 2019).

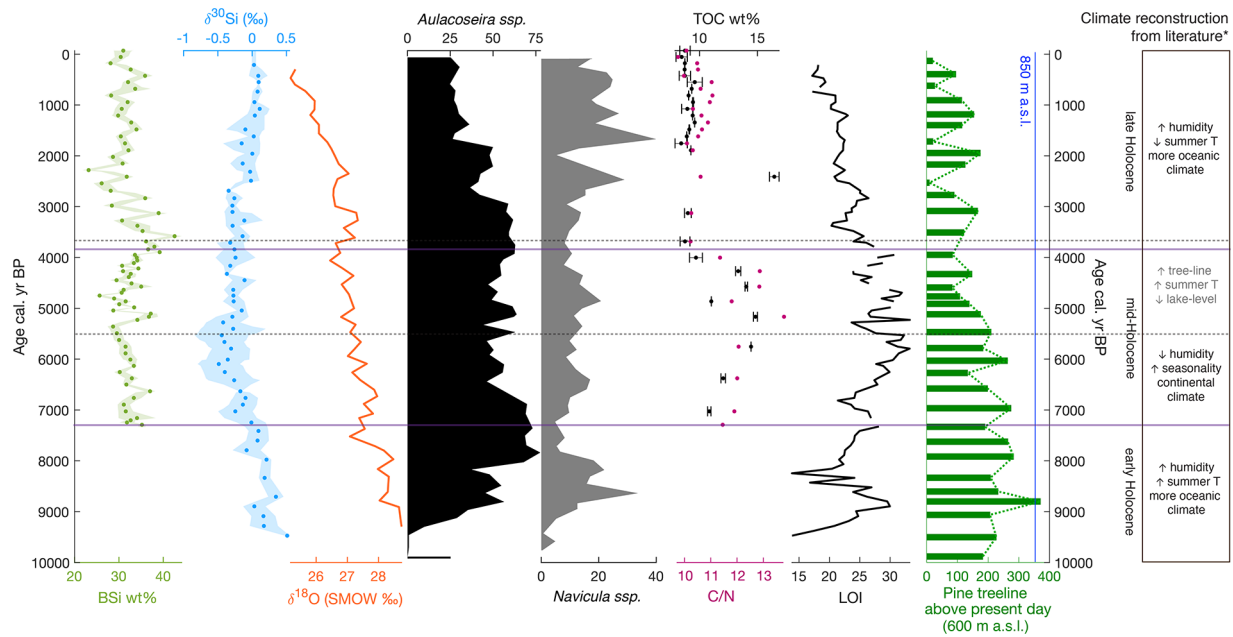


Figure 4. Summary of BSi wt%, TOC wt% and C/N ratios from the 2019 core and $\delta^{30}\text{Si}$ (‰) from the 1999 core according to ages and compared with previously published $\delta^{18}\text{O}$ (‰ VSMOW), *Aulacoseira ssp.* and *Navicula ssp.* abundances from the 1999 core (Shemesh et al., 2001). Further, pine treeline reconstruction at northern Scandinavia (Shemesh et al., 2001).

*Climate and vegetation reconstruction based on Barnekow (1999); Berglund et al. (1996); Hammarlund et al. (2002); Rosén et al. (2001); Seppä and Hammarlund (2000).

In DSi-unlimited systems, such as Lake 850, diatom accumulation and $\delta^{30}\text{Si}_{\text{BSi}}$ are likely reflecting several processes acting together. More specifically:

1. changes in diatom production and species composition derived from changes in summer temperatures and the length of the ice-free period, and
2. relative changes of DSi sources (groundwater vs surficial streams).

The Holocene lake history suggests that a combination of low detrital input (Ti, MS) and high BSi production are governing the diatom-rich sediment formation (Figure 3). The detrital input is low and thus, minerogenic particles do not dilute the amount of diatom settled in the lake floor, and BSi concentrations as high as 42 wt% were found. Additionally, many of the processes influencing $\delta^{30}\text{Si}_{\text{BSi}}$ and BSi concentration are partially controlled by changes in climate (Figure 4). Even though, the core age-depth model shows non-linear behaviour, the majority of the record is linear. Thus, we assume that BSi concentrations represents the changes in BSi accumulation (flux), and we used BSi concentration instead of flux in our interpretation.

Climate reconstructions in the Abisko area suggest a warmer and more humid early Holocene influenced by Atlantic air masses (Berglund et al., 1996; Rosén et al., 2001), more continental climate with pronounced seasonality and increased elevation of the pine treeline during the mid-Holocene (Barnekow, 1999; Berglund et al., 1996; Hammarlund et al., 2002; Rosén et al., 2001), cooling connected with decreasing insolation (Barnekow, 1999) reflected in $\delta^{18}\text{O}$ (Figure 4, Shemesh et al., 2001) in the late Holocene. Although $\delta^{30}\text{Si}_{\text{BSi}}$ data show a pattern that coincides with intervals of change in Holocene climate as described above, the $\delta^{18}\text{O}$ data from Lake 850 do not consistently follow the same patterns (Figure 4, Shemesh et al., 2001). In addition, $\delta^{30}\text{Si}_{\text{BSi}}$ does not show substantive changes coincident with fluctuations in treeline (Figure 4), which suggests that vegetation is not driving changes in $\delta^{30}\text{Si}_{\text{BSi}}$.

Diatom production and species composition

No significant correlation of BSi and $\delta^{30}\text{Si}_{\text{BSi}}$ was found throughout the Holocene, therefore, diatom production is not the only factor driving $\delta^{30}\text{Si}_{\text{BSi}}$, which is in agreement with the hypothesis that $\delta^{30}\text{Si}_{\text{BSi}}$ formed in DSi-unlimited lakes does not reflect DSi utilization. However, reported changes in diatom production and species composition (Shemesh et al., 2001) may influence variation in $\delta^{30}\text{Si}_{\text{BSi}}$ and BSi concentration, as indicated by the similarity in trends. Planktic *Aulacoseira ssp.* have lower relative abundance in the early and late Holocene, when benthic *Navicula ssp.* abundances increased (Figure 4, Shemesh et al., 2001). BSi concentrations are higher during the mid-Holocene in comparison with a decreasing trend in the late Holocene, which coincides with changes in the abundance of heavily silicified *Aulacoseira ssp.* Additionally, increased diatom production and thus the BSi concentration during mid-Holocene was possibly triggered by a longer growing season, which is suggested by large abundance of planktic species (*Aulacoseira ssp.*, Figure 4) and lower amounts of benthic species (*Fragilaria ssp.*, see Shemesh et al., 2001) (Lotter and Bigler, 2000; Smol, 1988), as well as by warmer and drier climate indicated in the heavier $\delta^{18}\text{O}$ values (Figure 4, Shemesh et al., 2001). Stable $\delta^{30}\text{Si}_{\text{BSi}}$ has heavier values during the early and late Holocene, which may be driven by differential species-specific fractionation (Panizzo et al., 2016; Sutton et al., 2013; Schmidtbauer et al., in review) or heavier DSi sources, such as pore waters at the water-sediment interface (Ehlert et al., 2016; Geilert et al., 2020; Ng et al., 2020; Schmidtbauer et al., in review) for benthic species, whereas a lighter isotopic signature can be connected to the higher relative abundance of planktic *Aulacoseira ssp.* (Figure 4). However, there are no direct measurements showing the species-specific fractionation in Lake 850, and further studies need to be done in order to support this interpretation. Further, based on a recent study of the Si budget of Lake 850, which shows a large contribution of an additional DSi source in form of groundwater (Zahajská et al., 2021b), we assume that the variation in $\delta^{30}\text{Si}_{\text{BSi}}$ is driven by relative changes in DSi sources rather than species-specific fractionation.

Relative changes in DSi sources

Changes in DSi sources have been demonstrated to influence $\delta^{30}\text{Si}_{\text{BSi}}$, however, in other studies those changes were connected to changes in watershed vegetation or soil development (Fontorbe et al., 2013; Frings et al., 2016; Nantke et al., 2019). Analysis of recent temporal dynamics in Lake 850 revealed the importance of groundwater supply as a source of DSi fuelling high diatom growth (Zahajská et al., 2021b). The isotopic signature of the diatoms in Lake 850 shows no significant correlation with the wt% of BSi or BSi accumulation rate, which supports the hypothesis that the $\delta^{30}\text{Si}_{\text{BSi}}$ is not connected to production but instead is driven by changes in DSi sources (Nantke et al., 2019). Relative changes in DSi sources during the diatom growing season measured in groundwater (ranging from -0.55‰ to 0.24‰ , mean -0.07‰) and surficial run-off (ranging from 0.02‰ to 0.77‰ , mean 0.5‰) in the modern Lake 850 (Zahajská et al., 2021b) are suggested to be responsible for contemporary $\delta^{30}\text{Si}_{\text{BSi}}$ values. Moreover, the $\delta^{30}\text{Si}_{\text{BSi}}$ values are within a narrow range throughout the Holocene, therefore, a stable DSi source must be present through time. The light $\delta^{30}\text{Si}_{\text{BSi}}$ during the Holocene reflects the absence of processes that fractionate Si in the lake watershed, such as soils or vegetation.

Holocene climate reconstructions of the Abisko area suggest changes in summer temperatures (Barnekow, 1999; Berglund et al., 1996; Bigler et al., 2003; Hammarlund et al., 2002; Rosén et al., 2001; Seppä and Hammarlund, 2000), which may have influenced the length of the ice-free period and in turn diatom growth. Thus, Holocene $\delta^{30}\text{Si}_{\text{BSi}}$ may reflect changes in the duration of the ice-free period and associated changes in the relative proportion of surface inputs versus groundwater as DSi sources. Changes in summer temperatures are connected with changes in vegetation cover, where shrub dominance increases during warmer periods (Myers-Smith et al., 2015). Even though the lake was shown to be above the pine treeline in last 8000 cal. yr BP (Shemesh et al., 2001), the expansion or contraction of shrub vegetation can trigger feedback processes, such as development of soils and/or localized snow patches (Myers-Smith et al., 2011), which can influence the DSi delivery to the lake.

Holocene development of Lake 850

Early Holocene—Shift in DSi sources. No BSi and XRF data are available for the early Holocene period, however, the $\delta^{30}\text{Si}_{\text{BSi}}$ record combined with LOI data and the treeline increase (Figure 4) are consistent with a warm and humid climate. Climate reconstructions from diatoms, pollen and chironomids for the Abisko region suggest the influence of Atlantic air masses creating more humid and oceanic climate (Berglund et al., 1996; Rosén et al., 2001). The decreasing $\delta^{30}\text{Si}_{\text{BSi}}$ (Figure 4) can be associated with prolonged ice-free periods, as recorded in an increase of planktic diatom species (Figure 4) and in the $\delta^{18}\text{O}$ data reflecting changes in isotopic composition of the influx water originating from precipitation or changes in water balance (Shemesh et al., 2001). Thus, changes in the relative proportion of DSi sources from surficial-dominated DSi input relative to groundwater-dominated DSi sources, would result in the isotopic shift towards lighter $\delta^{30}\text{Si}_{\text{BSi}}$ based on recent mass balance of the lake (Zahajská et al., 2021b). Indeed, groundwater represents three times more water input compared to ephemeral streams today and has a modelled isotopic signature between -0.55‰ and 0.24‰ (Zahajská et al., 2021b). The relative proportion of DSi sources are connected to changes in summer temperature and precipitation.

Mid-Holocene—lake dominated by diatom production and groundwater. Climate reconstructions from other sites in the region suggest a reduced influence of Atlantic air masses, thus a more

continental climate with lower humidity, more pronounced seasonality and colder winters (Barnekow, 1999; Berglund et al., 1996; Hammarlund et al., 2002; Rosén et al., 2001). Yet, $\delta^{18}\text{O}$ data from Lake 850 suggest cooling and higher precipitation in summer (Figure 4, Shemesh et al., 2001).

High and variable TOC together with increased C/N (Figure 4) suggest more terrestrial organic carbon input, especially during the altitudinal increase of treeline in the Abisko area between 5500 and 3400 cal. yr BP (Barnekow, 1999; Berglund et al., 1996; Hammarlund et al., 2002; Rosén et al., 2001; Seppä and Hammarlund, 2000). This increase in treeline corresponds with an increase of summer temperature documented in nearby Lake Tibetanus (Barnekow, 1999; Seppä and Hammarlund, 2000) and Lake Sjuodjijäure (Rosén et al., 2001). However, the pine treeline did not reach an altitude of 850 m a.s.l. (Figure 4), and the detrital input (Ti, MS, Fe) during this period is at a minimum and negatively correlated with TOC. Together these data suggest that the total organic carbon likely originates from the shoreline and littoral areas of the lake, supported by the $\delta^{13}\text{C}$ values characteristic for lacustrine algal production (Meyers, 1994; Meyers and Lallier-Vergés, 1999).

Additionally, lake-level changes evident in other lakes in Scandinavia (Seppä and Birks, 2002, and references therein) suggest the possibility that lake shallowing and expanded growth of terrestrial mosses in littoral areas could account for both increased TOC and C/N but low detrital input. A slight increase in benthic *Navicula* ssp. is consistent with the hypothesis of moderate lake-level change and expanded littoral habitat. Alternatively, the low detrital input could be explained by lower run-off due to a generally drier period.

The diatom assemblage (Figure 4) does not show substantial changes, and the prolonged dominance of heavy silicified *Aulacoseira* ssp. may be responsible for the high BSi concentrations as a result of a prolonged ice-free period (Bigler et al., 2002). The recent Lake 850 mass-balance models showed that a prolonged ice-free period increases the relative contribution of isotopically lighter groundwater DSi to the lake water balance and produces isotopically lighter $\delta^{30}\text{Si}_{\text{BSi}}$ (Zahajská et al., 2021b). Therefore, BSi concentration can be connected to an unlimited isotopically light DSi source for diatom uptake (reflected in $\delta^{30}\text{Si}_{\text{BSi}}$), coupled with a long ice-free period, as indicated by the dominance of *Aulacoseira* ssp. (Figure 4), associated with higher mid-Holocene summer temperatures.

Late-Holocene—increased run-off and shortening of ice-free period. The oscillations of BSi concentration during the late Holocene reflect changes in sedimentation due to a slight increase in detrital input (Ti, Fe, MS, Figure 3), coincident with a C/N ratio that suggests increased autochthonous production. Thus, detrital input is likely to be of mineral composition originating from enhanced physical weathering during spring snowmelt with little organic carbon. A positive correlation was found between BSi concentration and the OC proxy ($R^2 = 0.32$, p -value = $7 \cdot 10^{-3}$), which indicates that BSi concentration is correlated with autochthonous algal production during this period, supported by C/N and $\delta^{13}\text{C}$ values. However, BSi and $\delta^{30}\text{Si}_{\text{BSi}}$ in this period show no significant correlation, therefore, other processes, such as changes in DSi sources, changes in diatom production connected to relative species abundance and preservation potential are responsible for the $\delta^{30}\text{Si}_{\text{BSi}}$ signature. The decreasing trend in OC and LOI, similar to BSi, suggest changes in diatom production likely driven by a shorter ice-free period.

During the late Holocene, a gradual shift towards heavy $\delta^{30}\text{Si}_{\text{BSi}}$ shows a trend similar to that of the cooling climate reflected in $\delta^{18}\text{O}$ (Figure 4), driven by decreasing summer insolation (Barnekow, 1999) and a more oceanic climate (Berglund et al., 1996). These changes in climate are responsible for a

shorter ice-free period, as reflected in decreasing abundances of *Aulacoseira* spp., which grows during longer ice-free periods in a well-mixed water column (Figure 4). Longer winters are responsible for more meltwater, a higher contribution of spring stream water and increased erosion (Rosén et al., 2001; Snowball et al., 1999). A shorter ice-free period is connected with the input of relatively heavier groundwater $\delta^{30}\text{Si}_{\text{BSi}}$ (Zahajská et al., 2021b).

Additionally, during the spring snow melt a higher relative contribution of run-off and stream DSi supply is expected to influence the lake $\delta^{30}\text{Si}$ signature. Further, if the BSi fluxes to the sediment are similar to the longer ice-free period, to accumulate the same BSi during the short ice-free period, the diatom production must be enhanced. Thus, higher DSi uptake during the shorter diatom growth can be reflected in the increased $\delta^{30}\text{Si}_{\text{BSi}}$ (Zahajská et al., 2021b). Alternatively, the higher relative abundance of benthic species *Navicula* spp. can be hypothetically responsible for the trend in $\delta^{30}\text{Si}_{\text{BSi}}$.

Sparse $\delta^{30}\text{Si}$ sediment records. Approximately, 100 lakes in Northern Sweden have been investigated for BSi concentrations in surface sediments (Rosén et al., 2010), including Lake 850, but no $\delta^{30}\text{Si}$ analyses have been made on any of these lakes. Lake 850 has a surface sediment BSi concentration of 40.3wt%, which places this lake into the top 10% of high BSi concentrations among the 100 lakes studied, and it is the only lake above current treeline with high BSi concentrations (based on data from Rosén et al., (2010)). The closest lake in Lappland with a stable silicon isotopic record is Lake Kuutsjärvi, situated below the pine treeline at 341 m a.s.l. (Tallberg et al., 2015). The top 25 cm of sediment from Lake Kuutsjärvi has a stable silicon isotopic signature of sedimentary diatoms similar to the uppermost sediment of Lake 850, although unlike this study, for Lake Kuutsjärvi the $\delta^{30}\text{Si}_{\text{BSi}}$ was calculated and not measured directly on diatoms.

The mean $\delta^{30}\text{Si}_{\text{BSi}}$ in Lake 850 is at the lower end of the global marine and freshwater diatom isotopic composition range (Frings et al., 2016). The $\delta^{30}\text{Si}_{\text{BSi}}$ values throughout the core in this study are generally lighter than published lacustrine $\delta^{30}\text{Si}_{\text{BSi}}$ from the Arctic Lake El'gygytgyn, where $\delta^{30}\text{Si}_{\text{BSi}}$ ranges from 0.9‰ to 1.4‰ (Swann et al., 2010), and Lake Baikal $\delta^{30}\text{Si}_{\text{BSi}}$, where values range from 1‰ to 1.5‰ (Panizzo et al., 2016).

The isotopically light signature of fossil diatoms from Lake 850 (mean -0.19 ± 0.16 ‰) is comparable with $\delta^{30}\text{Si}_{\text{BSi}}$ in volcanic lakes (Chen et al., 2012; Street-Perrott et al., 2008). The more similar isotopic signature of diatoms from the sediments of volcanic Lake Huguangyan, where $\delta^{30}\text{Si}_{\text{BSi}}$ ranges from -0.6 ‰ to 1.1‰ (Chen et al., 2012), and Lake Rutundu, where values span from -1.3 ‰ to 0.5‰ (Street-Perrott et al., 2008), suggest that the DSi sources carry isotopically lighter signature originating from freshly weathered bedrock and lack of processes that would fractionate the DSi.

In Lake Huguangyan, the $\delta^{30}\text{Si}_{\text{BSi}}$ is driven by diatom production rather than changes in DSi supply (Chen et al., 2012), however, as it lies in a crater and has small watershed, the DSi likely carries isotopically light DSi due to weathering of basaltic bedrock. Lake Rutundu is a small maar lake at a high-altitude with an isotopically light DSi source originating in lavas and tuffs (-0.4 ‰ to -0.5 ‰, Ding et al., 1996). In Lake Rutundu, the changes in maximal $\delta^{30}\text{Si}_{\text{BSi}}$ are explained by full DSi utilization by diatom production, and the minimal $\delta^{30}\text{Si}_{\text{BSi}} \sim -1.3$ missing ‰ is interpreted to originate from isotopically light DSi enriched groundwater supply during low lake-level (Street-Perrott et al., 2008). Therefore, the interpretations of these two lakes are consistent with the hypothesis that the $\delta^{30}\text{Si}_{\text{BSi}}$ in Lake 850, ranging from -0.49 ‰ to 0.51‰, is influenced by an isotopically light DSi source originating from groundwater (Zahajská et al., 2021b).

Conclusions

Changes in detrital input and TOC suggest that increased run-off and changes in summer temperatures are responsible for the long-term variation in BSi concentration. The diatom-rich sediment in Lake 850 is formed due to low sedimentation rates and a stable DSi source that supports diatom growth, which is not DSi-limited. The Holocene environmental changes in the catchment, such as increases in treeline recorded by other proxies, are not reflected in $\delta^{30}\text{Si}_{\text{BSi}}$. Instead, diatom production, species composition and changes in the relative proportions of DSi sources driven by climate forcing influence the $\delta^{30}\text{Si}_{\text{BSi}}$.

The Holocene history of Lake 850 is influenced by climate-inducing changes in the relative proportions of DSi sources, as well as changes in the length of growing season, and thus diatom species composition and diatom production. The lighter Si isotopic signature of the diatoms during the mid-Holocene suggests higher groundwater input bringing isotopically lighter DSi into the lake compared to the early- and late Holocene. The late Holocene Si isotopic composition of the diatoms is consistent with changes in climate documented in $\delta^{18}\text{O}$, driving shortening of the growing season (reflected in diatom species composition) and increased surface run-off that dilutes groundwater input, resulting in heavier Si isotopes.

Additionally, the $\delta^{30}\text{Si}_{\text{BSi}}$ signature throughout the Holocene varies within a narrow range suggesting stable environmental conditions and a continuous sufficient supply of DSi. The $\delta^{30}\text{Si}_{\text{BSi}}$ record of Holocene diatoms has a light signature (mean -0.19 ± 0.16 ‰), comparable to the diatom Si signature in volcanic lakes (ranging from -1.3 ‰ to 1.1‰; Chen et al., 2012; Street-Perrott et al., 2008). This suggests that Lake 850 is fed by isotopically lighter DSi sources, such as a water source with time-limited contact with bedrock and that is transported over a short distance (mean of measured samples ~ 0.5 ‰; Zahajská et al., 2021b) and groundwater (mean of modelled $\delta^{30}\text{Si}_{\text{BSi}} \sim -0.07$ ‰; Zahajská et al., 2021b) compared to mean freshwater $\delta^{30}\text{Si}$ (1.26‰; Sutton et al., 2018) during the Holocene.

This study shows that combining BSi concentration and the $\delta^{30}\text{Si}_{\text{BSi}}$ in the sediment has the potential to identify DSi sources responsible for diatom production and accumulation. Comparing our data with other, still sparse, lacustrine sedimentary $\delta^{30}\text{Si}_{\text{BSi}}$ data reveals the importance of local factors and processes that affect the lake Si cycle. More studies on lacustrine sedimentary $\delta^{30}\text{Si}_{\text{BSi}}$ are needed to improve the estimates of the sinks and sources in aquatic ecosystems.

Acknowledgements

We thank the organizations and the individuals who helped with the fieldwork and/or provided us with equipment and/or advice: Thomas Westin, Keith W. Larson, Erik Lundin, Svante Zachrisson, CIRC, Christian Bigler, Lena Rubensdotter, Gunhild Rosqvist, field assistants Carla Nantke and Ethan Silvester. We thank Mats Rundgren for help with ^{14}C samples evaluation and species identification. We acknowledge Hans Schöberg and Melanie Kielman for assistance during sample preparation and isotope data acquisition. This is Vegacenter contribution number # 043.

Funding

The author(s) disclosed receipt of the following financial support for the research, authorship, and/or publication of this article: This work was supported by The Royal Physiographic Society in Lund and by the Center for Geosphere Dynamics (UNCE/SCI/006) to PZ, the Swedish Research Council to DJC, the Swedish Research Council Tage Erlander Professorship to SCF, and the “Fonds National de la Recherche Scientifique” (FNRS, Belgium, FC69480) to SO.

Data accessibility

All data, if not directly available in tables and supplementary materials, are available in the PANGAEA database DOI:10.1594/PANGAEA.929941.

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Supplemental material

Supplemental material for this article is available online.

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