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# Tracking mineral and geochemical characteristics of Holocene lake sediments: the case of Hotagen, west-central Sweden

Hussein Jaafar Kanbar<sup>1</sup> · Thai Tran Le<sup>1</sup> · Fredrik Olajos<sup>2</sup> · Göran Englund<sup>2</sup> · Michael Holmboe<sup>1</sup>

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## Abstract

**Purpose** Intact lake sediments reflect the development of terrestrial ecosystems. This development can be understood by decoding mineral and geochemical information of sedimentary archives. Therefore, we characterized a Holocene lake sediment core and revealed bulk to micro-scale variations via a combination of geochemical techniques and statistical methods.

**Methods** A 2.3 m sediment core was collected from Hotagen, a lake in west-central Sweden; a sediment sample was collected every 5 cm. A part of each sediment sample was kept untreated (named bulk) and another part was size-fractionated into <4, 4–16, 16–64, and >64 μm subsamples. Characterization was then made with respect to grain size distribution (GSD), physico-chemical parameters, geochemical properties, organic composition, and mineralogy. The sediments were investigated at bulk, micro-, and elemental scales using powder X-ray diffraction (XRD), diffuse reflectance infrared Fourier transform spectroscopy (DRIFT), and scanning electron microscopy coupled to energy-dispersive X-ray spectroscopy (SEM–EDX).

**Results** The deepest sediment was identified as glacial till dating back to the Late Pleistocene. The bulk sediments showed a clear distinction between 0–195 cm (unit 1, U1) and 200–225 cm (unit 2, U2) depths. Quartz and feldspar minerals decreased and organic matter and clay minerals increased from the till towards the lower limit of U1. The development in the sedimentary properties marked the transformation of the terrestrial ecosystem from glacier-covered land to vegetated areas. This development was also well reflected by the appearance of X-ray amorphous materials and the formation of distinct organo-mineral aggregates; chlorite was the predominant clay mineral in these aggregates. The geochemical variation between U2 and U1 sediments was further established by resolving the DRIFT spectral components through multivariate curve resolution alternating least square (MCR-ALS). The U1 sediments settled over a period of ~7500 years and showed comparable mineral, geochemical, and organic composition. However, the size-fractionated sediments, mainly <4 μm, showed diverse mineral and geochemical composition. Indeed, these sediments were distinct by containing relatively higher amounts of X-ray amorphous materials and clay minerals, the latter had variable Na, Mg, and K contents.

**Conclusion** The combined use of geochemical and statistical approaches used in this study followed the mineral and geochemical development of sediments that had settled during the Late Pleistocene and Early Holocene Epochs. Finally, the U2 sediments marked the terrestrial ecosystem development that occurred during the late glaciation, deglaciation, and post-glaciation periods.

**Keywords** Sediment core · Size fraction · Post-glaciation · XRD · DRIFT · Ecosystem development

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## 1 Introduction

Lake sediments are recognized as storage zones for lithogenic and anthropogenic materials that are generated in catchments. These materials settle in lakes over long periods and form lacustrine archives (e.g., Schnurrenberger et al. 2003; Kalis et al. 2003). The inorganic and organic sedimentary composition reflect watershed lithology, terrestrial characteristics, ecosystem development, climate

change, mineralogy, and geochemical properties (e.g., Engstrom et al. 2000; Lacey et al. 2015; Panagiotopoulos et al. 2020; Strakhovenko et al. 2020). Therefore, lake sediment cores can reveal past environmental changes that had occurred approximately 10,000 years ago in Scandinavia during the transition from the Pleistocene to the Holocene Epochs (Mörner 2004; Stroeven et al. 2016). During the Late Pleistocene in Scandinavia, the Fennoscandian ice sheets eroded the landscape by plucking and abrasion, which resulted in fine clastic sediments. These fine sediments, as well as other clasts, rocks, and debris, were transported with glaciers and eventually settled when reaching lowland settings (Marshak 2011; Stroeven et al. 2016). Towards the Holocene, substantial changes occurred to the ecosystem as glaciers thawed, bedrock was exposed, and vegetation covered the land. Subsequently, primary producers facilitated weathering through the introduction of plant- and microbial-based organic acids.

Ecosystem development during the Pleistocene–Holocene transition and the Holocene can be revealed in sediments using organic proxies, such as pollen, micro- and macrofossils, and sedimentary ancient DNA (e.g., Pedersen et al. 2013; Capo et al. 2021), elemental proxies, such as carbon and nitrogen contents and isotopes (e.g., Pérez et al. 2014; Pulley et al. 2015), as well as mineral and geochemical ones (e.g., Alizai et al. 2012; Thorpe et al. 2019). The inorganic materials stored in sediments mainly reflect the catchment's bedrock, soil, and land cover. Therefore, sediments mark the development of terrestrial ecosystems and weathering magnitudes that were prominent during the Pleistocene–Holocene. Subsequently, more clay minerals and X-ray amorphous materials are enriched in fine sediments (Hillier 1995; Thorpe and Hurowitz 2020). The X-ray amorphous materials include allophanes, ferrihydrites, diatom frustules, and organic matter (Pérez et al. 2016; Thorpe et al. 2019). Those materials, in addition to clay minerals, (hydro)oxides, microorganisms, and organic matter (OM), represent cementing materials for aggregates (e.g., organo-mineral, mineral–mineral, and bio-organomineral). So, and due to increasing deposition of such phases, mainly during the Pleistocene–Holocene transition and the Holocene, aggregates are expected to form (e.g., Zimmermann-Timm 2002; Tombácz et al. 2004). The formation and size of aggregates depend on the mineral and organic composition (e.g., surface charge and area), as well as the physico-chemical conditions of the aqueous media (e.g., pH and ionic strength), water hydrodynamics, and turbulence (e.g., Lartiges et al. 2001; Tombácz et al. 2004; Bouyer et al. 2005; Tran and Strom 2019; Kanbar et al. 2020a). For example, distinct clay minerals (e.g., mica and chlorite minerals) are expected to behave differently in aggregates based on surface charge and area. Moreover, sedimentary organic matter reflects ecosystem development directly through its increasing deposition in lakes with time, especially after the Pleistocene,

and indirectly by the formation of organo-mineral aggregates (Schaaf et al. 2011). Vegetation covers bedrock and soil, thus reducing the erosion of particles and debris on the one hand, and promoting the erosion and weathering of fine particles on the other, such as clay minerals and X-ray amorphous materials. These changes can be monitored and studied through mineral and geochemical approaches.

To characterize mineral composition, variation, and evolution in sediment cores, powder X-ray diffraction (XRD) is commonly used. It is a fast and reliable technique to identify major crystalline minerals, quantify them using Rietveld refinement, and give indications about X-ray amorphous phases (e.g., Doebelin and Kleeberg 2015; Pérez et al. 2016; Thorpe et al. 2019). Additionally, infrared spectroscopy techniques, such as Fourier transform infrared (FTIR) and diffuse reflectance infrared Fourier transform (DRIFT), can reveal mineral (regardless of crystallinity), geochemical (or chemical), and organic characteristics of sediments. Indeed, infrared techniques proved to be fast, cheap, and effective techniques to study such characteristics (e.g., Madejová et al. 2017; Margenot et al. 2019). Using infrared techniques to reveal organic information has been proven to be accurate and faster in comparison to more time-consuming total organic carbon (TOC) or loss on ignition (LoI) measurements (Rosén et al. 2010; Ancin-Murguzur et al. 2020). Furthermore, DRIFT, and other vibrational spectroscopy data, can be treated by decomposing spectra into resolved components (Felten et al. 2019), which can then be used to detect minor mineral, chemical, and organic variations. Moreover, XRD and infrared techniques can be used in combination to follow changes in mineral properties, such as precipitation, intercalation of water molecules and ions, dissolution, swelling, and weathering (e.g., Lindholm et al. 2019; Kanbar and Kaouk 2019). Infrared spectroscopy is commonly used in connection to other geochemical techniques, such as XRD and electron microscopy, to reveal further information (e.g., Ammar et al. 2016; Le Meur et al. 2016). Scanning electron microscopy coupled to energy-dispersive X-ray (SEM-EDX) complements XRD and DRIFT by determining the elemental composition of particle surfaces. The variation in elemental composition, and more importantly elemental ratios, provided by SEM-EDX can detect minute elemental variations (Ewing and Nater 2003; Courtin-Nomade et al. 2015; Kanbar et al. 2017). Therefore, the combination of the abovementioned geochemical techniques can provide information at bulk, micro-, and elemental scales in sediments as well as other matrices (e.g., soils, technosols, polymers, and geo-polymeric materials). Hence, such analytical approaches are useful in studies where bulk and micro-scale variations are important to note, such as ecology, paleo-ecology, limnology, paleo-limnology, geology, and sedimentology.

The last glacial period in the northern hemisphere ended approximately 10,000 years ago. Subsequently, ecosystem

evolution prevailed during the late glaciation, deglaciation, and post-glaciation periods (e.g., Gibbard and Van Kolfshoten 2004; Öberg and Kullman 2011; Zimmermann et al. 2021). These changes are chronologically stored in undisturbed (intact) lacustrine sediments. The developments of the ecosystem during those periods have been recently recorded through sedimentary ancient DNA (sedaDNA) and organic proxies (e.g., Snowball et al. 2002; Zale et al. 2018; Zimmermann et al. 2021). Nonetheless, the mineral and geochemical composition of sediments that accompanied ecosystem development in Sweden, especially during the deglaciation and post-glacial periods, have not been reported. Therefore, this study aims to characterize the mineral, geochemical, and organic properties of sedimentary materials that have been deposited since the Late Pleistocene. This was done through a combination of geochemical techniques and statistical approaches that revealed information at different scales. Accordingly, bulk, micro-scale, and elemental analyses were carried out on initial and size-fractionated deposits of a sediment core.

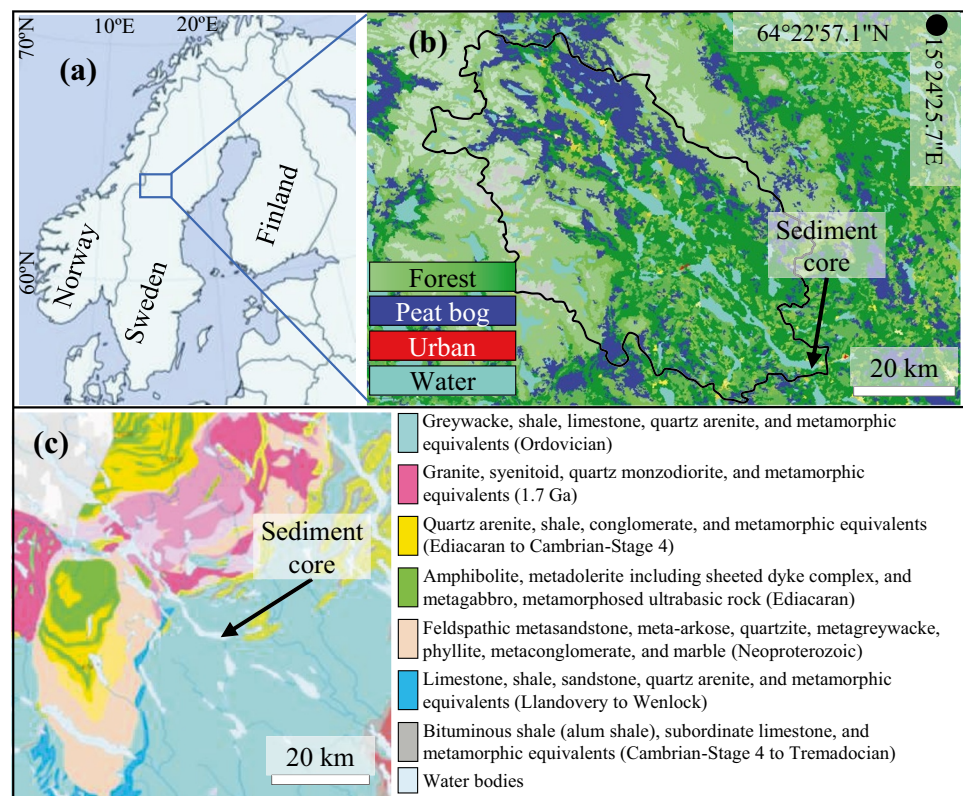
## 2 Area description, sampling site, and sediment coring

A sediment core was collected from a lake in Hotagen, a tributary of the River Indalsälven, located in west-central Sweden (Fig. 1a). The watershed covers a 202-km<sup>2</sup> surface area and has an 850 mm average annual precipitation; its

land cover is dominated by boreal forests (~57%) and has very few urban areas (Fig. 1b). The catchment emerged from beneath the Fennoscandian ice-sheets approximately 10,000 years ago (Öberg and Kullman 2011; Stroeven et al. 2016). Due to limited anthropogenic activity in the region, the sediments in lake Hotagen are expected to reflect the Pleistocene–Holocene transition that occurred during the late-glaciation, deglaciation, and post-glaciation periods. The sediment core was collected from the smaller basin called Laxviken (east part of the lake) which was covered by a 20-m water column; the maximum depth in the lake was 71 m. The sampling area is located in the Caledonian orogen and the bedrock is mainly composed of graywacke, granite, quartz arenite, granitoid, feldspathic metasandstone, amphibolite, phyllite, and limestone (Fig. 1c) (ESDAC - European Commission 1963; Andersson et al. 1985; SGU 2019). Consequently, the main mineral composition of the bedrock is quartz, amphiboles, chlorites, alkali feldspars, plagioclase feldspars, mica, calcite, aragonite, and dolomite.

The 2.3 m sediment core was collected using an 86-mm inner-diameter piston corer (Olajos et al. 2018). The top 6 cm of the core was not collected due to possible disturbance of the loose and muddy surface sediments during coring. Therefore, the depths indicated in this study are in reference to the uppermost sediment (i.e., 6 cm below the top of the core).

**Fig. 1** **a** Overview map of part of northern Europe (modified from Feher (2020)), **b** land cover map showing the main land cover classes (Corine Land Cover 2018) and the watershed's boundary (<https://vattenwebb.smhi.se/hydronu/>), and **c** geological map showing the main bedrock composition (SGU 2019). The location of the sediment core is indicated by arrows



### 3 Materials and methods

#### 3.1 Sample preparation, size fractionation, and treatment

Radiocarbon dating was done to three wood macrofossils that were found at different depths in the core using an accelerator mass spectrometry (AMS) at the Beta Analytic Radiocarbon Dating Laboratory, Miami Florida. The depth estimates were translated to age estimates using the R-based program Clam 2.2 (Blaauw 2010). For mineral and geochemical analyses, a sediment sample was collected every 5 cm along the 2.3 m core. The sediments on the periphery of the core were discarded due to possible lateral movement during coring. Each of the 45 samples was divided into two parts, an intact and untreated part (named bulk) and a size-fractionated part (having sizes of <4, 4–16, 16–64, and >64  $\mu\text{m}$ ). Before fractionation via particle settling (using Stokes' Law, e.g., see Bergaya and Lagaly (2013)), the samples were mixed in ultrapure water overnight. To maintain the characteristics of the sediment fractions, the aggregates (organo-mineral and mineral–mineral) were not disaggregated. This was an important part because DNA sorption was assessed on lake Hotagen sediments based on the geochemical, mineral, and organic compositions of each size fraction, including the aggregates (Kanbar et al. 2020b). Therefore, each size fraction contained aggregates of the relatively smaller fraction(s), except the <4  $\mu\text{m}$  fraction, i.e., the clay-sized fraction, since it is the smallest one. Unless indicated otherwise, the bulk and size-fractionated sediments were freeze-dried and ground, using an agate mortar and pestle, before analyses.

It is worth noting that all the bulk sediments were characterized by XRD and DRIFT ( $n = 45$ ). Due to similar mineral and geochemical compositions throughout large parts of the core (as described later in the text), size-fractionated samples of selected sediment depths were chosen for further investigation. Additionally, elemental composition (by X-ray fluorescence, XRF), cation exchange capacity (CEC), and pH were determined for selected bulk sediments (SM 1).

#### 3.2 Grain size distribution

Grain size distribution (GSD) analysis was performed on the bulk sediments by laser diffraction (Partica LA-950V2 laser scattering particle size distribution analyzer from Horiba). The samples were neither freeze-dried nor disaggregated prior to GSD analysis. The bulk sediments were mixed in a sample bath with distilled water and grain size measurements were done before and after applying sonication for 1 min. Measurements were done before and after sonication

to study aggregate size. Non-sonicated measurements indicate aggregate sizes (or sediment sizes), while measurements after sonication indicate grain sizes (or particle sizes), as sonication breaks aggregates.

#### 3.3 Powder X-ray diffraction

The crystalline minerals of the bulk and size-fractionated sediments were identified by randomly-oriented powder X-ray diffraction (XRD) measurements using PANalytical X'pert<sup>3</sup> diffractometer (CuK $\alpha$  radiation 1.54187 Å). The ground sediments were front-loaded onto 16 mm stainless-steel circular cavity holders and were run using a spinner stage (for better randomization) with the following parameters: 4–64° 2 $\theta$ , 0.026° step size, 1 s collecting time, 4 s/spin, 45 kV, 40 mA, 0.04 rad Soller slits, 1/8° incident divergence slit, 5 mm anti-scattering receiving slit, and 10 mm mask. The crystalline minerals were identified with the HighScore Plus software (Degen et al. 2014) and the Crystallography Open Database (COD), and then quantified from the experimental diffractograms by Rietveld refinement using Profex-BGMN (Post and Bish 1989; Doebelin and Kleeberg 2015; Döbelin 2021) and structural data from the COD. For accurate peak shape modeling, the fundamental parameters approach (FPA) was set to match the instrument's configurations (e.g., divergence slits, Soller slits, and optical elements in the beam path), and the parameters k2 (micro-strain broadening), B1 (crystallite size), and GEWICHT (scale factor that quantifies the phase's weight fraction in the sample as well as corrects preferred orientation) were refined. Subsequently, the goodness-of-fit (GoF) and chi-squared values ( $\chi^2$ ) were used to assess the quality of the fit (Döbelin 2021); the  $\chi^2$  and GoF values are reported in SM 2. To verify the reproducibility of mineral quantification due to sample preparation (i.e., mounting and possible preferred orientation), two sets of samples were mounted and measured four times and gave similar results (SM 2).

For selected bulk sediments, oriented mounts were prepared on glass slides to characterize clay minerals using the protocol developed at the Laboratoire Interdisciplinaire des Environnements Continentaux (LIEC), Nancy, France (Bihannic and Gley 2021). Due to similar clay mineral composition in all the sediments, two samples were chosen for oriented XRD measurements. Briefly, the sediments were disaggregated via sonication and the particles smaller than 4  $\mu\text{m}$  were collected by applying Stokes' law. These clay-mineral rich suspensions were then transferred to three glass slides and left to settle with preferred orientation along the c-axis. The samples were air-dried, treated with ethylene glycol, and calcinated at 550 °C. The vapor treatment was used for the ethylene glycol-treated samples (e.g., Poppe et al. 2001). Subsequently, XRD measurements were done with the following parameters: 3–30° 2 $\theta$ , 0.016° step size, 1.7 s collecting time, 45 kV, 40 mA, 0.04 rad

Soller slits, 1/8° incident divergence slit, 5 mm anti-scattering receiving slit, and 10 mm mask.

### 3.4 Diffuse reflectance infrared Fourier transform spectroscopy

Diffuse reflectance infrared Fourier transform spectroscopy (DRIFT, IFS 66v/s by Bruker) was used to determine the functional groups of all bulk and selected size-fractionated sediments. Well-homogenized and dry mixtures of sediments were prepared with KBr for DRIFT measurements. For each sample, 128 spectra were recorded under vacuum (<7 mbar) in the 5200–400 cm<sup>-1</sup> range with a 4 cm<sup>-1</sup> spectral resolution. In order to decompose the complex multi-component spectra into pure phase components, the raw infrared (IR) spectra were treated using the open-source MCR-ALS GUI (multivariate curve resolution alternating least square gentle user interface) provided by the Vibration Spectroscopy Core Facility at Umeå University (Felten et al. 2015, 2019; [www.umu.se/en/research/infrastructure/visp/downloads/](http://www.umu.se/en/research/infrastructure/visp/downloads/)). Multiple regression was used to estimate the organic matter contents based on LoI (calculated from XRF) and organic IR signatures (e.g., Rosén et al. 2010). Loss on ignition or organic matter content was calculated by the complementarity of the sum of major oxides to 100; this method has been shown to accurately represent sedimentary organic matter in various lakes (Rydberg 2014).

#### 3.4.1 Chemical and mineral identification of sediments after organic matter removal

Sedimentary organic matter was chemically removed by sodium hypochlorite (NaOCl) from the same samples that were analyzed by XRD and DRIFT; this method was used since it does not modify mineral structures, unlike ashing and H<sub>2</sub>O<sub>2</sub> treatment (e.g., Mikutta et al. 2005; Margenot et al. 2015). Detailed information on organic matter removal by NaOCl can be found elsewhere (Margenot et al. 2019 and SM 3). The sediments subjected to organic matter removal are hereafter denoted Sed<sub>inorg</sub>, indicating the inorganic nature; non-treated sediments are denoted Sed. The Sed<sub>inorg</sub> samples (bulk and size-fractionated) were then analyzed by DRIFT. Consequently, and due to the disappearance of OM IR peaks which partly interfere with mineral IR peaks, some of the DRIFT chemical and mineral fingerprints were clearer for Sed<sub>inorg</sub> in comparison to Sed.

### 3.5 Scanning electron microscopy

A scanning electron microscopy coupled to energy-dispersive X-ray spectroscopy (SEM-EDX) was used to investigate the elemental composition of the predominant mineral surfaces

(Carl Zeiss Evo SEM with LaB6 electron source, UCEM). Non-metal coated dry sediments were mounted onto double-adhesive carbon tapes and measured under low vacuum conditions (LV, 60 Pa). Quantitative EDX spectra were collected using Aztec 3.3 (Oxford instruments) with dead time below 10%, EDX signals between 8000 and 10,000 counts/s, and using oxygen for stoichiometry and carbon as a deconvolution element (not used in elemental quantification).

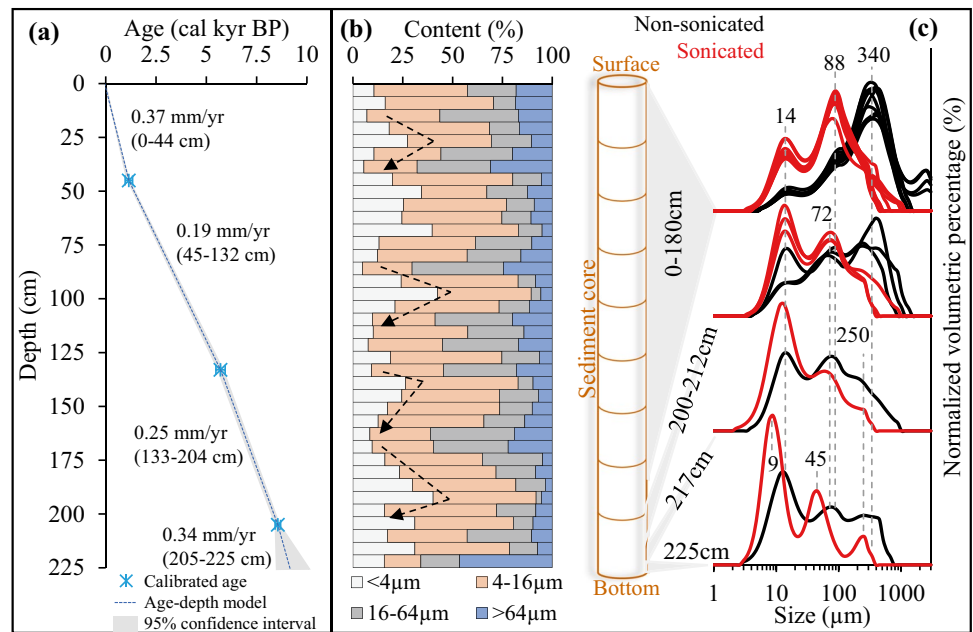
## 4 Results

### 4.1 Sediment age, accumulation, size, and aggregation

The deepest sediment (225 cm) dated to the Late Pleistocene, approximately 10,000 years ago, and was identified as glacial till that had deposited during the end of the glacial period (Fig. 2a). This sediment had a distinct gray color. The color then faded into a brown color just above the till (i.e., at depth 220 cm) until 180 cm. No noticeable color variation was seen for the 0–180 cm sediments, except for a lighter brown color for the 1, 5, 35, and 135 cm sediments. The color transformation in the till–180 cm reflects a change in the sedimentary materials that had settled during approximately 2000 years (Fig. 2a). Due to limited human interference in the area, the sediments are unlikely to have been dredged or disturbed. The sediment accumulation rate in the lake ranged between 0.19 and 0.37 mm/yr, as inferred by the three AMS radiocarbon dates (Fig. 2a). The summary of the radiocarbon dates is included in Table 1 and the calibration of radiocarbon ages to calendar ages is included in SM 4. The sediment accumulation rates roughly indicate that each 5 cm, the distance between the sampled sediments in the core, is a product of 125–250 years of deposition. The relatively low sedimentation rates are likely due to the small catchment area, low primary production as a result of cold and nutrient-poor environments (Olli 2008; Lawson et al. 2012), and the presence of larger lakes upstream of the sampling site that function as sedimentation basins.

The sediment sizes did not show a clear trend as a function of depth (Fig. 2b). Nonetheless, there is a noticeable pattern every ~25 cm, as indicated in the <4 μm fractions (e.g., 20–45, 90–110, and 170–200 cm; Fig. 2b and SM 5a). The abundance of the 4–16-μm fraction varied slightly with depth in comparison to the other fractions. The till had approximately 50% of sedimentary materials coarser than 64 μm. Moreover, even without disaggregation (e.g., sonication), the samples showed a considerable fraction below 4 μm (5–42%), indicating an overall clayey nature of the sediments.

**Fig. 2** **a** Sediment age-depth model with 95% confidence interval (shaded area) and average sedimentation rates, **b** sediment size based on Stokes' law (dashed arrows indicate patterns; explained in the text), and **c** grain size distribution (GSD) of bulk sediments with non-sonicated samples in black (representing aggregates) and sonicated samples in red (representing grains or particles); the cylinder represents the sediment core and follows the same vertical scale as part "a" of this figure. Each plotted profile represents the GSD of one sediment sample, where the number of sediment samples is 9 and 3 for 0–180 and 200–212 cm, respectively



The bulk sediments showed multimodal GSD (Fig. 2c). Based on the sonicated (representing grain or particle sizes) and non-sonicated samples (representing aggregate or sediment sizes), the sediment core can be divided into distinct sections, which correspond to 0–180 cm, 200–212 cm, 217 cm, and 225 cm (till). For the sonicated samples, the till particles were predominantly centered at 9 µm, followed by relatively coarser 45 µm (silty) and 250 µm (fine-medium sand) populations. This coarsest population was also reflected by a relatively high >64 µm fraction (Fig. 2b). From the till towards later sediments (i.e., sediments that had settled afterward), the finest population shifted towards coarser sizes, i.e., from 9 µm in the till to 12 µm in 217 cm and to 14 µm in 200–212 cm and 0–180 cm (Fig. 2c). The 45-µm population of the till behaved in a similar manner; it increased to 62, 72, and 88 µm in the 217 cm, 200–212 cm, and 0–180 cm sediments, respectively. The non-sonicated samples also showed a gradual size increase from the till towards later sediments (Fig. 2c). Indeed, there is a clear shift of the predominant sediment population from the till (14 µm) towards the 0–180 cm sediments (340 µm). Also, the coarse populations of the non-sonicated samples shifted

to smaller sizes upon sonication; these smaller populations were already observed in the sediments (non-sonicated), but became more obvious after sonication. For example, the 14 and 88 µm populations of the 0–180 cm sediments were slightly visible in the non-sonicated samples, but became clear after sonication. This indicates that those particles were initially present as aggregates, possibly organo-mineral and mineral–mineral aggregates. Therefore, it is expected that these sediments, i.e., 0–180 cm, contain higher amounts of cementing materials (such as organic matter, carbonates, and clay minerals). Furthermore, the variation between aggregate and particle sizes is more significant in the 0–180 cm sediments, while it was reduced in the underlying 200–212 cm sediments, and relatively insignificant in the 217 and 225 cm sediments. This shows that more aggregations had formed during the later stages of the Holocene, i.e., in the 0–180 cm sediments that had settled in the last ~7500 years (Fig. 2a, c). Finally, the GSD profiles shifted from finely skewed in the 0–180 cm sediments to symmetrical and coarsely skewed in the deepest sediments, and the till showed to be very poorly sorted in comparison to the other sediments (based on Blott and Pye (2001), SM 5b).

**Table 1** Summary of the radiocarbon data obtained from the sediment core

Sediment depth (cm)	Sample ID (Beta Lab ID*)	<sup>14</sup> C age (BP) ± 1σ	Calibrated age range (BP)	Dating material
45	Beta-517571	1220 ± 30	1258–1063	Wood
133	Beta-517572	4980 ± 30	5862–5612	Wood
205	Beta-517573	7780 ± 30	8609–8455	Wood

\*The permanent sample ID assigned by Beta Analytic Inc

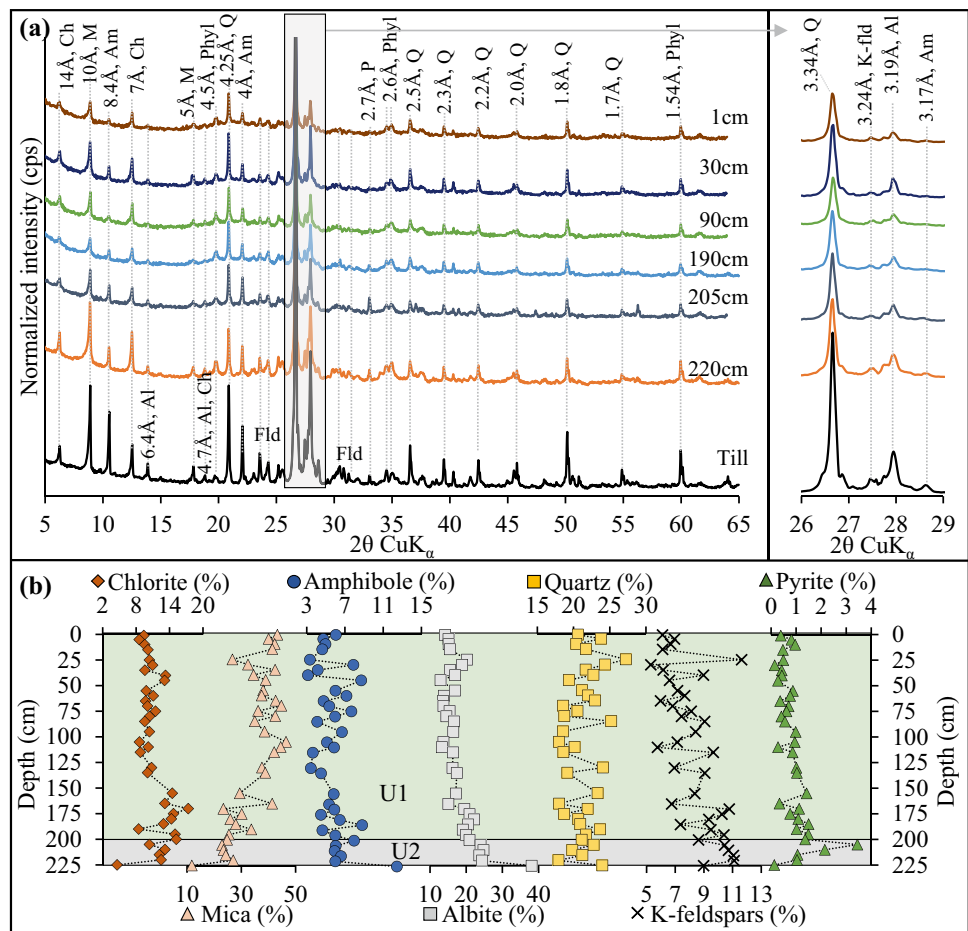
## 4.2 Bulk sediments

### 4.2.1 Crystalline mineralogy as a function of depth

Lake Hotagen sediments were mainly composed of silicate minerals, such as quartz, amphiboles (inosilicates), feldspars (tectosilicates), and clay minerals (phyllosilicates), which were present throughout the core (Fig. 3a). Amphibole minerals were mainly identified by the diffraction peak at 8.4 Å. The XRD patterns and watershed's lithology suggest that the amphibole mineral is tremolite, yet it cannot be easily distinguished from actinolite or hornblende, which are constituents of the parent bedrock in the watershed. Therefore, the mineral(s) identified by the 8.4 Å peak will be addressed as amphibole. The phyllosilicate minerals detected in the sediments were chlorite and mica, as mainly identified by the 14- and 10 Å diffraction peaks, respectively. The 7.14- and 3.58 Å peaks could be assigned to kaolinite (001 and 002 reflections, respectively) as well as chlorite (002 and 004 reflections, respectively). However, kaolinite did not appear to be present in the sediments due to the absence of the 060 peak at 1.50 Å. Further explanation is included in the oriented XRD patterns (SM 6). The 10- and 5 Å diffraction

peaks belong to micaceous minerals, such as illite, muscovite, and biotite. The relative broad 001 peak suggests illite since muscovite and biotite have relatively sharper 001 peaks. Nonetheless, due to the common origin and complexity of clear mineral identification in heterogeneous sediments, and based on lithology, biotite, and muscovite might also be present. Indeed, such micaceous minerals result from glacial grinding which can further form illite (Poppe et al. 2001; Galán 2006). Therefore, these minerals will be addressed as mica minerals. Additionally, the phyllosilicates were further noted by the diffraction peaks at 4.5, 2.6, and 1.54 Å (Fig. 3a). The 060 reflections between 1.50 and 1.54 Å, as in this case, indicate tri-octahedral phyllosilicates, while di-octahedral silicates have the 060 reflections between 1.48 and 1.50 Å (Brown and Brindley 1980). All these minerals, in addition to quartz (3.34 Å) and feldspars, originated from the parent bedrock (Fig. 1c) as suggested by the watershed's lithology (SGU 2019). The feldspar minerals identified in the sediments were Na-feldspars (albite, 3.19 Å), K-feldspars (orthoclase and microcline, ~3.2 Å), and possibly Na, K-feldspars (anorthoclase, ~3.2 Å). Due to the heterogeneity of the sediments, the feldspar minerals will be addressed as albite (Na-feldspars) and K-feldspars. Unlike

**Fig. 3** **a** XRD patterns of bulk sediments. Ch, chlorite; M, mica; Am, amphibole; Phyl, phyllosilicates; Q, quartz; P, pyrite; Fld, feldspars; K-flid, K-rich feldspars (orthoclase and microcline); and Al, albite. The inset to the right highlights the change in quartz and feldspar peaks. **b** Mineral contents of the bulk sediments as a function of depth





the other minerals, pyrite (2.7 Å) was mainly detected in sediments below 190 cm (Fig. 3a). Although the watershed contains carbonate bedrock, no crystalline carbonates were detected by XRD (e.g., aragonite, calcite, or dolomite); swelling clay minerals were also not detected. More information about sheet silicates obtained from oriented XRD patterns is included in SM 6. Furthermore, the diffractogram of the 220 cm sediment showed a small bump in the 15–25° 2 $\theta$  region (Fig. 3a). This bump gradually increased towards 210 cm and appeared to be similar for the rest of the sediments. The bump indicates X-ray amorphous materials, such as allophanes, diatoms, and organic matter (Pérez et al. 2016; Thorpe et al. 2019).

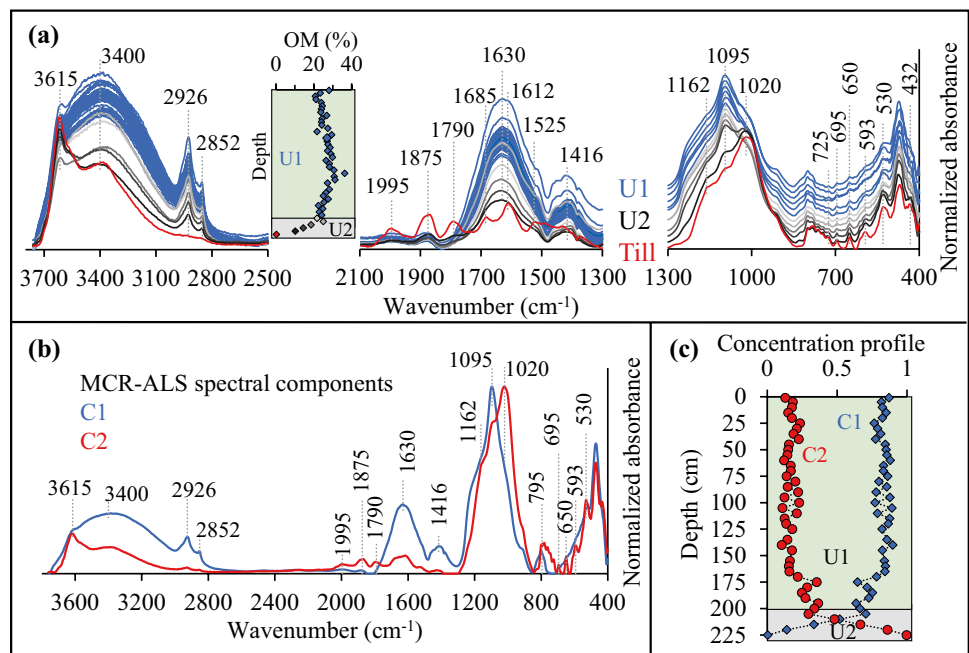
Quartz, mica, and albite minerals were predominant throughout the core, while chlorite, amphibole, and K-feldspar minerals were less abundant; pyrite was only detected in certain sediments (Fig. 3b). The abundance of quartz (24%), albite (38%), and amphiboles (13%) in the till reflected the watershed's lithology (ESDAC - European Commission 1963; Andersson et al. 1985; SGU 2019). Above the till (i.e., 220 cm), these minerals were less abundant and phyllosilicates (chlorite and mica) were more prominent. Mica mineral contents increased from the till towards 200 cm, whereas albite showed an opposite trend. Based on those minerals, it is clear that the till is distinct, the 200–220 cm sediments showed unique trends, and the overlying and younger 0–195 cm sediments showed variable mineral composition. Amphibole minerals did not display a depth-dependence trend; pyrite generally increased from 225 to 205 cm and then decreased and was negligible throughout the core. According to the XRD findings, the sediments can

be categorized into unit 1 (U1 0–195 cm) and unit 2 (U2 200–225 cm). Even though GSD suggested more distinct sections (Fig. 2c), the last three sections (i.e., 200–212, 217, and 225 cm) can be considered as the transition from the till towards the more homogeneous U1 sediments. More information about sediment categorization is included later in the text and in SM 7.

#### 4.2.2 Mineral, chemical, and organic variation as a function of depth: DRIFT

The IR spectra of the bulk sediments showed chemical variations mainly among the U2 sediments on the one hand, and between the U1 and U2 sediments on the other (Fig. 4a). These variations reflect the composition of various minerals and are mainly based on the Si–O stretching and bending vibrations in the 1200–950- and 600–400 cm<sup>-1</sup> regions, respectively, and on the OH stretching and bending vibrations (of silicates and aluminosilicates) in the 3700–3500- and 950–650 cm<sup>-1</sup> regions, respectively (Fig. 4a). The OH stretching peaks of kaolinite octahedral layers (at 3697 and 3620 cm<sup>-1</sup>) were absent, thus confirming the XRD findings that kaolinite is not present in the sediments. The Si–O stretching peaks identified at 1095 and 1020 cm<sup>-1</sup> were attributed to silicate minerals, such as quartz, feldspars, and phyllosilicates (Parikh et al. 2014; Madejová et al. 2017). Due to the heterogeneity of the bulk sediments, i.e., they contain all these minerals (Fig. 3), the assignment of the peaks to their respective minerals was not possible. Quartz was readily identified in the till by the quartz overtone combination bands at 1995, 1875, and 1790 cm<sup>-1</sup> as well as 1685, 1612, and 1525 cm<sup>-1</sup> (Nguyen et al. 1991; Padilla et al. 2014).

**Fig. 4** **a** DRIFT spectra of bulk sediments. The U1 sediments (0–195 cm) are marked in blue and the U2 sediments (200–225 cm) are marked in black-gray, except the till, which is marked in red; 220 cm is marked in black and the color changes to lighter gray for the above U2 sediments. The spectra in the 1300–400 cm<sup>-1</sup> region are stacked for clearer visualization of the Si–O peak shift (1095–1020 cm<sup>-1</sup>). The inset shows the variation of organic matter (OM) with depth. **b** The resolved MCR-ALS spectral components of the bulk sediments (C1 and C2 are component 1 and component 2, respectively) and **c** the resolved concentration profiles of the two components



Some of these bands were obvious in the till due to the absence of the overlapping OM peak at  $1630\text{ cm}^{-1}$ . Furthermore, the till showed the Si–O stretching peak at  $1020\text{ cm}^{-1}$  and a shoulder at  $1095\text{ cm}^{-1}$  (red spectrum in Fig. 4a). The shoulder at  $1095\text{ cm}^{-1}$  then protruded into a peak in the 220 cm sediment, and eventually two peaks were seen in 215 cm (i.e., at  $1020$  and  $1095\text{ cm}^{-1}$ ). The overlying sediments (i.e., 0–210 cm) had the Si–O stretching peak predominantly at  $1095\text{ cm}^{-1}$  and a shoulder at  $1020\text{ cm}^{-1}$ . Additionally, the 0–215 cm sediments showed a shoulder at  $1250$ – $1170\text{ cm}^{-1}$ , suggesting quartz, humic substances, allophanes, amorphous silicates, or diatom frustules (Rosén et al. 2010; Meyer-Jacob et al. 2014). The change in the OH and Si–O bending vibrations in the  $800$ – $690\text{ cm}^{-1}$  and  $600$ – $400\text{ cm}^{-1}$  regions, respectively, evolved like the Si–O stretching bands (i.e., from the till towards 215 cm). This suggests that these chemical signatures are related to the same minerals. Moreover, the intensity and position of the OH stretching and water bending vibrations ( $\sim 3400$  and  $1640\text{ cm}^{-1}$ , respectively) are affected by interlayer cations (Madejová et al. 2017 and references therein), indicating a change in the abundant mineral(s), especially between the U1 and U2 sediments.

As for the organic IR signature, the variation was based on the peaks at  $3400\text{ cm}^{-1}$  (amine N–H stretching and OH stretching of humic substances),  $2926$  and  $2852\text{ cm}^{-1}$  (aliphatic C–H stretching),  $1630\text{ cm}^{-1}$  (aromatic C=C stretching and/or amide C=O),  $1614\text{ cm}^{-1}$  (C–O stretching in carboxyl groups of humic substances; –COOH), and  $1416$ – $1400\text{ cm}^{-1}$  (aliphatic C–H bending) (Parikh et al. 2014; Madejová et al. 2017). The link between these peaks and organic matter content has been described elsewhere (e.g., Rosén et al. 2010). Furthermore, the bands at  $3400$  and  $1640\text{ cm}^{-1}$  also indicate OH stretching and bending of water molecules, respectively. These organic IR signatures were quasi-absent in the till which contained only 0.27% organic matter (inset in Fig. 4a and SM 1). The organic matter contents increased from the till to the U1–U2 transition region at 195–200 cm (except for 205 cm) and fluctuated between 20 and 35% in the U1 sediments. Moreover, the main IR variation of the U1 sediments was due to OM. This is supported by similar DRIFT spectra for the U1 sediments after OM removal, i.e., for  $\text{Sed}_{\text{inorg}}$ , as indicated by MCR-ALS (SM 8).

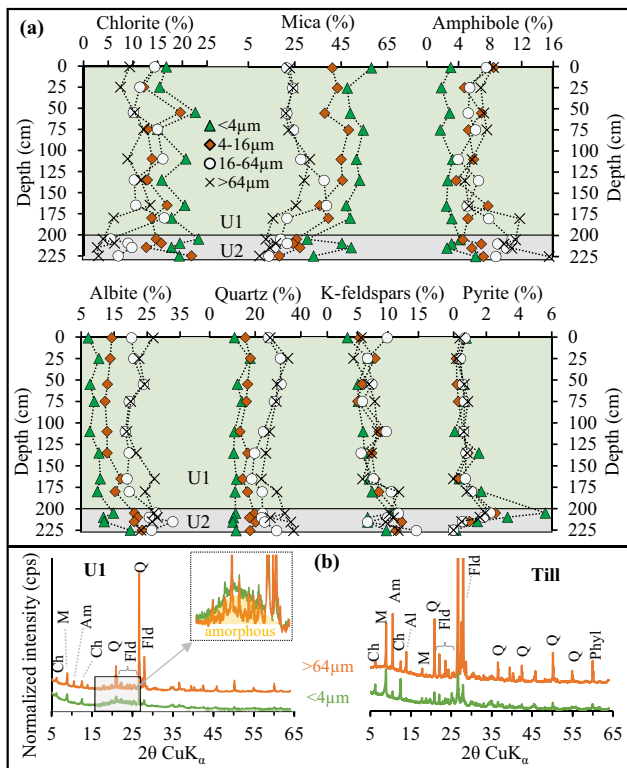
Two MCR-ALS spectral components explained the variation of the bulk sediments throughout the core (Fig. 4b), each representing a distinct mineral and geochemical composition. The MCR-ALS approach was used to decompose the experimental IR spectra by first computing the most probable number of pure components by single value decomposition, and secondly by finding the pure spectral components and their respective weights, which are shown as concentration profiles (Fig. 4c). It should be noted that the changes within the U2 sediments, as seen by GSD, XRD, and DRIFT (Figs. 2c, 3b, and 4a), can further be divided into more MCR components; this is

addressed when size-fractionated sediments are discussed (Section 4.3.2). The MCR spectral component C1 was characterized by the presence of organic matter (according to the peaks described in the previous paragraph) and an Si–O stretching peak at  $1095\text{ cm}^{-1}$  (Fig. 4b); C2 was distinct by lacking OM bands and having unique peaks at  $1020\text{ cm}^{-1}$  (Si–O stretching),  $800$ – $600\text{ cm}^{-1}$  (OH bending),  $600$ – $400\text{ cm}^{-1}$  (Si–O bending), and  $2000$ – $1600\text{ cm}^{-1}$  (quartz overtone). The first MCR spectral component (C1) predominated the 0–205 cm sediments, as indicated by the resolved MCR-ALS concentration profile (Fig. 4c). However, this signature gradually declined from 175 towards 210 cm, where the latter sediment had almost equal contributions of C1 and C2. Subsequently, the MCR component C2 predominated the deepest sediments (215–225 cm).

### 4.3 Size-fractionated sediments

#### 4.3.1 Crystalline mineralogy based on sediment size

In most U1 sediments, the  $<4$  and  $4$ – $16\text{ }\mu\text{m}$  size fractions and the  $16$ – $64$ - and  $>64$ - $\mu\text{m}$  size fractions showed distinct mica, albite, and quartz mineral contents; mica minerals were enriched in the former fractions while albite and quartz were enriched in the latter fractions (Fig. 5a). As for chlorite, although enriched in the  $<4\text{ }\mu\text{m}$  fractions in some sediments (e.g., 5, 25, 110, and 170 cm), it did not show a clear enrichment in the finest sediment fractions as mica did. Furthermore, the distinction of minerals based on size-fractions did not necessarily apply in the U2 sediments. For instance, albite and mica mineral contents were distinct between the  $<4\text{ }\mu\text{m}$  fractions on the one hand and the  $>4\text{ }\mu\text{m}$  fractions on the other (i.e.,  $4$ – $16$ ,  $16$ – $64$ , and  $>64\text{ }\mu\text{m}$ ). Clay minerals, as well as organic matter, are part of cementing materials for aggregates and are therefore included in the coarse fractions (Fig. 2c). Thus, clay minerals are not necessarily enriched in the fine fractions. Amphibole mineral contents were lowest in the  $<4\text{ }\mu\text{m}$  sediments, while the other size fractions showed fluctuating contents. Nonetheless, more amphibole was detected in the U2 sediments with increasing size fractions. Pyrite and K-feldspar contents did not show a clear trend with sediment size. Pyrite showed a noticeable increase from the till towards 205 cm, as mainly seen in the  $<4\text{ }\mu\text{m}$  sediments. Interestingly, the finest fraction of the till displayed similar mineral characteristics to the  $16$ – $64$ - and  $>64$ - $\mu\text{m}$  fractions of the U1 sediments, e.g., mica, amphibole, and albite (Fig. 5a). The X-ray amorphous materials indicated by the bump in the  $15$ – $25^\circ 2\theta$  region were more noticeable in the U1 sediments in comparison to the U2 sediments (Fig. 5b). Moreover, this signature was higher for the  $<4\text{ }\mu\text{m}$  fractions in comparison to the coarser fractions ( $4$ – $16$ ,  $16$ – $64$ , and  $>64\text{ }\mu\text{m}$ ); the latter showed similar bump signatures. Similar to the bulk sample, the size-fractionated till samples did not display a bump in that region (Fig. 5b).



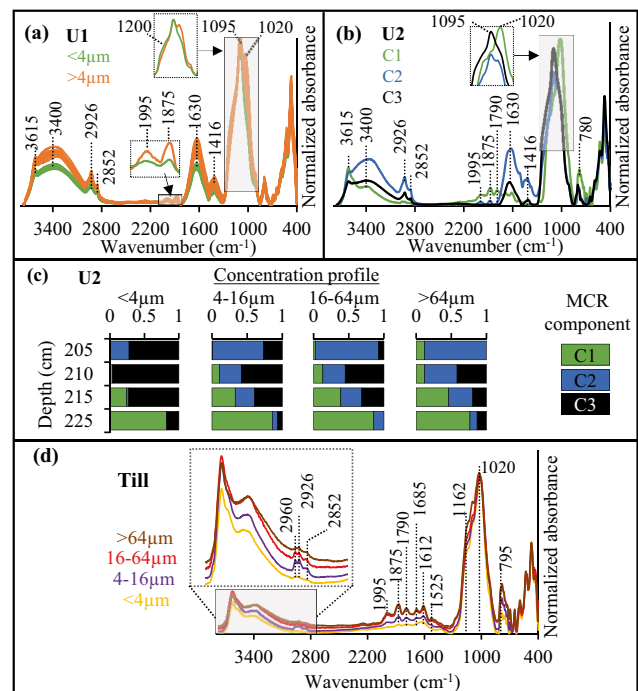
**Fig. 5** **a** Mineral composition of the size-fractionated sediments as a function of depth and **b** X-ray diffractograms of  $<4$  and  $>64$   $\mu\text{m}$  size fractions for 1 cm (representing U1) and the till; both diffractograms have the same Y-axis scale. Ch: chlorite, M: mica, Am: amphibole, Q: quartz, Fld: feldspars. The inset shows the bump that indicates X-ray amorphous materials; the diffractograms are superimposed for clearer visualization of the bump. All peaks and d-spacings are included in Fig. 3

### 4.3.2 Mineral, chemical, and organic variation based on sediment size: DRIFT

Bulk samples showed the distinction between U1 and U2 sediments (Figs. 3 and 4). Therefore, MCR-ALS analyses of the size-fractionated sediments were done for these two units separately. Accordingly, the MCR-ALS results showed the geochemical variations firstly based on sediment size and secondly on depth. Two MCR spectral components explained the IR variation in the U1 sediments (Fig. 6a). The first MCR component applied to the  $<4$   $\mu\text{m}$  fractions of all the U1 sediments and showed an Si–O peak at  $1095$   $\text{cm}^{-1}$  and had relatively low OM IR signatures. Therefore, the Si–O stretching peak at  $1095$   $\text{cm}^{-1}$  was assigned to clay minerals (chlorite and/or mica in this case). Additionally, the shoulder at  $\sim 1200$   $\text{cm}^{-1}$  was more obvious for the  $<4$   $\mu\text{m}$  fractions (inset in Fig. 6a), suggesting higher contents of amorphous materials in that fraction. This was also evidenced by the bump in the X-ray diffractograms (Fig. 5b). The second MCR spectral component was characterized by

an additional Si–O stretching peak at  $1020$   $\text{cm}^{-1}$  and relatively high OM and quartz overtone peaks (insets in Fig. 6a). This component applied to the fractions greater than  $4$   $\mu\text{m}$  (i.e.,  $4$ – $16$ ,  $16$ – $64$ , and  $>64$   $\mu\text{m}$ ). In the U1 sediments, the resolved MCR components were easily distinguished between  $<4$   $\mu\text{m}$  (C1) and  $>4$   $\mu\text{m}$  (C2) sediment, but they did not change as a function of depth. Therefore, the concentration profile is not shown.

The U2 sediments showed geochemical variations based on size fraction as well as depth. The size-fractionated U2 sediments showed three resolved MCR spectral components (Fig. 6b). The first component (C1) mainly described the till and to a lesser extent 215 cm (Fig. 6c). This component was characterized by a clear Si–O peak at  $1020$   $\text{cm}^{-1}$ , quartz peaks ( $1995$ – $1525$   $\text{cm}^{-1}$ ), specific bands at  $950$ – $650$   $\text{cm}^{-1}$ , and the absence of OM IR signatures, and predominantly applied to all the till fractions (Fig. 6c) since the quartz overtone peaks were clearly visible (Fig. 6d). The second MCR component (C2) was characterized by two Si–O peaks ( $1095$  and  $1020$   $\text{cm}^{-1}$ , inset in Fig. 6b), relatively high OM peaks, and some quartz overtone peaks. This component mainly described the 205 cm sediments greater than  $4$   $\mu\text{m}$ . The bulk 205 cm sediment showed relatively high OM content as well (Fig. 4a); it is worth noting that the high OM IR signatures,



**Fig. 6** **a** DRIFT spectra of the U1 sediments showing the distinction between  $<4$  and  $>4$   $\mu\text{m}$  size fractions (indicating the two MCR spectral components). **b** The MCR spectral components and **c** concentration profiles of the size-fractionated U2 sediments. **d** The DRIFT spectra of the size-fractionated till with focus on the OM peaks (inset)

in this case, might have overwhelmed the signatures of the other MCR components. The third MCR component (C3) showed a relatively sharp Si–O peak at  $1095\text{ cm}^{-1}$ , the absence of quartz overtone peaks, and small OM peaks; this component predominantly described the  $<4\text{ }\mu\text{m}$  fractions of the U2 sediments except the till. Furthermore, the  $<4\text{ }\mu\text{m}$  fraction of the till showed comparable IR peaks to the coarser fractions of the other sediments, i.e., quartz overtone peaks at  $2000\text{--}1600\text{ cm}^{-1}$  and an Si–O stretching peak at  $1020\text{ cm}^{-1}$  (Fig. 6a, d).

The change in the predominance of C1 in the till to C3 in 210 cm and then C2 in 205 cm highlights the evolution of the sediments (Fig. 6c). Indeed, the MCR components showed the transition from OM-depleted and quartz and feldspar-rich samples (represented by C1, Fig. 6b), to sediments with relatively moderate OM and clay mineral contents (C3), and finally to OM-rich and clay mineral-rich samples (C2). The component C2 held the characteristics of the U1 sediments (Fig. 6a, b), regardless of size. According to these components, such as the presence of the Si–O peak at  $1020\text{ cm}^{-1}$  in the till and in the coarse fractions of the other sediments, and in combination with XRD findings, this peak was assigned to feldspar and/or quartz. Feldspar was also characterized by the  $950\text{--}650\text{ cm}^{-1}$  and  $725\text{ cm}^{-1}$  fingerprints and quartz was identified by the bands between  $2000$  and  $1800\text{ cm}^{-1}$  (Padilla et al. 2014; Meyer-Jacob et al. 2014). The change in quartz and/or feldspar on the one hand, and clay mineral peaks ( $1020\text{--}1095\text{ cm}^{-1}$ ) on the other, was seen in the bulk U2 sediments (Fig. 4a). The Si–O bending (at  $593$ ,  $530$ , and  $432\text{ cm}^{-1}$ ) and OH bending (at  $725$ ,  $695$ , and  $650\text{ cm}^{-1}$ ) vibrations also identified feldspar minerals in the coarse U2 sediments. These are the characteristics of C1 (Fig. 6b) which decreased from the till towards 180 cm (Fig. 4a). Interestingly, the till showed organic matter signatures in the  $3000\text{--}2800\text{ cm}^{-1}$  region (inset in Fig. 6d). Nonetheless, the intensities of these OM peaks were significantly lower than any other sediment in the core. In addition to the  $2852$  and  $2926\text{ cm}^{-1}$  OM IR peaks that were detected in all the other sediments, the till showed a unique peak at  $2960\text{ cm}^{-1}$  (indicating aliphatic C–H groups of organic matter), which was mainly detected in the  $4\text{--}16\text{ }\mu\text{m}$  fraction.

#### 4.4 Micro-scale variation: SEM–EDX

The elemental composition of the predominant mineral surfaces in the sediments (i.e., aluminosilicates: feldspar and clay minerals) was addressed at micro-scale levels using SEM–EDX (Fig. 7). Morphologically, the SEM images showed well-aggregated phases, even in the finest fractions (Fig. 7a), and relatively large particles in the  $>64\text{ }\mu\text{m}$  fractions (Fig. 7b). Regardless of sediment depth or size fraction, Si was the major element, followed by Al, and then Fe, Mg, Na, and K, while Ti, S, and P were only detected in trace amounts (EDX spectra in Fig. 7). Within the same sediment depth, the

elemental composition varied based on size fraction (e.g., the  $<4$  and  $>64\text{ }\mu\text{m}$  fractions of  $1\text{ cm}$  in Fig. 7a, b, respectively), which reflected mineral variation. The aluminosilicates in the  $<4\text{ }\mu\text{m}$  fractions contained higher Si contents and lower Al, Fe, Ca, K, Na, and Mg contents than the  $>64\text{ }\mu\text{m}$  fractions (Fig. 7a, b). This variation was consistent throughout similar phases within each sample, as compiled in Table 2. Furthermore, the change in the main elemental ratios, i.e., Si/Al, pointed to this variation. Indeed, the Si/Al ratios averaged  $6.9 (\pm 1.8)$  and  $2.4 (\pm 0.2)$  in the  $<4\text{ }\mu\text{m}$  and  $>64\text{ }\mu\text{m}$  fractions, respectively (Table 2). Similar to the mineral and geochemical variations identified by XRD and DRIFT (Figs. 5 and 6), the elemental composition of the  $<4\text{ }\mu\text{m}$  fraction of the till was comparable to the  $>64\text{ }\mu\text{m}$  sediments of U1 (Fig. 7b, d). Yet, the till was more enriched in Al, Na, Mg, and K, contained less Si, and consequently had lower Si/Al ratios (Table 2). The enrichment of these cations in the fine fraction of the till reflected the predominance of feldspar minerals and cation-rich aluminosilicates such as albite (Figs. 3b and 5a). In addition to the predominant aluminosilicates, some less significant minerals were identified, such as framboidal pyrite (SM 9) and zinc sulfide (Fig. 7b); pyrite was identified in the finest fractions of U2 (Fig. 5b). Diatom frustules were detected throughout the sediment core except the till (Fig. 7a–c).

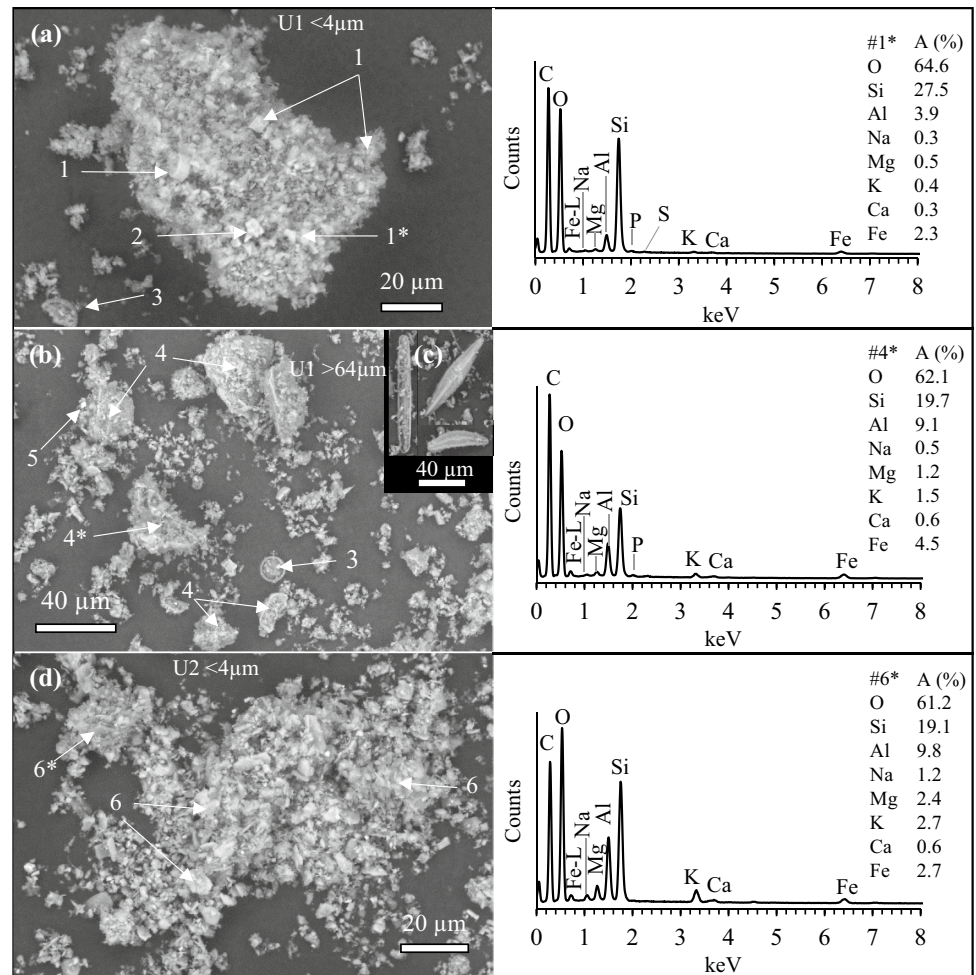
## 5 Discussion

### 5.1 Tracking sedimentary characteristics using a combination of geochemical techniques: application in this study and perspectives

The data obtained from the various geochemical techniques showed similar categorization of the bulk sediments. The sediments were divided into two units with inflection points roughly at 200 cm based on XRD (Fig. 3b) and OM (Fig. 4a), and at 210 cm based on DRIFT (Fig. 4c). In fact, more dissimilar categorization was expected since these techniques yield distinct findings; XRD mainly gives information on crystalline minerals while DRIFT considers chemical, organic, and mineral signatures (regardless of crystallinity). The similarity in the categorization of the sediments strongly suggests that the mineral and organic evolution occurred conjointly as a result of ecosystem development that had occurred in the late glaciation, deglaciation, and post-glaciation periods. Indeed, natural environmental changes that occurred during these periods can lead to such changes (e.g., Snowball et al. 2002; Voldstad et al. 2020; Zimmermann et al. 2021).

The application of the MCR–ALS GUI was initially intended for biological material (Felten et al. 2015, 2019). However, it showed significant findings when applied to the mineral, organic, and geochemical data in this study. Hence, this technique advances the categorization and interpretation of sediment

**Fig. 7** SEM back-scattered electron (BSE) micro-images for sediments. **a** Fine surface sediment ( $<4\ \mu\text{m}$ , case of 1 cm, U1) showing (1) aluminosilicates, (2) Mg-rich aluminosilicate, and (3) diatom frustule. **b** Coarse surface sediment ( $>64\ \mu\text{m}$ , case of 1 cm, U1) showing (4) aluminosilicates, and (5) zinc sulfide. **c** Diatom frustules were generally found in the sediments throughout the core except the till. **d** Fine fraction of the till ( $<4\ \mu\text{m}$ , 225 cm, U2) showing (6) aluminosilicates. Numbers on the images show where EDX spectra were collected; the EDX spectra next to the images belong to the data points marked with asterisks. Due to the similarity in elemental composition (A, %), the same number is assigned to similar phases (in each image). The average elemental composition of these points is included in Table 2. The carbon emission lines in the EDX spectra belong to carbon tape onto which the samples were deposited; carbon was not taken into consideration upon elemental quantification



properties, which is crucial in fields where sediment or soil cores are studied (e.g., paleo-limnology, paleo-ecology, sedimentology, and landscape ecology). Furthermore, the DRIFT data covered the mineral and organic variations of the sediments, which were further improved by linking it to XRD. For instance, amorphous materials were detected by a bump in the diffractograms ( $\sim 15\text{--}25^\circ 2\theta$ ; Figs. 3a and 5b) and by a shoulder in the IR spectra ( $\sim 1200\ \text{cm}^{-1}$ ; Figs. 4a and 6a). In both techniques (i.e., XRD and DRIFT), these signatures were more obvious in the U1 sediments and the  $<4\ \mu\text{m}$  fractions in comparison to the U2 sediments and the coarser fractions. Similar to using IR signatures as a fast, reliable, and cost-effective tool to semi-quantify total organic carbon, carbonates, and biogenic silica (e.g., Rosén et al. 2010; Ancin-Murguzur et al. 2020), IR might be used to quantify amorphous materials in heterogeneous environmental matrices, such as sediments and soils (e.g., Pérez et al. 2016). It should be noted that whether the amorphous material(s) detected in both techniques are the same has not been established. Either way, such findings would be less tedious and more timesaving than the conventional methods of amorphous phase quantification by XRD using internal standards (e.g., Zhou et al. 2018; Thorpe et al. 2019).

## 5.2 First sedimentary materials reaching the lake after the glacial period

Deglaciation in Fennoscandia occurred between 22,000 and 9700 years BP (Mörner 2004; Stroeven et al. 2016). During that period, mountainous areas witnessed steepening, degradation, and collapse of rockwalls, subsequently generating rock debris (Ballantyne 2002; Jarman 2002) that deposited and formed unsorted glacial sediments, i.e., the till (e.g., Hall et al. 2019). Indeed, the till in lake Hotagen was very poorly sorted (SM 5b) and reflected the lithogenic character of the watershed by high quartz (24%), albite (38%), and amphibole (12%) contents (Fig. 3). The till had a finer texture in comparison to the other sediments (Fig. 2c), possibly due to harsh mechanical weathering (e.g., grinding) that resulted from glacial abrasion during deglaciation (Riebe et al. 2004). Additionally, this weathering process also homogenized the sediments and made mineral variation per size fraction less obvious in comparison to the other sediments (Fig. 5a); this also explains the enrichment of feldspar minerals in the fine fraction of the till, whereas these minerals were abundant in the coarse fractions of the

**Table 2** The atomic composition of the aluminosilicates in the three sediment samples presented in Fig. 7; n=4, 5, and 4 for U1 <4 µm, U1 >64 µm, and U2 <4 µm, respectively

	Atomic percentage (%)								Ratio
	O	Si	Al	Na	Mg	K	Ca	Fe	Si/Al
U1 <4 µm	64.5 ± 0.5	27.1 ± 1.3	4.1 ± 0.8	0.2 ± 0.1	0.5 ± 0.1	0.6 ± 0.2	0.3 ± 0.1	2.4 ± 0.5	6.9 ± 1.8
U1 >64 µm	62.0 ± 0.3	20.3 ± 0.5	8.4 ± 0.6	0.8 ± 0.8	0.9 ± 0.3	2.0 ± 1.3	0.6 ± 0.3	4.4 ± 1.5	2.4 ± 0.2
U2 <4 µm	60.9 ± 0.4	19.1 ± 1.2	9.1 ± 0.7	1.4 ± 0.8	2.5 ± 0.5	2.8 ± 0.6	0.6 ± 0.2	3.2 ± 0.9	2.1 ± 0.2

other sediments (Fig. 5a). Moreover, the <4 µm fraction of the till and the >64 µm fractions of the other sediments showed similar geochemical characteristics, such as the presence of quartz overtone peaks, an Si–O stretching peak at 1020 cm<sup>-1</sup>, and Si–O and OH bending peaks (Fig. 6). Furthermore, the bulk and size-fractionated till did not show signatures of X-ray amorphous materials (Figs. 3a and 5b). This indicates the absence of materials such as organic matter, allophanes, and diatom frustules on the one hand, and/or the limitation or absence of chemical weathering of silicate minerals on the other.

Organic matter IR signatures were identified mainly in the 4–16 and 16–64 µm fractions of the till (Fig. 6d). This OM might have been preserved in ice-sheets, possibly in interglacial soils that deposited during deglaciation and post-glaciation, or might have come from organisms that formed during the Late Pleistocene (late glaciation or deglaciation periods) or Early Holocene (e.g., Zale et al. 2018; Zimmermann et al. 2021). Indeed, it was shown that vegetation was able to penetrate thin ice sheets during Late glaciation 14,000–11,000 years ago (Kullman 2002). However, the OM IR peak at 2960 cm<sup>-1</sup> was only detected in the till, and not in the other sediments. This suggests that the unique OM IR band belongs to material(s) that was stored in ice-sheets that thawed in the vicinity of lake Hotagen, was present pre-deglaciation and discontinued post-glaciation (e.g., Clarke et al. 2019; Zimmermann et al. 2021), or originated from another watershed (e.g., Sadori 2013). Radiocarbon dating, organic identification, pollen records, and sedimentary ancient DNA can give more information in this regard (e.g., Öberg and Kullman 2011; Swärd et al. 2018). Although the dating records were only based on three wood samples, the sedimentation rate was higher in the 205–225 cm sediments in comparison to the above 133–205 cm sediments (Fig. 2a). Relatively higher sedimentation rates are due to paraglacial landscape response, i.e., the absence of vegetation, high discharge, and elevated erosion and transportation of deposits (e.g., Ballantyne 2002), which are thought to have predominated during the Late Pleistocene (late glaciation and deglaciation periods) and Early Holocene (post-glaciation).

### 5.3 Sedimentary materials marked ecosystem development during the Late Pleistocene–Holocene

Primary succession evolved during the Late Pleistocene–Early Holocene due to deglaciation, which caused biological, chemical, and physical changes to the ecosystem (e.g., Engstrom et al. 2000; Ballantyne 2019). Terrestrial and aquatic vegetations subsequently increased OM deposition in lakes (Righi and Meunier 1995), as shown by the gradual increase in sedimentary OM in the U2 sediments (Fig. 8a). Furthermore, the gradual change from glacier-covered land (during late glaciation–deglaciation), to bare soil and exposed bedrock (deglaciation–post-glaciation), and eventually vegetated land (Holocene) caused a variation in the materials that were transported in the watershed and finally reached lakes. Such organic and inorganic changes were noted as a function of sediment size and depth (Figs. 3, 4, 5, and 6). Based on the watershed's bedrock and soil coverage in west-central Sweden, the predominant minerals are amphibole, quartz, feldspar, chlorite, and mica (Andersson et al. 1985; Allen et al. 2001; SGU 2019). The sum of quartz, albite, and K-feldspars on the one hand (i.e., the minerals that predominated the coarse fraction) and the sum of the clay minerals (i.e., chlorite and mica minerals that predominated the fine fraction) and OM on the other hand showed opposite trends with depth (Fig. 8a). These trends suggest that OM and mineral variation conjointly reflect ecosystem development. Also, the U2 sediments showed the transition from glacially derived sediments to more common lacustrine sediments, represented by the drastic changes from the till to the overlying U2 sediments (Figs. 3, 4, 5, and 6).

Pyrite might have also reflected the watershed's development (e.g., through change in land cover). Pyrite contents gradually increased from the till to 205 cm (Fig 3b). Since the Fe contents in the sediments were relatively low, pyrite is thought to have been inherited from parent bedrock rather than being formed within the sediments (e.g., Ohfuji and Rickard 2005). So, the variation in pyrite contents might be due to change in land cover (from exposed bedrock to vegetated areas) or modification in the particles that reached the lake as a result of size-dependent transportation caused by vegetation. The latter case might be plausible since pyrite was significantly enriched in

the  $<4\ \mu\text{m}$  size fractions of the U2 sediments, while it showed similar contents in the other fractions (Fig. 5a). Carbonates were not detected in the sediments despite their presence in the bedrock (SGU 2019); the absence of crystalline carbonates was established by XRD (Fig. 3a). Furthermore, it is clear to say that these minerals, if present, were not below the XRD detection limit since they could not be detected by SEM-EDX. In this case, the bedrock containing carbonates might not have been exposed and therefore could not be transported to the lake, or carbonates were dissolved as a result of weathering (e.g., Hanna et al. 2018). Although the former might be more plausible, the latter cannot be ignored due to the acidic pH of the sediments and nearby lakes (SM 1).

The absence of X-ray amorphous materials in the till and their presence in the overlying sediments reflected ecosystem change as well (Figs. 3a and 5b). This change was marked by increasing contents of clay minerals and organic matter (especially between the till and the overlying U2 sediments, Fig. 8a), however, it might also be due to increased contents of allophanes, ferrihydrites, diatom frustules, and weathered silicate minerals (Pérez et al. 2016; Thorpe et al. 2019). Sedimentary aggregates also indicate ecosystem development (e.g., Schaaf et al. 2011). Indeed, sedimentary aggregates showed twofold variations in our case. The first is the gradual increase in the difference between aggregate and particle sizes from the till to the overlying sediments (Fig. 2c). The second variation was noted as a function of depth; with time, larger aggregates were detected in the sediments. The main aggregate size population shifted from  $14\ \mu\text{m}$  in the till, to  $72$  and  $250\ \mu\text{m}$  in the other U2 sediments, and finally to a clear  $340\ \mu\text{m}$  population in all the U1 sediments (Fig. 2c). Both variations are linked to higher contents of cementing materials, which are clay minerals, OM, and X-ray amorphous phases (Fig. 8a). These materials were enriched in the U1 sediments that showed the highest difference between particle and sediment sizes on the one hand, and the biggest aggregate sizes on the other (Fig. 2c). Although not studied here, microorganisms are also linked to aggregate formation via biofilms (Lartiges et al. 2001; Zimmermann-Timm 2002). Therefore, sedimentary aggregates and their composition can be used as proxies to study ecosystem development. Aggregates are affected by river hydrodynamics (e.g., turbulence), physico-chemical properties of the media (e.g., pH, redox conditions, and ionic strength), and suspended matter concentration (e.g., Tran and Strom 2019). Therefore, the trend in aggregate size of the  $<4\ \mu\text{m}$  fraction (Fig. 2b) might be connected to such parameters.

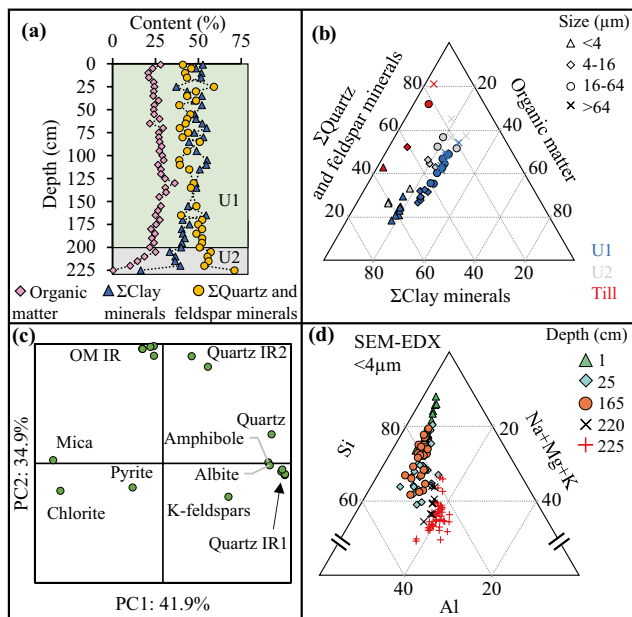
#### 5.4 More insights into ecosystem development through size-fractionated sediments and correlations

The size-fractionated sediments showed a clear variation between OM, the sum of quartz and feldspar minerals, and the sum of clay minerals (Fig. 8b). This was primarily seen

between the till and the other sediments (red symbols in Fig. 8b) and secondly between the U1 and U2 sediments, and reflects ecosystem development marked by important mineral, organic, and geochemical transformations. The U2–U1 transition was recorded in terms of GSD, mineral, organic, chemical, and elemental compositions (Figs. 2c, 3b, 4c, and 8). Unfortunately, the till–220 cm transformation was not well-recorded in this study since sediments were collected every 5 cm; sampling at smaller intervals would have shown this transition better. However, among the different size fractions, the increase in quartz and feldspar minerals and decline in clay minerals was gradually observed with increasing sediment sizes, yet it was more significant between the  $<4\ \mu\text{m}$  fraction and the other fractions combined (Fig. 8b); OM variation was only observed between the  $<4$  and  $>4\ \mu\text{m}$  size fractions, as established by multivariate analysis (Fig. 6a).

The correlation of the mineral and organic properties of the sediments by principal component analysis (PCA) highlighted ecosystem development (Fig. 8c). The first principal component (PC1) explained 41.9% of the variance with positive loadings for quartz, albite, amphibole, and to a lesser extent K-feldspars, and negative loadings for the clay minerals (the correlation matrices and PCA components are included in SM 10). The second component (PC2) described 34.9% of the variance and is directly related to organic matter. As discussed in the previous section, the transformation of those variables marked the changes within the U2 sediments and between U2 and U1 sediments. Moreover, there is a link between the quartz IR data and quartz contents from XRD (Fig. 8c and SM 10). It should be noted that the quartz DRIFT fingerprint was obtained from  $\text{Sed}_{\text{inorg}}$  samples since organic matter bands interfere with parts of the quartz fingerprint (at  $1525\ \text{cm}^{-1}$ ). This correlation was valid for the quartz IR peaks at  $1995$ ,  $1791$ ,  $1875$ , and  $1525\ \text{cm}^{-1}$ , but not for  $1612$  and  $1685\ \text{cm}^{-1}$  (Fig. 8c and SM 10). Indeed, recent studies used quartz IR fingerprints, mainly the Si–O stretching and bending vibrations, to quantify quartz in sediments (Vogel et al. 2016; Hahn et al. 2018).

The aluminosilicates in the  $<4\ \mu\text{m}$  fractions showed variable elemental composition based on four groups, which are 1 cm, 25 and 165 cm, 220 cm, and the till (Fig. 8d). Therefore, even though the sediments were categorized into two units based on major mineral and geochemical characteristics, more distinction was possible at micro-scales. The decline in cations (sum of Na, Mg, and K) from the till towards 220 cm indicated, at least partially, the transition from cation-rich aluminosilicates (e.g., K-feldspar and albite) to mica and chlorite minerals. This transition was more obvious between U2 (220 and 225 cm) and U1 (25 and 165 cm), i.e., when significant sedimentary variation occurred. Interestingly, the abundance of the major cations and ratios did not change in U1, except for the surface



**Fig. 8** Change of OM, the sum of clay minerals (mica and chlorite), and the sum of quartz and feldspar minerals (K-feldspars and albite) for **a** bulk sediments as a function of depth and **b** size-fractionated sediments ( $n=12$  for each size fraction). **b** The U1 sediments (0–195 cm) are marked in blue and the U2 sediments (200–225 cm) are marked in gray, except the till, which is marked in red; each symbol represents a size fraction. **c** Principal component analysis (PCA) matrix for the sediments (bulk and size-fractionated);  $n=60$  for each variable. The data points represent the mineral percentages (XRD-Profex) except for those marked by IR, which were taken from DRIFT data. OM IR represents the DRIFT intensity peaks at 1416, 1630, 2852, 2927, and  $3400 \text{ cm}^{-1}$ . Quartz IR1 and Quartz IR2 represent the DRIFT intensity peaks at 1525, 1790, 1875, and  $1995 \text{ cm}^{-1}$ , and 1612 and  $1685 \text{ cm}^{-1}$ , respectively; since OM interferes with some quartz IR peaks, the quartz IR data were taken from  $\text{Sed}_{\text{inorg}}$  samples, i.e., sediments after OM removal. **d** Atomic abundance (SEM-EDX) of the  $< 4 \mu\text{m}$  fractions of 1, 25, 165, 220, and 225 cm sediments, with  $n=19, 32, 25, 8,$  and 46, respectively

sediment (Fig. 8d). The similarity in the elemental composition of the predominant mineral surfaces in U1 (e.g., 25 and 165 cm) suggests that insignificant mineral variation occurred over a period of roughly 6400 years, i.e., the period during which these sediments had deposited.

In general, the most weathered minerals and mineraloids, such as clay minerals, poorly crystalline minerals, and amorphous materials, are enriched in fine sediments (e.g., Velde and Meunier 2008; Thorpe et al. 2019). Despite the prevailing cold climate conditions in Hotagen that limit chemical weathering and although we did not directly prove that chemical weathering occurred, the enrichment of possibly weathered materials in the  $< 4 \mu\text{m}$  sediments, i.e., clay minerals and X-ray amorphous material (Fig. 5), suggest chemical weathering of silicates. Indeed, chemical weathering was proven to be possible in conditions similar to this study (Thorpe

et al. 2019). In any case, X-ray amorphous materials and clay minerals promote aggregate formation. The formation and size of aggregates depend on surface charge and area of the aggregate-forming materials (e.g., Tombácz and Szekeres 2006). Since chlorite minerals have higher surface charges than mica minerals, chlorite is expected to be the predominant clay mineral in the organo-mineral or mineral–mineral aggregates (Poirier et al. 2020), followed by mica. Interestingly, this was noticeable based on chlorite and mica contents in the different size fractions. Unlike mica, chlorite was not noticeably enriched in the fine fraction (Fig. 5a). Additionally, and in contrast to mica, the difference in chlorite contents between the size fractions was not clear. This strongly suggests that chlorite is the main clay mineral involved in the aggregates. Indeed, it has been statistically proven that mica minerals had more available reactive sites than chlorite, since the latter was part of organo-mineral and mineral–mineral complexes (Kanbar et al. 2020b).

Finally, the combined use of the geochemical techniques, i.e., XRD, DRIFT, and SEM-EDX, and statistical approaches showed sedimentary variations at different scales. Firstly, XRD showed bulk variation in mineralogy, which was used to track major ecosystem changes that occurred during the Late Pleistocene–Early Holocene. This was mainly seen by the change in the abundance of crystalline minerals, the appearance of X-ray amorphous materials (Figs. 3 and 5), and the formation of aggregates (Fig. 2c). Secondly, DRIFT showed mineral, chemical, and organic variations (Fig. 4), which better marked the transition during the deglaciation and post-glaciation periods. Moreover, applying MCR-ALS to the size-fractionated sediments tracked sedimentary evolution with depth as well as sediment size (Fig. 6a–c). Finally, SEM-EDX further identified micro-scale variations of the predominant minerals (Fig. 8b).

## 6 Conclusion

A 2.3 m sediment core collected from lake Hotagen, west-central Sweden, revealed ~10,000 years of ecosystem development that occurred after the last glacial period. The bulk sediments were categorized into two distinct units based on grain size distribution (GSD), bulk mineralogy (XRD), chemical and organic compositions (DRIFT), and micro-scale analyses (SEM-EDX). The first unit (0–195 cm, U1) presented similar mineral and organic characteristics at a bulk scale and the second unit (200–225 cm, U2) marked the development of the catchment during the Late Pleistocene–early Holocene (i.e., during late glaciation, deglaciation, and post-glaciation). The mineral variation was marked by decreasing quartz, feldspar, and amphibole contents and increasing organic matter and clay mineral contents (mica and chlorite) with time. The chemical and organic variations were



clearly shown by multivariate analysis (MCR-ALS) of the DRIFT spectra that further showed distinct subunits. Indeed, the organic and mineral transitions within the U2 sediments and between U1 and U2, especially for the size-fractionated sediments, reflected the progression of the ecosystem post-glaciation. Ecosystem development was also denoted by the development of X-ray amorphous materials as well as the formation of distinct organo-mineral and mineral–mineral aggregates. Chlorite was proven to be the predominant clay mineral included in these aggregates. Furthermore, SEM-EDX revealed micro-scale variations (Na, K, and Mg) of the predominant aluminosilicates. The U1 sediments were shown to be different only at the micro-scale, while the U2 sediments were different at the mineral, chemical, organic, and micro-scales. Additionally, the 0–195 cm sediments, which had deposited over a period of roughly 7500 years, showed similar mineral and organic characteristics. This suggests that the sedimentary materials mainly changed during the early stages of the Pleistocene–Holocene transition that are archived in the U2 sediments. Only minor elemental changes occurred once major ecosystem development ceased, i.e., when organic deposition in the sediments reached a constant level. The geochemical techniques used in this study, especially when applied to the clay-sized fractions, demonstrated bulk, micro-, and elemental scale variations that marked mineral, organic, and elemental progression of the sediments as the ecosystem evolved post-glaciation.

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## Declarations

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## Supplementary Material

# Tracking mineral and geochemical characteristics of Holocene lake sediments: the case of Hotagen, west-central Sweden

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## **SM 1: General geochemical characterization**

### **Methodology**

The elemental composition of selected bulk sediments (5, 130, 220, and 225 cm) was determined by X-ray fluorescence (Bruker S8-Tiger WD-XRF analyzer, EMG, Umeå University, Sweden). Information about the measurement parameters, QA/QC, detection limits, matrix effect, and others is indicated elsewhere (Rydberg 2014). The organic matter content (or Loss on Ignition, LoI) was calculated by the complementary of the sum of major oxides to 100; this value was proven to be representative of LoI<sub>550</sub> (Rydberg 2014). The cation exchange capacity (CEC) was determined by the Cu-trien method and was quantified by UV/Vis spectroscopy at 578 nm (Lambda 750 UV/Vis/NIR spectrometer by Perkin Elmer). Detailed information is found elsewhere (Meier and Kahr 1999; Ammann et al. 2005). The pH was determined using 0.01 M CaCl<sub>2</sub> (e.g., Hendershot et al. 2008).

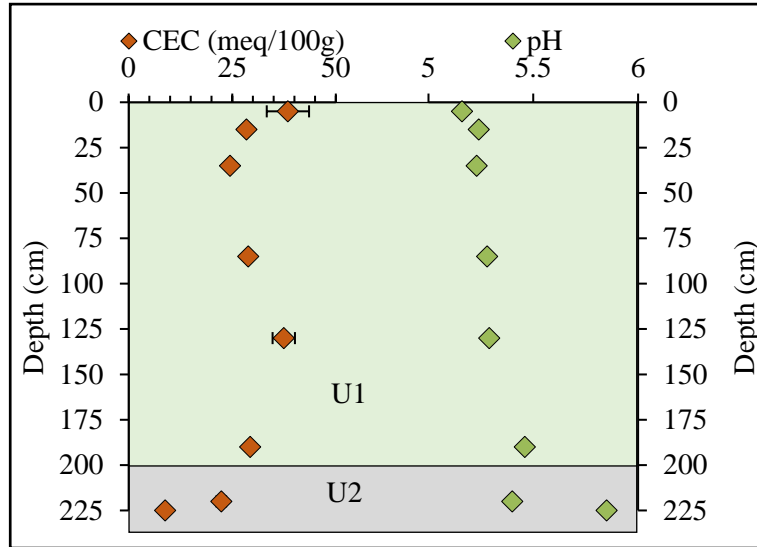
### **Results**

The major elemental variation between the sediments was seen between 220 cm and the till (225 cm) on the one hand, and between 5 and 130 cm on the other (table below). The 220 and 225 cm sediments had relatively high Al, Ba, Ca, K, Mg, Na, Rb, Si, Sr, Ti, V, and Zr contents. The enrichment of Al, Ca, K, Mg, Na, and Si is linked to minerals deposited from the watershed. Indeed, the watershed's lithology is abundant in silicates (e.g., quartz, amphibole, phyllosilicates, and tectosilicates that originate from greywacke, granite, and amphibolite) and carbonatite (ESDAC - European Commission 1963; SGU 2019); the latter might contain Ba, Rb, Sr, Ti, and Zr intrusions (Singer 1992). Furthermore, it has been recently discovered that the nearby city, Håggån, lies in a Vanadium rich zone (Webb 2018; World Nuclear News 2019), thus explaining V enrichment in the deepest sediment. On the other hand, Br, Cl, Fe, Mn, P, S, W, and Zn were generally enriched in the 5 and 130 cm sediments. These metals might originate from detrital material after ecosystem development. Indeed, the development of the ecosystem and the build-up of vegetation increase the deposition of those metals by inducing mineral weathering by organic acids (e.g., Huang and Keller 1972). The watershed does not include industrial areas or highly urbanized regions. This can be seen by the absence of Pb and Zn enrichments towards surface sediments. Organic matter showed a clear variation, especially between the till and the other sediments. The organic matter content was insignificant in the till (0.27%) and then increased to 17% and 25% in the 220 and 130 cm sediments, respectively. The distinct rise in OM marked the development of the ecosystem during the deglaciation – post-glaciation periods (during the Late Pleistocene – Holocene). The CEC increased from the till

towards 130 cm, then fluctuated for the above sediments. In lacustrine or riverine sediments with relatively high OM contents, such as the case of lake Hotagen sediments (except for the till), the CEC is mainly explained by the organic matter content. Moreover, the sediments were acidic and the pH ranged between 5.1 and 5.9. Other lakes in the vicinity of Hotagen were also acidic (Almer et al. 1974; Borg and Johansson 1989; Warfvinge and Bertills 1999), mainly caused by the deposition of nitrogen and sulfur gases emitted from trees inhabiting the region, such as spruce, birch, and pine (e.g., Iwald 2016).

Elemental composition of selected bulk sediments (XRF)

	<b>5cm</b>	<b>130cm</b>	<b>220cm</b>	<b>Till (same as 225cm)</b>
<b>Al (%)</b>	5.3	4.8	7.3	7.3
<b>Ca (%)</b>	0.9	1.3	1.1	2.4
<b>Fe (%)</b>	6.6	4.8	5.2	3.8
<b>K (%)</b>	1.4	1.1	2.9	2.6
<b>Mg (%)</b>	0.6	0.5	1.6	1.5
<b>Na (%)</b>	0.5	0.4	1.2	2.1
<b>P (%)</b>	0.2	0.2	0.1	0.1
<b>S (%)</b>	0.4	0.8	0.5	0.1
<b>Si (%)</b>	23.3	19.8	23.9	31.4
<b>Ti (%)</b>	0.3	0.2	0.4	0.6
<b>As (mg/kg)</b>	7	17.9	17.9	12.9
<b>Ba (mg/kg)</b>	374	267	788	559
<b>Br (mg/kg)</b>	47.3	36.3	18.8	1.7
<b>Cl (mg/kg)</b>	140	158	14	n.d.
<b>Cu (mg/kg)</b>	39	67	132	28
<b>Mn (mg/kg)</b>	694	950	816	698
<b>Ni (mg/kg)</b>	28	53	56	27
<b>Pb (mg/kg)</b>	22	17	34	22
<b>Rb (mg/kg)</b>	85	62	168	102
<b>Sr (mg/kg)</b>	72	78	136	282
<b>V (mg/kg)</b>	71	63	97	104
<b>W (mg/kg)</b>	7.5	4.5	5.4	2.8
<b>Zn (mg/kg)</b>	218	305	242	66
<b>Zr (mg/kg)</b>	162	142	194	356
<b>LoI (%)</b>	24.5	35.4	16.7	0.27



Variation of cation exchange capacity (CEC) and pH of bulk sediments as a function of depth.



**SM 2: Quality of the Rietveld refinement fit and reproducibility of mineral  
quantification (XRD-Profex)**

Profex-BGMN was used for mineral quantification (Doebelin and Kleeberg 2015; Döbelin 2021). The quality of the fit was assessed by changing the refinement parameters (mainly k2, B1, and GEWICHT) and subsequently by following the goodness-of-fit (GoF) and Chi-squared values ( $\chi^2$ ). The  $\chi^2$  and GoF values are reported below for the bulk and size-fractionated sediments.

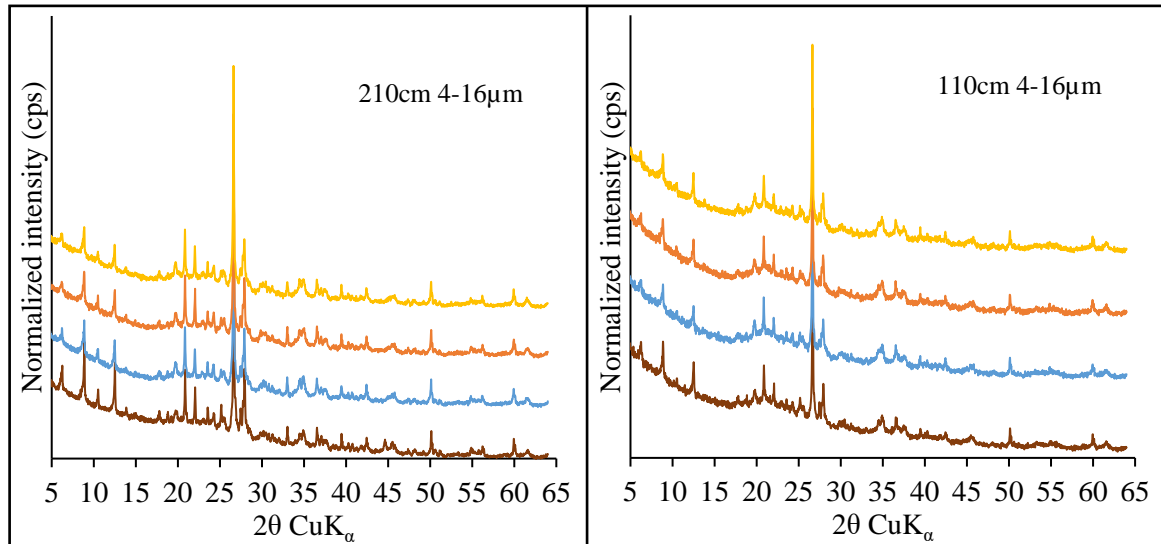
The Chi squared values ( $\chi^2$ ) and goodness-of-fit (GoF) obtained after Rietveld refinement using Profex-BGMN for bulk sediments

Sediment depth (cm)	$\chi^2$	GoF
1	2.34	1.53
5	2.26	1.50
10	2.60	1.61
15	2.26	1.50
25	2.51	1.58
30	2.14	1.46
35	2.43	1.56
40	1.49	1.22
45	1.59	1.26
55	2.41	1.55
60	1.54	1.24
65	2.14	1.46
70	2.33	1.53
75	1.69	1.30
80	2.28	1.51
85	2.62	1.62
95	1.83	1.35
105	1.77	1.33
110	2.06	1.44
115	2.22	1.49
130	1.97	1.40
135	1.62	1.27
155	1.87	1.37
165	1.87	1.37
170	2.12	1.46
175	1.72	1.31
180	2.24	1.50
185	2.42	1.56
190	1.75	1.32
195	1.62	1.27
200	2.32	1.52
205	1.46	1.21
210	2.29	1.51
215	2.45	1.56
220	2.38	1.54
225 (till)	9.65	3.11

The Chi-squared values ( $\chi^2$ ) and goodness-of-fit (GoF) obtained after Rietveld refinement using Profex-BGMN for size-fractionated sediments

	Sediment depth (cm)	$\chi^2$	GoF
<4 $\mu\text{m}$	1	1.69	1.30
	25	3.04	1.74
	55	2.38	1.54
	75	2.48	1.57
	110	2.50	1.58
	135	1.62	1.27
	165	1.77	1.33
	180	1.49	1.22
	205	1.57	1.25
	210	1.23	1.11
	215	1.53	1.24
	225	3.09	1.76
4-16 $\mu\text{m}$	1	1.66	1.29
	25	1.96	1.40
	55	2.45	1.56
	75	1.62	1.27
	110	1.26	1.12
	135	1.55	1.24
	165	1.42	1.19
	180	1.80	1.34
	205	1.30	1.14
	210	1.83	1.35
	215	1.45	1.20
	225	3.27	1.81
16-64 $\mu\text{m}$	1	2.87	1.70
	25	2.26	1.50
	55	2.17	1.47
	75	1.84	1.36
	110	1.56	1.25
	135	1.64	1.28
	165	1.79	1.34
	180	1.94	1.39
	205	4.58	2.14
	210	2.28	1.51
	215	5.49	2.34
	225	3.03	1.74
>64 $\mu\text{m}$	1	2.94	1.71
	25	2.97	1.72
	55	2.99	1.73
	75	2.23	1.49
	110	1.41	1.19
	135	1.48	1.21
	165	1.67	1.29
	180	5.94	2.44
	205	2.76	1.66
	210	4.34	2.08
	215	5.39	2.32
	225	19.46	4.41

To check the reproducibility of mineral quantification using Profex-BGMN, two samples were mounted and run four times. Each sample was unloaded and loaded between the four measurements. On another note, it should be mentioned that the divergence slit and mask (included in the main text) were chosen so that the X-ray irradiated the area occupied by the sample.



The diffractograms of the four measurements (replicates) for each of the two samples (210 cm 4-16 µm and 110 cm 4-16 µm). The diffractograms are stacked for clearer visualization.

Mineral contents of the two sediment samples (average  $\pm$  standard deviation, n=4)

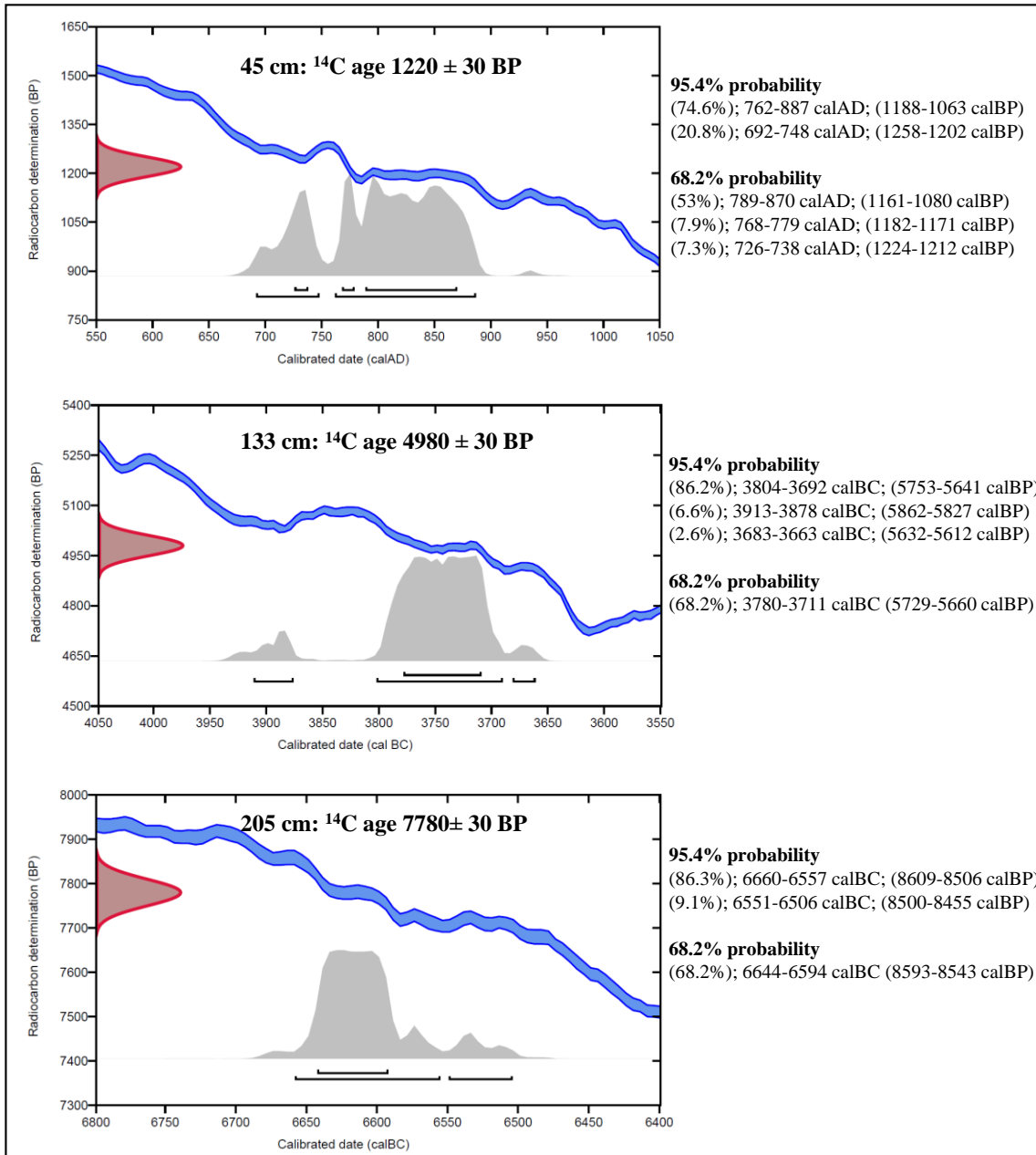
	Chlorite	Mica	Amphibole	Albite	Quartz	K-Feldspars	Pyrite
210cm, 4-16µm	11.7 $\pm$ 0.6	49.6 $\pm$ 0.4	3.8 $\pm$ 0.4	13.3 $\pm$ 0.4	14.2 $\pm$ 0.6	6.9 $\pm$ 0.7	0.5 $\pm$ 0.1
110cm, 4-16µm	13.1 $\pm$ 1.7	26.1 $\pm$ 1.2	5.6 $\pm$ 0.6	22.8 $\pm$ 0.9	19.0 $\pm$ 0.8	11.5 $\pm$ 0.1	1.9 $\pm$ 0.1

### **SM 3: Organic matter removal and DRIFT notes**

Sedimentary organic matter was removed from bulk and size-fractionated sediments. The initial sediments are named Sed and the ones after organic matter removal are named Sed<sub>inorg</sub>. Several methods are found in the literature to remove organic matter from sediments and soils (e.g., Mikutta et al. 2005). In our case, we are aiming for a method that will result in minimal mineral alteration. Therefore, we used a relatively gentle method, which is NaOCl treatment. Even though chemical treatment by NaOCl does not remove all the organic matter, it does not cause a change in the mineral structure of the matrix, like ashing (thermal combustion) or H<sub>2</sub>O<sub>2</sub> treatment would do (Mikutta et al. 2005; Margenot et al. 2015). The detailed steps of using NaOCl to remove OM are included in Margenot et al. (2019).

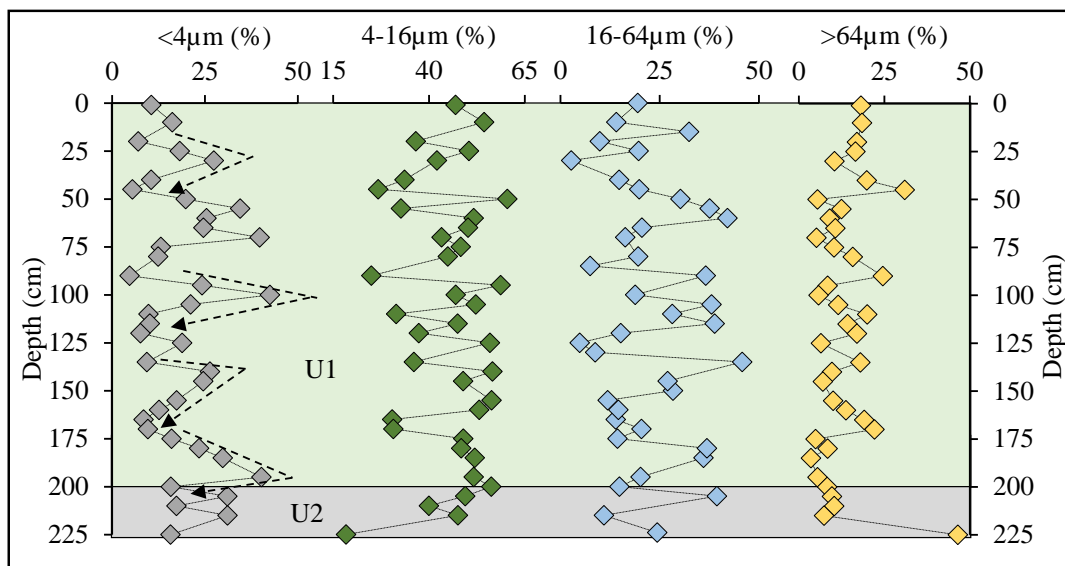
On another note, the peak intensities of OM obtained from the DRIFT spectra can be compared without needing to divide by another normalizing peak (e.g., quartz at 1874 cm<sup>-1</sup> in Padilla et al. (2014)) since the raw spectra were treated and normalized by the multivariate curve resolution alternating least square gentle user interface (MCR-ALS GUI) provided by the Vibration Spectroscopy Core Facility at Umeå University ([www.umu.se/en/research/infrastructure/visp/downloads/](http://www.umu.se/en/research/infrastructure/visp/downloads/)).

## SM 4: The calibration of radiocarbon ages to calendar ages



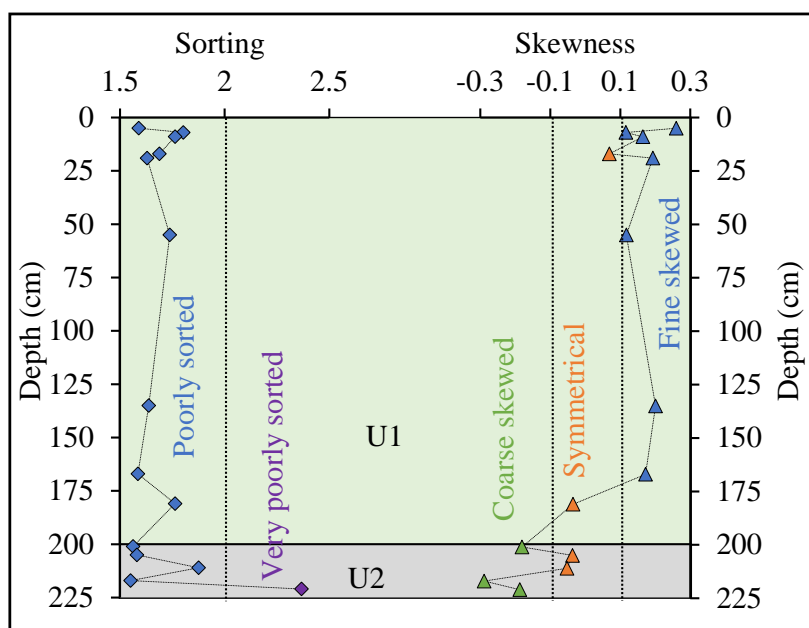
## SM 5: Sediment size characteristics

a) Sediment size as a function of depth (separation according to Stokes' law).



Content of sediment sizes (including aggregates) as a function of depth. Particle sizes are reported in Fig. 2c of the main text.

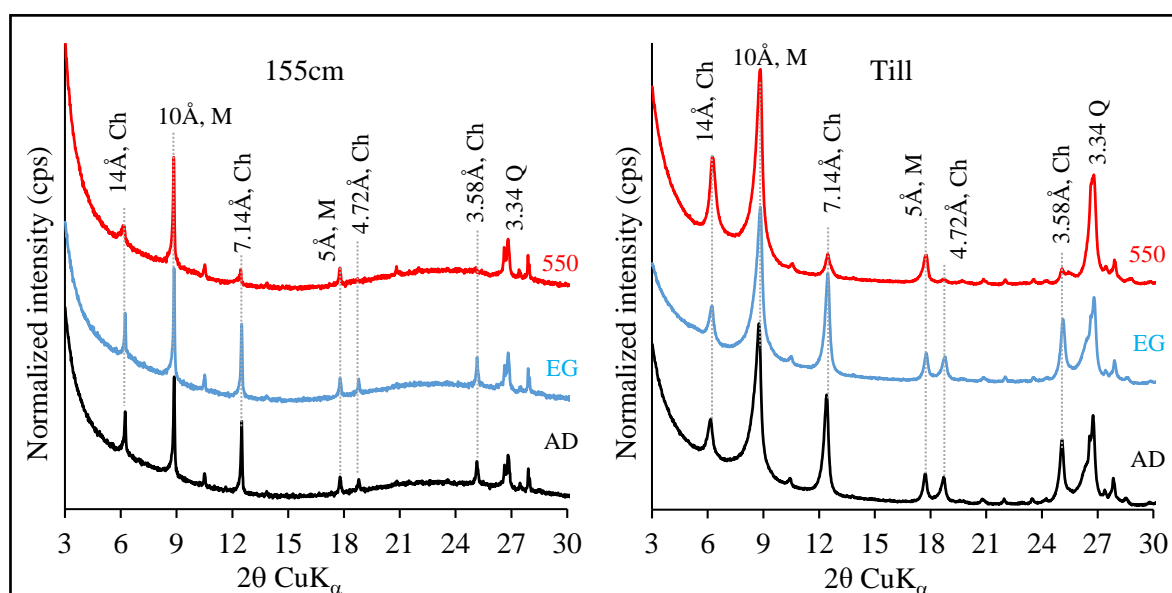
b) Grain size characteristics according to Blott and Pye (2001), calculations were made according to Folk and Ward (1957).



Sorting and skewness of lake Hotagen sediments as a function of depth.

## SM 6: Oriented XRD patterns

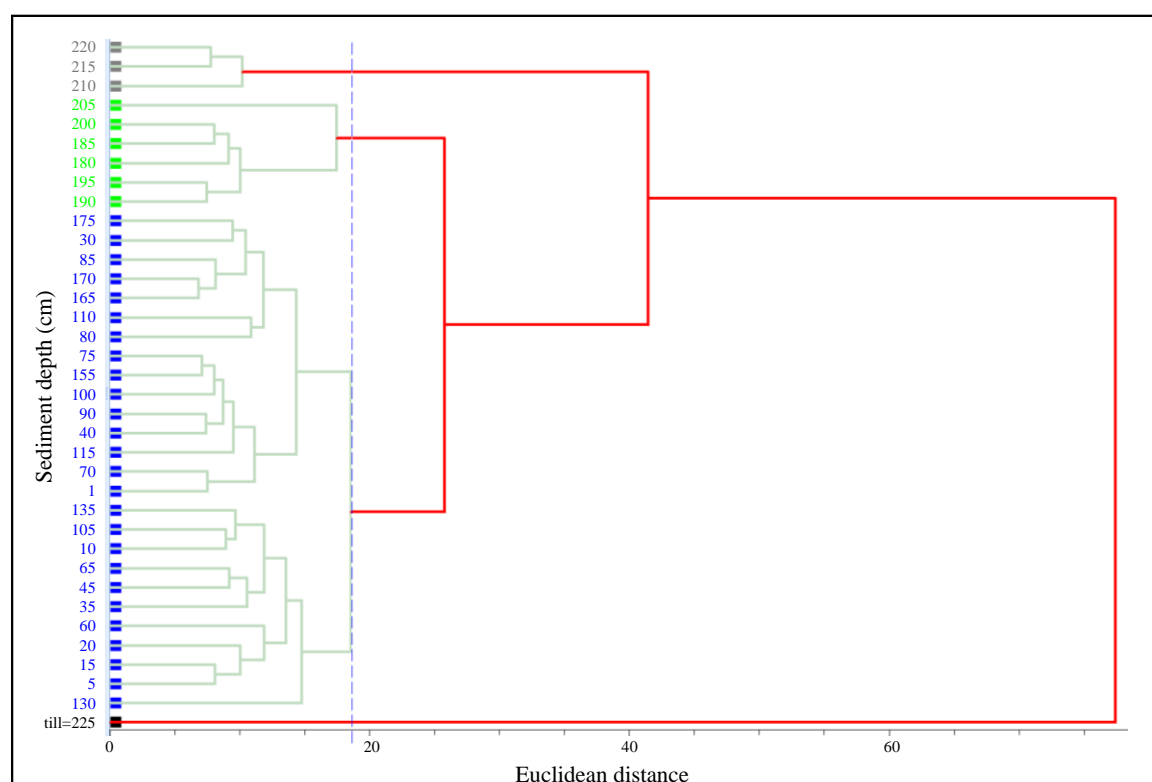
The persistence of the 14 and 10 Å peaks for all the treatments indicates the presence of chlorite and mica minerals. Chlorite minerals are further identified by the increase in the 14 Å peak after heat treatment (> 500°C), as clearly seen in the till (figure below), while higher-order peaks, such as 7.14, 4.72, and 3.85 Å noticeably decrease (Poppe et al. 2001). Nonetheless, the 7.14 Å peak represents the 002 reflection of chlorite as well as the 001 reflection of kaolinite. Since the 060 reflection of kaolinite (1.50 Å) was not visible in the diffractograms (Fig. 3 in the main text), it is safe to say that kaolinite is not present in the sediments. Rather, the 7.14 Å peak strictly belongs to chlorite. The absence of kaolinite was also confirmed by the nonappearance of the OH stretching peaks of kaolinite octahedral layers (at 3697 and 3620 cm<sup>-1</sup>, Fig. 4a in the main text). Furthermore, only the 004 and 060 reflections of tri-octahedral chlorites were visible (3.58 and 1.54 Å). The 060 reflection between 1.50 and 1.54 Å, as in this case, indicates tri-octahedral phyllosilicates; di-octahedral silicates have the 060 reflection between 1.48 and 1.50 Å (Brown and Brindley 1980). The sediments do not contain any swelling clay minerals (e.g., smectites), as indicated by the EG-oriented slides. It is worth noting that all the sediments showed comparable mineral composition (i.e., the same minerals were found with depth). Hence, we assume that the findings of the oriented XRD slides for these two sediments apply throughout the core.



Oriented XRD patterns for the 155 cm sediment and the till (225 cm). For both samples, the air-dried (AD in black), ethylene glycol treated (EG in blue), and calcinated (550°C in red) samples are shown. Ch: chlorite, M: mica, and Q: quartz.

## **SM 7: Categorization of the sediments**

Hierarchical clustering of the XRD data roughly showed 3 clusters which are 0-175 cm, 180-205 cm, and 210-220 cm (figure below). The till (225 cm) showed unique attributes and was not categorized among the other sediments. This highlights the uniqueness of the till and the transition in the mineral properties until 175 cm. The mineral composition of the 0-175 cm sediments was rather similar. Nonetheless, the transition in 180-200 cm sediments seemed minor in comparison to the 200-225 cm sediments, as mainly seen by the predominant minerals (clay and feldspar minerals, Fig. 3b in the main text). Infrared characteristics (DRIFT, Fig. 4 in the main text) showed similar variation with depth, yet the inflection point of the organic infrared signature was at 200 cm (inset in Fig. 4a) while the inflection point of the whole IR spectral region was at 210 cm (Fig. 4c); another less significant inflection point was detected at 200 cm. Therefore, categorizing the sediments might not comply with the different data presented in the paper. However, based on the overall mineral and geochemical data of the bulk and size-fractionated sediments, the sediment core was sectioned into the units U1 (0-195 cm) and U2 (200-225 cm).

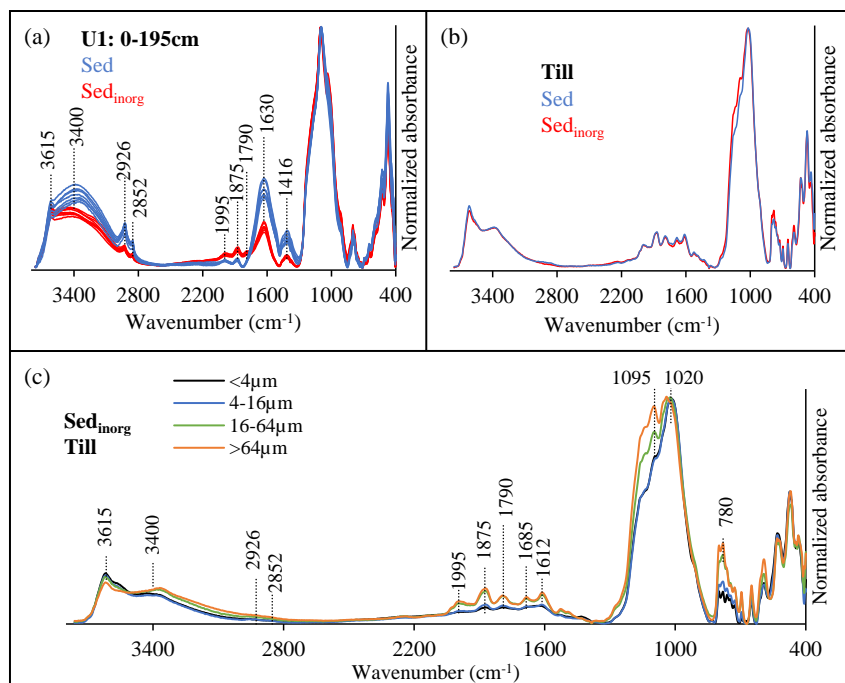


Dendrogram from hierarchical cluster analysis of the bulk sediments based on XRD data. HighScore Plus version 3.0e (PANalytical) was used for cluster analysis (Degen et al. 2014).



**SM 8: DRIFT characteristics of Hotagen lake sediments before (Sed) and after organic matter removal (Sed<sub>inorg</sub>)**

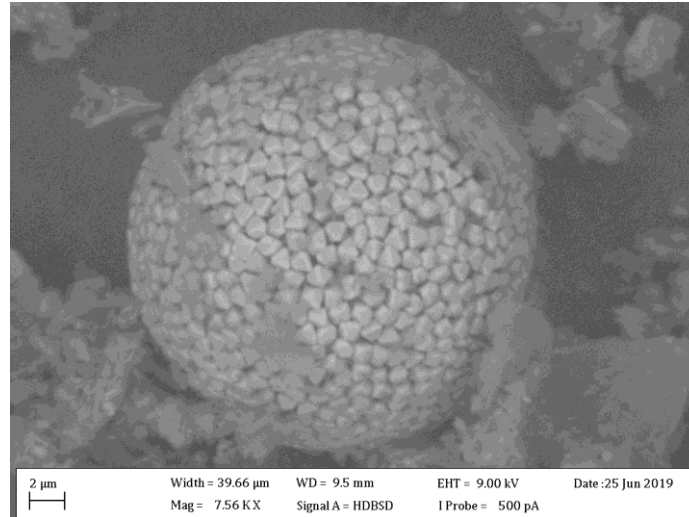
Sed<sub>inorg</sub> IR spectra showed lower peaks at 3400, 2926, 2852, 1630, and 1416 cm<sup>-1</sup> in comparison to Sed, indicating the removal of OM, although not completely (part “a” in the figure below). The similarity in the bulk Sed<sub>inorg</sub> DRIFT spectra of U1 sediments indicates that organic matter was the main variable, while mineral composition did not seem to vary at a bulk scale. In the U1 Sed<sub>inorg</sub>, MCR-ALS only showed one component. It should be noted that minerals were not affected by NaOCl treatment, as the Sed and Sed<sub>inorg</sub> spectra are similar (e.g., the till in part “b”). The quartz fingerprint in the bulk and size-fractionated sediments was clearer after OM removal. Indeed, the Si-O stretching peak at 1020 cm<sup>-1</sup> and the quartz fingerprint at 600-400, 1995, and 1875 cm<sup>-1</sup> became more obvious in Sed<sub>inorg</sub> (part “c” below). Additionally, the shoulder at 1250-1170 cm<sup>-1</sup> was more prominent after organic matter removal, indicating that the peak might be due to quartz, diatom frustules, or amorphous biogenic silica, but not organic matter. Since this shoulder was more prominent in the coarse fraction, it was assigned to quartz. The quartz overtone peaks are indicated in the 2000-1600 cm<sup>-1</sup> range. Higher contents of quartz are also indicated by the protrusion of the 1020 cm<sup>-1</sup> peak and shoulder at 1250-1170 cm<sup>-1</sup> for the coarse fractions.



a) Sed and Sed<sub>inorg</sub> DRIFT spectra of bulk U1 sediments (0-195 cm). b) Sed and Sed<sub>inorg</sub> DRIFT spectra of the till (225 cm). c) DRIFT spectra of the till after OM removal (Sed<sub>inorg</sub>) showing the quartz overtone peaks.

## **SM 9: Scanning electron microscopy**

Framboidal pyrite detected in the U2 sediments.



SEM image of the till (225 cm, <4  $\mu\text{m}$ ).

## SM 10: Correlation between the minerals of the bulk and size-fractionated sediments

Spearman correlation for the minerals of the bulk and size-fractionated sediments (XRD-Profex, n=60)

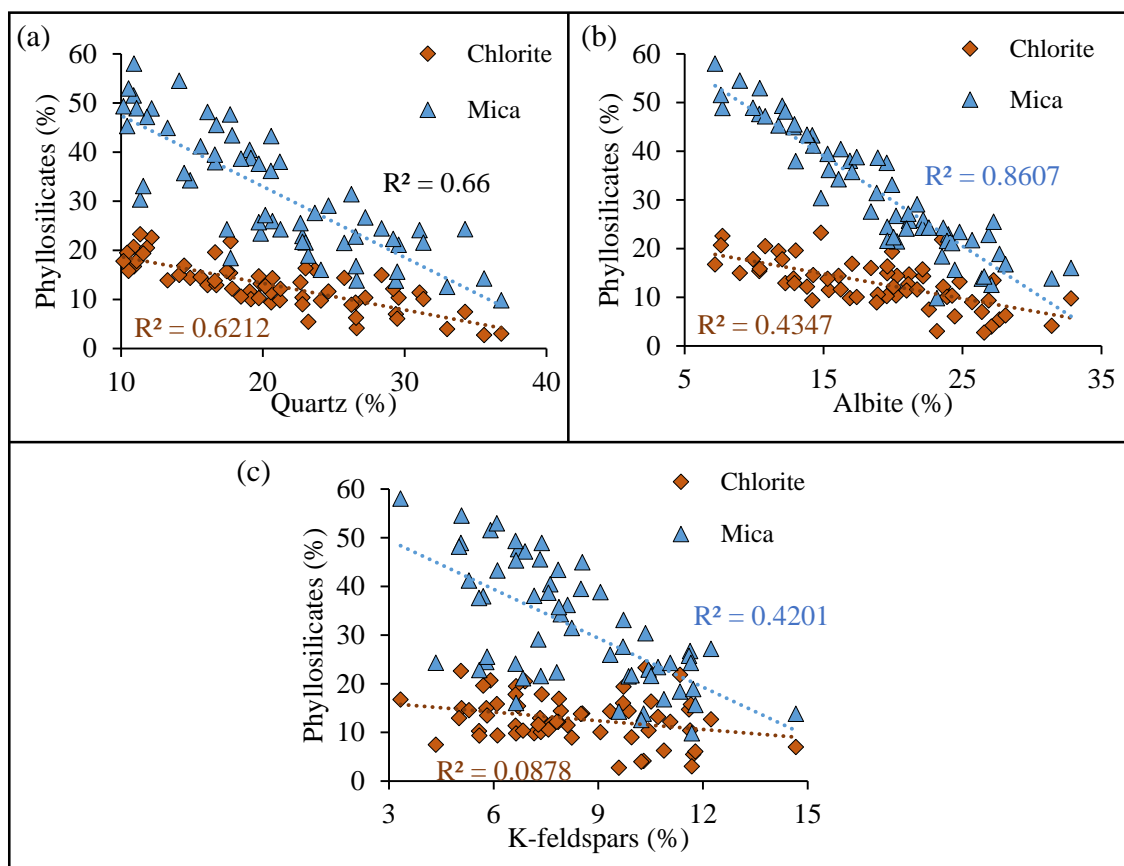
Spearman correlation		Mica	Amphibole	Albite	Quartz	K-feldspars	Pyrite
Chlorite	Rho ( $\rho$ )	.594**	-.568**	-.665**	-.781**	-.263*	0.046
	P	0.000	0.000	0.000	0.000	0.042	0.727
Mica	Rho ( $\rho$ )		-.780**	-.928**	-.826**	-.600**	0.007
	P		0.000	0.000	0.000	0.000	0.957
Amphibole	Rho ( $\rho$ )			.707**	.588**	.395**	-0.132
	P			0.000	0.000	0.002	0.313
Albite	Rho ( $\rho$ )				.776**	.524**	0.033
	P				0.000	0.000	0.804
Quartz	Rho ( $\rho$ )					.319*	-0.121
	P					0.013	0.355
K-feldspars	Rho ( $\rho$ )						0.254
	P						0.051

Rho ( $\rho$ ): Spearman's rho.

P: P-value. The P-value is the probability of finding the observed results when the null hypothesis ( $H_0$ ) is true.  $H_0$ : there does not exist a correlation between X and Y.

\*\* : Correlation is significant at the 0.01 level (2-tailed).

\* : Correlation is significant at the 0.05 level (2-tailed).



Correlation between clay minerals (chlorite and mica) and a) quartz, b) albite, and c) K-feldspars for bulk and size-fractionated sediments, n=60 for each plot.

Spearman correlation for quartz data based on the bulk and size-fractionated sediments (XRD and DRIFT, n=60). Due to the interference of organic matter IR peaks, the quartz IR intensities were obtained from Sed<sub>inorg</sub> data (i.e., sediment after the removal of organic matter).

		Quartz IR peak intensity (DRIFT)					
Spearman correlation		1525cm <sup>-1</sup>	1612cm <sup>-1</sup>	1685cm <sup>-1</sup>	1790cm <sup>-1</sup>	1875cm <sup>-1</sup>	1995cm <sup>-1</sup>
<b>Quartz (XRD)</b>	Rho (ρ)	.693**	0.244	.295*	.759**	.854**	.804**
	<i>P</i>	0.000	0.060	0.022	0.000	0.000	0.000
<b>1525cm<sup>-1</sup></b>	Rho (ρ)		.278*	.343**	.833**	.761**	.752**
	<i>P</i>		0.031	0.007	0.000	0.000	0.000
<b>1612cm<sup>-1</sup></b>	Rho (ρ)			.968**	.257*	0.215	.312*
	<i>P</i>			0.000	0.048	0.100	0.015
<b>1685cm<sup>-1</sup></b>	Rho (ρ)				.379**	.324*	.430**
	<i>P</i>				0.003	0.012	0.001
<b>1790cm<sup>-1</sup></b>	Rho (ρ)					.957**	.947**
	<i>P</i>					0.000	0.000
<b>1875cm<sup>-1</sup></b>	Rho (ρ)						.972**
	<i>P</i>						0.000

Rho (ρ): Spearman's rho.

*P*: P-value. The P-value is the probability of finding the observed results when the null hypothesis (H<sub>0</sub>) is true. H<sub>0</sub>: there does not exist a correlation between X and Y.

\*\* : Correlation is significant at the 0.01 level (2-tailed).

\* : Correlation is significant at the 0.05 level (2-tailed).

Component matrix for the PCA data presented in Fig. 8c.

	PCA Component	
	PC1	PC2
$\Sigma$ Clay minerals (mica+chlorite)	-0.949	-0.102
$\Sigma$ Quartz+feldspars	0.937	0.123
Quartz Sed <sub>inorg</sub> 1875cm <sup>-1</sup>	0.934	
Quartz Sed <sub>inorg</sub> 1790cm <sup>-1</sup>	0.927	
Quartz Sed <sub>inorg</sub> 1525cm <sup>-1</sup>	0.903	
Quartz Sed <sub>inorg</sub> 1995cm <sup>-1</sup>	0.903	
Mica	-0.895	
Albite	0.868	
Quartz	0.855	0.289
Amphibole	0.817	
Chlorite	-0.791	-0.264
K-feldspars	0.555	-0.246
Pyrite	-0.182	-0.175
OM 3400cm <sup>-1</sup>	-0.108	0.981
OM 1630cm <sup>-1</sup>	-0.128	0.977
OM 2927cm <sup>-1</sup>	-0.188	0.962
OM 2852cm <sup>-1</sup>	-0.193	0.96
OM (%)	-0.172	0.942
OM 1416 cm <sup>-1</sup>		0.905
Quartz Sed <sub>inorg</sub> 1612cm <sup>-1</sup>	0.166	0.888
Quartz Sed <sub>inorg</sub> 1685cm <sup>-1</sup>	0.288	0.799

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