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Long-term development and trajectories of inferred lake-water organic carbon and pH in naturally acidic boreal lakes

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Abstract

Monitoring of surface waters in the boreal region over the last decades shows that waters are becoming browner. This timeframe may not, however, be sufficient to capture underlying trajectories and driving mechanisms of lake-water quality, important for prediction of future trajectories. Here we synthesize data from seven lakes in the Swedish boreal landscape, with contemporary lake-water total organic carbon (TOC) concentrations of 1.4–14.4 mg L^{-1}, to conceptualize how natural and particularly human-driven processes at the landscape scale have regulated lake-water TOC levels over the Holocene. Sediment-inferred trends in TOC are supported by several proxies, including diatom-inferred pH. Before $\sim$ 700 CE, all lakes were naturally acidic (pH 4.7–5.4) and the concentrations of inferred lake-water TOC were high (10–23 mg L^{-1}). The introduction of traditional human land use from $\sim$ 700 CE led to a decrease in lake-water TOC in all lakes (to 5–14 mg L^{-1}), and in four poorly buffered lakes, also to an increase in pH by > 1 unit. During the 20th century, industrial acid deposition was superimposed on centuries of land use, which resulted in unprecedentedly low lake-water TOC in all lakes (3–11 mg L^{-1}) and severely reduced pH in the four poorly buffered lakes. The other lakes resisted pH changes, likely due to close connections to peatlands. Our results indicate that an important part of the recent browning of boreal lakes is a recovery from human impacts. Furthermore, on a conceptual level we stress that contemporary environmental changes occur within the context of past, long-term disturbances.

Long-term, human activities can have wide-ranging, enduring impacts on terrestrial landscapes and the lakes embedded within them. Perhaps the most important long-term human activity in the boreal forest region of northern Europe is historical agriculture, which was very different to contemporary, industrial farming (Emanuelsson 2009). In the context of Scandinavia, the limited availability of arable soils meant that forests and mires played a crucial part in food production and their use was shaped by the available resources (Frödin 1952; Bele and Norderhaug 2013). In southern Sweden livestock, including cattle, sheep, and goats, were grazed in forest outfields surrounding the farms, and winter fodder came mainly from leaf-hay harvesting from the coppicing and pollarding of broadleaved trees and shrubs (Slotte 2001). Further north, farmers relied extensively on a transhumant system where livestock were moved from the home farms to remote forest farms (*fäbod*) during the summer months (Supporting Information Fig. S1, Larsson 2012). At the summer farms, livestock were grazed in the forest by day and hay (sedges) was cut from surrounding wetlands for winter fodder (Frödin 1952). Due to the varied nature of these historical activities, we refer to them collectively as traditional land use.

Traditional land use has long been associated with lake-water pH increases (i.e., cultural alkalization; Renberg et al. 1993*b*), but more recent studies also show a link to carbon sequestration and export. Human actions to improve grazing (i.e., thinning and burning) and the grazing and harvesting itself create more open forests with decreased aboveground biomass (Moen et al. 1999; Gimmi et al. 2013; Fredh et al. 2019) leading to less litter transferred to soils and thus decreased soil carbon pools (Bowden et al. 2014; Lo et al. 2015). The abandonment of these traditional land-use practices during the late 19th and early 20th centuries in favor of modern forestry has led to large-scale reforestation and

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Additional Supporting Information may be found in the online version of this article.

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increase (i.e., recovery) in the standing forest biomass (Lindbladh et al. 2014).

Over the past decades, environmental monitoring programs have registered increasing concentrations of colored organic carbon in lakes across the boreal region (Monteith et al. 2007), a trend colloquially referred to as surface-water browning. Browning has a strong influence on lake ecosystems through stronger lake stratification and changes in both nutrient dynamics and habitat availability (Solomon et al. 2015). Organic carbon in boreal lakes is dominated (> 95%) by terrestrial, allochthonous material (Meili 1992; Jonsson et al. 2001), and the majority of the total organic carbon (TOC) pool consists of dissolved organic carbon (DOC; 97%, Kortelainen et al. 2006). With such large terrestrial influence, lakes are sensitive to changes in their surrounding landscape. The large-scale reforestation associated with the transition from traditional land-use to industrial forestry is therefore considered as one of the important drivers of browning (Finstad et al. 2016; Škerlep et al. 2020). However, other important drivers are also recognized, including the recovery from acid deposition (Monteith et al. 2007; Meyer-Jacob et al. 2019) due to the effects of acidity on terrestrial organic matter solubility and mobility (Evans et al. 2012), and climate change due to its effects on terrestrial productivity and hydrology (Finstad et al. 2016; Weyhenmeyer et al. 2016).

The drivers of surface-water browning have mainly been studied on decadal timescales corresponding to the timeframe of environmental monitoring. Truly long-term drivers are rarely considered, even though the long-term perspective has proved highly valuable for assessment of future ecosystem vulnerability and establishment of management goals with respect to other contemporary environmental issues (Bennion et al. 2011). An example is acidification, where long-term paleolimnological studies established that lake-water pH during the 1980s were significantly lower than preindustrialization levels (Battarbee 1984), but also that even earlier human activities, in this case traditional land use, had produced a cultural alkalization of many naturally acid lakes (Renberg et al. 1993*b*).

Here we contribute to the understanding of current monitoring trends in surface-water carbon concentrations by presenting long-term (Holocene) trajectories of sediment-inferred, lake-water TOC in seven boreal lakes in Sweden, with a range of contemporary TOC concentrations from 1.4 to 14.4 mg L^{-1}. Previous research has established a reliable conceptual model for the Holocene development of lake-water pH in Sweden (Renberg et al. 1993*a*), which we expand on and present a similar model for lake-water TOC based on our multiple Holocene lake-water TOC profiles. Changes in lake-water TOC are supported by diatom-inferred pH trends, pollen-inferred vegetation cover, and catchment-disturbance indicators based on elemental geochemistry and charcoal particles, parts of which have been previously published. The synthesis of new lake-water TOC data and previously published data allows us to comprehensively interpret how long-term, landscape scale processes, including natural landscape and vegetation development, traditional land-use-driven vegetation change and modern industrial acid deposition, have influenced carbon dynamics. The seven study lakes are divided into two groups, "southern" and "central" lakes, based on their geographic locations in Sweden (Fig. 1). These groups also broadly represent distinct ecosystems, with different modern, measured lake-water TOC states (1.4–8.7 mg L^{-1} for the southern and 13.6–14.4 mg L^{-1} for the central lakes; Table 1), different documented pH responses to acid deposition (Ek and Korsman 2001), and different long-term developments of lake-water TOC. The southern lakes have previously been used as examples of typical "clear" boreal forest lakes (Renberg et al. 1993*a*; Boyle 2007), and we aim to critically examine this notion in light of our new lake-water TOC reconstructions.

Methods and sites

Study areas

Four of the seven study lakes (Måkevatten, Lysevatten, Härsvatten, and Lilla Öresjön, Fig. 1a) are located ~ 30 km inland from the Swedish southwest coast at 106–143 m above sea level (a.s.l.). The climate is maritime (Köppen Cfc) with annual mean temperatures of +6.7–6.9°C and precipitation of 850–1100 mm yr^{-1} (smhi.se, last accessed 17 December 2019). The other three lakes (Dragsjön, Lång-Älgsjön, and Tryssjön) are located 350 km NE of the southern lakes in central Sweden at 344–435 m a.s.l. (Fig. 1b). Here the climate is continental to subarctic (Köppen Dfb/c) with an annual mean temperature of +4.1°C and precipitation of 850 mm yr^{-1}. Common for both regions are catchments dominated by felsic bedrock with minor, or no, mafic intrusions. The bedrock is overlain by thin glacial till, although deeper pockets occur within the catchments of Lilla-Öresjön and Tryssjön, and bedrock outcrops are frequent. The vegetation is typical for shallow, nutrient-poor soils in the nemoboreal and boreal forest regions and is dominated by Scots pine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* [L.] H. Karst) and birch (*Betula* spp.), with a field layer of dwarf shrubs (*Empetrum* and *Vaccinium* spp.). Two of the central lakes, Lång-Älgsjön and Dragsjön, have peatlands and floating *Sphagnum* mats along much of their shorelines (Fig. 1d), features which are strongly limited at the third central lake, Tryssjön, and contrast the rocky shorelines of the southern lakes (Fig. 1e). Table 1 provides locations, characteristics, and water chemistry for the seven lakes.

Historical records show the presence of summer farms at the central lakes (Hillerström 1984) and previous pollen analysis have documented signs of grazing at both the central and southern lakes (Renberg 1990; Guhrén et al. 2007; Rosén et al. 2011; Meyer-Jacob et al. 2015). However, due to the domination of glacial till in all of the lake catchments, the sites were likely never subject to extensive human settlement or crop cultivation.

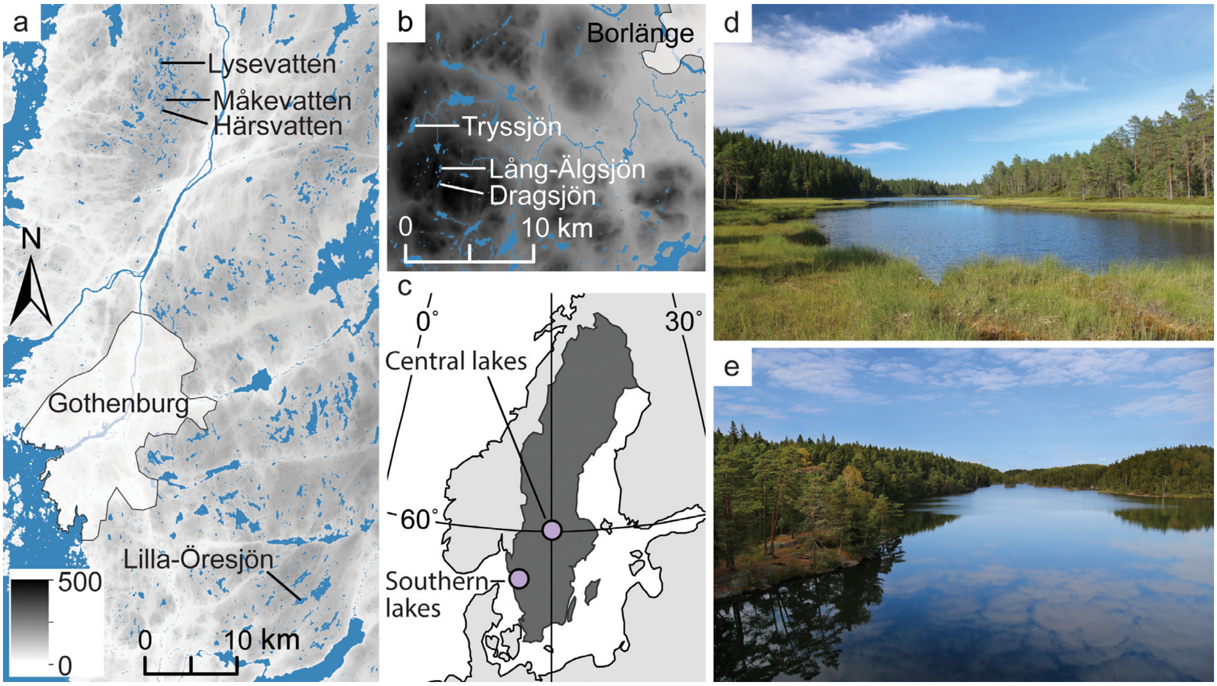

Fig. 1. Location of the southern (**a**) and central study lakes (**b**) in Sweden (**c**). Dragsjön (**d**) belongs to the central lakes and Härsvatten (**e**) belongs to the southern lakes. Note the extent of peatland at Dragsjön (**d**) and the lack thereof at Härsvatten (**e**). The grayscale shading indicates elevation (m) for maps (**a**, **b**). Detailed maps for each lake catchment can be found in Supporting Information Figs. S2–S8.

Table 1. Catchment and lake characteristics, and water chemistry for the seven study lakes.

Lake	Lilla-Öresjön	Härsvatten	Måkevatten	Lysevatten	Dragsjön	Lång-Älgsjön	Tryssjön
Coordinates	57°33′2″N 12°19′30″E	58°1′26″N 12°2′2″E	58°2′8″N 12°2′25″E	58°4′3″N 12°1′40″E	60°23′41″N 15°7′16″E	60°24′18″N 15°7′21″E	60°26′8″N 15°5′1″E
Elevation (m)	106	131	142	113	432	435	344
Maximum lake depth (m)	17	25	12	18	6	12	20
Lake area (ha)	62	19	14	41	5	5	30
Catchment area (ha)	392	149	76	69	88	66	1245
Catchment/lake area	7	10	5	2	22	13	42
Peatlands (%)	15	11	6	6	15	35	16
Lake-water TOC (mg L^{-1})	8.7[*]	5.8[†]	1.4–4.5[‡]		13.6[‖]	14.4[‖]	13.8[¶]
Color (mg Pt L^{-1})	75[*,#]	15[†,#]		25[§]	154[‖]	172[‖]	130[¶,#]
Absorbance 420 nm	0.15[*]	0.03[†]					0.26[¶]
pH	5.7[*]	5.1[†]	4.4–4.8[‡]	7.0[§]	4.7[‖]	4.5[‖]	6.4[¶]
Total phosphorus (μg L^{-1})	9.9[*]	4.1[†]	4–7[‡]		13[‖]	11[‖]	7.0[¶]

[*]Lilla Öresjön. Data represent averages of 41 measurements made 2010–2019. Data from the Swedish national monitoring program ("Sjöar trendstationer"; miljodata.slu.se, last accessed 17 December 2019).
[†]Härsvatten. Lake-water TOC and pH data represent averages of 50 measurements made 2010–2019, and total phosphorus and absorbance 420 nm data averages of 28 measurements made 2010–2013. Data from the Swedish national monitoring program ("IKEU"; miljodata.slu.se, last accessed 17 December 2019).
[‡]Måkevatten. Data represent the range of four measurements made in 1990, 1995, 2000, and 2005. Data from the Swedish national monitoring program ("Omdrevssjöar"; miljodata.slu.se, last accessed 17 December 2019).
[§]Lysevatten. Data represent averages of five measurements made 2010–2015. Lake-water TOC and total phosphorus were not measured. Data from Nationella Kalkdatabasen (kaldatabasen.se, last accessed 18 August 2019).
[‖]Dragsjön and Lång-Älgsjön. Data based on single measurements made in September 1998. Data from Malmestrand (2000).
[¶]Tryssjön. Data represent averages of 51 measurements made 2010–2019. Data from the Swedish national monitoring program ("IKEU"; miljodata.slu.se, last accessed 17 December 2019).
[#]Color calculated from absorbance at 420 nm as follows: 500·Absorbance 420.

Data

Results presented in this study build on analyses of both new empirical data and previously published data. Supporting Information Table S1 details the year of sampling and which data are primary or secondary in this article. Within each section below, we briefly summarize methods for secondary data and then describe the analyses and methods for the primary data.

Chronology

Sediment chronologies were either created or updated for all lakes using both measured ages in the form of radiocarbon dates (^{14}C; Supporting Information Table S2) and ^{210}Pb-dating, and inferred ages based on spheroidal carbonaceous particle (SCP) counts (Supporting Information Table S1), Pb concentrations, $^{206}Pb/^{207}Pb$ stable isotopic ratios, and pollen curves. Lead stratigraphic markers were assigned ages by matching our lake records with the varved sequence from Kalven in central Sweden (Bindler et al. 2011). Pollen markers in central Sweden were assigned ages by matching against the well-dated record from Holtjärnen (Giesecke 2005). All radiocarbon-dated samples were calibrated using the IntCal13 calibration curve (Reimer et al. 2013), except for the two samples for Lilla-Öresjön where we used already published dates (Renberg 1990). Age-depth models were developed with CLAM 2.2 in R 3.5.2 (Supporting Information Table S2, Figs. S2–S8). Three radiocarbon dates were corrected for old carbon (Oldfield et al. 1997). Our chronologies are similar to the previously published models for Måkevatten, Lång-Älgsjön and Dragsjön, and the age-depth models are new for Härsvatten, Lilla Öresjön and Tryssjön (Supporting Information Figs. S2, S4–S8). For Lysevatten, we added two new radiocarbon dates and constrained the sequence to 6400 BCE (Supporting Information Fig. S3).

Analyses

Lake-water TOC concentrations (mg TOC L^{-1}) are inferred for all seven lakes from visible near-infrared (400–2500 nm) absorbance spectra of sediment samples measured on a FOSS XDS Rapid Content Analyzer operated in diffuse reflectance mode at a spectral resolution of 0.5 nm using the method and training set by Meyer-Jacob et al. (2017). This approach uses an orthogonal partial least squares regression model, in which absorbance spectra of surface-sediment samples are calibrated against the corresponding measured surface-water TOC concentration. The training set consists of 345 arctic, boreal, and northern temperate lakes, with a lake-water TOC range from 0.5 to 41 mg L^{-1} and the model has a cross-validated R^2 of 0.57 and a prediction error of 4.4 mg TOC L^{-1} (Meyer-Jacob et al. 2017). The model uses lake-water TOC rather than DOC because monitoring programs in Sweden have measured lake-water TOC since implementation of these programs in the 1980s. Furthermore, in boreal lakes DOC represents almost 100% of the lake-water TOC pool (e.g., Kortelainen et al. 2006). Previously published profiles from Lysevatten (Rosén et al. 2011) and Lång-Älgsjön (Meyer-Jacob et al. 2015) were reanalyzed using the above model for the purpose of this study.

Diatom sample preparation and lake-water pH reconstructions are described in full in the primary publications (Supporting Information Table S1). Lake-water pH was reconstructed using weighted averaging and regression based on the SWAP (in the case of Måkevatten a simplified genus-based model) and Nordic data sets, with a RMSEP of 0.3–0.36 (Supporting Information Table S1). For presentation in this article, we have modernized nomenclature for all shown taxa (algaebase.org; last accessed 20 August 2019) and grouped them based on pH optima in the SWAP data set (Stevenson et al. 1991), similar to the Hustedt pH categories.

A full description of the pollen sample preparation can be found in the original publications (Supporting Information Table S1). In this study, pollen sums are reported as a percentage of the total terrestrial pollen sum and we highlight the main patterns of vegetation change at Lysevatten, Lång-Älgsjön, Dragsjön, and Tryssjön using the REVEALS model for quantitative vegetation reconstruction (Sugita 2007) with DISQOVER (Theuerkauf et al. 2016) in R. We use the taxa, pollen productivity estimates and fall speeds compiled by Alenius et al. (2017) and have added two additional taxa (*Empetrum* and *Fagus*) and associated pollen productivity estimates and fall speeds from Mazier et al. (2012). Pollen taxa were assigned to four different land-cover categories: forest, peatland, open land, and cropland (Supporting Information Table S3). Additionally, we show thermophilic trees as a subgroup of forest. It should be noted that the categories "open land" and "forest" do not necessarily represent discrete entities in these ecosystems, particularly during the period of traditional land use, and therefore, references are given to specific taxa to exemplify the changes behind the REVEALS output. Relative abundances for important pollen taxa are found in Supporting Information Figs. S3, S6–S8 .

Major and trace element concentrations for Måkevatten, Härsvatten, Lilla Öresjön, Dragsjön and Tryssjön were analyzed on 0.05 or 0.2 g powdered sediment using a Bruker S8-Tiger wavelength-dispersive X-ray fluorescence spectrometer with the method described by Rydberg (2014). Lysevatten was reanalyzed using this method for consistency between lake data sets and particularly because the current instrument and calibration give lower detection limits (namely for Pb) than the original instrumentation used (cf. Rosén et al. 2011), important for improving the age-depth model. Lång-Älgsjön was analyzed by Meyer-Jacob et al. (2015) using the same method. We use the potassium to aluminum ratio (K/Al) as a simple weathering index of silicate minerals based on the preferential loss of K from soils during chemical weathering (Kauppila and Salonen 1997). The ratio thus indicates the relative importance of chemical weathering (lower ratio) vs. material export to the lake (erosion, higher ratio).

Results

Vegetation and landscape development

Following deglaciation, the landscape in southern Sweden was colonized by pioneer plant taxa, including *Betula*, *Salix*, *Empetrum*, *Hippophaë*, and *Artemisia*, as shown by high-resolution pollen records from two lakes within 120 km of our southern lakes (Seppä et al. 2005; Antonsson and Seppä 2007). Over the following ~2000 yr, the landscape transitioned to a closed *Betula* and *Pinus* forest with increasing abundances of thermophilic trees (*Tilia*, *Ulmus*). At Lysevatten, abundances of thermophilic trees began to decline again from ~4000 BCE (Fig. 2). In Måkevatten, our longest record, the K/Al ratio decreased sharply from 0.3 to 0.1 over the first ~3300 yr following deglaciation (until 7300 BCE, Supporting Information Fig. S2). This represents the common pattern of postglacial primary succession where an open landscape with fresh, exposed soils is colonized by a progressively more closed forest that leads to soil stabilization, decreased erosion rates, and depletion of more-easily weathered minerals (Engstrom et al. 2000; Boyle 2007). The K/Al ratio increased again to ~0.2 between 5400 and 4800 BCE in Måkevatten, which is similar to the K/Al ratio in the earliest samples from Lysevatten, dating to 6200 BCE (Fig. 2). This indicates an increase in less-weathered mineral matter through an erosion event, supported by the presence of a visible layer of biotite in Måkevatten. After 4800 BCE, the K/Al ratio decreased to low and stable levels, ~0.05 in Måkevatten and ~0.16 in Lysevatten, indicative of a return to a state of slow, but consistent input of more-weathered mineral matter (i.e., low erosion).

The earliest indication of human impact on the landscape around the southern lakes is an increase in inferred open land cover at Lysevatten from ~300 CE (Fig. 2), due to increases in taxa typically associated with human disturbance, including Poaceae, Chenopodiaceae, *Rumex*, *Urtica*, and eventually *Juniperus* (Supporting Information Fig. S3; Behre 1981, Fredh et al. 2019). This change occurs around the same time as the first cereal pollen appears at 670 CE in Lilla Öresjön (Renberg 1990). At Lysevatten, charcoal and the fire-resistant dwarf-shrub *Calluna* increased from ~1280 CE and the first cereal

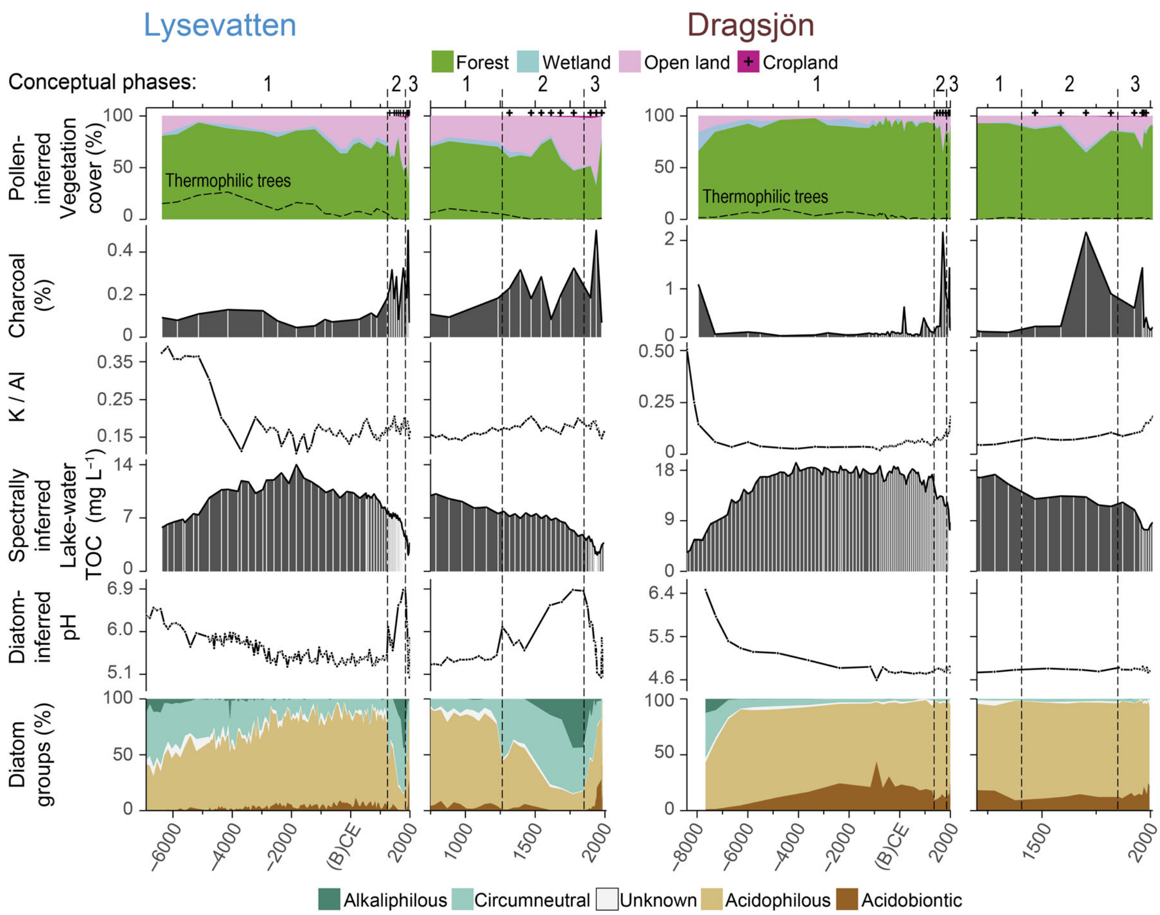

Fig. 2. Summary data for Lysevatten and Dragsjön, which characterize the southern and central lake groups, respectively. Samples with cropland > 0% are highlighted for visibility (+). Conceptual phases are separated by dashed lines and numbered above: (1) natural lake development, (2) impacts associated with traditional land use, starting at ~1270 CE in Lysevatten and ~1410 CE in Dragsjön, (3) impacts associated with industrialization, starting at 1850 CE in both lakes. For each lake, the left panel shows the entire sequence while the right panel focus on phase 2 and 3.

pollen appear at ~ 1320 CE (*Secale cereale* and *Hordeum vulgare*), concurrent with the disappearance or major decline in several broadleaved taxa, including *Ulmus*, *Quercus*, *Tilia*, and *Corylus*. By 1780 CE, the estimated total forest cover had decreased to 50% (compared to 75% prior to evidence of land use) and the area of cropland increased to 1.3% (Fig. 2). In Måkevatten, the K/Al ratio increased from 0.06 to 0.13 between 920 and 1800 CE. A slight increasing K/Al trend is also seen in Lysevatten. The vegetation development described by the pollen data is typical for upland areas in southern Sweden (Antonsson and Seppä 2007; Fredh et al. 2019) and represents forest thinning for grazing and heathland development (Atlestam 1942) and a regional presence of cropland. This drives the increased fire frequency as seen in the increase in *Calluna* pollen and charcoal particle counts, and increased erosion as indicated by the K/Al ratio.

After ~ 1850 CE and the transition toward industrial human activities and impacts, traditional land use at the southern lakes remained intensive for another ~ 100 yr. The estimated area of cropland peaked at 2.4% at ~ 1900 CE and the estimated total forest cover was historically low, with 36% at ~ 1940 CE (Fig. 2, Supporting Information Fig. S3). By ~ 1980 CE, the total forest cover had increased to 78% due to the recovery of *Pinus* and *Picea*, while the area of cropland and the charcoal counts decreased. This followed the widespread trend in Sweden during the 20th century, when small-scale agriculture, including all forest grazing, mire haymaking and much of the small-scale crop cultivation was abandoned and subsequently replaced with modern forestry (Lindbladh et al. 2014).

Central Sweden was deglaciated later than southern Sweden due to the south-to-north glacial regression. The records from these lakes therefore only date back to ~ 8500 BCE, but similar to the southern lakes, the basal samples have high abundances of pollen from taxa indicative of more open, wetland-dominated vegetation, including Cyperaceae, *Calluna*, *Salix*, and Poaceae. After a few hundred years, these taxa disappeared, or were substantially reduced, and the catchments became dominated by closed *Betula* and *Pinus* forests. The K/Al ratios declined rapidly from 0.5 to 0.6 at ~ 8500 BCE to low, stable background levels of 0.05–0.08 already after ~ 400–1200 yr (Fig. 2, Supporting Information Figs. S6–S8). As with the southern lakes, the primary succession from fresh, exposed soils to a closed forest led to decreased erosion and thus lower K/Al ratios.

The earliest pollen evidence of human land use in proximity to the central lakes is an increase in inferred open land cover (mainly through an increase in Poaceae and *Rumex*) in Tryssjön from ~ 750 CE (Supporting Information Fig. S8). A similar increase in open land occurred at Lång-Älgsjön and Dragsjön at 1120 and 1440 CE, respectively (Fig. 2, Supporting Information Figs. S6, S7). The extent of open land continued to increase over the next few hundred years and forest cover correspondingly decreased, from 89–93% prior to human land use, to minima of ~ 65–70% by 1530–1710 CE. A continuous presence of cereal pollen was established at 1030–1470 CE in all three lakes. K/Al ratios increased in all lakes from 1400 –1530 CE, reaching 0.1–0.18 by 1750–1820 CE (Supporting Information Figs. S6–S8). In general, the development at the central lakes is similar to the southern lakes—with pollen evidence for forest thinning, which led to increased erosion as indicated by the K/Al ratio—albeit occurred later.

From 1850 CE, inferred forest cover showed a generally increasing trend at the central lakes and in the most recent samples (after ~ 1980 CE) forest cover increased to 81–91%, due to recovery of *Picea* and *Pinus*, with corresponding decreases in open land and cropland. This is similar to the forest cover of 89–93% before the onset of traditional land use (Fig. 2; Supporting Information Figs. S6–S8). This indicates an abandonment of traditional land use in favor of modern forestry and reforestation, similar to the southern lakes and the general trend in Sweden at this time (Lindbladh et al. 2014). The K/Al ratio remained at 0.13–0.19 from 1850 CE until the most recent samples. This is higher than the levels before traditional land use, which suggests that soil conditions did not fully return to predisturbance conditions even though forest cover increased, possibly as an effect of forestry (Finstad et al. 2016).

Development of lake-water pH

Immediately following deglaciation at the southern lakes (~ 10,500 BCE) inferred pH was 6.3–7.4 and diatom assemblages composed of alkalophilic (*Pseudostaurosia brevistriata*, *Pantocsekiella comensis*, *Asterionella formosa*) and circumneutral (*Pantocsekiella kuetzingiana* agg., *Achnanthidium minutissimum*) taxa (Fig. 3; Supporting Information Figs. S2–S5). Over the following ~ 11,500 yr (i.e., until 390–1220 CE), pH decreased gradually to 5.0–5.4 and the diatom assemblage was replaced by more acidophilic (*Aulacoseira distans* agg., *Eunotia incisa*, *Frustulia rhomboides* agg.) and acidobiontic (*Eunotia naegelii*, *Tabellaria quadriseptata*) taxa. This development in the diatom assemblage is well described in the literature (Battarbee 1984; Renberg et al. 1993*a*) and is associated mainly with pH changes driven by vegetation succession and long-term soil development (Engstrom et al. 2000; Boyle 2007).

Coinciding with the pollen evidence of land use, inferred pH began to increase in the southern lakes, earliest in Lilla Öresjön at ~ 670 CE, while in Lysevatten, Måkevatten, and Härsvatten pH increased from ~ 1050–1270 CE (Fig. 3). Over 220–560 yr, pH increased by 1.0–2.2 units (to 6.3–7.2 at peak values) in all the southern lakes and these elevated levels were maintained until 1850–1890 CE. The pH increases reflect changes in lake ecology, where all acidobiontic diatoms disappeared and many acidophiles (e.g., *F. rhomboides* agg., *Eunotia* spp.) decreased or disappeared (Supporting Information Figs. S2–S5). They were replaced by many taxa common in the early Holocene (phase 1), such as *P. kuetzingiana* agg., *A. minutissimum*, and *A. formosa*.

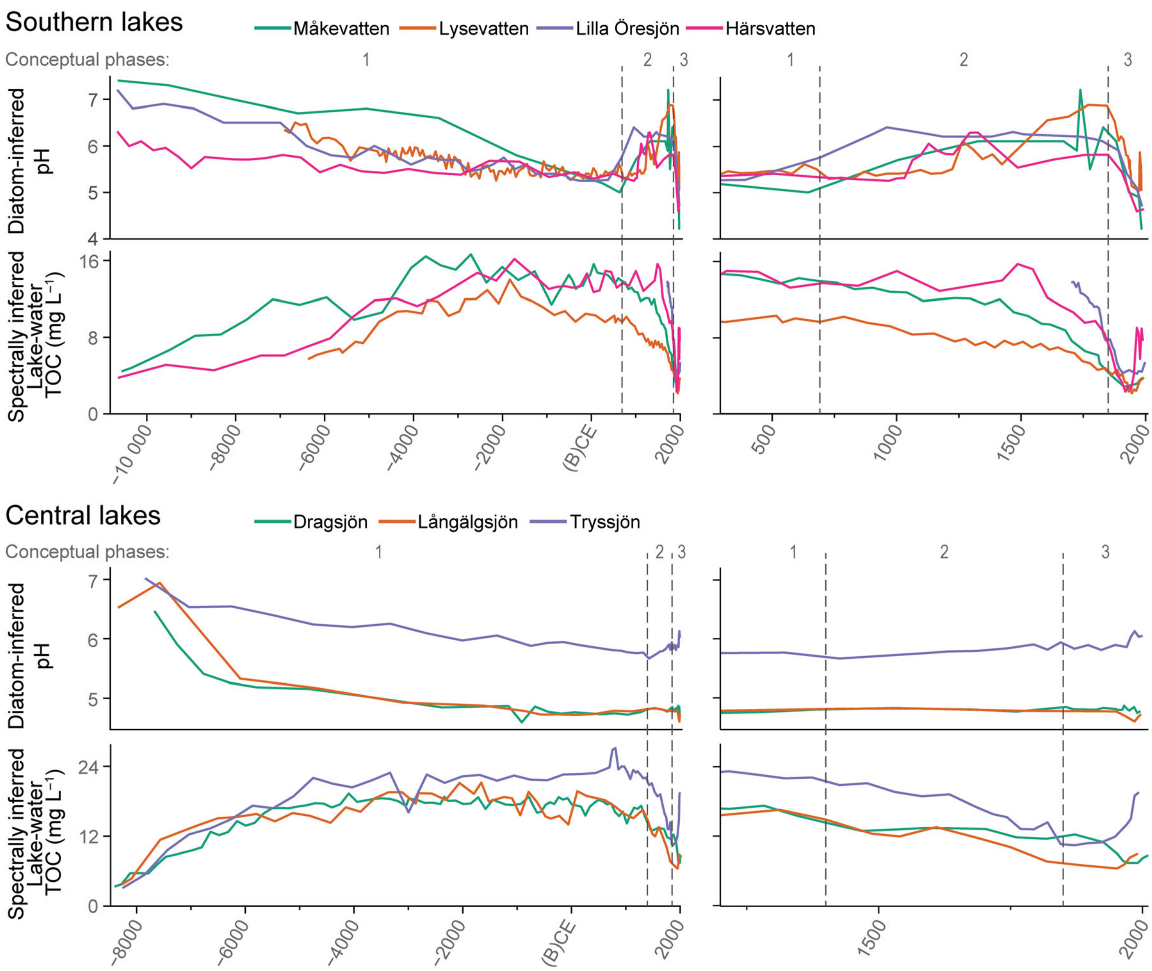

Fig. 3. Diatom-inferred pH and spectrally inferred lake-water TOC for the southern (top panels) and the central lakes (bottom panels). Conceptual phases are separated by dashed lines and numbered above: (1) natural lake development, (2) impacts associated with traditional human land-use, starting at ~ 690 CE in the southern lakes and ~ 1400 CE in the central lakes, (3) impacts associated with industrialization, starting at ~ 1850 CE in all lakes. Note that the transitions from phase 1 to 2 are based on the earliest signs of human presence in any lake in each group, see Supporting Information Figs. S2–S8 for individual transitions. Left panels show the entire sequence while the right panels focus on phase 2 and 3.

Inferred pH started decreasing in the southern lakes from 1850–1890 CE, reaching prealkalization levels of 5.0–5.4 by 1910–1940 CE and historical minima of 4.2–5.0 during ~ 1965–1985 CE. This is reflected by a change in diatom assemblages, initially as a decrease in the circumneutral taxa associated with the period of cultural alkalization (e.g., *P. kuetzingiana* agg.) in favor of acidophilic taxa common before the alkalization (e.g., *F. rhomboides* agg. and *E. incisa*; Supporting Information Figs. S2–S5). As pH decreased below the natural minima, the diatom assemblages became dominated by acidophilic (*N. leptostriata*) and acidobiontic (*T. quadriceptata*, *E. naegelii*, and *Oxyneis binalis*) taxa. The decrease in pH coincides with sharply increasing spheroidal carbonaceous particle counts and sulfur concentrations in the sediments (Supporting Information Figs. S2–S5), associated with the large-scale emissions of sulfur (Mylona 1996) and the resulting lake-water acidification (Battarbee 1984; Ek and Korsman 2001). Lysevatten was limed in 1974 CE, which resulted in a temporary increase of 0.9 units over a 5-yr period between 1974 and 1983 CE (Renberg and Hultberg 1992) and produced a novel ecosystem (Renberg et al. 1993*a*) dominated by *A. minutissimum*. Meanwhile, there was no discernable change in pH in the remaining, untreated lakes. However, monitoring data from after our sampling of these lakes indicate that the untreated lakes (Måkevatten, Härsvatten, and Lilla Öresjön) have recovered to within 0.2 units from their naturally acidic pH and Lysevatten remains 1.6 units above natural background due to liming.

The lakes in central Sweden were characterized by a circumneutral inferred pH of 6.5–7.0 immediately following deglaciation (prior to ~ 7700 BCE; Fig. 3). Subsequently, pH in Lång-Älgsjön and Dragsjön decreased rapidly and within only ~ 2000 yr declined to 5.3 and then further to ~ 4.8 over the following ~ 5500 yr (i.e., until 960–430 BCE). The initial diatom assemblage was composed of alkalophilic and circumneutral taxa (*P. brevistriata*, *P. kuetzingiana* agg., and *Discostetlla glomerata*) then progressively more acidophilic (*Encyonema perpusillum*, *Brachysira vitrea*, *B. brebissonii*, *F. rhomboides* agg., *Eunotia* spp., and *Navicula leptostriata*) and acidobiontic (*T. quadriceptata*, *Semiorbis hemicyclus*, and *Kobayasiella* spp.) taxa (Fig. 2; Supporting Information Figs. S6, S7). In Tryssjön, lake-water pH decreased from 7.0 to 5.7 over ~ 9100 yr (1430 CE) (Fig. 3), as the diatom assemblage gradually changed to acidophilic taxa (*Aulacoseira* spp: *A.* [*subarctica* type 2], *A. distans* agg., *A. alpigena*, *B. vitrea*, *Tabellaria flocculosa*, *F. rhomboides* agg. and *Eunotia* spp., Supporting Information Fig. S8). The gradually decreasing pH in Tryssjön is more similar to the southern lakes and other lakes in the literature (e.g., Battarbee 1984; Renberg et al. 1993*a*). The postglacial acidification in Dragsjön and Lång-Älgsjön is much more rapid, but similar examples exist (Korhola and Tikkanen 1991).

In two of the central lakes, Dragsjön and Lång-Älgsjön, inferred pH remained stable at 4.8 throughout the period of traditional land use (~ 1120–1980 CE; Fig. 3). However, there were strong diatom assemblage shifts already from ~ 440–550 CE when many epipelic (living on soft sediment) and acidophilic or acidobiontic taxa decreased or disappeared (e.g., *F. rhomboides* agg., *N. leptostriata*, and *Kobayasiella* spp.) and were replaced by tychoplanktonic (living sometimes pelagic, sometimes benthic), acidophilic taxa, for example, *A. distans* agg. and *A. perglabra* (Supporting Information Figs. S6, S7). This change in the diatom assemblage precedes evidence of land use by ~ 500 yr, and may instead be linked to an expansion of the surrounding floating sphagnum-mats over the soft-sediment littoral zones following peatland expansion along the shorelines (Meyer-Jacob et al. 2015; Ninnes et al. 2017). pH remained at 4.7–4.8 in Dragsjön and Lång-Älgsjön throughout the period of traditional land use and later industrial activities. The diatom species remained largely unchanged; however, some epipelic diatom taxa (*F. rhomboides* agg. and *Kobayasiella* spp.), which decreased or disappeared at 440–550 CE, recovered, while the tychoplanktonic *Aulacoseira* spp. decreased (Supporting Information Figs. S6, S7). The stable pH, despite atmospheric acid deposition, is different from the southern lakes but not uncommon in other naturally acidic lakes in central Sweden (Norberg et al. 2008; Myrstener et al. 2019). The return of epipelic taxa can be interpreted as a deepening of the photic zone and increased habitat for epipelon, which coincides with the historically low concentrations of lake-water TOC (presented in the following section).

In Tryssjön, which lacks the immediate peatland connectivity of the other two central Swedish lakes, inferred pH was initially stable at 5.7 following the establishment of traditional land use at the lake. However, pH increased by 0.2 units during 1580–1970 CE and then by a further 0.3 due to liming (Fig. 3; Norberg et al. 2008). The initial 0.2 pH increase corresponds roughly to the most intensive period of human land use as indicated by the pollen and the K/Al ratio. The only substantial changes in the diatom ecology throughout this period occurred after the liming, which resulted in an increase in *P. kuetzingiana* agg. (Supporting Information Fig. S8). The pH response in Tryssjön due to the human activities appears intermediate compared to the other central lakes and the southern lakes, with a small increase following the traditional land use but no response to the industrial acid deposition.

Development of lake-water TOC

Inferred lake-water TOC was low (< 5 mg L^{-1}) in the lakes in southern Sweden immediately following deglaciation (before ~ 10,500 BCE; Fig. 3). Concentrations then increased gradually over several thousand years and peak concentrations of 14–17 mg L^{-1} were reached after 8000–9000 yr (i.e., at 2700–1700 BCE; Fig. 3). This is consistent with the early vegetation and soil development, which would increase the soil carbon pool and thus carbon export to the lakes (Engstrom et al. 2000). After 2700–1700 BCE, lake-water TOC declined slightly, most notably in Lysevatten. Between ~ 1000 and 1850 CE inferred lake-water TOC decreased more rapidly in Lysevatten and Måkevatten, reaching 5–10 mg L^{-1}, coinciding with the pollen evidence for land use. Most of the decrease occurred after 1350–1650 CE, when land use intensified. Following the onset of modern industrialization (i.e., from ~ 1850 CE), lake-water TOC continued the decreasing trend, reaching only 3–4 mg L^{-1} at 1900–1950 CE—concentrations not seen since lake formation (Fig. 3). Thus the lowest concentrations occurred during a period when traditional land use, as seen in the pollen data, overlapped with industrial acid deposition, recognized as an important driver of lake-water organic carbon dynamics (Monteith et al. 2007; Meyer-Jacob et al. 2019). For the most recent samples (~ 1985–1993 CE) lake-water TOC increased slightly to 4–8 mg L^{-1} (Fig. 3), similar to many other acid-sensitive lakes in Sweden over this time period (miljodata.slu.se, last accessed 17 December 2019; Futter et al. 2014).

Similar to the southern lakes, inferred lake-water TOC in the lakes in central Sweden was initially low, 3–4 mg L^{-1} (before ~ 8300 BCE), but then increased rapidly to and plateaued at 18–23 mg L^{-1} after 2500–3500 yr (i.e., at 5800–4800 BCE; Fig. 3). These concentrations were maintained until 1270–1400 CE. After this, lake-water TOC decreased to 10–14 mg L^{-1} by ~ 1820 CE. This is concurrent with the establishment of traditional land use and the majority of the decrease occurred after 1610–1700 CE, when pollen indicate an intensification of the land use in the landscape surrounding the lakes. The lowest concentrations since deglaciation occurred by ~ 1950–1990 CE when lake-water TOC fell to 7–8 mg L^{-1} in Lång-Älgsjön and Dragsjön, and ~ 10 mg L^{-1} in Tryssjön. In the most recent samples (~ 1990–2010 CE) concentrations have increased to 9 mg L^{-1} in Dragsjön and Lång-Älgsjön and 20 mg L^{-1} in Tryssjön (Fig. 3). Similar to the southern lakes, the historically low concentrations of lake-water TOC occurred during the overlap of traditional land use and industrial acid deposition, and the recent increase follows the same trend as environmental monitoring data from Tryssjön and numerous other lakes in the region (miljodata.slu.se, last accessed 17 December 2019).

Discussion

We organize our discussion within a conceptual model dividing the lake-sediment records into three broad developmental phases (Fig. 4), based on a framework previously established for base-poor, "clear" lakes in Sweden (Renberg et al. 1993*a*). The first phase encompasses natural lake conditions where development was linked to ontogenetic and climatic changes over a period that began at lake formation and lasted ~ 9000–11,000 yr. The establishment of a closed forest decreased soil weathering rates and the warming climate of the early Holocene drove changes in both terrestrial and aquatic ecosystems, which resulted in a natural lake acidification and an increase in lake-water TOC (Engstrom et al. 2000). All of our lakes follow the same basic trajectory, but with the important difference that lake-water TOC increased and pH decreased more rapidly in the central lakes (Fig. 3).

The second phase encompasses preindustrial human activities since ~ 700 CE. The start of phase 2 is defined by the

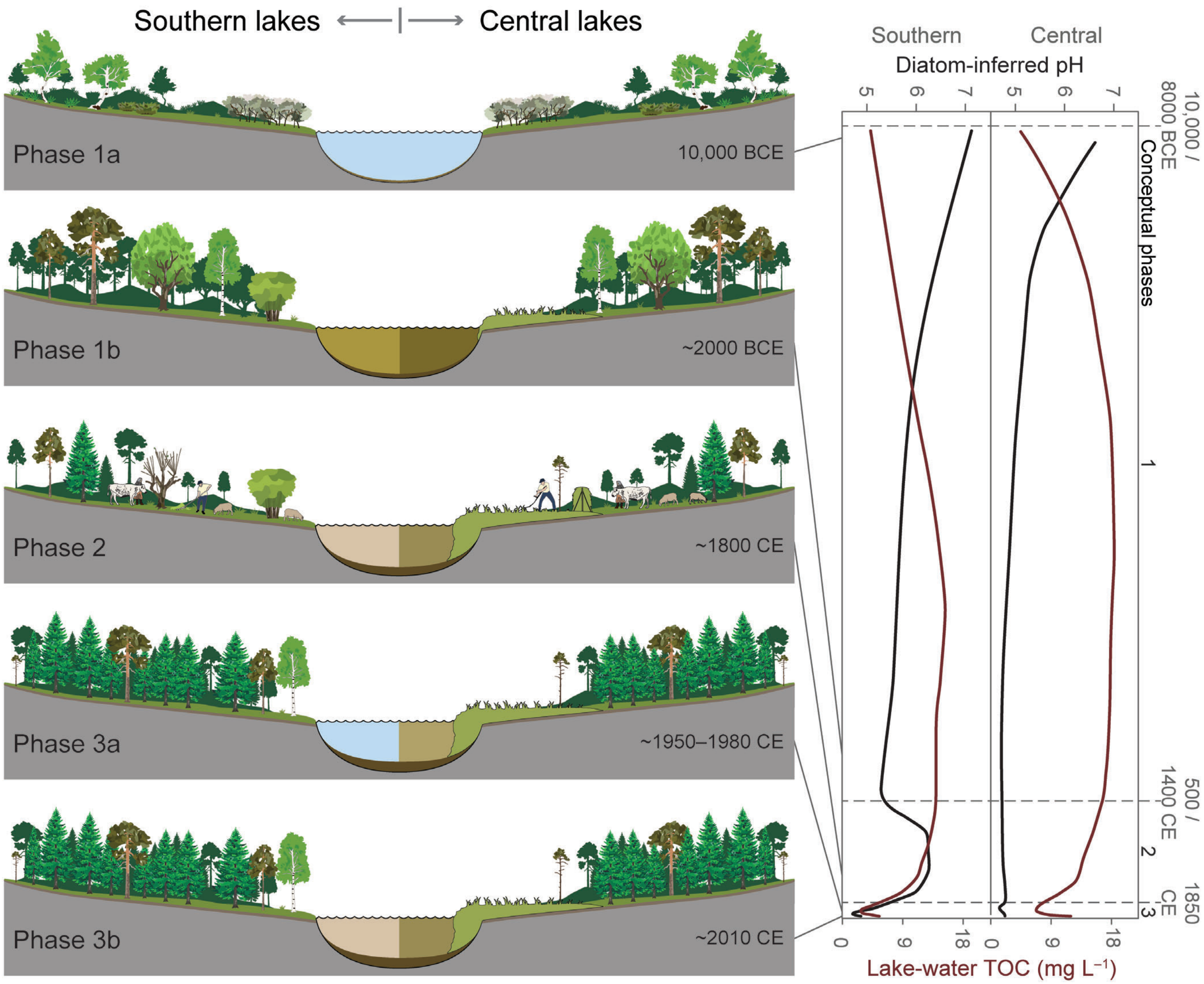

Fig. 4. Conceptual model for the development of the southern and central lakes in boreal Sweden over the past ~ 10,000–12,000 yr. The left diagrams represent snapshots of the landscape at five characteristic periods in time: phase 1a—after deglaciation and before major soil and vegetation development; phase 1b—the "background" period before discernible human impact; phase 2—during the maximum impact of traditional land-use activities and before any discernible impact of acid deposition; middle to phase 3a—after most of the remote land-use activities have been abandoned and during maximum impact of acid deposition; phase 3b—the "recovery" from traditional land use and acid deposition. This phase is interpreted from trends of inferred lake-water TOC data and ~ 30 yr of environmental monitoring data (miljodata.slu.se, last accessed 17 December 2019). The two panels to the right represent a conceptual summary of diatom-inferred pH and lake-water TOC in each lake type.

point of the first discernible human impact on each respective lake, and therefore the start date differs between sites. These activities encompass several traditional agricultural methods—such as forest grazing, harvesting of hay, and cereal cultivation. This led to a restructuring of catchment vegetation, increasing erosion, decreasing lake-water TOC, and, in some lakes, alkalization.

We define the start of the third phase as ~ 1850 CE and thus it encompasses the complex impacts on boreal lakes associated with the transition to a modern industrial society. The most important changes were the abandonment of traditional land-use practices in favor of modern forestry and the industrial pollution, for example, acid deposition.

Phase 1: Natural landscape development and lake ontogeny

The well-established, postglacial vegetation succession and development of a closed canopy forest dominated by *Betula* and *Pinus* (Giesecke 2005; Seppä et al. 2005; Antonsson and Seppä 2007) led to a stabilization of catchment soils, which would include a decrease in erosion, depletion of more-easily weathered minerals and loss of base cations through vegetation uptake (Engstrom et al. 2000; Boyle 2007). This soil development is reflected in the observed declines in K/Al ratios in all the lakes. However, there is an excursion with increasing K/Al ratios at ~ 6000 BCE in Måkevatten and Lysevatten (Supporting Information Figs. S2, S3), which was likely an erosion phase in these two nearby lakes. In contrast to the southern lakes, K/Al ratios decline more rapidly in Dragsjön and Lång-Älgsjön in central Sweden during the early Holocene (Supporting Information Figs. S6, S7). While this more rapid decline could be interpreted as a more rapid soil development at these sites, an increase in *Sphagnum* "leaf" counts and the presence of 4-isopropenylphenol (a polyphenol specific for *Sphagnum*; Schellekens et al. 2015) in both Lång-Älgsjön and Dragjön (Ninnes et al. 2017) indicate development of adjoining peatlands at this time. The peatlands would serve to filter runoff from mineral soils and reduce inputs of mineral particles to the lake sediments. The early development of the peatlands adjoining Lång-Älgsjön is supported by a limited survey of ages of the basal peat layer at three locations, with dates of 9800, 5900, and 4200 yr BP (Meyer-Jacob et al. 2015).

The postglacial pH development in all lakes follows a pattern well established in the literature and appears characteristic of boreal lakes in general (Renberg et al. 1993*a*). The declines in lake-water pH were driven by vegetation and soil development that contributed to decreasing inputs of base cations (e.g., declining K/Al ratios) as well as an increasing supply of organic acids derived from the buildup of soil organic matter (Engstrom et al. 2000; Boyle 2007; Rosén et al. 2011). In the southern lakes and in Tryssjön, pH decreased gradually over ~ 9300 yr, ultimately reaching to 5.0–5.7. In Dragsjön and Lång-Älgsjön, the decline in pH was more rapid and reached lower values—declining to pH 5.3 already within the first 2000 yr and then further to ~ 4.8 over the following ~ 6500 yr. While this is in contrast with the established model for natural acidification of acid sensitive lakes (Renberg et al. 1993*a*; Boyle 2007), a similar pattern has been shown for boreal forest lakes in Finland (Korhola and Tikkanen 1991). For these lakes, the development of adjoining peatlands disconnected the lakes from the surrounding mineral soils and thus the influence of soil weathering—as seen in the K/Al ratios—and instead their lake-water chemistry was controlled by infiltration through the peatlands and supply of organic acids.

The early Holocene development of lake-water TOC is broadly similar in all lakes (Fig. 3), and consistent with the postglacial vegetation and soil development, which led to increased soil carbon pools and thus carbon export to the lakes (Engstrom et al. 2000). However, one key difference between the two lake groups is that lake-water TOC increases faster and to higher concentrations in the central lakes (18–23 mg L^{-1} after 2500–3000 yr; Figs. 3, 4) than the southern lakes (14–17 mg L^{-1} after 8000–9000 yr; Figs. 3, 4). This is expected given their present values, but more fundamentally because the central lakes have larger percentages of peatland in their catchments (Table 1) than the southern lakes (for which long-term lake-water TOC data is available). Furthermore, in Dragsjön and Lång-Älgsjön the early increase in lake-water TOC coincides with the evidence of nearshore peatland development as an important driver of lake-water TOC. The rapid increase in lake-water TOC is also similar to other lakes that we have studied at Moshyttan in central Sweden, where concentrations plateaued at 14–17 mg L^{-1} after 3000–5300 yr (Myrstener et al. 2019). Independent support for the early increase in lake-water TOC in these lakes is found in the diatom record, where most of the acidophilic diatom taxa (*E. incisa* and *F. rhomboides* agg.) that increase during the early lake development are also associated with high lake-water TOC concentrations (Stevenson et al. 1991). It is noteworthy that all our studied lakes show a relatively high inferred lake-water TOC (> 14 mg L^{-1}) during the late phase 1 (i.e., before any discernable human impacts). This is in sharp contrast to the perception of the southern lakes as "clear-water" lakes, a description established when they were initially sampled.

We observe that lake-water TOC concentrations in several of our lakes are somewhat higher during the period from ~ 4000 to 1000 BCE. Thereafter, lake-water TOC decreases slightly, coinciding with the late Holocene climate cooling as indicated by the declines in pollen from thermophilic trees (e.g., *Tilia* and *Ulmus*; Fig. 2; Seppä et al. 2005). We can speculate that a cooling climate may have affected the production and export of organic matter from the lake catchments, and led to the observed declines in lake-water TOC, the inverse of how the warming climate over the last decades has been suggested to drive increasing lake-water TOC (Finstad et al. 2016).

Phase 2: Historical human impacts as a driver of declining lake-water TOC

The period of human land use is associated with increasing inputs of less-weathered mineral matter, which is indicated by increasing K/Al ratios in all lakes, and increased fire frequency, indicated by increasing charcoal particle counts and fire-resistant taxa such as *Calluna*. In the southern lakes, this is associated with an increase in inferred pH by > 1 unit and a change in the diatom assemblage. This cultural alkalization has been described previously for lakes in southern and central Sweden where catchment disturbance and increased transfer of base cations to the lakes led to an increase in lake-water pH (Renberg et al. 1993*b*). Inclusion of organic acids based on inferred lake-water TOC concentrations into chemical models of acidification for the Lysevatten record shows that an increase in base cation supply in combination with the decline in lake-water TOC—and thus also organic acids—would together account for the pH increase in these lakes (Rosén et al. 2011). In the central lakes, pH increased in Tryssjön by ~ 0.2 units during the most intensive land use, while pH remained stable in Dragsjön and Lång-Älgsjön throughout the period of traditional land use. The central lakes are likely more resistant to acidification due to their higher lake-water TOC concentrations (and thus more organic acids), which buffer against pH decreases (Ek and Korsman 2001). We further suggest that Dragsjön and Lång-Älgsjön were more resistant to alkalization because of the extensive peatland development adjoining the lakes, which physically buffers against the disturbance driving the pH change (e.g., by intercepting eroded soil material).

All of our study lakes exhibit a decline in lake-water TOC, starting at ~ 1000 CE in the southern lakes and ~ 1200 CE in the central lakes. This decline coincides with evidence in the pollen record of early land use, which consisted of low-intensive forest grazing indicated by increasing open land taxa such as Poaceae (Behre 1981), followed later (1300 and 1700 CE) by more widespread land use with forest thinning, increased fire frequency, heathland development, and, at least within the pollen source area, cultivation. By ~ 1820 CE, lake-water TOC had decreased by ~ 50% in the southern and ~ 40% in the lakes, compared to background levels.

From a process perspective, these landscape-scale activities had important long-term implications for lake-water chemistry (i.e., lake-water TOC and pH). In Fig. 4, we illustrate the traditional land-use practices in the two regions and briefly describe relevant aspects. Forest grazing led to a more open forest structure both through the grazing activities themselves and from human actions to improve grazing (i.e., thinning and burning). A thinner forest, grazing of the field-layer vegetation and extensive removal of leaf-hay would in turn reduce the transfer of litter to soils and reduce soil carbon pools (Bowden et al. 2014; Lo et al. 2015). In upland and alpine ecosystems, these traditional land-use practices have been shown to contribute to a long-term (centennial) drawdown in soil carbon pools (Gimmi et al. 2013; Lo et al. 2015). Field studies in Norway have shown regular cutting of mires for hay (sedges) not only removes aboveground biomass, but also reduces belowground storage of carbon because plant resources are reallocated to new shoot growth rather than root growth (Moen et al. 1999).

Direct quantitative measures on the proportions of biomass removed through traditional land-use activities in our study areas unfortunately do not exist, but collectively there is ample circumstantial evidence that indicates the scale of their impact. For example, based on ethnological records, Slotte (2001) estimated that during the 19th century 1 million ha of deciduous forests in Sweden were exploited for leaf-hay to cover winter fodder requirements for sheep and goats alone (estimates could not be made for horse, cattle, and swine). Historical maps—such as the map from 1816 of the area adjoining Måkevatten from the national land reforms (*Storskiftet*, Supporting Information Fig. S9)—shows the extent to which land was divided among landowners along with descriptions of the utility and value of all parts of the land. For the 1816 map, the forested land was depicted as still quite open, which is consistent with historical studies for the lake Gårdsjön project (2 km to the north) that showed that in the mid-1800s low-intensity grazing and dimension cutting of timber resulted in relatively open forests, with low canopies and irregular tree density (Olsson 1985). The eastern and southern shoreline of Måkevatten was bounded by the outfield forests of farms lying further in those directions (beyond the boundaries of this map). The significance of this is that prior to the 20th century the landscape as a whole was extensively utilized. Though different in details, this widespread landscape utilization was similar in central Sweden. There were four farmers sharing the summer farm closest to Dragsjön, 26 farmers sharing the summer farm closest to Lång-Älgsjön, and 31 farmers at Tryssjön (Hillerström 1984) in the early 1800s. These are only three of approximately 100 summer farms registered in the Swedish National Heritage Board database (raa.se, last accessed 15 January 2020) within the surrounding 100-km^2 area, with more than 1400 summer farms in the county. Stocking rates at the summer farms were not documented, but Larsson (2012) calculated from tax records that the average number of cows, sheep, and goats per home farm in three parishes in central Sweden were 6, 3, and 3, respectively, in the late 1500s/1600s and 5, 11, and 6, respectively, in the mid-1800s. Multiplied by the number of farmers relocating their livestock to each summer farm, the collective impact of forest grazing was extensive. Taken together, we suggest that traditional land-use activities in both these regions reduced above- and belowground carbon stocks to such extent that the transfer of labile carbon to surface waters was reduced.

The decline in lake-water TOC during ~ 1500–1850 CE coincides not only with the relatively more intensive traditional land use observed from pollen data, but also with the climate anomaly known as the Little Ice Age. The potential impact of the Little Ice Age climate cooling of ~ 1°C (Matskovsky and Helama 2014) on lake-water TOC in a central Swedish setting was discussed by Meyer-Jacob et al. (2015), who argued that while the Little Ice Age potentially led to a small direct decrease in lake-water TOC its largest impact was likely indirect. The colder climate decreased cultivated crop yields, which drove a further expansion in forest grazing and haymaking in the marginal lands of Sweden (Larsson 2012), thus indirectly driving the reduction of lake-water TOC.

Phase 3: Industrialization and modern changes in lake-water chemistry

By the time of industrialization, lake-water TOC had already decreased by ~ 50% in the southern lakes and ~ 40% in the central lakes compared to background values. This trend continued in phase three and during the lowest concentrations of lake-water TOC at 1900–1990 CE, the southern lakes had decreased by 75% and the central by 55%, compared to background values (Figs. 3, 4). These low concentrations occurred during a period when the influences from traditional land-use activities driving the long-term decrease overlapped with the effects of atmospheric acid deposition.

From sediment concentrations of sulfur, spheroidal carbonaceous particle counts and modeled historical sulfur deposition (Mylona 1996), it is clear that sulfur deposition already occurred by the mid-19th century, and that it notably intensified from the mid-20th century to a large-scale acidification of surface waters in northwestern Europe (Battarbee 1984). As a result, inferred pH decreased to unprecedentedly low values and the diatom assemblage changed to a novel system dominated by acidobiontic taxa in the southern lakes during ~ 1965–1985 CE. Acid deposition is further recognized as an important driver of lake-water organic carbon dynamics (Monteith et al. 2007) and has contributed to declines in lake-water TOC in northwestern Europe and North America since ~ 1850–1900 CE (Valinia et al. 2015; Meyer-Jacob et al. 2017, 2019). In central Sweden, although lake-water TOC also declined further, pH remained stable (excluding the liming-induced increase in Tryssjön), likely because surrounding peatlands and a decline in organic acids (i.e., lower lake-water TOC) may have balanced the lower levels of acid deposition.

The pollen data indicate that traditional land use remained active at both the southern and central lakes until ~ 1950 CE. By this time, traditional practices were largely abandoned and replaced with modern forestry, which registers as an increase in inferred forest cover in all lakes. The timing of this process is known from local sources (Olsson 1985), nationally from pollen-based studies (e.g., Fredh et al. 2019) and from analysis of historical documents and forest inventory data (Lindbladh et al. 2014). Consistent with our linking of traditional land use and the resulting decrease in forest cover with the centennial declines in inferred lake-water TOC, recent research has demonstrated this process in reverse. Based on long-term (decadal) monitoring data in Norway and Sweden, Finstad et al. (2016) and Škerlep et al. (2020) identified an increase in forest cover—"greening"—since the mid/late-20th century as one of the key factors driving the measured increases in surface-water organic carbon concentrations (browning). By the same reasoning, a less-green past would have likewise had less-brown waters.

Because of the complexity of human activities in southern and central Sweden, it is difficult to distinguish the relative contributions of different drivers to the decrease in lake-water TOC during phase 3, but both changes in land use and acid deposition are likely to be important (Škerlep et al. 2020). However, some insights on the relative importance of each driver is indicated by lake-water TOC levels having remained higher in the central lakes, likely due to their naturally higher concentrations but also due to the lower industrial acid deposition in central Sweden (Mylona 1996). In the most recent samples (~ 1985–2010 CE), inferred lake-water TOC has increased slightly in most lakes and this overlaps in time with the start of environmental monitoring, which shows increasing lake-water carbon trends in Härsvatten, Lilla Öresjön, and Tryssjön (miljodata.slu.se, last accessed 17 December 2019) and in many other acid sensitive lakes across Sweden since the mid-1980s (Futter et al. 2014).

Conclusions

Our studied lakes show broadly similar developments in lake-water TOC and pH over the Holocene, but we also observe some key differences. The central lakes show relatively smaller decreases in lake-water TOC in response to the traditional land-use and recent industrial acid deposition compared to the southern lakes. We suggest that this is driven by the presence of nearshore peatlands at the central lakes due to their function as constant sources of organic material. An early development of peatlands at the central lakes is also likely connected to the more rapid increase in lake-water TOC during the early Holocene and the subsequent stable background concentrations before the establishment of human land-use. Similar to previous findings that boreal lakes with higher lake-water TOC are more resistant to pH change (Ek and Korsman 2001), the peatlands at the central lakes also acted as pH buffers against cultural alkalization and acidification, whereas pH changed in response to both land-use and acid deposition at the poorly buffered southern lakes.

We can only infer a connection between the historically low concentrations of lake-water TOC and the traditional land-use practices and industrial acid deposition, but our results are in line with recent studies linking the current browning (i.e., increase in lake-water TOC) to recovery not just from acidification (Monteith et al. 2007) but also due to reforestation (Finstad et al. 2016; Škerlep et al. 2020). While our studied lakes show a wide range of contemporary lake-water TOC values (1.4–14.4 mg L^{-1}), our results indicate that all lakes had much higher lake-water TOC before the advent of traditional land-use and the later industrial acid deposition, and all of the lakes would likely be considered as brown-water lakes if natural landscape conditions still prevailed today. In other words, environmental monitoring of boreal lakes in Sweden started during an extreme and unique phase of their development history, which is important to recognize if we are to establish appropriate reference levels. Notably, our results do not rule out the potential for a further lake-water TOC increase due to the continuous recovery from past ecosystems disturbances and ongoing climate change.

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Conflict of Interest

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