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Chemical responses of small boreal lakes to atmospheric and catchment drivers over four decades

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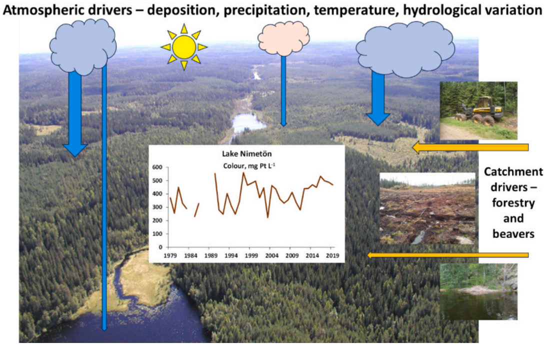
HIGHLIGHTS

- Water colour increased parallel with most rapid decrease in acid deposition.
- Water colour and iron responded in parallel to hydrological variation.
- Major cations declined and as well as electrical conductivity and alkalinity.
- Chemical recovery of the lakes from earlier acidification is still continuing.
- Groundwater lakes differed from lakes dominated by surface runoff.

GRAPHICAL ABSTRACT



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ABSTRACT

During the last few decades organic matter concentrations and water colour values have increased in a large number of lakes and rivers in Eurasia and North America. The upward shift in colour, often called water browning, and shortage of mobile cations have been linked to the recovery of catchments and lakes from acid deposition and increased precipitation. Here, long-term water chemistry responses of 33 boreal forest lakes to atmospheric and catchment scale drivers were studied in a small drainage basin in southern Finland. The longest data series cover four decades starting in 1979, and thus include the period of highest acid deposition in the middle of the 1980s and its dramatic decline during the next decade. The water quality data was taken during the autumn mixing, and in this long-term data set water colour increased significantly in 23 lakes, and the most uniform increase took place in the 1990s. In lakes fed predominantly by surface-waters, colour and iron have largely behaved in parallel, both responding to the variation in hydrology with higher concentrations after wet summers. Seepage lakes, in contrast, have responded to rainy periods less noticeably. In accordance with previous studies, the results indicate that most recent changes in colour have been hydrologically driven and are lake-specific rather than consistent among the lakes. In the long-term, the base cation concentrations have

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declined in most lakes, resulting in electrolyte dilution, loss of alkalinity, and decreased pH. In the uppermost headwater surface water lakes, iron concentrations showed an increasing trend since 1990, but in this century the trends have been less clear. Overall, the results suggest that 25 years after the most rapid reduction in atmospheric deposition, the studied lakes are still undergoing chemical recovery. Forestry practices and beavers may have impacted on many lakes as well, complicating interpretation of the chemistry patterns caused by changes in deposition and hydrological conditions, and lake-specific characteristics.

1. Introduction

Since the end of the 1980s, extensive lake and river water browning has been reported across Europe and North America (Roulet and Moore, 2006; Monteith et al., 2007; Yallop et al., 2010; Räike et al., 2012, 2024; Garmo et al., 2014; de Wit et al., 2016), a phenomenon often interpreted as a result of recovery from acidification (de Wit et al., 2021), and warmer and wetter climate as well as changes in land-use (Kritzberg et al., 2020). According to De Wit et al. (2016), in Fennoscandia increasing concentrations of coloured dissolved organic matter (CDOM), mainly comprised of humic substances, have appeared in surface waters across climatic gradients and catchments, and these trends have been most distinct in those areas with the strongest reduction in sulphur (S) deposition. In addition to humic substances, iron concentrations have also increased in many boreal freshwaters (Sarkkola et al., 2013), which can contribute to water colour (Kritzberg and Ekström, 2012; Xiao et al., 2015).

In Finland, a slight, but consistent, recovery in lake water pH in many of the acidified lakes has been reported (Mannio, 2001; Vuorenmaa et al., 2006; Vuorenmaa and Forsius, 2008). As a result, reproduction of acid-stressed fish populations has improved compared to the time before the reduction in acid deposition (Nyberg et al., 2010; Tammi et al., 2004; Rask et al., 2001, 2014). A similar recovery of acidified surface waters has been reported elsewhere in Europe and North America (Stoddard et al., 1999; Evans et al., 2001; Skjelkvåle et al., 2005; Keller et al., 2007; Baldigo et al., 2016).

A decrease in the acidity and ionic strength of soil solution has been shown to increase dissolved organic carbon (DOC) release from the soil (Tipping and Hurley, 1988; Evans Jr et al., 1988; Vance and David, 1989; Kalbitz et al., 2000), explaining, at least partly, the elevated DOC concentrations in freshwaters (see Krug and Frink, 1983; Evans et al., 2012). Besides S deposition, nitrogen (N; Pregitzer et al., 2004) and base cation depositions also affect soil water chemistry and DOC release from soil. Cation deposition increases buffering capacity of soil and mitigates the effects of acid deposition (Watmough et al., 2014) by enhancing the ionic strength of soil solution.

Besides large-scale drivers such as atmospheric deposition and climate, vegetation and soil in the surrounding catchment and hydrological connectivity influence lake pH, alkalinity and dissolved organic carbon (DOC) concentration (Arvola et al., 2004; Erlandsson et al., 2008; Ågren et al., 2014; Finér et al., 2021; Siefert and Santos, 2021; Räike et al., 2024). Drainage basin scale hydrological inter-connectivity is a vital link between terrestrial and aquatic ecosystems (Worral and Burt, 2007; Tranvik and Jansson, 2002; Erlandsson et al., 2008; Einola et al., 2011), and aquatic organic carbon and base cations, for example, usually originate overwhelmingly from terrestrial sources (Arvola et al., 1990a, 1990b; Siefert and Santos, 2021).

In areas with a cool and humid climate, such as in southern Finland, precipitation promotes runoff, which is governing the load of nutrients and DOC in rivers and lakes (Arvola et al., 2010; Räike et al., 2012, 2024; see also Laudon et al., 2011). Besides hydrologically driven processes, land-use practices related to forestry, including clear-cutting, afforestation, site preparation, ditching, stump-removal and burning, are possible influences on DOC and colour (Huotari et al., 2013; Kritzberg et al., 2020; Škerlep et al., 2020), as well as their fluxes from riparian zones due to floods caused by beavers (Vehkaoja et al., 2015; Nummi et al., 2018).

Our aim was to assess the factors that influence the chemical properties of small lakes and cause variations among them within a small geographical area where climate and atmospheric deposition are uniform. This study addresses the following questions. Does browning progress uniformly in the studied lakes as several previous studies have indicated (Q1)? Have base cation concentrations decreased as changes in their deposition suggest (Q2)? How have alkalinity, electrical conductivity, pH, nitrogen, phosphorus, potassium, and iron, in particularly, changed over time (Q3)? And finally, is the acidification still proceeding, or do the lakes indicate any chemical recovery from acidification as some recent results from North America and northern Europe suggest (Q4)?

2. Material and methods

2.1. *The study area*

In this study, we examine the variability and trends in water chemistry of 33 headwater lakes since the end of the 1970s until 2019. All the lakes are situated in a small, forested drainage basin (with a total area of 62 km^2) in the Evo area, in the uppermost reaches of the fifth largest river basin in Finland, the River Kokemäenjoki (Fig. 1). The study area belongs to the Long-term Ecological Research (LTER) network.

This study extends the timespan and number of variables in comparison to previous study (Arvola et al., 2010). The Evo area lies on the Precambrian Shield bedrock and its soils are affected podzolization, a result of the boreal coniferous forest and the cool, humid climate (Jylhä et al., 2014). In the eastern part of the study area, there is a black schist zone which goes from southeast to northwest and another zone extends in the west. Both zones are narrow and covered by sandy and/or podzol soils.

In the study area small peatlands dominated by *Sphagnum* are common both in the forest as well as around the lakes, while some nutrient-poor sandy and gravel deposits exist in the lower part of the drainage basin. Due to the small study area, the catchments and lakes share the same weather and climate while their landscape, landcover, and hydrology vary. Most of the lakes are fed predominantly by surface water runoff, but a few are seepage lakes fed by groundwater inflow and precipitation.

The area is sparsely populated, and besides atmospheric deposition forestry is the only major direct human disturbance on the lake catchments (Arzel et al., 2020). All the lakes are small (0.4–50 ha) and many of them are inter-connected hydrologically (see Arvola et al., 1990b, 2010; Järvinen et al., 2002) by streams or via groundwater. Lakes from Sorsajärvi to Alinen Rautjärvi (See Fig. 1 and Table 1) form a lake-chain. All the other lakes contribute to the lake-chain, e.g. the uppermost headwater lakes such as Nimetön, Tavilampi and Valkjärvi. Two groundwater-fed seepage lakes, Vähä Valkjärvi and Iso Valkjärvi, are hydrologically connected to the outflowing river from the Evo area rather than to Alinen Rautjärvi (Fig. 1, Table 1).

Most studied lakes ($N = 28$ lakes) have one or more inflows and one outflow, which means they are predominately fed by surface-water runoff. Headwater surface-water runoff lakes (HSW; $N = 11$) were distinguished from those lower in the lake-chain, which are referred to as downstream surface-water fed lakes (DSW; $N = 17$). Lakes without any visible inflowing stream or outflow, and thus predominantly fed by groundwater and precipitation to the lake surface, are referred to as groundwater (GW) lakes ($N = 5$). Among this category, Syrjänalunen is exceptional, because of large springs on the bottom due to its location beside an esker. On the other hand, Iso Mustajärvi belongs to HSW group of lakes, even though the lake is lying on sandy deposits (see Fig. 1), because the lake has a large catchment with many man-made ditches, which are promoting surface runoff. The characteristics and typology of the lakes and their catchments are given in Table 1, as well as the atmospheric deposition of sulphur (S), nitrous oxide (NOx), calcium (Ca), and magnesium (Mg) since the late 1980s in Fig. S1.

The surrounding soil types vary between the lakes. GW lakes are typically situated on the glacial-fluvial sandy or gravel deposits in the lower part of the study area, while HSW lakes are located on the podzol deposits in the upper areas. Although all the studied lakes are situated below 160 m a.s.l., which is the lowest level of supra-aquatic areas in southern Finland (Okko, 1962; Saarnisto and Saarinen, 2001), the catchments (or the major proportion of them) of six lakes are located above this zone. That means their soils have not been washed out after the last ice age. Basic information about the study lakes and their catchments is given in Table 1 and Table S1.

In addition, introduced American beaver (*Castor canadensis*) is active in the area (Vehkaoja et al., 2015). The beaver population has consisted of 20–40 individuals and beavers have occupied from time to time 27 of the lakes (Vehkaoja et al., 2015; Johnston, 2017; Kivinen et al., 2020). However, exact information about their activity in each lake is not very accurate, for instance regarding the date of arrival and departure, as well as their defined impact on the water table of the lakes.

2.2. Sampling and analytical methods

From three lakes water chemistry data is available since 1979, whilst in most lakes the data goes back to 1985. The last samples for this study were taken from each lake in autumn 2019. The sampling time has been always during the second half of October or during the first week of November, when the lakes are well mixed after autumnal turnover (Salonen et al., 1984). Water samples were taken from the shore with a small scoop attached to a 2 m long rod, and 3 subsamples from each lake (i.e. sampling site) were put into larger bottles, which were kept dark and cold, and after each sampling day they were transported to the laboratory of Lammi Biological Station, University of Helsinki, ~20 km from the study area. Because of the sampling, water samples represent the uppermost water layer (0–0.5 m). Each year all lakes were sampled within one week, usually within 2–3 days. Analyses were carried out according to the standard methods (see Arvola et al., 2014). One of the lakes, Valkea-Kotinen, belongs to the ICP IM programme (Forsius et al., 2021), and thus has a more intensive sampling scheme.

Colour (Col) was used as a proxy for dissolved organic carbon (DOC). Col measurements were carried out until 2000 by comparator colour discs, and after that by a spectrophotometer, except in Lake Valkea-Kotinen, where the spectrophotometer was used in parallel with comparator since 1990 (see Arvola et al., 2014). Water colour was determined by a spectrophotometer at 420 nm after filtration through 0.2 μm membrane or GF/C glass fibre filters (no significant difference in the filtration efficiency between the filters was found) and against Pt—Co standards (APHA 2000). DOC was determined by combustion at 900- -950 °C (Salonen, 1979) or, since 2001, at 680 °C with a Shimadzu

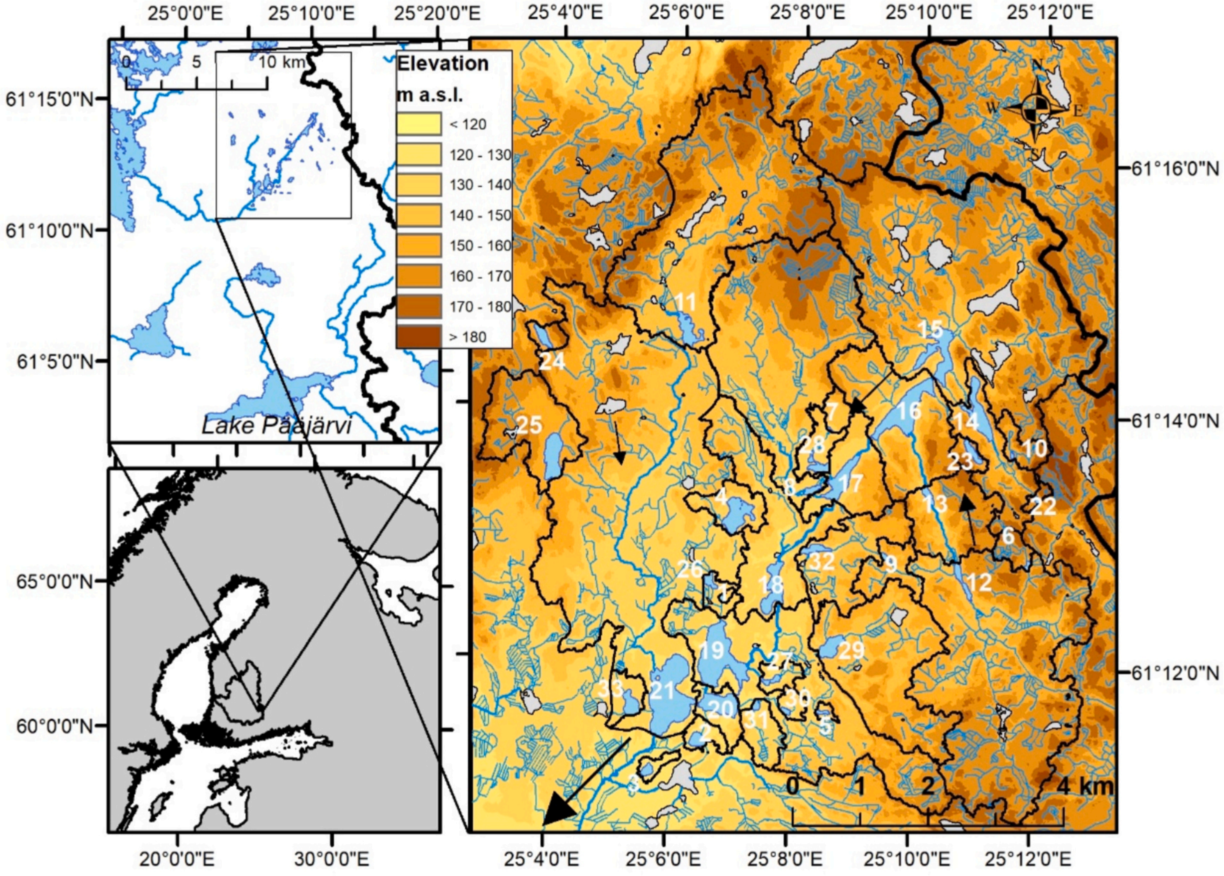

Fig. 1. Location of the Evo study area in Finland and in northern Europe, as well as the individual studied lakes shown in blue. The numbers refer to Table 1. Lake numbers from 14 to 21 represent the lake chain referred to in the text, and arrows indicate the directions of water flow. Lakes not included in the study are shown in grey. The map on the left shows the location of the study area in the greater River Kokemäenjoki catchment area. The arrows indicate water flow direction. Obs. The lakes locate on one drainage basin, but they have their own catchment areas, and some of those are also nested.

TOC 5000 analyzer.

The comparator results were made comparable with the spectrophotometer results by multiplying those by a factor 1.55 (Fig. S2). That was achieved by comparing the colour values measured by the two devices in lakes Valkea-Kotinen and Pääjärvi, a medium-size deep, humic lake in the vicinity of the Evo region. For both lakes, the same correction factor was obtained. Besides those measurements, simultaneous Col and DOC determinations were run two times from 15 Evo lakes and 39 times from lake Valkea-Kotinen. Both data sets showed a strong linear relationship ($R^2 > 0.97$; $\mathrm{Col} = 19.202*\mathrm{DOC} - 60.123$; $n = 30$) between the two variables (see Fig. S3), and slopes of the regression lines were close to each other. The other variables of interest were water pH, alkalinity (Alk), electrical conductivity (EC), total nitrogen (N), total phosphorus (P), natrium (Na), potassium (K), calcium (Ca), magnesium (Mg), manganese (Mn) and iron (Fe). Water pH, Alk, and EC were determined at the laboratory after sampling or during the following day. P and N concentrations were determined after wet oxidation (Koroleff, 1983), and cation concentrations by AAS as described by Keskitalo and Salonen (1994). If the samples couldn't be determined immediately after return to the laboratory, they were kept in cold (+4 °C) and dark before the determinations. The details of the analytical methods can be found from Arvola et al. (2014) and Vuorenmaa et al. (2014).

It is notable that cation data is lacking from several lakes since 1990 until 1998.

2.3. Local deposition and other measurements

The Finnish Meteorological Institute provided local deposition and meteorological data. The meteorological data are based on two meteorological stations, one situated in the Evo study area and the other at Lammi Biological Station (https://en.ilmatieteenlaitos.fi/observations-in-finland?station=101154). For precipitation, the sum from June to October was used from Lammi Biological Station and wet and dry years were defined as the uppermost and lowermost quartiles of the data sets, which means that wet years had precipitation higher than 410 mm and dry years lower than 250 mm, respectively.

The deposition data come from the Valkea-Kotinen research area (see Ruoho-Airola et al., 2014). The Finnish Environment Institute provided the hydrological data from a permanent hydrological measuring station in the outlet of Valkea-Kotinen (https://wwwp2.ymparisto.fi/scripts/hearts/welcome.asp). Another set of long-term hydrological data comes from the River Mustajoki, which originates from the eastern part of Evo area and flows into Lake Pääjärvi, about 10 km NE from Lammi Biological Station. For runoff, the average of October runoff was used, and for precipitation, the sum since the beginning of June until the end of October. The information about soil types is based on the soil maps prepared by the Geological Survey of Finland.

The study area has experienced a rapid decrease in sulphur (S) deposition since the end of the 1980s, soon after the protocols to the Convention on Long-Range Transboundary Air Pollution of the UNs Economic Commission for Europe and legislation of the European Union (Forsius et al., 2003) were adopted. In <10 years SO_4 deposition decreased by 60 % (Fig. S1), and at the beginning of the 2000s it was 70 % lower than in 1988, and 15 years later 80 %, respectively. A very similar decreasing trend has been found in nitrogen oxides (NOx) deposition, although the rate of decrease was lower (Fig. S1). After the

Table 1

The study lakes with their basic characteristics. Number refers to the map in Fig. 1; LA = Lake area (ha); Elev = Lake elevation above sea level (m); CA = Catchment area (ha); LA/CA = Lake area/Catchment area; Soil = Dominant soil type(s); Hydrology = Dominant water source(s); GW = Groundwater runoff lake; HSW = Headwater surface runoff lake; DSW = Downstream surface runoff lake; Stressor = key land-use factor(s) affecting lake, B = beaver present/impact during the study, PB = Prescribed burning during the last 50 years, NL = No forest logging during the last 50 years. Note: Different forestry practices, including ditching of peat soils, are not given. Only two catchments (Valkea-Kotinen and Karhujärvi) have had no forestry practices during the last 50 years. In addition, forestry practices have varied a lot in terms of intensity, extent, and location relative to a lake basin, but considering the entire study area it has been the only major man-made stressor since the middle of 1850s, excluding atmospheric deposition.

Lake	Number	LA	Elev	CA	LA/CA	Soil	Hydrology	Stressor
Alinen Mustajärvi	1	0.7	129.4	1.2	0.59	Sand/Glacial gravel	GW	
Iso Valkjärvi	2	3.9	126.3	13	0.30	Sand	GW	
Vähä Valkjärvi	3	2.4	125.9	10	0.23	Sand	GW	
Valkea Mustajärvi	4	13.9	135.9	41	0.34	Glacial till/Sand	GW	
Syrjänalunen	5	1.0	137.5	4.3	0.22	Sand/Glacial gravel	GW	PB
Karhujärvi	6	0.9	157.9	21	0.04	Glacial till/Rock/Peat	HSW	NL
Mekkojärvi	7	0.4	136.4	19	0.02	Glacial till	HSW	B
Haukijärvi	8	2.2	131.4	580	0.00	Glacial till/Peat	HSW	B
Horkkajärvi	9	1.2	142.7	52	0.02	Glacial till	HSW	PB
Nimetön	10	0.4	151.7	32	0.01	Glacial till	HSW	B,PB
Hokajärvi	11	9.3	141.2	556	0.02	Glacial till/gravel	DSW	B
Iso Keltajärvi	12	3.9	139.9	762	0.01	Glacial till/Rock	DSW	B,PB
Vähä Keltajärvi	13	2.5	136.1	894	0.00	Glacial till/Rock	DSW	B
Haarajärvi	14	15.0	141.4	155	0.10	Glacial till/Rock	DSW	B,NL
Sorsajärvi	15	19.2	133.9	1080	0.02	Glacial till/Rock	DSW	B
Savijärvi	16	27.3	133.9	2190	0.01	Glacial till/Rock	DSW	B
Rahtijärvi	17	12.9	131.1	2922	0.00	Glacial till/Rock	DSW	B
Pitkäniemenjärvi	18	13.9	126.8	3340	0.00	Glacial till/Rock	DSW	B
Ylinen Rautjärvi	19	37.4	125.5	3875	0.01	Glacial till/Sand/Rock	DSW	B
Keskinen Rautjärvi	20	14.3	125.5	4115	0.00	Glacial till/Sand	DSW	B
Alinen Rautjärvi	21	49.9	125.5	6198	0.01	Glacial till/Sand	DSW	B
Tavilammi	22	0.8	157.9	9.2	0.09	Glacial till/Rock	HSW	B
Valkjärvi	23	3.4	155.5	18	0.19	Glacial till/Rock	HSW	B
Valkea-Kotinen	24	4.2	155.9	22	0.19	Glacial till/Peat	HSW	B,NL
Iso Ruuhijärvi	25	14.2	149.0	149	0.10	Glacial till/Sand	HSW	B,PB
Iso Mustajärvi	26	2.6	129.6	125	0.02	Sand/Glacial till	HSW	
Kaitalammi	27	2.1	125.7	20	0.11	Sand	HSW	B
Halsjärvi	28	4.4	131.2	60	0.07	Glacial till/Sand	DSW	B
Hautajärvi	29	7.7	137.4	300	0.03	Sand/Glacial till	DSW	B,PB
Huhmari	30	1.1	128.2	14	0.08	Sand/Glacial till	DSW	B,PB
Särkijärvi	31	1.5	126.0	44	0.03	Sand/Rock	DSW	B
Majajärvi	32	3.9	133.3	186	0.02	Glacial till/Rock	DSW	B
Onkimanjärvi	33	6.3	129.4	109	0.06	Sand/Glacial till	DSW	B

first 10 years of the study, approximately 50 % of NOx deposition remained. Parallel with the reductions in acid deposition, a reduction was found in cation deposition; calcium (Ca) deposition has declined slightly more in comparison to magnesium (Mg), and the reduction of Ca followed tightly that of S ($R^2 = 0.735$, $p < 0.001$), and Mg that of NOx ($R^2 = 0.810$, $p < 0.001$).

The long-term chemistry data records were also looked at shorter 10 years long periods.

2.4. Statistical analyses

Original non-transformed data was used for the Mann-Kendall trend analyses (MK trend), which were carried out by Spreadsheet for Excel provided by the Finnish Meteorological Institute. Trends were calculated for all lakes for different time periods since 1979, depending on the lake and chemistry variable. The longest data sets (38 years) were available from two HSW lakes (Horkkajärvi and Nimetön), followed (35 years) by one GW lake (Alinen Mustajärvi), and the statistical differences in the chemistry variables between the three lakes were tested separately by one-way ANOVA. In addition, correlation of Col and Fe values of two lakes, Valkea-Kotinen and Pääjärvi, both with extensive data sets was calculated. $P < 0.05$ was used as a criterion for significance; the analyses were made using SPSS Statistics 28.

The chemistry trends over time of different lake types were investigated using General Additive Models (GAM) and Generalized Linear Models (GLMM). We fitted both model types with an interaction

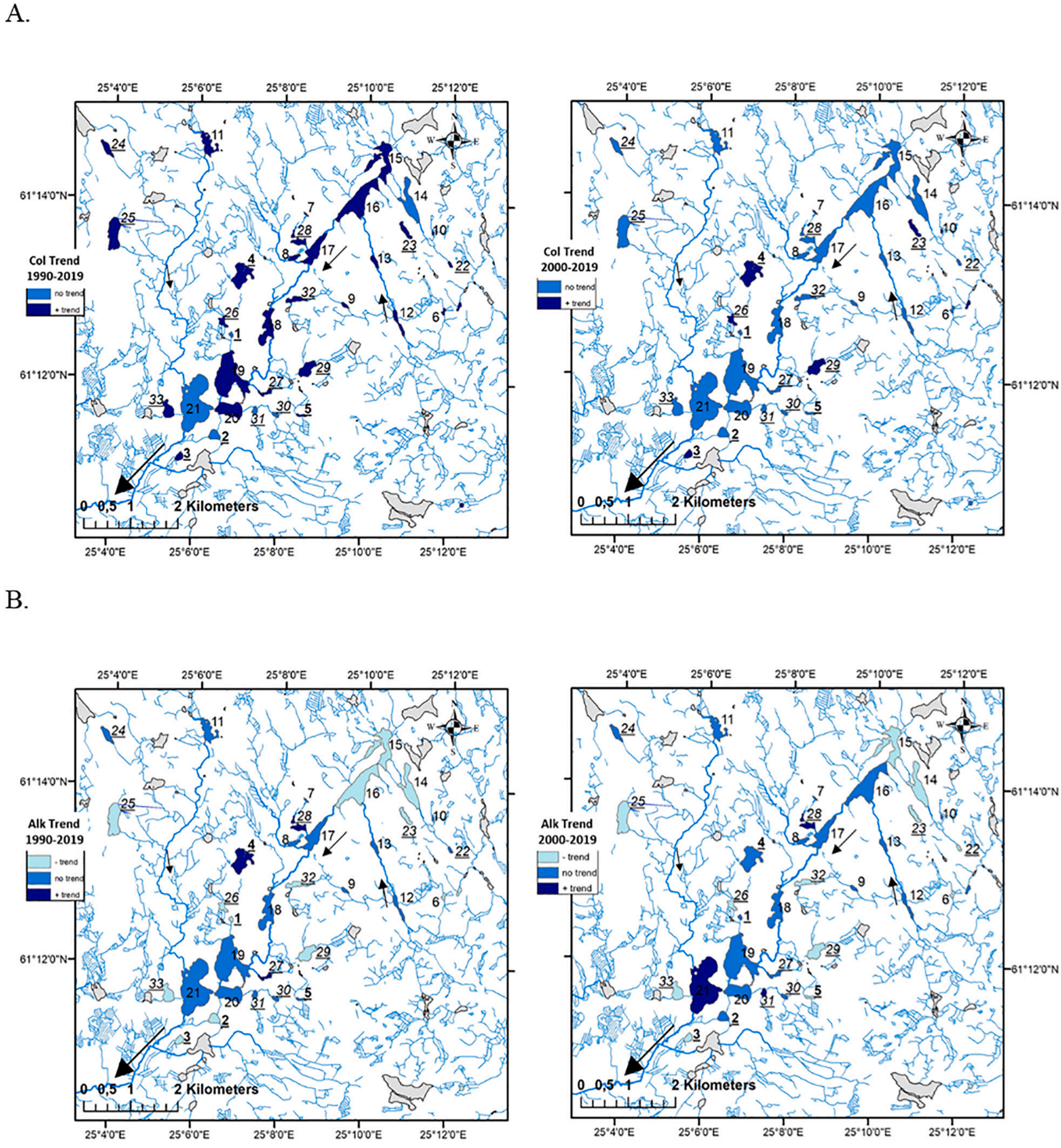

Fig. 2. Lake specific MK trends of colour (A) and alkalinity (B) since 1990 (left) and since 2000 (right). Dark blue colour indicates significant trend ($p < 0.05$). In Fig. 2 B. decreasing trends are also given. The arrows indicate water flow direction.

between Year and LakeType (DSW, HSW, GW), and Lake as a random factor. Before the GAM and GLMM analyses, we correlated the chemistry variables (see Suppl.) and dropped from the analysis the variables correlated to avoid analysis redundancy, but also meaning that the results from one can be interpreted for the other variable depending on the correlation level.

All GAMs and GLMMs analyses were performed using program R (R version 3.6.2 (2019-04-26), R CoreTeam, 2019), and R package by Sarkar (2008) was used for graphical edition, mgcv R package (Wood, 2017) was used for GAM analyses, and that by Bates et al. (2015) for GLMM analyses (for details, see Supplement for Material and methods).

For the GAM and GLMM analyses log-transformed data was used to meet better the assumption of normal distribution.

3. Results

Most chemistry variables differed between the GW lakes and SW lakes (Fig. 2, Table A1), with typically lower values in GW lakes. The variation within the lake categories was large, however, and in GW lakes the high variation was mostly dependent on one lake (Syrjänalunen) with very high base cation concentrations. If only humic lakes (Col values >100 mg Pt L^{-1}) with an average pH >5.6 were considered, thus excluding the most clearwater lakes and the three most acidic lakes, the relationship between pH and Col ($r = 0.823$, $p < 0.001$, $n = 26$) indicated that a Col increase by 100 mg Pt L^{-1} corresponded a decrease in pH by 0.24 unit.

3.1. Water colour and iron

Col values varied among the study lakes between 3 and 873 mg Pt L^{-1}. Within the lake categories, the highest values were in HSW lakes (mean value 288 mg Pt L^{-1}) and lowest in GW lakes (mean value of 43 mg Pt L^{-1}). The Col values (Table S1) clearly showed that most lakes were heavily loaded by allochthonous humic substances. Only in four GW lakes did the Col values remain constantly below 100 mg Pt L^{-1}. Since 1990 Col values increased significantly (MK trend, $p < 0.05$) in 24 out of 33 lakes (Fig. 2A; Table 2), and the highest values were often measured during the last or second last 5-yr period (Figs. S2, S3). By contrast, the lowest Col values were found in the 1980s in the three lakes with the longest data series (Fig. 3). Since 2000, however, only seven lakes had a significant increasing Col trend ($p < 0.05$). Col values and MK trends were usually strongest in HSW lakes (Figs. 2, 4; Tables S2, S3).

According to the GAM results, Col levels between HSW and DSW lakes did not differ (Table 3), however, and their trends did not show any difference (Fig. 4). Relative to the GW lakes, Col values of SW lakes were significantly higher, and in SW lakes the GAM results indicated a slight increase in Col over the last 30 years (Fig. 4).

The GAM results (Table 3) showed that the parametric part of the model only included LakeType. When the relationship between Col and Year (according to lake types) was significant ($p < 0.05$), the relationship was non-linear for all the groups, including the random effect (Lake), and the model explained a significant amount of the variability of Col in the data.

$$\text{We fitted each time the formula : } Y = \alpha\text{LakeType} + s(\text{Year*LakeType}) + s(\text{Lake}) + \varepsilon$$

Table 2
Number (N) of lakes and their share (%) of the studied lakes with statistically significant (SS) increasing and decreasing Mann-Kendall Trends of different variables between 1990 and 2019, and 2001 and 2019. GW = Groundwater runoff lake; HSW = Headwater surface runoff lake; DSW = Downstream surface runoff lake; pH = water acidity; alk = alkalinity; EC = electrical conductivity; Col = water colour; N = total nitrogen; P = total phosphorus; Na = sodium; K = potassium; Ca = calcium; Mg = magnesium; Mn = manganese; Fe = iron.

1990–2019		pH	Alk	EC	Col	N	P	K	Na	Ca	Mg	Mn	Fe
GW	N of significance	3	4	5	1	1	1	2	1	4	3	1	3
	% of all	60	80	100	20	20	20	40	20	80	60	20	60
	inc. (%)	20	25	0	100	0	0	0	0	0	0	0	67
	dec. (%)	80	75	100	0	100	100	100	100	100	100	100	33
HSW	N of significance	8	5	8	10	2	3	3	2	6	6	3	4
	% of all	73	45	73	91	18	27	27	18	55	55	27	36
	inc. (%)	0	0	0	100	100	100	0	50	0	0	0	100
	dec. (%)	100	100	100	0	0	0	100	50	100	100	100	0
DSW	N of significance	6	9	15	12	3	6	9	7	10	11	7	7
	% of all	35	53	88	71	18	35	53	41	59	65	41	41
	inc. (%)	0	22	0	100	65	50	0	72	0	0	0	100
	dec. (%)	100	78	100	0	35	50	100	28	100	100	100	0
GW + SW	N of significance	17	18	28	23	6	10	14	10	20	20	11	14
	%	52	55	85	70	18	30	42	30	61	61	33	42
2000–2019		pH	Alk	EC	Col	N	P	K	Na	Ca	Mg	Mn	Fe
GW	N of significance	3	3	4	1	1	0	0	1	3	2	1	1
	% of all	60	60	80	20	20	0	0	20	60	40	20	20
	inc. (%)	0	33	0	100	100	0	0	0	0	0	0	100
	dec. (%)	100	67	100	0	0	0	0	100	100	100	100	0
HSW	N of significance	6	5	4	4	2	1	2	2	6	5	2	1
	% of all	55	45	36	36	18	9	18	18	55	45	18	9
	inc. (%)	0	0	0	100	100	0	0	50	0	0	0	0
	dec. (%)	100	100	100	0	0	100	100	50	100	100	100	100
DSW	N of significance	6	7	9	1	2	5	8	5	8	9	8	2
	% of all	35	41	53	6	12	29	47	29	47	53	47	12
	inc. (%)	0	15	0	100	100	20	0	60	0	0	0	0
	dec. (%)	100	85	100	0	0	80	100	40	100	100	100	100
GW + SW	N of significance	15	15	17	6	5	6	10	8	17	16	11	4
	%	45	45	52	18	15	18	30	24	52	48	33	12

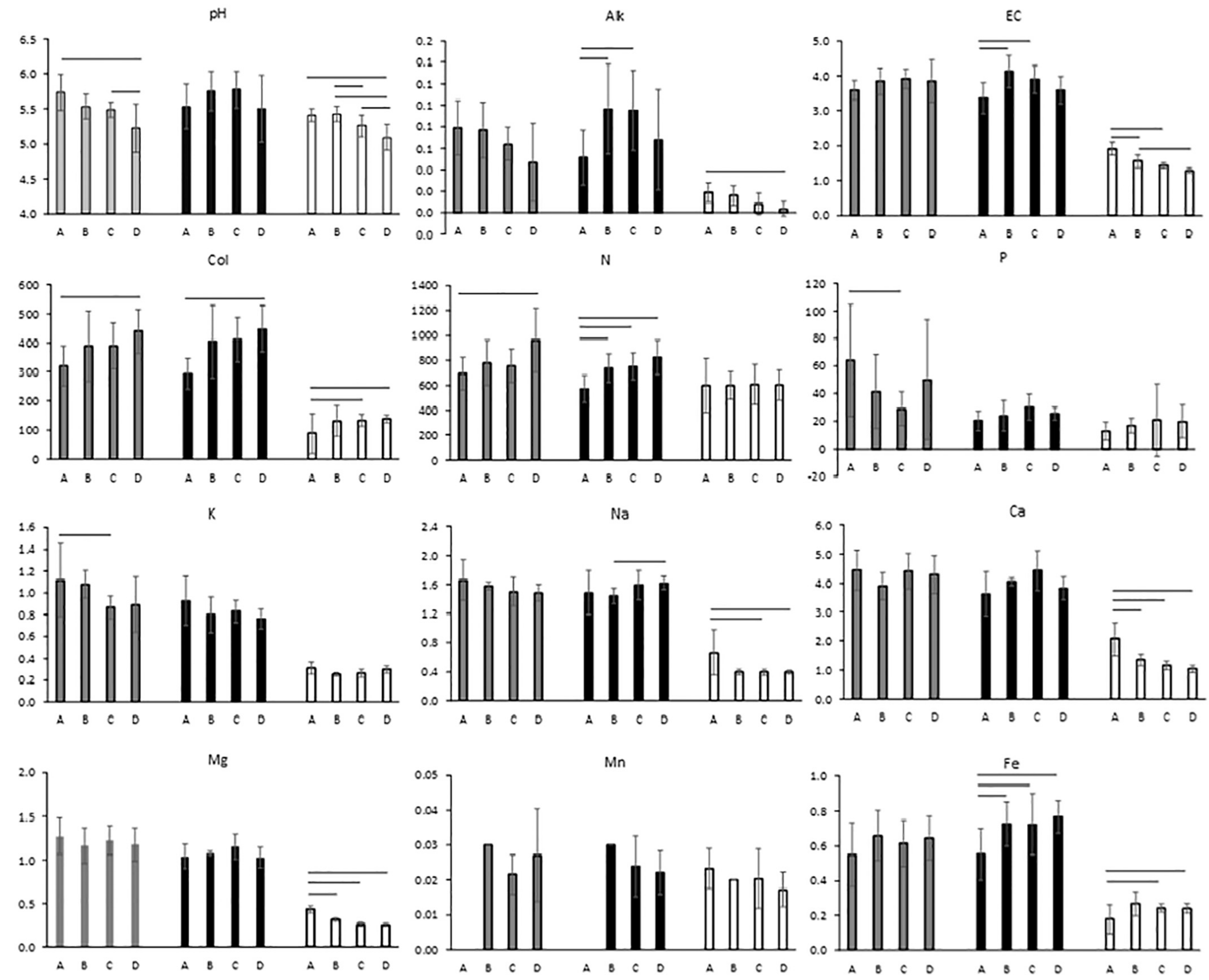

Fig. 3. The average (±SD) chemistry values of three lakes with longest data records. Grey columns indicate lake Nimetön, black columns lake Horkkajärvi, and white columns lake Alinen Mustajärvi. A = 1979–1989 (for pH, Alk, EC, Col, N and P $n = 8$, for the rest n = 5); B = 1990–1999 (for pH, Alk, EC, Col, N, P and Fe $n = 10$, except for cations n = 3); C = 2000–2009 (n = 10), and D = 2010–2019 (n = 10). n = number of annual data records. The line above two column indicates statistical significance (One-way ANOVA, $p < 0.05$). The endpoints of the horizontal lines indicate the columns in question.

where Y is the response variable, αLakeType is the intercept for each type of lake, s(Year*LakeType) is a smoother of Year with an interaction with LakeType with a thin plate regression spline, s(Lake) is a random effect to control spatial correlation, and ε is the error. We fitted the same formula for the GLMMs, without including a smoother.

Fe and discharge were the only variables which explained the long-term variation of Col both in SW and GW lakes (Table 4), while the impact of acid deposition was significant only in SW lakes. In the main lake-chain, the highest Fe concentrations existed in Rahtijärvi followed by the next two downstream lakes. In other lakes of the lake-chain, Fe concentrations were 20 % lower, and did not indicate similar decrease along the lake-chain as was found in Col.

In the longest data sets since 1979, the correlation coefficients between Fe and Col varied between 0.64 and 0.70, being highest in Horkkajärvi and lowest in Nimetön and Alinen Mustajärvi. A strong relationship between Fe and Col ($r = 0.68$) was also found in Valkea-Kotinen with an extensive data set over the ice-free seasons since 1990. During the wet seasons Fe concentrations and Col values were 40–55 % and 38–46 % higher in that lake than during the dry seasons.

In GW lakes, Fe concentrations did not show any clear trend over the time (Fig. 4), although in two of them the concentrations slightly increased (Tables S2, S3). In SW lakes, the increase in Fe concentrations was generally stronger in 1990s than in this century (see Fig. 4).

The ratio between the mean values of Col and Fe (Col/Fe; Table S3) was higher in 26 lakes during this century than in the 1990s, and the ratio had a strong dependence on water acidity. Since 1990 the correlation coefficient (r) between Col/Fe and pH was 0.83 for all lakes, and if we dropped out one outlier (Vähä Valkjärvi with an exceptional high Fe concentration relative to Col), the lakes followed closely a common line ($r = 0.91$, Fe = −358.53*pH + 2475.2).

3.2. Ca, Mg, K, EC, Alk, pH and nutrients

During the last three decades, Ca and Mg concentrations have decreased ($p < 0.05$) in 20 lakes, EC in 28 lakes, and pH and Alk in respectively 17 and 18 lakes (Table 2; Table S1; Fig. 2B; Fig. S4). Since 2000, the number of statistically significant cation, EC, Alk, and pH trends was almost the same for each variable as in the longer data set, except for EC because the number of lakes with a significant decrease in EC was 17.

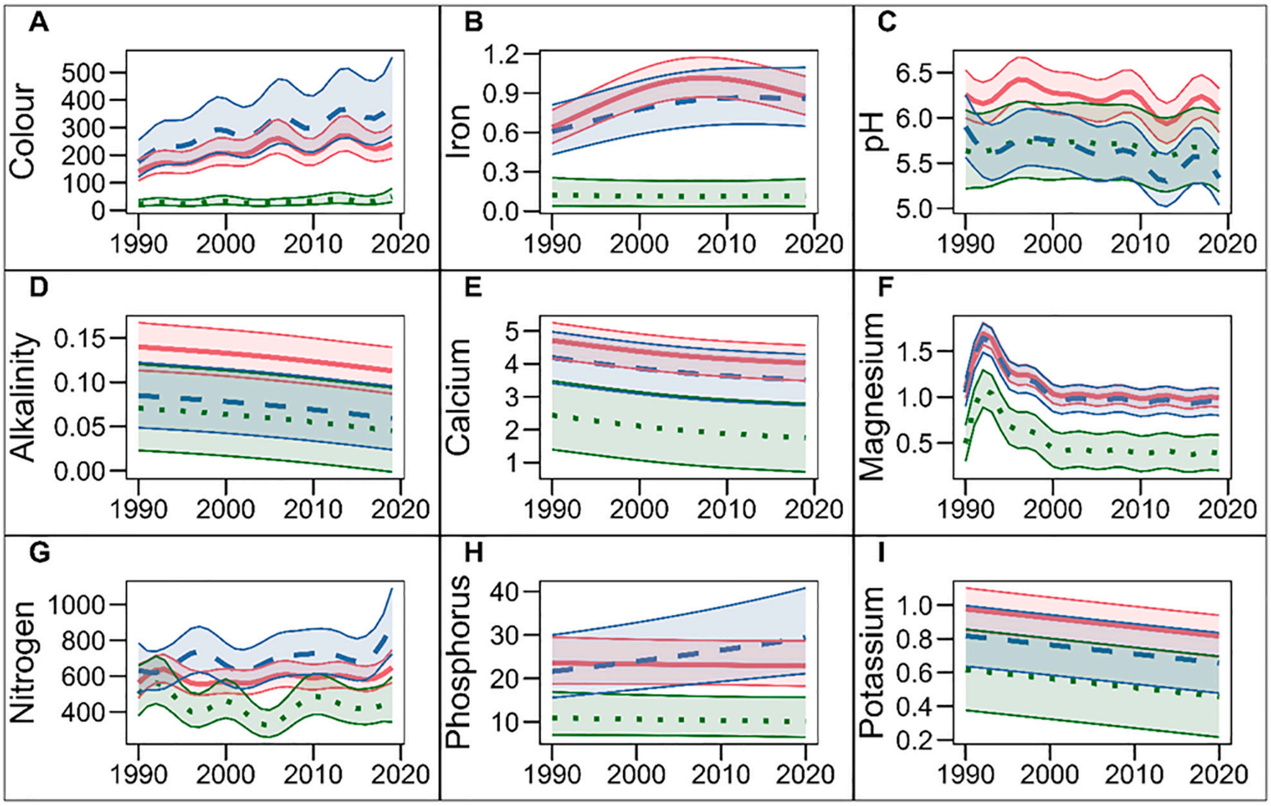

Fig. 4. General additive model (GAM) predictions for water colour (Col, mg Pt L^{-1}), iron (mg L^{-1}), pH (pH-unit), alkalinity (mmol L^{-1}), calcium (mg L^{-1}), magnesium (mg L^{-1}), total nitrogen (μg L^{-1}), total phosphorus (μg L^{-1}), and potassium (mg L^{-1}) of the three lake categories. Blue lines indicate HSW lakes, red lines DSW lakes, and green lines GW lakes. The coloured areas show deviation of the variables (95 % confidence interval).

Fe was the only cation for which concentrations increased in a considerable number (39 %) of the lakes. The statistically significant chemistry trends were slightly more abundant during the three decades long data set than during this century (Table 2). Since 2000, Ca, Mg, K, EC, and pH were the variables with the highest number of significant decreasing trends, and three lakes a decreasing Fe trend was also found.

GW lakes had, on average, lower pH, Alk, EC, and cation concentrations in comparison to the SW lakes (Fig. 3; Table S3). Between the GW lakes and SW lakes, the most evident difference was found in Fe concentrations, however. Ca, Mg and Na concentrations were also lower in GW lakes in comparison to HSW lakes, where Ca and Mg correlated significantly with alkalinity in nearly half of the lakes.

When June–October precipitation was >411 mm, the average Ca and Mg concentrations were 1.05 and 1.03 times to those with low precipitation (<245 mm). All significant K trends were decreasing over the whole study, and no difference between the lake types was found (Table 2; Table S1). Significant Na trends were fewer, and an equal number of increasing and decreasing trends was found.

The MK trend results did not reveal any uniform change over time in the concentration of total N and total P, the two major nutrients. Regarding N, only six lakes had significant trend, four an increasing trend and two a decreasing trend. Since 1990, 10 lakes had a significant trend in P concentration, five an increase and five a decrease. During this century, however, five decreasing trends appeared and one increasing trend, and all of those were in SW lakes.

The model results did not indicate any statistically significant difference in Ca, Mg, K, N and P concentrations between the HSW and DSW lakes, while pH and Alk were different between the two lake categories (Table 3). However, Ca, Mg, K, N K concentrations were different between the DSW and GW lakes, and P concentrations increased over time only in HSW lakes (Fig. 4). By contrast, EC values did not differ between the three lake categories due to the large variation within each lake group, but the values have a decreasing trend. For example, all GW lakes tend to have low EC except Syrjänalunen with the highest EC among the 33 lakes.

3.3. Lake-specific trends

In two HSW lakes, Mekkojärvi and Valkea-Kotinen, as well as in Hautajärvi, Col increase was most steady when the periods since 1990 and since 2000 were taken into consideration (see Table A2). In GW lakes Valkea Mustajärvi and Vähä Valkjärvi, an increasing Col trend was found during both periods, while in DSW lakes from Sorsajärvi to Keskinen Rautjärvi, Col trends increased significantly only when the period since 1990 was considered, and in the lowermost DSW chain-lake, Alinen Rautjärvi, there was no trend. Along the lake-chain downstream from Savijärvi, Col values decreased by ca. 35 %, and the lowest values were found in the three lowermost lakes, Ylinen Rautjärvi, Keskinen Rautjärvi and Alinen Rautjärvi (Table 2; Fig. S4).

Since 1979 Col values have increased by 45–60 % in two HSW lakes (Nimetön and Horkkajärvi) and by 39 % in the GW lake Alinen Mustajärvi. In all three, the Col values varied substantially between different years and time periods, however, being lowest in the 1980s (Fig. 3).

Among the other chemistry variables, Ca and Mg concentrations increased in the lake chain from Haarajärvi to Rahtijärvi, after which their concentrations declined towards Alinen Rautjärvi. A similar change along the lake-chain was found in pH, Alk, EC and P, while K and Na increased from the topmost lake to the lowest one by >30 %.

Relative to the other headwater lakes, Karhujärvi was exceptional due to its low pH and Alk, both consistent with its low base cation concentrations. K and Fe concentrations were also lower there than in any other HSW lakes (Table S2). Regarding the studied lakes, Syrjänalunen differed from all others due to its high cation concentrations, high Alk, high EC and high pH, but low Col and Fe concentrations

Table 3

Summary of the General additive model (GAM) (A) and Generalized Linear Models (GLMM) (B) results, revealing differences between the lake groups.

A. GAM					
Response variable	Family	Variables	*p*	$R_{sq(adj.)}$	Deviance expl. (%)
log10(Col+1)	Gaussian (identity)	**Smooth terms**		0.891	89.9
		s(Year)			
		HSW	<0.001		
		DSW	<0.001		
		GW	<0.001		
		s(Lake)	<0.001		
		Parametric terms			
		Lake type			
		HSW-DSW	0.495		
		HSW-GW	<0.001		
		DSW-GW	<0.001		
Ca	Gamma (identity)	**Smooth terms**		0.777	84.0
		s(Year)	<0.001		
		HSW			
		DSW			
		GW			
		s(Lake)	<0.001		
		Parametric terms			
		Lake type			
		HSW-DSW	0.533		
		HSW-GW	0.022		
		DSW-GW	<0.001		
Mg	Gamma (identity)	**Smooth terms**		0.662	83.6
		s(Year)	<0.001		
		HSW			
		DSW			
		GW			
		s(Lake)	<0.001		
		Parametric terms			
		Lake type			
		HSW-DSW	0.861		
		HSW-GW	<0.001		
		DSW-GW	<0.001		
log10(pH + 1)	Gaussian (identity)	**Smooth terms**		0.819	83.0
		s(Year)			
		HSW	<0.001		
		DSW	<0.001		
		GW	0.696		
		s(Lake)	<0.001		
		Parametric terms			
		Lake type			
		HSW-DSW	0.008		
		HSW-GW	0.890		
		DSW-GW	0.140		
Sqrt(Fe)	Gaussian (identity)	**Smooth terms**		0.820	82.7
		s(Year)			
		HSW	<0.001		
		DSW	<0.001		
		GW	0.732		
		s(Lake)	<0.001		
		Parametric terms			
		Lake type			
		HSW-DSW	0.404		
		HSW-GW	<0.001		
		DSW-GW	<0.001		
log10(Alk+1)	Gaussian (identity)	**Smooth terms**		0.773	78.1
		s(Year)	<0.001		
		HSW			
		DSW			
		GW			
		s(Lake)	<0.001		
		Parametric terms			
		Lake type			
		HSW-DSW	0.047		
		HSW-GW	0.888		
		DSW-GW	0.041		
N	Gamma(log)	**Smooth terms**		0.465	54.7
		s(Year)	<0.001		
		HSW	0.005		
		DSW	0.118		
		GW	<0.001		
		s(Lake)	<0.001		
		Parametric terms			

(*continued on next page*)

Table 3 (*continued*)

A. GAM

Response variable	Family	Variables	p	$R_{sq(adj.)}$	Deviance expl. (%)
		Lake type			
		HSW-DSW	0.352		
		HSW-GW	<0.001		
		DSW-GW	<0.001		

B.

Response variable	Family		Variance	SD
log10(P)	Gamma(log) GLMM	**Random effects**		
		Lake (Intercept)	0.0428	0.2071
		Residual	0.0363	0.1906
		Number of obs.	973	
		Number of lakes	33	
		Fixed effects	t value	Pr(>t)
		Intercept	28.338	<0.001
		scale(Year)	−0.441	0.659
		LakeTypeGW	−3.269	0.003
		LaketypeHSW	0.404	0.689
		Scale(Year):LakeTypeGW	−0.346	0.730
		Scale(Year):LakeTypeHSW	2.934	0.003
		Contrast	t.ratio	*p*
		DSW - GW	3.269	0.007
		HSW - DSW	−0.404	−0.914
		GW - HSW	−3.239	0.008
K	REML	**Random effects**		
		Lake (Intercept)	0.074	0.2719
		Residual	0.0166	0.129
		Number of obs.	752	
		Number of lakes	33	
		Fixed effects	t value	Pr(>t)
		Intercept	8.689	<0.001
		Year	−8.04	<0.001
		LakeTypeGW	−2.619	0.014
		LaketypeHSW	−1.438	0.161
		Contrast	t.ratio	*p*
		DSW - GW	2.619	0.036
		HSW - DSW	1.438	0.335
		GW - HSW	−1.317	0.397

Table 4

Generalized linear mixed effects model (GLMM) results showing the drivers/variables explaining best the variation of water colour in 1990–2019. Because the General additive model (GAM) results did not indicate any significant difference of Col between the HSW and DSW lakes, we grouped them together for the analysis.

	Model type	N obs & N lake	Response variable	Model results			
SW	LMM	N obs = 714	Sqrt(Col)	**Fixed effects**	**Estimate**	**SE**	***p***
		N lake = 27		Intercept	15.755	0.489	<0.001
				Q	0.721	0.072	<0.001
				T	0.158	0.076	0.038
				Fe	1.267	0.102	<0.001
				pH	−1.233	0.123	<0.001
				SO_4 + NOx	−0.430	0.079	<0.001
				Random effects	**Variance**	**SD**	
				Lake	6.344	2.519	
				$\mathbf{R^2_{LMM}}$	**m**	**c**	
					0.330	0.824	
GW	LMM	N obs = 132	Sqrt(Col)	**Fixed effects**	**Estimate**	**SE**	***p***
		N lake = 5		Intercept	6.197	1.094	0.005
				Q	0.210	0.088	0.019
				T	0.208	0.107	0.054
				Fe	0.750	0.132	<0.001
				pH	−0.224	0.292	0.445
				SO_4 + NOx	−0.048	0.106	0.650
				Random effects	**Variance**	**SD**	
				Lake	5.954	2.44	
				$\mathbf{R^2_{LMM}}$	**m**	**c**	
					0.091	0.881	

(Table S2).

When average values of the chemistry variables (see Table S2) were compared between the lake groups, none of the variables differed ($p > 0.05$) between the lakes situated higher (>150 m a.s.l.) and lower (<130 m a.s.l., excluding the lake-chain from Haarajärvi to Alinen Rautjärvi) in the landscape. However, in the seven uppermost lakes the average (and median) Col values were 1.7 (1.6), P 1.3 (2.0) and Mn 3.2 (1.1) times higher than in lowermost seven lakes. In contrast, Alk and Fe were 2.2 (1.5) and 1.2 (1.7) times higher in the lowermost lakes in comparison to the uppermost ones. Alk values declined by 69 % in the lakes highest in the drainage basin (catchments >150 m a.s.l.) when average Alk values from last century (1990–2000) were compared with those of the last ten years (2010–2019), and a rather similar decrease (45 %) was found in the lakes lowest in the drainage basin. In contrast, Col values increased by 90 % and 83 %, respectively. Changes were, however, lake-specific rather than constant, and not always statistically significant.

3.4. *Differences between lake categories*

The values of six variables (Col, Mg, Mn, Fe, N, P) differed significantly between SW and GW lakes, while only two variables (pH, Alk) were different between HSW and DSW lakes (Table 3; Table S4). The variation of the chemistry variables within the lake categories was rather large (see also Fig. 4), however.

Until the end of the 1990s the chemistry trends of the lakes showed a rather consistent pattern, with an increase in Col and a decrease in cation concentrations, Alk and EC. In 21st century, however, Col trends have varied substantially within the lake groups. In contrast, cation concentrations, Alk, pH, and EC continued to decrease in most lakes regardless of lake category (Tables 2, 3; Fig. 4).

4. Discussion

4.1. *Browning*

The results clearly demonstrated high colour variability among the lakes and over the study period, with distinctly lower values in the GW lakes than in SW lakes. In the SW lake category, most lakes had an increasing colour trend over the years, and at the end of last century colour values increased parallel with a rapid decrease in acid deposition, in agreement with several other studies (de Wit et al., 2021; Räike et al., 2024). Since the beginning of 21st century, Col values have not been increased as uniformly among the SW lakes as before, indicating that catchment and lake specific properties may have become more important in determining colour. The upward trend is still visible especially in many HSW lakes, however.

Earlier, Eklöf et al. (2021) and Bragée et al. (2015) have concluded that the effects of S deposition have already subsided in Sweden, and according to Arvola et al. (2017) TOC concentrations in Finnish surface waters have already reached the pre-industrial level. Later changes in Col may have been predominantly impacted by other factors influencing organic matter loading (see Schelker et al., 2012; de Wit et al., 2016; Finér et al., 2021), such as climatic and hydrological conditions, vegetation, and various forestry practices common in the study area. In our data set, the impact of temperature on Col remained uncertain, however, despite its statistical significance (cf. Räike et al., 2024). The effect of temperature may be difficult to find out because it has changed gradually over the last decades (Ruoho-Airola et al., 2014), and it may therefore influence vegetation and soil properties in the long-term with a delay in response rather than in the short-term.

4.2. *Iron*

Our results support previous observations that there is a close linkage between Fe and dissolved organic matter (DOM), as well as Fe and Col, in boreal lakes (Jones et al., 1988; Schindler et al., 1992; Heikkinen et al., 2022) and rivers (Kritzberg and Ekström, 2012). Fe and Col correlated significantly in 26 out of the studied 33 lakes, and in Valkea-Kotinen, Nimetön, Horkkajärvi and Alinen Mustajärvi the relationship between iron and colour was noticeably stronger than in Swedish rivers (Kritzberg and Ekström, 2012).

The results did not clearly indicate, however, that the contribution of Fe to the water Col was vitally important, although ferric iron has a very pale violet or brown colour, and therefore also impact on Col of a water body (e.g. Riise et al., 2023). Rather it seemed that the impact of dissolved humic substances become stronger as the higher Col/Fe ratios in this century suggested. The ratio showed a strong dependence on water pH. Humic substances may have masked the impact of Fe in the most stained lakes parallel with two nearby rivers and a medium-size humic lake (Tulonen et al., 1992; Huotari et al., 2013; Arvola et al., 2017).

In principle, one might expect that in GW lakes with distinctly lower OM concentrations and colour values than in SW lakes, Fe could have more relevance in determining water colour, although its role may be difficult to specify as our GLMM results indicate. According to the model, only <10 % of the variation of Col could be explained if the random effects were excluded while 88 % was explained if we added the lake variability. In SW lakes, the corresponding numbers were 33 % and 82 %, respectively, showing that a third of the variance of Col was explained by the fixed effects of the model. Even in acid sandy soils OM and Fe transport by the hydrological pathway are closely integrated (Moens et al., 2022), suggesting that they are shaping water colour parallel. All these results imply that properties of catchments and in-lake processes are crucially important.

4.3. *Base cations, acidity, and alkalinity*

According to Lydersen (1997), organic acids modify both the acidity of surface waters and their response to changes in strong acid inputs when low or no bicarbonate alkalinity exists, a situation which appeared in many of the Evo lakes, where an inverse relationship between Alk, pH and Col existed. However, this was slightly unexpected because organic matter decomposition and anoxia may generate alkalinity in acid waters by the reduction of nitrate and sulphate, and production of iron(II) and ammonium, and consequent release of calcium (Davison and Woof, 1990). Our results suggest that in lakes with moderate or high Col concentration, CO_2 production through organic matter mineralization and organic acids regulated water acidity. In the most acidic HSW lakes, especially in Karhujärvi, strong acids may have influenced as well. To what extent the impact of acid deposition on soil and surface water was mitigated by the deposition of base cations (see Ruoho-Airola et al., 2014) and does the possible release of calcium into a lake system, affect alkalinity, remains unsolved, however.

GW lakes with small lake area relative to their catchment (LA:CA) are more sensitive than SW lakes to direct deposition on the lake surface. In most studied lakes, direct deposition on the lake surface had only a minor effect on water chemistry, as the example from Valkea-Kotinen indicates, where Ca and Mg direct depositions were estimated to contribute only 2–3 % of their amount in the uppermost 1 m water layer, and even less (1–1.5 %) in the total volume of the lake. In SW lakes, the element depositions to the catchments are usually several times higher than direct deposition to the lake surface (see Table 1), suggesting the importance of the nearby catchment. GW lakes, in turn, are located on sandy soil with lower buffering capacity, which makes them vulnerable to the effects of acid and base cation deposition (Salminen et al., 2004; Hazlett et al., 2020, and references therein). Sandy soils are not able to retain nutrients efficiently, and a high drainage rate may also reduce its buffering capacity.

In summary, the results showed that Ca, Mg and K concentrations decreased during the last three decades in line with the reduction in acid and cation deposition, and a decrease of alkalinity of the lakes.

4.4. Hydrology and landscape

In Finland, forest soils are predominantly made by podzols (Tamminen and Tomppo, 2008), which are also widely distributed over the study area (Table S1). The topmost layer of podzol is usually rich in organic matter (humus), while many elements such as aluminium and Fe move gradually deeper in the soil profile. Water percolation with vertical and horizontal transport of elements finally transmits part of them to the lakes. We propose that surface water transport of chemical constituents together with underground hydrological processes explained the observed higher element concentrations during and after wet seasons, when saturated soil moisture conditions interconnect lakes closely with their surroundings (Strock et al., 2016). In the study area, organic soils cover significantly smaller area than podzol soils and sand plus gravel soils (Table S1).

In the lake-chain downstream from Rahtijärvi, concentrations of several elements declined, presumably due to in-lake processes such as sedimentation when the residence time became longer, and due to dilution by groundwater. In addition, DOM degradation by microbes (Tulonen et al., 1992) and UV radiation (Vähätalo and Wetzel, 2004), as well as sedimentation (Curtis, 1998) may have decreased Col values along the hydrological pathway. The decrease in Col was in line with the results of Rasmussen et al. (1989), who noted that large and deep lakes with significant indirect drainage from other lakes were less coloured than headwater lakes.

Although the quantity of groundwater remains open in GW lakes, a small catchment area relative to the lake area suggests that precipitation dilutes water chemistry especially in the seepage lakes. Allochthonous OM and element loadings are substantially lower there than in SW lakes. Ca and Mg trends were surprisingly similar in all lakes, although their concentrations were substantially lower in GW lakes compared to SW lakes, except in Syrjänalunen, a spring lake, thus implying that drivers behind the trends were common irrespective of the lake type. In such a small study area, the lakes and catchments share, in principle, identical deposition chemistry and climate, whereas soil texture and catchment disturbances are lake specific.

Although for many variables GAM analyses did not show any difference between HSW and DSW lakes, it is obvious that lake's position in the landscape does matter which became evident when the uppermost lakes in the landscape were compared to the lakes lower in the system. Because the latter receive water through several lakes and catchments, their water chemistry is influenced more by processes higher in the drainage basin, including lakes therein, rather than in a lake itself or in its neighbouring catchment. Hydrologically interconnected lakes, common in Finland, often have similar patterns in chemistry (Kankaala et al., 2023). Therefore, it was not a surprise that in our data set discharge was a key driver explaining the variation of Col in SW lakes.

Due to the few GW lakes, we must be careful with their data interpretation and especially when make conclusions and generalizations based on the results.

4.5. Beavers and forestry

Among the studied lakes, only six lakes (Syrjänalunen, Iso Valkjärvi, Vähä Valkjärvi, Alinen Mustajärvi, Karhujärvi, Iso Mustajärvi) did not have any indication of beaver occupation, and in four lakes (Valkea Mustajärvi, Horkkajärvi, Ylinen Rautjärvi, Keskinen Rautjärvi) they were not able to regulate water level by damming. This means that beavers had an opportunity to influence hydrological and chemical properties as well as the biota of 23 lakes by regulating their water table (Vehkaoja et al., 2015; Kivinen et al., 2020; Nummi et al., 2021). Their effects were often difficult to evaluate, however, due to their intermittent presence at a lake. Another difficulty is that the effects on water quality can have time lags and may depend strongly on the length and intensity of the occupation, morphometry of a lake, and the surrounding catchment.

The results of Vehkaoja et al. (2015) showed that during the first three beaver-impoundment years, Col values increased with a simultaneous decrease in dissolved oxygen (DO) concentrations, after which both returned to the background level within 4–6 years of beaverś disappearance. Poor DO conditions caused by beavers may have influenced Fe concentrations in several lakes and explain, at least partly, why Fe trends developed in those lakes parallel with Col (see also Kazanjian et al. 2021). Lakes surrounded by a *Sphagnum* margin, common in the study area, are especially vulnerable to the effects of elevated watertable in riparian areas on water quality (Vehkaoja et al., 2015).

Lakes impounded/influenced by beavers did not show any common features except, that beavers did not occupy any of the GW lakes. Beaver lakes tended to show high variability in Col, such as in lakes Huhmari, Valkjärvi, Särkijärvi and Mekkojärvi, which were among the five lakes with highest coefficient of variation (CV > 0.45) in Col. Only Vähä Valkjärvi, a GW lake heavily impacted by forestry, had higher Col variation. During the study period, in nine lakes Col values exceeded, at least once, 500 mg Pt L^{-1} and in eight of those beavers were present. In addition, the highest Col value ($>$800 mg Pt L^{-1}) also coincided with a beaver flood. Consistent with the above, Col increase in Valkea-Kotinen during the last decade could be a result of beaver occupation, which was rather persistent unlike during the previous years. Unfortunately, the beaver observations don't allow us to run any further analyses.

Forest management, including clear-cutting, soil scarification and artificial regeneration, pre-commercial and commercial thinning, and ditching of peatlands, have also caused disturbance of soil, vegetation, and hydrology in the study area (Rask et al., 1993; Lindberg et al., 2020). Although intense forestry has continued for over a century, forestry practices have never covered the entire catchment of any lake. Because various forestry treatments, also including prescribed burning and ditching, have been carried out at different times, and sometimes have been repeated at varying spatial scales and intensity, their impact is difficult to distinguish from other drivers. Only two catchments and lakes (Karhujärvi and Valkea-Kotinen) have avoided any forestry treatments at least during the last fifty years.

The most evident forestry effects were found in Vähä Valkjärvi, where logging practices took place at the beginning of the 2010s. As a result, Col and Fe concentrations were nearly three times higher after the treatments, and no return to the pre-treatment concentrations was found before the end of this study. Water quality effects in Vähä Valkjärvi were very similar to those in two other GW lakes (Iso Valkjärvi, Valkea Mustajärvi; unpublished data, Lammi Biological Station, University of Helsinki) in the 1980s, when intensity of Col also increased after logging.

Schelker et al. (2012) have noticed up to 35 % higher DOC concentrations in headwater streams impacted by clear-cutting and site preparation than in the control, and riverine carbon fluxes increased by 100 % and 79 %, respectively. Harvesting affects groundwater level and consequently water quality by increasing DOC and mobile ion concentrations (Laudon et al., 2009; Kreutzweiser et al., 2008). In one HSW lake, Nimetön, catchment deforestation and burning of logging remains in the early 1980s resulted in an increase in pH, Alk, and total N and total P, followed by an increase in algal primary production and decomposition of organic matter (Rask et al., 1993). However, the fertilization effect lasted $<$5 years, which was in line with the observation of Carignan et al. (2000).

Topsoil disturbance together with climate change related issues such as warmer winters, higher rainfalls, and lower soil frost (Venäläinen et al., 2001; Finér et al., 2021), may accelerate decomposition of organic matter (Schelker et al., 2012) and transport of DOC to the lakes. Thus, even though the impacts of forestry and prescribed burning can be distinguished, the effects gradually fade. In eastern Canadian Shield lakes, Winkler et al. (2009) found that natural variability, due to various reasons, is more important in determining the ecological patterns of lakes than short-term impacts of forest harvesting. In comparison to many other areas in Finland, however, forestry has been less intense in this century in the Evo area, because a major part of it belongs to the Natura 2000 network covering Europe's most valuable and threatened habitats.

4.6. Concluding remarks

The results showed that colour values increased consistently in the 1990s in synchrony with a decline in atmospheric deposition. Later Col changes among the lakes were less coherent, however, indicating that the effects of acid deposition have subsided, and other localised drivers became more important. An increase in organic acids apparently underpinned the decrease in pH while organic matter decomposition and anoxic conditions did not generate alkalinity and release of calcium enough to compensate their decline.

It is obvious that hydrological conditions impacted on both Col and Fe. Low Alk coherence within the lake categories was a surprise, implying that regardless of common trends the chemical properties of individual lakes can vary considerably. Hypolimnetic anoxia in some lakes and differences in autumnal mixing patterns among the lakes are factors which may cause lake-to-lake variation in alkalinity. High Col coherence, in turn, suggested that the transport of organic matter from the drainage basin was spatially rather synchronized. The warmer climate and possible increase in precipitation in future may change the hydrology and biogeochemistry of the catchments with major consequences of the lakes. Long-term chemistry data sets therefore are important regarding lake management and sustainable forestry, an issue especially important in Finland, a country with most intense forestry.

Regarding the original four questions addressed by the study, the results demonstrated a consistent increase in Col irrespective of the landscape and hydrological setting in 1990s along decreasing acid deposition, while later Col trends varied more among the lakes (Q1). Col values of SW lakes were also influenced by other factors such as hydrological conditions, Fe concentrations, and air temperature. Cation concentrations decreased over the last three decades as was suggested (Q2), and in agreement with that Alk, EC and pH decreased in all three lake categories (Q3). The results suggest that the studied lakes are still recovering from acidification although the impact of multiple forestry operations, and occurrence of beavers, must be considered as potentially important factors as well (Q4). In addition, the results implied that besides the meteorological drivers, other disturbances such as possible beaver occupation and forestry operations may impact in long-term on the lakes, in particularly the small ones.

CRediT authorship contribution statement

Lauri Arvola: Writing – review & editing, Writing – original draft, Visualization, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Martti Rask:** Writing – review & editing, Writing – original draft, Validation, Resources, Project administration, Methodology, Investigation, Data curation, Conceptualization. **Jussi Huotari:** Writing – review & editing, Writing – original draft, Visualization, Validation, Formal analysis. **Tiina Tulonen:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Data curation. **Kimmo K. Kahilainen:** Writing – review & editing, Validation. **Jukka Ruuhijärvi:** Writing – review & editing, Resources, Project administration, Investigation, Data curation, Conceptualization. **Henrik Lindberg:** Writing – review & editing, Resources, Methodology. **Risto Viitala:** Writing – review & editing, Resources, Methodology. **Clarisse Blanchet:** Writing – review & editing, Methodology, Formal analysis. **Celine Arzel:** Writing – review & editing, Methodology. **Petri Nummi:** Writing – review & editing, Resources, Data curation. **Kalevi Salonen:** Writing – review & editing, Supervision, Resources, Methodology, Investigation, Data curation, Conceptualization.

Declaration of competing interest

We confirm that the authors do not have any potential competing interests regarding the manuscript STOTEN 178696 (Article reference: STOTEN_STOTEN-D-24-17801). On behalf of all authors (Lauri Arvola, Martti Rask, Jussi Huotari, Tiina Tulonen, Kimmo K. Kahilainen, Jukka Ruuhijärvi, Henrik Lindberg, Risto Viitala, Clarisse Blanchet, Celine Arzel, Petri Nummi, Kalevi Salonen).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2025.178696.

Data availability

Data will be made available on request.

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