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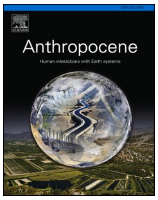

Recent increase in sediment dry matter, carbon, and phosphorus accumulation in small boreal lakes with clayey catchments

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ABSTRACT

This study estimated the mass accumulation rates of sediment (MAR), carbon (CMAR), and phosphorus (PMAR) in small Finnish lakes with agricultural clayey catchments over a 25-year period (1986–2011) and compared these with the conditions before major agricultural land use. Twenty-two lakes were cored for recent and reference (pre-disturbance) sediments. The recent sediment section was selected based on the 1986 ^{137}Cs fallout peak (TOP), whereas the pre-disturbance section (REF) was selected immediately below the first signs of human-induced erosion. The 50-cm reference section was dated with ^{14}C at both ends. The mass accumulation rates were estimated based on dating, weighing, and chemical analysis for both sediment sections. Furthermore, sediment-penetrating echo soundings were used to estimate the amount of sediment in the whole lake basins. These data were used to examine area-specific loading from clay-rich catchments. The average whole-basin pre-disturbance MAR, PMAR, and CMAR were 62 g m^{-2} a^{-1}, 0.06 g m^{-2} a^{-1}, and 4.7 g m^{-2} a^{-1}, respectively. The corresponding recent rates were 11, 13, and 8-fold (693 g m^{-2} a^{-1}, 0.79 g m^{-2} a^{-1}, and 37 g m^{-2} a^{-1}). In the recent conditions, sediments were generally more minerogenic and MAR, PMAR, and CMAR were higher in lakes with more arable fields in their catchments. Average area-specific suspended sediment load from the catchment for the region (~39% clayey soils) was approximately 69–137 kg ha^{-1} a^{-1} in the undisturbed state and 767–1534 kg ha^{-1} a^{-1} in recent conditions based on 100–50% retention. The results demonstrate that the increases in sediment, nutrient, and organic matter accumulation due to agriculture can be several fold over undisturbed state.

1. Introduction

Agriculture and anthropogenic deforestation are among the most prevalent land use types in the Baltic Sea basin, particularly in the highly populated agricultural southern part (Reckermann et al., 2022). Agricultural activity and the resulting land disturbance have been intensive during the industrialized period of the last ~150 years and, in the recent decades, their impacts on water systems have gained increasing attention. Although the sparsely populated northern part of the Baltic Sea basin is considered mostly pristine (Reckermann et al., 2022), a considerable part of the recent sediment and nutrient load entering the Baltic Sea comes from the cultivated, clayey coastal regions of Finland (Aakkula et al., 2010; Räike et al., 2020). In addition to the magnitude of human impact, the geologic and climatic contexts differ between the northern and southern parts of the Baltic basin. Glacial and post-glacial Quaternary sediments overlie Precambrian crystalline bedrock in the colder northern part of the basin, in contrast to the temperate southern part, where sedimentary rocks underlie the more recent sediments (Rosentau et al., 2017; Meier et al., 2022). In the future, climate change will likely exacerbate both agricultural and background nutrient loading from Finnish catchments via increasing river discharge during autumn and winter (Huttunen et al., 2015; Meier et al., 2022).

An important part of assessing the impacts on surface waters is the determination of the natural or reference state (Mills et al., 2017). HELCOM (2018) estimates that background export comprises approximately one third of the total nutrient load into the Baltic Sea and forms a significant part of the total load from Finnish catchments. Background loading is known to be higher in the clay-rich southern Finland than in till-rich inland, but more reliable estimates of the amount of export are still needed (Finér et al., 2021). However, the clayey regions in southern Finland are problematic in this respect, as most are used for agriculture (Ekholm and Mitikka, 2006), making it difficult to find reference basins to estimate natural sediment and nutrient loading based on current conditions (Bennion and Battarbee, 2007). This hampers lake and

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watershed management efforts as their design and dimensioning should take the background loading of sediment and nutrients into account (e.g. Bennion et al., 2001, Adams et al., 2014). Thus, the assessment of natural conditions in the clay-rich southern Finnish catchments is timely. The topic is also relevant to the global theme of soil erosion (Jenny et al., 2019; Baud et al., 2021) and carbon sinks (Mendonça et al., 2017).

In the absence of suitable reference catchments and basins, studies of sediment cores can provide information on the past conditions in the watershed and the lake basin and the subsequent human-induced changes (e.g. Foster and Walling, 1994, Foucher et al., 2020, 2021a, Chassiot et al., 2022). Sediment cores record changes in the composition and net accumulation of matter in the basins and allow studies of pre-disturbance reference sections (REF) in areas where modern reference basins are not available. Sediment records can also be used to infer pre-disturbance lake water nutrient concentrations using methods such as paleoecological modelling (e.g. Fritz et al., 1991, Anderson et al., 1993, Bennion et al., 1996, Liu et al., 2023). Extending the studies to undisturbed sediments also helps eliminate artefacts caused by within-basin sedimentation patterns as human-impacted sediment sections are compared with REF sections from the exact same location in the basin (e.g. Keck et al., 2020, Vähäkuopus et al., 2020).

In addition to the need to define background loading rates, the rapid increase in mechanized agriculture combined with the surficial geology of the catchments, makes Southwestern Finland well-suited for sediment-based studies on the effects of land disturbance on erosion and transport of nutrients and sediment from fine-grained soils to watercourses in both reference and disturbed conditions. Although the first signs of farming can occasionally be dated to the Stone Age (Lahtinen et al., 2017), development of the land use began mainly in the medieval times and is linked to the number of population. The number of inhabitants in Finland was only about 65,000 in the 13th century, 300,000 in the 16th century, 800,000 in the beginning of the 19th century, and 5.5 million in the late 20th century (Lehtosalo-Hilander, 1985). Tillage methods were still rudimentary in the 18th and early 19th centuries making the use of the heavy clay soils unattractive (Orrman, 1991). Arable land and meadows were either completely undrained or the network of ditches was limited, resulting in little direct runoff from the fields to watercourses (Soininen, 1974). With the development of tillage methods in the early 20th century, most of the areas containing fine-grained soils were drained and taken into cultivation. Mechanization enabled the efficient use of all types of arable land suitable for farming, which could lead to a significant increase in soil erosion and ensuing transport of sediment and nutrients into waterbodies. An important step in this regard was the transition from open ditches to drains. Only 1.4% of Finland was drained in the 1920s, but in the 1990s, the proportion of drained fields was already 53% (Aarrevaara, 1993). About 200,000 ha of cultivated land existed in southern and southwestern Finland in the 18th century, 440,000 ha at the end of the 19th century (62% of total cultivated area) and 978,000 ha in the 21st century (40%) (Soininen, 1974; Statistics Finland, 2022).

This study sought to determine and compare REF and recent (TOP) sediment, phosphorus (P), and carbon (C) loading conditions in 22 boreal catchments containing variable percentages of fine-grained soils (i.e., along a till-clay gradient) using lake sedimentological methods of coring, weighing, radiometric dating, and analyses of chemical composition. A TOP-REF, or after-before, approach was used in which REF sediment sections, taken below the signs of marked changes in sediment magnetic properties, are studied and compared with recent sediment sections from the same locations (AD 1986–2011). This allows extending the studies to more basins than otherwise feasible (e.g. Vähäkuopus et al., 2020) to select lake basins along a continuum of catchment geologies (here: clay-till gradient) and agricultural intensity. The within-basin sedimentological effects were considered by selecting simple-shaped basins of similar size and by studying the dimensions of the lacustrine sediment units from sediment-penetrating sounding profiles (Chassiot et al., 2022). Headwater lakes were selected to reduce the effects of upstream sedimentation. However, this type of TOP-REF research setting is less well suited for the documentation of the contribution of climatic effects, as all basins are in the same region and the modern sample sections represent the same time interval. The effects of climate change are therefore built in the results and recorded as part of the observed geological and land use gradients.

The aim of this sediment core-based study was to answer the following research questions. 1) Based on dating and the chemical composition of the core sections, what are the rates of sediment, C, and P accumulation (MAR, CMAR, PMAR) in the pre- and post-disturbance conditions of small boreal lakes with clay-rich catchments? Hypothesis: Accumulation rates of all three have increased due to human activities, including changes in the climate, but P retention may have decreased in some cases due to eutrophication-related hypolimnetic oxygen deficiencies. 2) How do the accumulation rates and sediment compositions differ along the till-clay gradient and between the pre- and post-disturbance conditions in the presently agricultural southern Finland? Hypothesis: There was a catchment geology-related gradient of mineral accumulation already in the post disturbance conditions but land disturbance, by especially agriculture corroborated by changes in climate, has resulted in a steepened gradient with high mineral matter accumulation in clay rich catchments. 3) Can we quantify area-specific suspended sediment loading from clay-rich boreal catchments under both recent and undisturbed conditions? Hypothesis: It is possible to obtain a realistic quantitative estimate of both background and recent loadings from the catchments that can be used in catchment management.

2. Materials and methods

2.1. Lake selection

This study comprises 22 mainly eutrophic lowland (14.8–110.6 m a.s.l.) lakes within a 230 × 90 km area in the clay region of southern Finland, extending to till-dominated soils (Fig. 1, Table 1, Supplementary material 1). Lakes with a high percentage of clayey soils in their catchment (30–76%) were mainly selected, but lakes with till-dominated catchments were also included to estimate changes in net sedimentation along a gradient of fine-grained soils. Quaternary glacial till is the most common surficial deposit overlying crystalline bedrock in Finland, whereas the clays are of post-glacial origin (Tikkanen, 2002). The selected lakes were morphologically as simple as possible and their median catchment to lake surface area ratio was 10. Headwater lakes were selected to reduce the effects of retention along the transport path before the basin. Data on lake basins, catchments, and water quality for the most recent period were obtained from public databases (Finnish Environment Institute 2022, Geological Survey of Finland 2022).

2.2. Sediment coring

The lakes were cored from ice in late winter 2011. The coring sites were located at the water quality monitoring sites of the environmental administration of Finland, mainly in the central parts of the basins. Recent, surficial sediments (~0–30 cm) were collected with a Limnos gravity corer (Kansanen et al., 1991) and REF section with a rod-operated piston corer (Livingstone, 1955) using 2-m plastic tubes (diam. 58 mm). The Limnos cores were sliced into 1-cm sections in the field, whereas the long cores were transported to the laboratory unopened for measurement and subsampling.

2.3. Sediment analyses

The 2-m cores were analyzed for magnetic susceptibility (10^{-6} SI) using a Bartington MS2E susceptibility meter with a MS2C loop sensor to aid in the selection of REF sediments below the first signs of human-induced erosion (Fig. 2). This intensification in erosion was seen as an

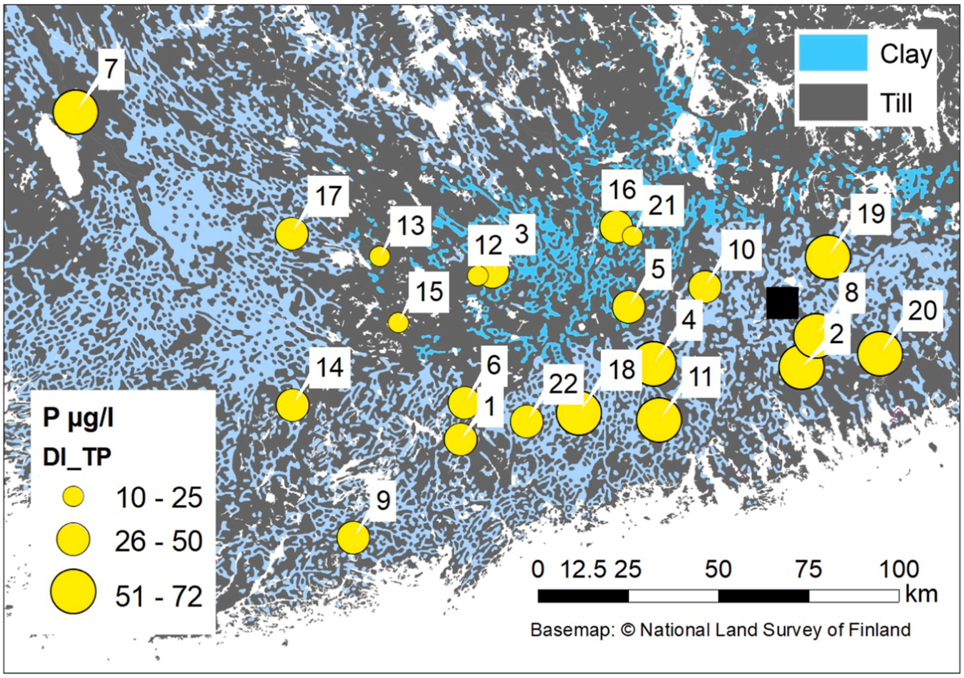

Fig. 1. The study sites and their diatom-inferred total phosphorus concentration ($\mu g\ l^{-1}$; Tammelin et al., manuscript in preparation, 2023) in pre-disturbance conditions. 1 = Enäjärvi, Vihti; 2 = Hopjärvi, Loviisa; 3 = Hyvälammi, Riihimäki; 4 = Isojärvi, Mäntsälä; 5 = Kilpijärvi, Mäntsälä; 6 = Kotojärvi, Vihti; 7 = Köyliönjärvi, Köyliö; 8 = Lapinjärvi, Lapinjärvi; 9 = Linkullassjön, Inkoo; 10 = Mallusjärvi, Orimattila; 11 = Niinijärvi, Pornainen; 12 = Ojajärvi, Loppi; 13 = Oksjärvi, Tammela; 14 = Omenojärvi, Kiikala; 15 = Onkimaanjärvi, Karkkila; 16 = Oriharonjärvi, Kärkölä; 17 = Rehtijärvi, Jokioinen; 18 = Rusutjärvi, Tuusula; 19 = Sääksjärvi, Iitti; 20 = Teutjärvi, Ruotsinpyhtää; 21 = Valkjärvi, Kärkölä; 22 = Valkjärvi, Klaukkala. Black square = Pyhäjärvi, Artjärvi (Itkonen and Salonen, 1994; Pajunen, 2004). The background image is based on the Quaternary Geology Map of Finland at 1:1 000 000 (GTK, http://geomaps2.gtk.fi/geo/) Blue = clay, gray = other deposits (mostly till), white = lakes.

increase in magnetic susceptibility, which mainly reflects the proportion of mineral matter in the sediment (Agricultural period, AGRIC, Fig. 2) (e.g., Thompson et al., 1975, Sandgren and Snowball, 2001, Valpola and Ojala, 2006). The 50-cm-long REF sediment section was taken below this susceptibility increase. One-centimeter-thick slices below and above the section (REF 1, REF 2) were taken for accelerator mass spectrometry ^{14}C age determination (AMS dating, University of Helsinki; Tikkanen et al., 2004). The dating results were calibrated to calendar years using the IntCal09 calibration curve (Reimer et al., 2009) and Oxcal 4.1 software (Bronk-Ramsey, 2009). Two lakes were excluded from the analysis of pre-disturbance conditions. Lake Onkimaanjärvi was excluded due to inadequate sample and Lake Köyliönjärvi for the long core not reaching undisturbed sediments at 193 cm depth.

The REF sections were weighed and a homogenized subsample was taken from each for geochemical analysis using the US EPA Method 3051 and a C/N analyzer by Labtium Oy (now Eurofins Labtium). These subsamples were weighed before and after drying at 105 °C to determine their wet and dry densities (D_W, D_D, g cm^{-3}; Håkanson and Jansson, 1983). The MAR_{REF}, CMAR_{REF}, and PMAR_{REF} (g m^{-2} a^{-1}) at the coring sites were calculated using the dating, D_D, and geochemical composition data. Accumulation calculations were based on the ^{14}C median CALBP-values, but calculation was performed also using maximum and minimum ^{14}C (± CALBP) values. This reflects the ^{14}C related maximum error in MAR. The piston cores penetrated the post-glacial gyttjas (high N/C organic lacustrine sediments, e.g. Wetzel, 2001) and reached the underlying clays in four lakes, allowing the sampling of the clays for geochemical analysis to estimate the composition of clays in the catchment. Additionally, 10-cm slices at the susceptibility minimum and immediately below and above it were sampled from two lakes for physical determinations. Furthermore, the AGRIC sediment sections above the REF section were dried and weighed to estimate the D_D and the amount of accumulated material in the agricultural period, but the D_D estimation was only based on weight and volumetric data, because the uppermost part of the cores dried in storage.

The short cores were weighed and dried for ^{137}Cs determinations at the Geological Survey of Finland (GTK) using EG&G Ortec ACE™– 2 K gamma spectrometer equipped with a four-inch NaI/TI detector. The ^{137}Cs-maximum value was used as a marker for the TOP layer to calculate the post-1986 Chernobyl accident MAR_{TOP}, CMAR_{TOP}, and PMAR_{TOP} at the coring sites (Fig. 3, Foucher et al., 2021b). The TOP samples were analyzed for chemical composition similar to the REF samples. It is emphasized, that the TOP-section is the uppermost part of the AGRIC-layer and represents only latest decades of the agricultural period in the catchment (Fig. 2).

2.4. Echo sounding

Sediment-penetrating dual-frequency echo sounding (200 and 24 kHz, Meridata MD 500 system) was used to determine sediment accumulation in the lake basins. Sounding lines were placed 100 m apart (Supplementary material 2). Observations obtained from the sediment cores aided in the interpretation of the echo-sounding profiles for the distribution of lacustrine gyttjas (in general sediments with C > 1%). Signal resolution was generally poor in nearshore areas. Such areas were not classified as gyttja, because they are apparently a mixture of gyttja and minerogenic material leached from the shores (Anderson and Martinez, 2015). Erosion, transportation, and accumulation (focusing) zones were identified when possible, and the thickness, area, and volume of the TOP and REF layers were calculated. The main emphasis in the interpretation was placed on the topmost ~1 m sediment sequence, which mainly comprises the AGRIC layer. Accumulation zones were defined as areas where the AGRIC layer was clearly visible. The bottom areas above it were defined as transportation zones and erosion zones that further continued into shallower areas. Because the thickness variations of overlapping sediment layers usually correlate relatively well

Table 1

The study sites, their location, and selected features. Catchm area = catchment area (km^2), Catchm clay = Catchment clay soils, % of surface area, Catchm field = Catchment field cover, % of surface area, Mean depth = basin mean depth (m), Max depth = basin max depth (m), Gyttja min depth = upper limit of gyttja (as water depth) determined by echo sounding (m), Lake area = lake surface area (m^2), Gyttja area = surface area of soft lacustrine sediments in the basin (m^2), P_{AQ} = observed modern lake water total phosphorus concentration calculated as growing season medians of 2000–2009 observations (μg l^{-1}), DI-TP = diatom-inferred total phosphorus concentration of lake water (μg l^{-1}) in reference period (Tammelin et al., manuscript in preparation, 2023), Gas = gas in sediment + the minimum depth of gas in the sediment, Vegetation = percentage emergent vegetation cover in the lake area based on aerial photographs (© National Land Survey of Finland).

Lake	Long	Lat	Catchm area (km^2)	Catchm clay (%)	Catchm fields (%)	Mean depth (m)	Max depth (m)	Gyttja min depth (m)
Enäjärvi	24.39531	60.35079	33.5	36.4	22	3.2	9.1	2.9
Hopjärvi	26.10026	60.55346	32.0	53.1	17	2.2	6.3	2.7
Hyvälammi	24.52305	60.77049	3.0	0	76	2	2.4	1.2
Isojärvi	25.35315	60.55456	15.0	34.8	23	1.9	2.7	1.4
Kilpijärvi	25.22065	60.69455	18.2	42.5	26	1.4	2.2	2.1
Kotojärvi	24.40903	60.44281	3.6	23.9	31	2	7.0	-
Köyliönjärvi	22.35246	61.10894	125.0	14	32	2.7	12.8	2.8
Lapinjärvi	26.17274	60.63082	37.0	46.9	20	2	2.6	2.4
Linkullasjön	23.88258	60.09591	11.5	33.9	35	4.2	6.6	3.0
Mallusjärvi	25.60382	60.74782	90.0	51.8	25	4.1	8.9	3.0
Niinijärvi	25.38915	60.41409	3.4	44.1	39	1.9	3.0	1.4
Ojajärvi	24.44864	60.75862	29.0	7	1.6	4.9	14.5	3.9
Oksjärvi	23.94915	60.79529	17.9	1.1	5	2.6	7.6	3.2
Omenojärvi	23.54466	60.41784	16.0	33.8	5	0.5	1.0	-
Onkimaanjärvi	24.05755	60.63372	27.1	0	0.2	3.7	15.7	3.4
Oriharonjärvi	25.14696	60.89184	12.8	13.4	13	2	3.0	1.8
Rehtijärvi	23.49357	60.84259	3.2	76.4	57	8	23.5	5.0
Rusutjärvi	24.98309	60.42699	13.4	39.2	27	2.5	3.6	-
Sääskjärvi	26.22669	60.82658	66.0	55.6	32	2.4	5.0	3.3
Teutjärvi	26.49643	60.58733	1755.0	63.4	2.4	0.7	2.1	-
Valkjärvi Kärkölä	25.23101	60.86946	20.0	29	30	3	11	2.2
Valkjärvi Klaukkala	24.72363	60.40211	7.7	71.7	23	7.2	12.2	5.6

Lake	Lake area (m^2)	Gyttja area (m^2)	P_{AQ} (μgl^{-1})	DI-TP (μgl^{-1})	Gas in sed. (y/n/cm)	Vegetation (%)
Enäjärvi	4,900,000	3,858,183	79	29	YES > 30 cm	NO
Hopjärvi	6,200,000	5,279,603	29	54	YES > 30 cm	YES 4%
Hyvälammi	119,400	109,956	125	29	YES > 0 cm	YES 5%
Isojärvi	3,100,000	2,665,465	75	54	YES > 30 cm	YES 4%
Kilpijärvi	2,600,000	2,193,973	77	34	YES > 0 cm	BAYS
Kotojärvi	300,000	240,000	67	31	-	NO
Köyliönjärvi	12,400,000	9,424,000	115	60	YES > 30 cm	BAYS
Lapinjärvi	5,200,000	3,785,522	83	63	YES > 30 cm	YES 10%
Linkullasjön	600,000	445,228	59	42	YES > 30 cm	BAYS
Mallusjärvi	5,400,000	4,156,626	99	46	YES > 100 cm	BAYS
Niinijärvi	300,000	227,236	114	54	YES > 30 cm	BAYS
Ojajärvi	4,500,000	2,705,928	10	14	NO	NO
Oksjärvi	3,000,000	1,973,372	14	22	PARTIALLY	NO
Omenojärvi	1,800,000	1,440,000	65	35	-	YES 25%
Onkimaanjärvi	600,000	2,367,326	19	10	NO	NO
Oriharonjärvi	500,000	354,341	49	32	YES > 30 cm	BAYS
Rehtijärvi	400,000	254,981	47	34	PARTIALLY	BAYS
Rusutjärvi	300,000	960,000	36	72	-	YES
Sääskjärvi	5,100,000	3,676,751	105	67	YES > 60 cm	BAYS
Teutjärvi	3,700,000	960,000	228	66	-	YES 25%
Valkjärvi Kärkölä	1,430,000	1,125,502	59	17	YES > 0 cm	YES 2%
Valkjärvi Klaukkala	500,000	919,876	25	31	YES > 60 cm	BAYS

in small basins (cf., Mäkinen and Ojala, 2013), the sounding profile interpretation of the AGRIC layer was considered to represent the thickness variations of both the REF and TOP sections in the accumulation zone. The sediment bed thicknesses determined in this study include sediment focusing via redeposition, thus portraying net sedimentation.

2.5. *Estimation of whole-lake accumulation rates and area-specific loading*

Whole-lake REF and TOP accumulation rates were estimated by combining the whole-basin sediment volumes with the site-specific dating, D_D, and chemical composition results. Therefore, we report MAR, CMAR, and PMAR both as site-specific rates and relative to the whole basin area. The whole-lake MAR was used for calculating area-specific suspended sediment loading from the catchment (kg ha^{-1} a^{-1}). A subset of 13 lakes (Enäjärvi, Hopjärvi, Isojärvi, Kilpijärvi, Kotojärvi, Köyliönjärvi, Lapinjärvi, Linkullasjön, Mallusjärvi, Niinijärvi, Rusutjärvi, Sääskjärvi, and Valkjärvi Kärkölä) were selected for this regional scale assessment because they had relatively similar sedimentation patterns and their sediment accumulation was considered to be described with sufficient accuracy. However, Lake Köyliönjärvi was only used for estimating the recent loading, as its long core did not reach the undisturbed sediments.

The estimation of area-specific suspended sediment load was made by calculating the specific load averages at varying retention rates (50% and 100%), because the trapping efficiency of the individual basins was not known. In addition to the trapping efficiency, the calculation of the specific load based on the MAR in the basin is affected by the production of autochthonous material (see Foster and Walling, 1994, Foucher et al., 2021a), which is more susceptible to decomposition than allochthonous material. Based on the average C concentration of 7% and the results of

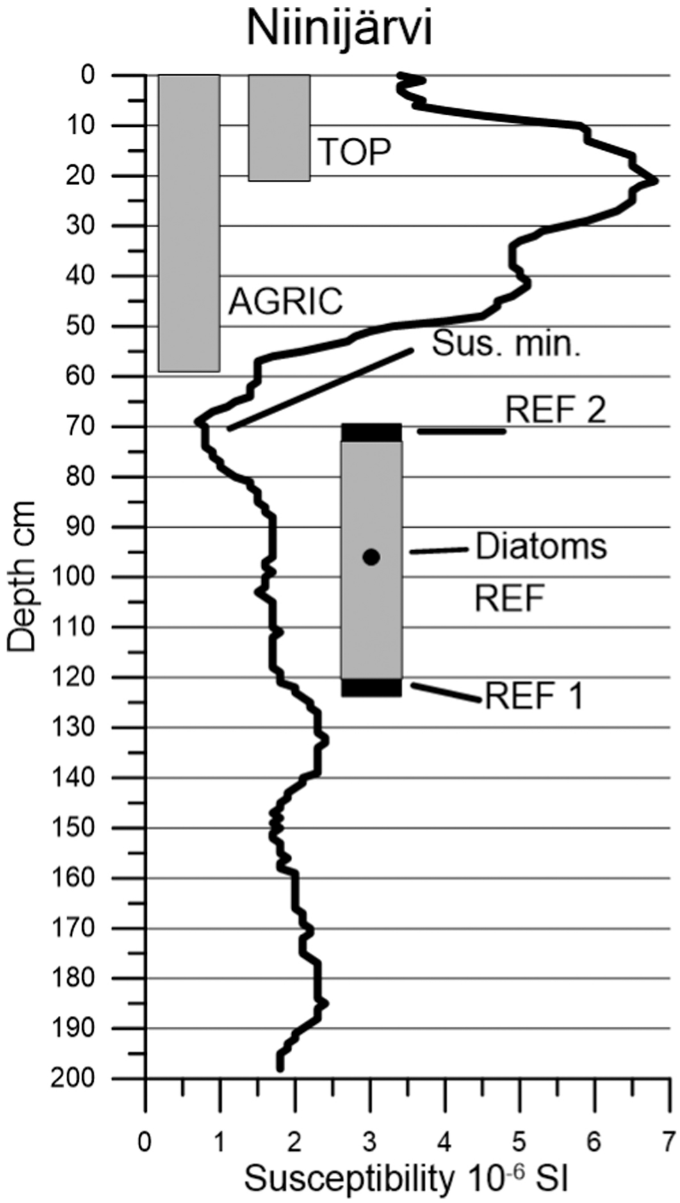

Fig. 2. Example susceptibility at the sampling point in Lake Niinijärvi and the sediment sampling scheme. The pre-disturbance sediment extends from the core bottom to a depth of about 70 cm and a 50-cm sample was taken from it for chemical analysis (one aggregate sample, REF). The middle sections of this pooled sample were sampled for diatom assays (two samples every 10 cm). Below and above the pre-disturbance sample, samples were taken for ^{14}C age determinations. Above about 55 cm, the sediment becomes clearly more clayey, reflecting the strong increase in land use intensity in the catchment area (AGRIC = Agriculture section). However, below this layer is a transition section that displays a susceptibility minimum (about 70 cm). At the core top is a 21 cm thick layer that has accumulated since 1986 (TOP). This section was also sampled for geochemical determinations. Other susceptibility curves are in the supplementary material 3.

Guillemette et al., (2017b) on the share of allochthonous organic matter in boreal lakes (57–87%), we estimated the share of autochthonous organic material to be 4% of the total sediment mass. This is because the amount of easily leachable and autochthonous inorganic material is minor compared to the total mass of sediment in Finnish lake sediments (see Fig. 23a in Mäkinen, 2019).

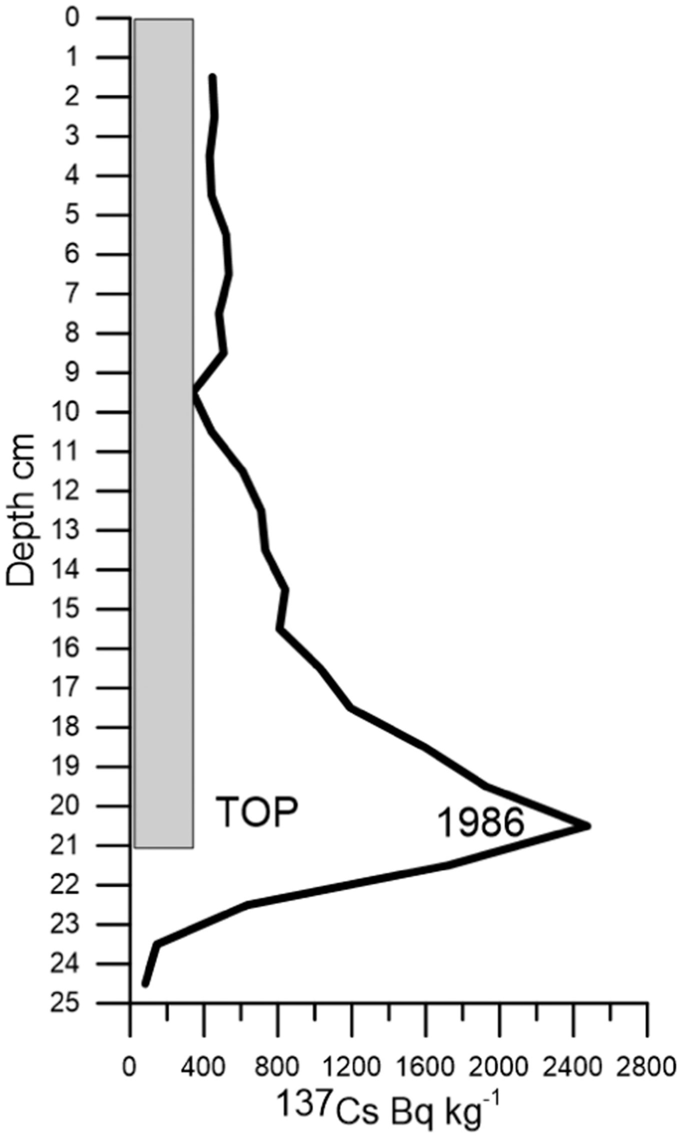

Fig. 3. ^{137}Cs activity from the Lake Mallusjärvi TOP sediment profile. The ^{137}Cs maximum is in the depth range of 20–21 cm, so the part above it has accumulated since 1986 (TOP). Other ^{137}Cs activity profiles are in the supplementary material 4.

2.6. Lake water phosphorus concentrations

Diatom-inferred lake water total phosphorus concentrations (DI-TP) were reconstructed for sediment subsamples taken from the middle parts of the REF sections (Fig. 2) in a separate study on the pre- to post-disturbance diatom assemblage and water quality change in naturally eutrophic boreal lakes (Tammelin et al., manuscript in preparation, 2023). Measured phosphorus concentrations retrieved from public environmental monitoring records were used as modern concentrations.

2.7. Correlations between variables

Relationships between the accumulation rate variables and the environmental (lake and watershed) variables and sediment chemical compositions were visualized with PCA ordinations performed separately for the reference and top accumulation rates. The ordinations were carried out with Past version 4.14 (Hammer et al., 2001) using correlation matrices and providing the environmental and sediment chemical variables as supplementary variables in the ordinations.

Finally, Spearman rank order correlations (r_S) were computed to identify statistically significant associations between accumulation rates, elemental concentrations, DI-TP concentrations, and variables describing the basins and catchments. Correlation coefficients statistically significant at the 0.05 level are indicated with one superscript asterisk (*), whereas two superscript asterisks (**) denote correlations significant at the 0.01 level (2-tailed). The reported correlation coefficients are for the site-specific accumulation rates. The numerical analyses were performed with IBM® SPSS® version 22 and Microsoft Excel.

3. Results

3.1. Sediment description

The REF sediments were in all cases visually homogeneous, mid- to dark brown gyttja or clayey gyttja. The long-cores of Lakes Kilpijärvi Mäntsälä, Oksjärvi, Onkimaanjärvi, and Teutjärvi reached the clays below the gyttjas and had gyttja thicknesses of 180 cm, 252 cm, 240 cm, and 120 cm, respectively. The D_W of the REF gyttjas varied between 1.03 and 1.25 g cm^{-3} and D_D between 0.07 and 0.45 g cm^{-3} (Table 2), depending on C concentration (3–22%; Table 3). These 50-cm REF sections represented an average of 1174 years (SD 573 yrs), during which sediment accumulation rate (SAR) was ~0.5 mm a^{-1}. Undisturbed, low magnetic susceptibility conditions (Fig. 2, Supplementary material 3) persisted, on average, until 1501 BP (SD 1116 yrs) according to the ^{14}C age determinations (Table 2). Almost all long cores displayed a rapid erosion-related increase in susceptibility at an average sediment depth of 60 cm. Magnetic susceptibility increased in the overlying AGRIC layer and typically reached its maxima at a depth of 10–50 cm. Between the REF and AGRIC sections, was a 10–20 -cm transitional zone characterized by a susceptibility minimum having 3% higher water content than the surrounding sediments.

The TOP sediment sections with a low C concentration of 2–4% were usually light brownish and unstructured. The color sometimes lightened towards the bottom of the profile. The TOP samples with a high C concentration (11–15%) were darker and quite homogeneous in appearance. Sulfide streaks sometimes occurred. The D_W of the short cores ranged between 1.03 and 1.22 g cm^{-3} and D_D between 0.06 and 0.37 g cm^{-3} (Table 2). The D_D of TOP and REF sections correlated

Table 2
REF 1 and REF 2 = lower and upper boundaries of the pre-disturbance sediment section (cm), REF Age 1 and REF Age 2 = sediment age at the upper and lower boundaries (^{14}C, CalBP), REF_D = dry density of the REF layer (g cm^{-3}), $AGRIC_D$ = dry density of the AGRIC layer (g cm^{-3}) + thickness of the layer (cm), TOP_D = dry density of the Top layer (g cm^{-3}), MAR_{REF} and MAR_{Top} = sediment accumulation (natural and recent; g m^{-2} a^{-1}, at the sampling point), $CMAR_{REF}$, $PMAR_{REF}$, $CMAR_{Top}$, $PMAR_{Top}$ = carbon and phosphorus accumulation natural and recent; g m^{-2} a^{-1}, at the sampling point. Columns MAR_{REF}, $CMAR_{REF}$ and $PMAR_{REF}$ contain also ^{14}C related maximum error (±).

Lake	REF 1	REF 2	REF Age 1	REF Age 2	REF_D	$AGRIC_D$	TOP_D	MAR_{REF}	MAR_{Top}	$CMAR_{REF}$	$PMAR_{REF}$	$CMAR_{Top}$	$PMAR_{Top}$
Enäjärvi	156	108	3945	2715	0.30	0.22 (0–60 cm)	0.13	118 ± 15	1367	6.4 ± 0.8	0.09 ± 0.01	108	2.10
Hopjärvi	111	60	1445	705	0.22	0.24 (0–40 cm)	0.22	153 ± 19	1148	10.7 ± 1.3	0.11 ± 0.01	41	1.27
Hyvälammi	146	95	3585	1845	0.16	0.10 (0–60 cm)	0.13	45 ± 4	751	6.3 ± 0.5	0.06 ± 0.01	61	0.89
Isojärvi	119	68	3328	2184	0.14	0.10 (0–30 cm)	0.08	62	472	6.9	0.04	49	0.48
Kilpijärvi	96	45	2730	0	0.12	0.05 (0–30 cm)	0.06	22 ± 1	427	2.3 ± 0.1	0.02 ± 0	65	0.58
Kotojärvi	122	69	1990	1210	0.22	0.24 (0–35 cm)	0.18	151 ± 30	1161	11.8 ± 2.3	0.15 ± 0.03	67	0.97
Köyliönjärvi	193	142	1305	995	0.52	0.27 (0–135 cm)	0.21	853 ± 258	2306	28.1 ± 8.5	0.59 ± 0.18	128	3.08
Lapinjärvi	173	122	3335	2045	0.17	0.24 (0–85 cm)	0.14	68 ± 8	658	7 ± 0.8	0.05 ± 0.01	56	0.64
Linkullasjön	141	89	2465	1216	0.26	0.36 (0–75 cm)	0.31	108 ± 14	2332	8.8 ± 1.2	0.22 ± 0.03	83	2.33
Mallusjärvi	146	100	1340	855	0.39	0.51 (0–85 cm)	0.37	371 ± 79	3087	11.5 ± 2.5	0.31 ± 0.07	61	3.05
Niinijärvi	123	72	1540	520	0.23	0.27 (0–60 cm)	0.30	113 ± 11	2482	9.3 ± 0.9	0.12 ± 0.01	108	2.01
Ojajärvi	92	40	3055	1660	0.20	0.15 (0–25 cm)	0.14	74 ± 9	442	3.9 ± 0.5	0.1 ± 0.01	-	-
Oksjärvi	157	91	5960	5315	0.10	0.03 (0–50 cm)	-	106 ± 36	-	23.6 ± 8	0.16 ± 0.05	-	-
Omenojärvi	153	102	1755	1170	0.17	0.15 (0–70 cm)	0.13	151 ± 52	413	14.2 ± 4.9	0.13 ± 0.04	39	0.37
Onkimaanjärvi	118	63	-	-	0.07	0.07 (0–30 cm)	0.07	-	499	-	-	-	-
Oriharonjärvi	108	67	2425	1395	0.20	0.35 (0–35 cm)	0.36	81 ± 12	1427	9.5 ± 1.4	0.19 ± 0.03	49	1.27
Rehtijärvi	107	54	2555	1230	0.30	0.37 (0–40 cm)	0.33	119 ± 17	1975	6.9 ± 1	0.1 ± 0.01	58	1.94
Rusutjärvi	118	67	1960	1305	0.13	0.12 (0–50 cm)	0.08	102 ± 14	678	11.1 ± 1.5	0.05 ± 0.01	76	0.82
Sääskjärvi	172	121	2230	820	0.45	0.57 (0–105 cm)	0.37	164 ± 17	2387	4.9 ± 0.5	0.11 ± 0.01	46	2.57
Teutjärvi	98	47	3340	1045	0.27	0.23 (0–40 cm)	0.20	61 ± 4	573	3.6 ± 0.2	0.04	27	0.48
Valkjärvi Kärkölä	157	109	3365	2235	0.29	0.3 (0–75 cm)	0.29	122 ± 17	2946	8.9 ± 1.3	0.17 ± 0.02	161	3.04
Valkjärvi Klaukkala	136	95	1150	555	0.33	0.37 (0–60 cm)	0.22	227 ± 55	1072	10.7 ± 2.6	0.19 ± 0.05	40	1.48

Table 3

Average element concentrations for the reference sediment sections (REF). The lowermost samples show average concentrations for original clays (CL) underlying the gyttjas. Al, Ca, Fe, K, Mg, C, and N are in % and other elements in mg kg^{-1}.

Lake	Al	As	Ba	Ca	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sr	Ti	V	Zn	Th	U	C	N
Enäjärvi	3.52	8	187	0.51	62	30	3.60	0.68	0.94	624	486	42	774	10	1130	31	1430	69	153	11	3	5.4	0.5
Hopjärvi	4.21	8	261	0.50	73	31	4.83	0.93	1.21	683	711	43	717	15	1140	34	1720	84	174	15	4	7.0	0.7
Hyvälammi	2.12	9	148	0.80	31	17	2.52	0.28	0.49	866	404	17	1300	6	1940	43	1140	46	156	5	3	13.8	1.0
Isojärvi	4.66	8	210	0.34	77	40	4.07	0.77	1.04	454	564	49	591	13	1750	29	1380	85	192	16	6	11.2	1.2
Kilpijärvi	3.47	8	189	0.48	55	25	3.60	0.53	0.75	449	316	35	1070	16	2830	31	1770	70	184	8	5	10.2	1.0
Kotojärvi	4.75	6	216	0.55	72	34	3.99	0.71	1.01	515	398	43	997	12	1210	32	1750	76	154	11	3	7.8	0.7
Köyliönjärvi	2.19	6	162	0.55	37	15	2.68	0.47	0.67	552	354	21	694	11	804	29	1390	47	71.1	9	4	3.3	0.4
Lapinjärvi	4.27	7	218	0.50	66	31	3.73	0.80	0.97	434	600	42	662	11	2030	35	1220	71	182	13	5	10.3	1.2
Linkullasjön	3.86	6	211	0.52	53	17	3.24	0.68	0.83	671	641	29	2030	11	1100	39	1640	68	154	8	3	8.1	0.6
Mallusjärvi	4.72	6	268	0.47	95	40	5.26	1.02	1.50	637	473	54	827	16	505	36	2300	93	178	15	4	3.1	0.4
Niinijärvi	4.71	6	238	0.45	81	29	4.30	1.01	1.17	473	654	45	1070	15	1260	37	840	80	182	12	4	8.2	0.8
Ojajärvi	2.31	8	98.9	0.50	45	19	3.55	0.40	0.69	516	312	21	1340	6	1020	23	1600	59	97.9	6	4	5.3	0.4
Oksjärvi	1.84	22	101	0.66	32	38	4.21	0.12	0.25	1330	143	21	1510	5	2490	36	550	51	130	5	25	22.3	1.3
Omenojärvi	3.51	8	171	0.35	61	29	3.64	0.53	0.78	527	336	39	861	10	1390	26	1480	69	178	11	4	9.4	1.0
Onkimaanjärvi	1.85	16	54.9	0.35	22	20	7.16	0.04	0.10	1180	94.5	10	2390	8	2330	24	405	57	125	3	6	19.5	1.3
Oriharonjärvi	3.75	4	181	0.58	60	22	3.04	0.55	0.79	493	319	32	2370	10	1380	40	1960	72	162	6	3	11.7	0.8
Rehtijärvi	4.81	7	221	0.33	86	43	4.11	0.98	1.19	499	446	49	880	14	666	29	957	79	177	17	6	5.8	0.5
Rusutjärvi	2.57	10	135	0.44	50	27	3.59	0.52	0.73	534	382	31	466	9	5460	23	1210	60	110	8	6	10.9	1.2
Sääskjärvi	4.31	6	234	0.49	83	33	4.55	0.87	1.28	556	553	47	661	13	430	37	1780	81	175	16	5	3.0	0.4
Teutjärvi	3.88	7	215	0.54	70	25	3.95	0.75	1.08	498	506	45	663	11	943	36	1800	70	226	14	3	5.9	0.6
Valkjärvi Kärkölä	3.38	5	190	0.51	69	23	3.68	0.72	1.01	697	362	36	1400	11	962	31	2160	85	163	8	10	7.3	0.5
Valkjärvi Klaukkala	4.27	8	254	0.48	84	40	4.66	0.96	1.32	661	590	47	840	17	774	32	1530	93	150	14	4	4.7	0.5
Kilpijärvi Mäntsälä CL	3.96	10	239	0.64	100	67	6.04	1.24	1.97	699	720	58	686	16	166	32	2750	109	161				
Oksjärvi CL	2.52	8	140	0.55	75	48	4.28	0.87	1.48	552	438	41	738	9	92	19	2610	83	111				
Onkimaanjärvi CL	2.11	8	121	0.52	58	26	3.9	0.67	1.14	984	472	29	696	7	236	18	2210	67	91				
Teutjärvi CL	3.3	7	231	0.63	77	48	5.03	1.06	1.55	593	797	54	663	17	159	34	2100	83	133				

positively (0.75 **). Both TOP (r_S= 0.95 **) and REF (r_S = 0.86 **) D_D also correlated with the D_D of the AGRIC layer, which ranged between 0.03 and 0.57 g cm^{-3} (Table 2). The 1986 increase in ^{137}Cs caused by the Chernobyl accident was detectable in all short cores at sediment depth 7–28 cm, even though the profile was smoothened out in some lakes (Fig. 3, Supplementary material 4).

3.2. *Echo sounding and sediment accumulation*

Echo sounding and the determination of accumulation area in the basin, employed to extend the coring results to the whole basin, was possible for 18 lakes (Fig. 4, Supplementary material 2, 5). Good quality sounding profiles allowed the identification of erosion, transportation, and accumulation zones in most basins. Only the erosion and accumulation zones could be interpreted from the profiles of Lakes Hyvälammi, Kilpijärvi, and Rehtijärvi (sediment C > 7%, P_{AQ} > 59 μg l^{-1}), because the sediment contained gas up to the sediment surface (Table 1). For Lakes Kotojärvi, Omenojärvi, Rusutjärvi, and Teutjärvi, the surface area of the accumulation zone had to be estimated assuming that the occurrence of accumulation zones is similar to the other lakes (see below: upper limit of gyttja). Because these four lakes are shallow, focusing was estimated to be relatively weak.

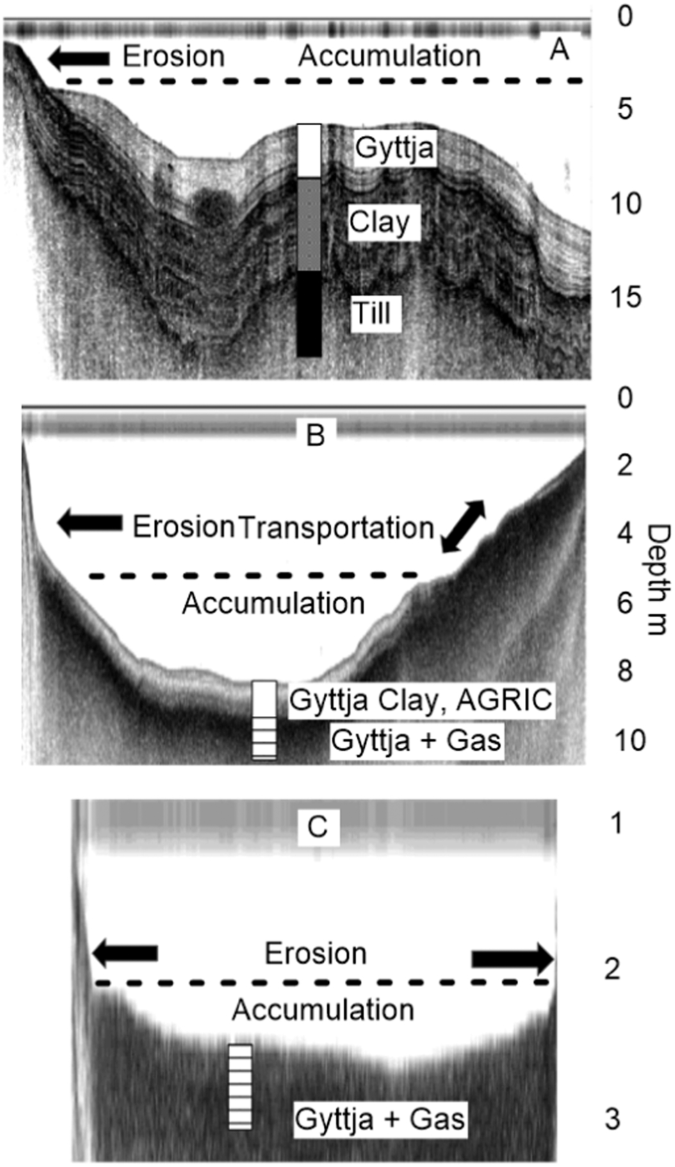

Fig. 4. Example echo-sounding profiles from Lakes Ojajärvi (A), Mallusjärvi (B) and Kilpijärvi (C). There has been no gas formation in the low-nutrient sediment (A), but in the more eutrophic lakes there has been a varying amount of gas formation, which slows down signal propagation. Sediment erosion, transport and accumulation zones can be distinguished with varying precision. In Lake Mallusjärvi, the uppermost gyttja clay layer in the middle of the accumulation zone represents the AGRIC layer (white bar). Below it is a layer containing gas (lined bar). Other echo-sounding profiles are in Supplementary material 5.

The best quality sonar signals were obtained from nutrient-poor lakes with little clay in their catchment area (Fig. 4). In these, the signal propagated all the way to the till, but the surface part of the sediment showed no distinguishable stratification despite a visible AGRIC layer in the susceptibility profiles. In more nutrient-rich lakes, gaseous sediment hampered signal propagation, but the uppermost part of the AGRIC layer could be distinguished as a light layer above the gas-containing material. The boundary between non-gaseous and gaseous sediment was partially gradual and was mainly located in the lower parts of the AGRIC layer or higher, at the location of the susceptibility maximum.

As to lake overgrowth, Lakes Lapinjärvi, Omenojärvi, and Teutjärvi were the most problematic to interpret. Overgrowth in the nearshore areas covered 10–25% of their entire lake areas (Table 1). Furthermore, the sampling point of Lake Teutjärvi is unlikely to represent the best possible accumulation site because of its location in a southern trough that is small relative to the size of the lake (Supplementary material 2).

According to the entire sonar data, the average upper limit of the gyttja (2.8 m) was close to the average mean depth of the lakes (3.0 m, r_S = 0.85 **). The accumulation zone covered, on average, 58% of the gyttja-covered area. In almost all cases, the strongest accumulation was located in the deepest part of the basin. This was particularly the case in circular-shaped lakes, in contrast to elongate or more complex basins. The accumulation zone under natural conditions was assumed to be approximately 6% larger than at present because of the approximately 1 m greater maximum water depth, which is reflected in a stronger sediment focusing compared to the current situation.

3.3. *Elemental composition and accumulation rates in the REF section*

The chemical compositions of the REF sediment samples are presented in Table 3. Based on duplicate analyses, the analytical variability was on average 2.6% from the mean value across all elements, being highest for As (6.5%) and Ti (7.0%) and < <2% for C and P. The C concentrations of the REF samples were on average 9.1%, being highest (>13%) in the three lakes with clay-poorest catchments. In the lakes with the highest reference period DI-TPs (> 45 μg l^{-1}; Table 1), REF sediments had P concentrations (~710 mg kg-1) similar to the clay samples below the gyttja (~700 mg kg^{-1}; Table 3).

Catchment clay percentage correlated positively with the pre-disturbance concentrations of several silicate bound elements (Table 3), such as Mg, K, Ni, Cr, and Al (r_S = 0.82**, 0.82**, 0.80**, 0.79**, and 0.68**).The REF sample P concentrations displayed strongest negative correlations with the sediment concentrations of Th, Ni, Cu, Na, Cr, and Mg (r_S = −0.75 **, −0.65 **, −0.52 *, −0.52 *, −0.51 *, −0.51 *), as well as with the catchment clay percentage (r_S = −0.65**) and DI-TP (r_S = −0.75**). In contrast, a positive relationship was found between P and C/N (r_S = 0.85**).

REF sediment C displayed strong positive correlations with N and S (r_S = 0.94 **, 0.89 **), whereas it correlated negatively with the catchment clay percentage (r_S = −0.64 **) and elements such as Mg, Cr, K, Ba, Th, and Ni (r_S = −0.72 **, −0.66 **, −0.62 **, −0.61 **, −0.60 **, −0.59 **).

The average pre-disturbance MAR_{REF}, $PMAR_{REF}$, and $CMAR_{REF}$ were 121 g m^{-2} a^{-1}, 0.12 g m^{-2} a^{-1}, and 8.9 g m^{-2} a^{-1} (coring site-specific; Table 2) and 62 g m^{-2} a^{-1}, 0.06 g m^{-2} a^{-1}, and 4.7 g m^{-2} a^{-1} (proportional to whole lake area), respectively. MAR_{REF} correlated positively with PMAR$_{REF}$ (r_S = 0.68 **) and CMAR$_{REF}$ (r_S = 0.55 *), as did PMAR$_{REF}$ and CMAR$_{REF}$ with each other (r_S = 0.64 **). MAR$_{REF}$ also correlated positively with certain REF sample elements, such as Fe, Mg, Cr, Ba, V, and K (r_S = 0.64 **, 0.64 **, 0.62**, 0.57 *, 0.57 *, 0.53*), and negatively with C, S, and N (r_S = −0.63**, −0.61**, −0.58 **). PMAR$_{REF}$ only correlated positively with Mn (r_S = 0.50 *) and P (r_S = 0.52 *), whereas no elemental correlations were found with CMAR$_{REF}$. None of the pre-disturbance accumulation rates correlated with the catchment clay percentage or DI-TP, but MAR$_{REF}$ and PMAR$_{REF}$ were generally higher in lakes with greater maximum depths (MAR$_{REF}$: r_S = 0.53 *, PMAR$_{REF}$: r_S = 0.46 *). In contrast, the DI-TP correlated negatively with maximum depth (r_S = −0.47 *) and positively with catchment clay (r_S = 0.59 **) and recent P$_{AQ}$ (r_S = 0.50 *).

The average area-specific suspended sediment loading from the catchment area varied from 69 to 137 kg ha^{-1} a^{-1} in the undisturbed conditions, depending on the amount of retention (100–50%).

When visualized through a PCA ordination run on the accumulation rate variables (Fig. 5a), the first component mainly separates between lakes of high natural accumulation rates (right) and low accumulation rates (left) while the second component differentiates high CMAR$_{REF}$ (upper quadrant) from high MAR$_{REF}$ and PMAR$_{REF}$ (lower quadrants). The high MAR$_{REF}$ lakes have a high percentage of clay in their catchment and are also deep (DEPTH$_{max}$) and large (LAKE$_A$). When selected sediment chemistries are plotted over the accumulation rates, the high CMAR$_{REF}$ lakes are also relatively high in C, N, S, and Mn but low in Al, K and Fe (Fig. 5b).

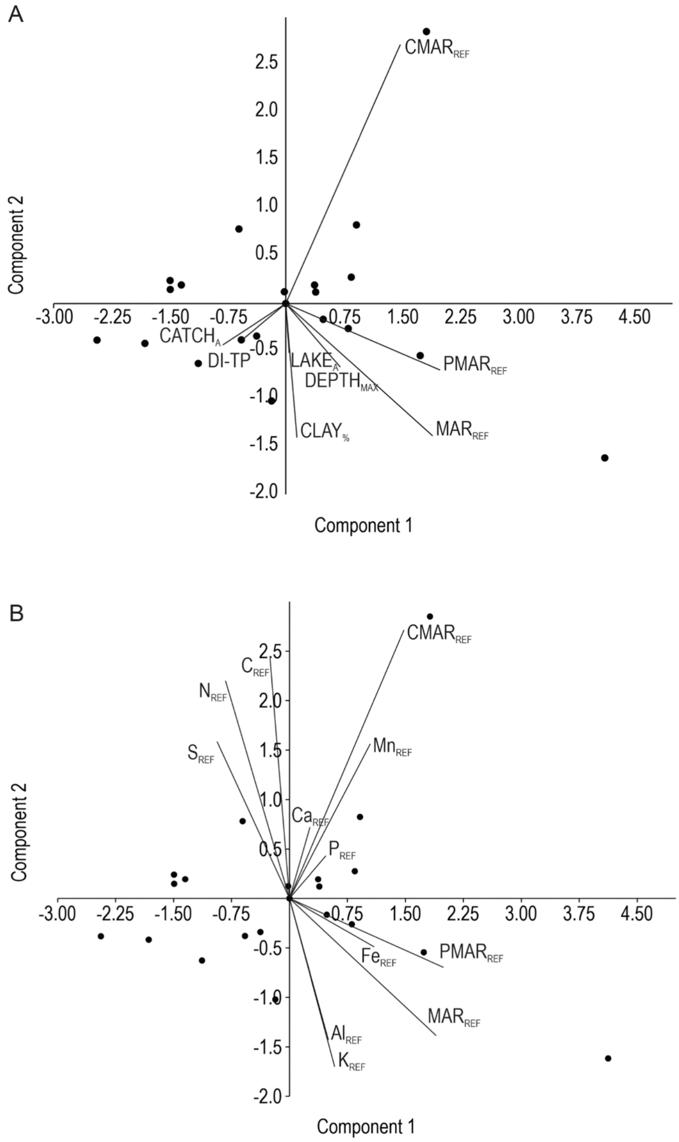

Fig. 5. a,b. PCA plots of the study lakes based on the accumulation rate variables (MAR, PMAR, CMAR) for the REF sediment sections. The plots also show the lake and watershed variables (panel a) and selected chemical variables (panel b) plotted as supplementary variables in the analysis. CATCH$_A$ = catchment area, LAKE$_A$ = lake area, CLAY$_{\%}$ = catchment clay soils, % of surface area.

3.4. Elemental composition and accumulation rates in the TOP section

Compared to the REF sediments, the concentrations of silicate-related elements, such as Al and K, were mainly higher in the TOP samples (Table 4) resembling the clay samples taken from the post-glacial clays underlying the lacustrine sediments in the cores of four lakes and occurring in the catchments (Table 3). P concentrations were also generally higher in the TOP samples (average ~1 100 mg kg^{-1}) and more even among the lakes. The average C concentration was slightly lower in the TOP sediments (6.2%) than in the REF sediments.

Many silicate-bound elements in the TOP sediments correlated with the clay percentage in the catchment (Table 4), but the correlations were generally weaker than in the REF sediments (e.g. Mg: r$_S$ = 0.58**, K: 0.56*, Ni: 0.64**, Al: 0.46*). TOP sediment P lacked a correlation with C/N ratio or lake water P (PAQ) and only correlated negatively with Ti (r$_S$ = -0.53*). TOP C correlated positively with N and S (r$_S$ = 0.99**, 0.87**), as well as with Pb (0.52*), and had negative correlations, for example, with Mg, Mn, V, and Fe (r$_S$ = -0.72**, -0.65**, -0.61**, -0.56*). Furthermore, there was a weak negative relationship between the recent C concentrations and mean and maximum lake depths (r$_S$ = -0.48*, -0.47*), while no such correlation was found between REF C and lake depth.

The site-specific averages of post-1986 MAR$_{TOP}$, PMAR$_{TOP}$, and CMAR$_{TOP}$ were 1 362 g m^{-2} a^{-1}, 1.55 g m^{-2} a^{-1}, and 69 g m^{-2} a^{-1} (Table 2), whereas the corresponding whole-lake values were and 693 g m^{-2} a^{-1}, 0.79 g m^{-2} a^{-1}, and 37 g m^{-2} a^{-1}. Hence, they were 11, 13, and 8-fold compared to the undisturbed conditions. Average SAR increased 13-fold from 0.5 mm a^{-1} to 6.4 mm a^{-1}, with the highest rate (11 mm a^{-1}) found in Lake Köyliönjärvi. The average area-specific suspended sediment loading from the catchment area varied between 767 and 1 534 kg ha^{-1} a^{-1}, depending on the amount of retention (100–50%).

Recent MAR$_{TOP}$ and PMAR$_{TOP}$ both correlated positively with their respective pre-disturbance rates (MAR$_{Top}$: r_S = 0.52 *, PMAR$_{Top}$: r_S = 0.64 **), but no correlation was found between TOP and REF CMAR. Compared to the undisturbed conditions, the correlation between MAR$_{TOP}$ and PMAR$_{TOP}$ was stronger (r_S = 0.93 **), whereas CMAR$_{TOP}$ showed slightly weaker correlations with MAR$_{TOP}$ (r_S = 0.53 *) and PMAR$_{TOP}$ (r_S = 0.56 *). MAR$_{TOP}$ and PMAR$_{TOP}$ both had a positive relationship with TOP Mn concentration (r_S = 0.72 **, 0.80 **) and negative ones with N (r_S = −0.75 **, −0.64 **), C (r_S = −0.70 **, −0.60 **), and Pb (r_S = −0.56 *, −0.51 *). No correlations were found between CMAR$_{TOP}$ and the elemental concentrations of the recent sediments. MAR$_{TOP,}$ PMAR$_{TOP,}$ and CMAR$_{TOP}$ were higher in lakes with a larger proportion of fields in their catchments (r_S = 0.59**, 0.48 *, 0.56 *). PMAR$_{TOP}$ also correlated positively with the maximum and mean lake depths (r_S = 0.79 **, 0.73 **) but not with the present-day epilimnetic P$_{AQ}$.

PCA ordination produced somewhat similar general results for the TOP samples than for the REF samples even though individual lakes are plotted differently. The first component again separates between high MAR$_{TOP}$ lakes (right) and low MAR$_{TOP}$ lakes (left) with high CMAR$_{TOP}$ lakes in the upper quadrants and high MAR$_{TOP}$ and PMAR$_{TOP}$ lakes in the lower right quadrant (Fig. 6a). The high accumulation rate lakes are deep but their catchments are not larger than average. The proportion of fields in the catchment (FIELD$_{\%}$) also correlates with high accumulation rates. The high MAR$_{TOP}$ and PMAR$_{TOP}$ lakes have high percentages of

Table 4

Average element concentrations for the recent (TOP) sediment sections. Al, Ca, Fe, K, Mg, C, and N are in % and other elements in mg kg^{-1}.

Lake	Al	As	B	Ba	Ca	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	P	Pb	S	Sr	Ti	V	Zn	C	N
Enajärvi	3.84	8	10	220	0.55	18	72	40	4.44	0.82	1.00	777	616	39	1451	28	3008	37	1553	79	159	7.70	1.01
Hopjärvi	4.52	8	11	297	0.55	22	84	43	5.75	1.17	1.37	1001	829	52	1109	30	1034	40	1930	91	174	3.54	0.47
Hyvalammi	2.43	8	<5	132	0.63	21	45	38	3.16	0.46	0.75	323	513	24	1186	16	4613	34	1715	59	104	8.11	0.88
Isojärvi	4.74	9	7	240	0.45	22	87	54	4.75	0.89	1.21	476	608	50	1021	49	2490	43	1665	87	193	10.33	1.30
Kilpijärvi	3.03	7	5	193	0.52	41	58	41	3.62	0.58	0.82	334	460	35	1348	34	9693	32	1580	66	182	15.13	1.86
Kotojärvi	4.22	7	<5	222	0.61	19	85	50	4.67	0.79	1.27	487	457	46	775	24	3158	37	2020	87	180	5.46	0.66
Köyliönjärvi	1.95	8	6	155	0.58	16	37	26	2.93	0.43	0.67	755	375	28	1335	16	4150	27	1153	45	96	5.54	0.73
Lapinjärvi	4.04	12	8	258	0.51	19	75	40	4.89	0.89	1.16	478	591	51	974	31	4666	38	1832	79	176	8.53	1.06
Linkullasjön	3.97	10	8	235	0.51	22	70	34	4.53	0.78	1.07	658	494	48	1000	21	2368	35	1485	76	164	3.55	0.43
Mallusjärvi	4.82	8	<5	307	0.54	21	99	47	5.89	1.24	1.66	839	711	55	989	21	642	46	2353	102	182	2.06	0.30
Niinijärvi	5.14	8	11	289	0.45	22	94	43	5.36	1.14	1.44	516	625	58	812	26	1733	43	1783	93	233	4.34	0.51
Omenojärvi	4.52	11	9	251	0.46	19	78	38	5.05	0.87	1.08	458	501	46	896	36	2510	38	1648	83	189	9.41	1.12
Oriharonjärvi	3.68	7	<5	215	0.48	17	82	31	4.46	0.88	1.33	482	435	39	891	16	1109	36	2565	89	163	3.44	0.39
Rehtijärvi	4.67	12	<5	225	0.40	22	97	63	5.79	1.04	1.47	561	517	54	984	23	698	38	2070	101	182	2.93	0.42
Rusutjärvi	3.10	11	6	172	0.45	27	62	43	4.45	0.60	0.90	493	471	38	1188	38	4524	29	1341	71	180	11.19	1.54
Sääskjärvi	3.68	9	<5	230	0.50	22	81	44	5.25	0.88	1.37	880	534	47	1077	20	534	41	1958	85	162	1.92	0.27
Teutjärvi	3.78	8	11	226	0.57	19	75	43	4.24	0.83	1.14	478	578	45	868	16	1448	42	1406	67	165	4.95	0.69
Valkjärvi Kärkölä	4.03	8	7	261	0.64	18	84	38	4.72	1.05	1.34	873	623	42	1031	19	1718	48	1341	91	169	5.48	0.54
Valkjärvi Klaukkala	5.17	12	13	318	0.51	19	98	54	5.76	1.26	1.53	792	860	55	1440	32	1948	42	1077	102	202	3.81	0.51

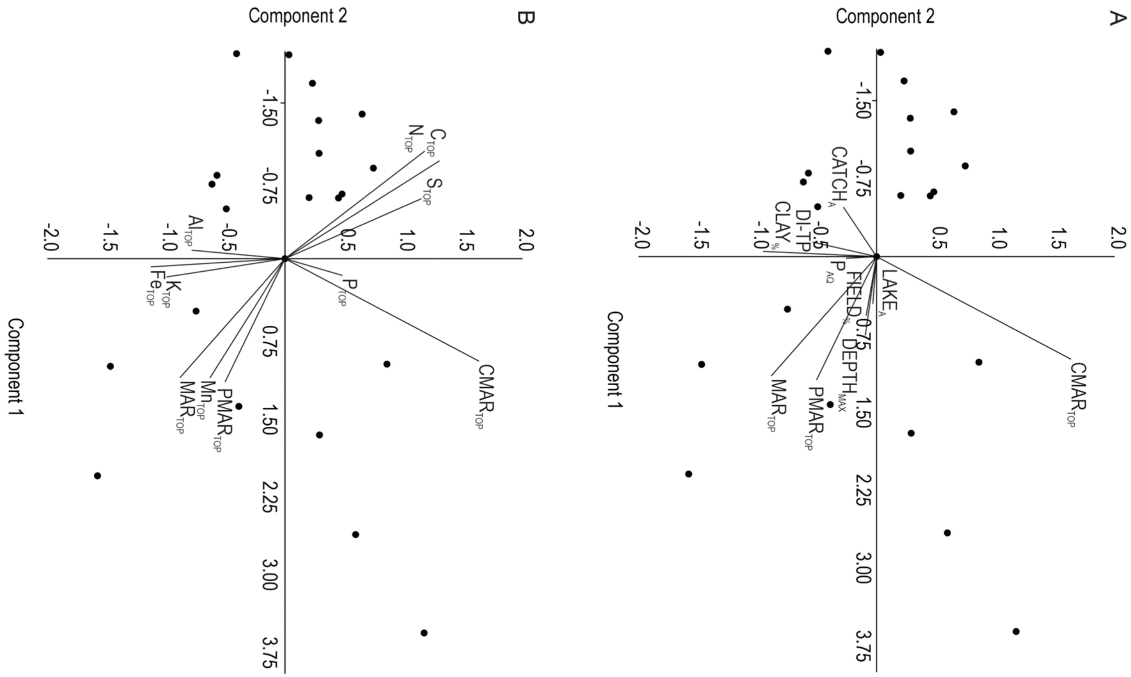

Fig. 6. a,b. PCA plots of the study lakes based on the accumulation rate variables (MAR, PMAR, CMAR) for the TOP sediment sections. The plots also show the lake and watershed variables (panel a) and selected chemical variables (panel b) plotted as supplementary variables in the analysis. FIELD$_\%$ = catchment field cover, % of surface area.

clay in their catchments, while CMAR$_{TOP}$ is negatively correlated with the proportion of clay. The sediment chemistries also plot fairly similar to the reference conditions, with the second component separating high C, N, S, and P samples (upper left) from the high Al, K, and Fe samples (Fig. 6b). In contrast to the reference lakes, the high MAR$_{TOP}$ and PMAR$_{TOP}$ top samples are also high Mn sediments.

4. Discussion

4.1. Pre- and post-disturbance accumulation rates

Sediment studies allowed estimates of pre-disturbance, or reference accumulation rates for the studied southern Finnish boreal lakes, located on the clay-rich coastal region of the Baltic Sea. These lakes had relatively low site specific MAR$_{REF}$ values in a global scale (121 g m^{-2} a^{-1}), regarding their nutrient-rich nature (mean DI-TP 41 µg l^{-1}; Table 1). A notably higher mean mid-19th century baseline value of 400 g m^{-2} a^{-1} was reported by Baud et al., (2021) based on a global set of lake sediment cores. According to the sediment cores of over 200 European lakes, lowland lakes had the highest (310 g m^{-2} a^{-1}) and mountain lakes the lowest (50 g m^{-2} a^{-1}) MARs in AD 1850, prior to the intensified land use of the 20th century (Rose et al., 2011). In contrast to the relatively low $MAR_{REF,}$ most of the studied lakes had a higher $CMAR_{REF}$ (average 8.9 g m^{-2} a^{-1}) than the average long-term CMAR estimate of 5.6 g m^{-2} a^{-1} determined from the sedimentary records of 228 European lakes (Kastowski et al., 2011). Compared to the long-term (Holocene) mean accumulation rate values in Finnish lakes in general (MAR 31 g m^{-2} a^{-1}, PMAR 0.05 g m^{-2} a^{-1}, CMAR 2.0 g m^{-2} a^{-1}), our estimates are (Table 2) higher but in in line with those previously found in small lakes on clay-rich regions (Pajunen and Mäkinen, 2003; Pajunen, 2004).

Notably elevated accumulation rates compared to background conditions were recorded for the recent, post-1986 sediments in the study region, deposited in the presence of human-induced stressors such as land use and climate change. As suspected, our recent accumulation rate estimates (Table 2) are comparable to those previously published for agriculturally-impacted, nutrient-rich boreal lakes (Tolonen et al., 1994, Tammeorg et al., 2018). The whole lake $CMAR_{TOP}$ values in most of the studied lakes (average 37 g m^{-2} a^{-1}) were higher than the median areal organic CMAR for boreal forest and taiga (18 g m^{-2} a^{-1}), but within or close to the global median C burial rate range of 46–88 g m^{-2} a^{-1} for lakes influenced by intensive agriculture and fertilizer use in different biomes (Anderson et al., 2020). Our average site-specific MAR_{TOP} (1 362 g m^{-2} a^{-1}) is remarkably similar to the mean post-1986 MAR (~1 300 g m^{-2} a^{-1}, single cores) of 21 meso–hypertrophic Finnish lakes studied by Tammeorg et al., (2018), whereas their mean PMAR (~2.9 g m^{-2} a^{-1}) is slightly higher than ours (1.55 g m^{-2} a^{-1}).

Although the magnitudes of our $MAR_{TOP,}$ $PMAR_{TOP,}$ and $CMAR_{TOP}$ estimates resemble those of other studies, the 11, 13, and 8-fold pre- to post-disturbance increases are high. It should also be noted that the REF to TOP change unlikely represents the greatest difference between undisturbed and human-impacted conditions, because the high D_D of the AGRIC section (Table 2) suggests even more intense sediment loading prior to the post-1986 TOP section. Baud et al., (2021) estimated a 3–4-fold MAR and SAR increase in a global lake set, from an average of 400–1 300 g m^{-2} a^{-1} and from 1.2 to 4.1 mm a^{-1}, respectively. A typical rise in sediment flux between undisturbed and human-impacted conditions has been 5–10-fold, although over 10-fold increases can be found in small and extensively impacted lowland catchments (Dearing and Jones, 2003). Contemporary CMARs that are only ~2–4.5-times higher than in long-term or pre-disturbance conditions appear common in the published literature concerning Northern Hemisphere lakes (Kastowski et al., 2011; Heathcote and Downing, 2012; Anderson et al., 2013, 2020). It is also worth to note, that some C and N degradation in sediment surface has probably affected the $CMAR_{TOP}$ /$CMAR_{REF}$ ratio (Gälman et al., 2008; Ferland et al., 2014).

The less pronounced 1.8-fold increase in epilimnetic P (Table 1) could be the result of P retention in the lakes. Engström et al., (2009) suggested increased sediment burial rate as a likely cause for the enhanced P retention in Lake Pepin (Minnesota, US), in which MAR increased approximately 10-fold, lake water P at least 4-fold, and sediment P 2-fold after European settlement in the early 19th century. Varying P retention can lead to a lack of correlation between sediment P burial flux and lake water P in long-term records, whereas recent sediments may also be influenced by diagenetic P enrichment of the sediment surface and time lags in P exchange at the sediment–water interface (Moyle and Boyle, 2021). The higher sediment P and the stronger correlation between MAR and PMAR in recent (r_S = 0.93 **) than in undisturbed sediments (r_S = 0.68 **), as well as the lack of correlation between sediment and epilimnetic P (P_{AQ} or DI-TP) all suggest that changes in P retention have occurred in the studied lakes (Søndergaard et al., 2003; Jilbert et al., 2020; Moyle and Boyle, 2021).

4.2. The influence of catchment geology and land use on sediment accumulation

Although catchment clay (Table 1) was clearly associated with the chemical composition of the pre-disturbance sediments (Table 3), the percentage of clayey soils in the catchment did not correlate statistically significantly with the accumulation rates (Table 2). This finding therefore partly contradicts our hypothesis on a geology-related pre-disturbance gradient in accumulation. However, there were indications of the effect of clay soils in the PCA ordination of the REF samples (Fig. 5a). Nevertheless, higher sediment concentrations of silicate-related elements and lower concentrations of organic elements characterize the clay-rich catchments compared to the till-rich catchments (Table 2, Fig. 5b). This relationship is partly spatial since the regions richest in clay are in the eastern parts of our study area (Fig. 1). The mean pre-disturbance sediment C concentration was somewhat lower than in small Finnish lakes on average (11.4%; Mäkinen and Pajunen, 2005), even though small and shallow lakes often have a higher proportion of organic matter in their sediments than large and deep lakes (Karjalainen et al., 2000). The sediment P concentrations of the naturally eutrophic lakes in these clay-rich regions were also lower than in small Finnish lakes in general (average 1 300 mg kg^{-1}, Mäkinen and Pajunen, 2005), corresponding to the concentrations found in the underlying clays and in other pre-isolation sediments (Mäkinen, 2005). This feature has been observed in earlier studies as well and is related to post-depositional mobility and inefficient retention of P in sediments (Carignan and Flett, 1981; Dillon and Evans, 1993; Tammeorg et al., 2018).

In addition to catchment deposits, basin depth was statistically significantly related to some of the observed differences among the lakes in their pre-disturbance state, indicating that a depth-related background gradient existed (Fig. 5a). Shallow lakes in our data set had generally lower MAR_{REF} and $PMAR_{REF}$ values, as well as higher DI-TPs than deeper lakes suggesting that shallow, wind-stressed lake basins were ineffective in removing phosphorus to the sediment already before recent disturbances. A notably high whole-basin MAR_{REF} of 184 g m^{-2} a^{-1} was estimated for the 8.9 m deep Lake Mallusjärvi, which is close to the 170 g m^{-2} a^{-1} evaluated by Pajunen (2004) for the much deeper Lake Pyhäjärvi (maximum depth 61 m; Fig. 1) located approximately 20 km to the east. Hhigh C burial is often related to high MAR and certain morphometric features, which impact exposure to oxygen and mineralization of the recently deposited material (Pajunen, 2004; Kastowski et al., 2011; Ferland et al., 2012, 2014; Kortelainen et al., 2013). However, PCA plots of both REF and TOP samples separated high-CMAR lakes from high-MAR lakes, the latter being typically deeper.

In our data set, higher $MAR_{TOP,}$ $CMAR_{TOP,}$ $PMAR_{TOP,}$ and lake water P_{AQ} were statistically significantly associated with lakes that had a larger percentage of fields on their catchments. This was also captured in the PCA ordination (Fig. 6a). Intensive MAR growth since the latter part of the 20th century has mainly been linked to enhanced catchment erosion (e.g. due to agriculture and urbanization) and elevated organic matter production in lakes, although climate change is likely to become an increasingly important factor controlling sediment accumulation in the future (Dearing and Jones, 2003; Brothers et al., 2008; Rose et al., 2011; Baud et al., 2021). Shallow lowland lakes are particularly susceptible to these changes with an augmented risk of lake overgrowth (Rose et al., 2011).

The elevated concentrations of silicate-related elements in the recent sediments (Table 4, Fig. 6b) suggest that tilling and ditching have promoted the erosion of unweathered clay-rich material from deeper soil layers compared to the undisturbed conditions (Mäkinen, 2005). The high specific surface area of fine-grained soils and their increased contact with water due to cultivation make these soils particularly susceptible to erosion (Rantakari and Kortelainen, 2008). Intensified land use since the Industrial Revolution is presumably a major driver behind the shift in sediment quality to more minerogenic in most of the studied lakes (Fig. 2, Supplementary material 3). However, the ~2-m long sediment sample of Lake Köyliönjärvi indicates millennial-scale agricultural loading and reflects the long history of constant settlement in Köyliö, which reaches back to the Stone Age (Salminen, 1905).

4.3. Area-specific suspended sediment loading

Improved understanding on the magnitude of natural loading from the fertile coastal areas of Finland is essential when assessing and reducing sediment and nutrient export into the Baltic Sea (Finér et al., 2021). Our estimates of the pre-disturbance area-specific suspended sediment loading for the clay-rich catchments of southern Finland (69–137 kg ha^{-1} a^{-1}) are significantly higher than the specific load of 5.1 kg ha^{-1} a^{-1} measured in minimally-disturbed, wooded mineral soil regions (Mattsson et al., 2003). However, the comparison between these two is challenging because of the differences in the research methods (flow measurement vs. sedimentology) and in the studied catchments. Practically all catchments in the fertile clay regions are agriculturally impacted and, thus, their present state cannot be used as a reference for the pre-disturbance conditions (Ekholm and Mitikka, 2006). Finér et al., (2021) estimated background exports over ~83 kg ha^{-1} a^{-1} for total organic carbon (TOC), ~2.2 kg ha^{-1} a^{-1} for N, and ~0.07 kg ha^{-1} a^{-1} for P in certain parts of southern Finland. Although our pre-disturbance results indicate higher background export from the clayey coastal catchments than previously anticipated, our recent area-specific suspended sediment load estimates fit well within the 50–5 000 kg ha^{-1} a^{-1} range of observations from Finnish field areas (Tikkanen et al., 1985; Tattari et al., 2015). For Lake Valkjärvi Klaukkala, however, our estimates (789–1 569 kg ha^{-1} a^{-1}) were higher than the approximately 600 kg ha^{-1} a^{-1} measured by Pajunen (2010). Studies from Southern Europe reported highly variable sediment loads ranging between 30 and 23 000 kg $ha^{-1}a^{-1}$ (Foucher et al., 2014; Foucher et al., 2021a; Chassiot et al., 2022).

4.4. Factors concerning the accumulation rate estimates

A pertinent aspect on our pre-disturbance accumulation rates is that the pre-disturbance (REF) sediment sections represent varying time periods prior to notable human influence in each individual catchment (Table 2). Furthermore, a gap exists between the upper limit of the REF section (REF 2) and the onset of human impact as REF 2 was taken well below the magnetic susceptibility rise (Fig. 2).

The reliability of the radiocarbon dating is a potential source of uncertainty in the MAR_{REF} estimates. Small, holomictic softwater lakes are considered relatively suitable for bulk sediment dating, but the approach is susceptible to possible contamination by old or young C or a non-constant reservoir effect (Björck and Wohlfarth, 2001; Ojala et al., 2019). Because we used the difference between REF 1 and REF 2 ages in the MAR calculations (Table 2), it is of primary interest that both are similarly influenced by the potential dating uncertainties. Both samples precede anthropogenic catchment disturbance and reworking of old organic soils (Guillemette et al., 2017a), as indicated by the magnetic susceptibility profiles (Supplementary material 3). However, Lakes Hopjärvi, Mallusjärvi, and Sääskjärvi were still undisturbed in the early 13th century according to our ^{14}C dating, whereas the varved sediments of Lake Pyhäjärvi (~20 km from these lakes, Fig. 1) indicate an onset of land use only in the 1670 s (Itkonen and Salonen, 1994). Other possible sources of temporal changes in the softwater reservoir effect include large fluctuations in lake level (e.g., in closed lakes in arid regions; Geyh et al., 1998) and permanent or seasonal hypoxia (e.g., in varved lakes; Ojala et al., 2019). The studied lakes are not varved, and large fluctuations in the lake level during the pre-disturbance period are unlikely (Saarnisto et al., 2000), but natural eutrophication may have led to occasional oxygen depletion in some of them. Nevertheless, the estimated pre-disturbance MAR_{REF} and SAR values are in the same range as in previous studies on small lakes with clayey catchments (Pajunen, 2004; Valpola and Ojala, 2006).

According to our results, the coring site-specific accumulation rates (Table 2) were, on average, ~2-fold compared to the values proportional to whole lake area due to sediment focusing. The relatively low maximum to mean depth ratio (average 2.4; Table 1) suggests a generally more modest influence of focusing than in the meso–hypertrophic lakes studied by Tammeorg (2018), which had a depth ratio average of 3.1. In Lake Kassjön in Sweden (depth ratio 2.2), the MAR, PMAR, and CMAR of a single central core were 33%, 50%, and 85% higher than the whole-lake values based on several cores (Rippey et al., 2008). Sediment focusing is, theoretically, most efficient during the early stages of sedimentation and lessens upwards in the sediment sequence as a basin becomes shallower and less steep (Blais and Kalff, 1995; Bennett and Buck, 2016). However, wind-induced bottom currents can affect the formation of accumulation-erosion zones in elongated or more complex lakes making sediment focusing less predictable (Blais and Kalff, 1995; Moyle and Boyle, 2021).

The vertical ^{137}Cs activity distributions (Fig. 3, Supplementary material 4) indicate generally more intense sediment redeposition in shallow lakes with low accumulation rates than in deeper lakes. This is seen as a prolonged rise in ^{137}Cs activity after the 1986 Chernobyl fallout or a broad peak instead of a more pronounced peak with a rapid post-fallout decline (cf., Ilus and Saxén, 2005). The gently upwards-decreasing post-1986 'tails' of ^{137}Cs, present in many of the profiles, have been described from several Fennoscandian lakes (including annually laminated sediments) and associated with the mobilization of ^{137}Cs from the catchment and littoral zone and its subsequent redeposition (Klaminder et al., 2012; Ojala et al., 2017; Vähäkuopus et al., 2020; Haltia et al., 2021).

Both post-depositional mobility (P and Mn) and mineralization of organic matter (C and N) alter the geochemistry of buried sediments, but their influence on the accumulation rate estimates was considered minor compared to the magnitude of the pre- to post-disturbance change. C is mainly of organic origin in Finnish lakes (Pajunen 2004, Rantakari and Kortelainen 2008) and organic C burial efficiency can vary considerably (3–93%) among lakes depending on sedimentation rate, sediment source, oxygen exposure time, and bottom water temperature (Sobek et al., 2009). However, C and N degradation slows down in deeper sediment layers until little loss takes place after a few decades (Gälman et al., 2008, Ferland et al., 2014). Sediment P, on the other hand, is known to migrate from deeper sediment layers and enrich in the oxygenated sediment surface (Søndergaard et al., 2003, Jilbert et al., 2020, Moyle and Boyle 2021).

The estimation of sediment volumes based on echo sounding and sediment cores is relatively simple and reliable but analyzing a larger number of cores would improve the interpretation especially if a large part of the basin is overgrown or gas hampers signal propagation in the sediment. The impact of vegetated areas on the sedimentation budgets of the basins could not be estimated using the echo-sounding approach, although it is significant because of the large amounts of organic and inorganic material typically bound to them (Madsen et al., 2001; Rooney et al., 2003). Thus, a single core from the central, open water area of an overgrown lake most likely represents a minimum estimate of MAR. This particularly concerns the recent MAR_{TOP} values, as the strongest overgrowth has likely occurred in the last 100–200 years simultaneously with anthropogenic eutrophication. More than a half of the annual bulk sedimentation may occur in the littoral zone of lakes in which macrophytes cover more than one-third of the sediment surface, whereas a similar portion of littoral PMAR would require a macrophyte coverage of approximately two-thirds (Madsen et al., 2001; Rooney et al., 2003).

5. Conclusions

According to the sediment cores and echo-sounding profiles of 22 small boreal lakes located in the currently agricultural, clay-rich lowland of southern Finland, MAR, CMAR, and PMAR were notably higher in the recent sediments compared to undisturbed conditions, confirming the first hypothesis of increased accumulation. MAR increased 11-fold, from 62 $g\ m^{-2}\ a^{-1}$ to 693 $g\ m^{-2}\ a^{-1}$. CMAR and PMAR are dependent on the overall accumulation rate, with CMAR showing an 8-fold increase from 4.7 $g\ m^{-2}\ a^{-}1$–37 $g\ m^{-2}\ a^{-1}$ and P a 13-fold increase from 0.06 $g\ m^{-2}\ a^{-1}$ to 0.79 $g\ m^{-2}\ a^{-1}$. These accumulation rates are proportional to the whole lake area and are approximately half of the site-specific values measured at the zones of high accumulation of the lakes. The patterns of pre- and post-disturbance mass accumulation rates were similar for P and the total mass but differed for carbon. Lakes with a larger proportion of fields in their catchments had statistically significantly (at the 5% level) higher recent accumulation rates. Human influence has also increased the sediment concentrations of silicate-related elements and epilimnetic P. In the undisturbed conditions, the lakes with clay-rich catchments were characterized by higher sediment concentrations of silicate-related elements, lower abundance of organic elements, and higher epilimnetic P than till-rich catchments (hypothesis 2). Deeper lakes had generally higher MAR_{REF} and $PMAR_{REF,}$ as well as lower DI-TPs. As hypothesized (3), it was possible to obtain realistic estimates of the area-specific suspended sediment load from these clay-rich catchments at 69–137 $kg\ ha^{-1}\ a^{-1}$ (undisturbed) and 767–1 534 $kg\ ha^{-1}\ a^{-1}$ (recent), depending on the amount of sediment retention in the lake basin (100–50%).

The results demonstrate that increases in sediment, nutrient, and organic matter accumulation due to agriculture can be several fold over undisturbed state even in formerly glaciated Boreal settings. The pre- to post-disturbance increases in the MAR, CMAR, and PMAR of the boreal clay region lakes are high when compared to prior research comprising a wider spectrum of lake types and ecoregions. Similarly, our estimates for area-specific sediment loads from the clay-rich lowland were notably higher than those previously reported for forested, till-rich boreal regions. Our results therefore highlight the role of clayey, agricultural catchments as sources of sediment and nutrient loading into the Baltic Sea, as well as emphasizes the sensitivity of small, naturally nutrient-rich boreal lakes to anthropogenic impact. The results further highlight the role of clay-rich boreal agricultural catchments as sources of sediment and nutrient loading into the Baltic Sea even though basins in the catchment capture part of this loading. Future multi-core studies would improve the interpretation on sediment accumulation and the MAR, CMAR, and PMAR estimates in heavily overgrown lakes, as overgrown areas have a significant impact on the sediment budget but are challenging to examine with our approach, which combines echo-sounding and single cores.

CRediT authorship contribution statement

Jari Mäkinen: Investigation. **Tommi Kauppila**: Formal analysis. **Mira Tammelin**: Formal analysis.

Declaration of Competing Interest

This work is part of a project called The effects of land use on diffuse loading (MAHA, 2010–2012), funded by the Ministry of Agriculture and Forestry of Finland.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ancene.2023.100421.

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