



# **P-cycling in sub-Arctic lakes** – the importance of sediment release and retention

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*P-cykler i sub-Arktiska sjöar – betydelsen av hur sedimentens sammansättning påverkar mobilisering- och retentionspotential*

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Water chemistry

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## Popular science summary

Lakes are important freshwater resources, hubs for life and sources of numerous ecosystem services. Even though lakes might seem to be isolated bodies of water their biological, physiological, and chemical properties are directly associated to the access of particles and nutrients within each individual watershed.

The nutrient which is a limiting factor to lake productivity is phosphorus (P). A negative P concentration within a waterbody will make it less productive; it is said to be subjected to oligotrophication. In an over-productive ecosystem this is usually not a problem, but it can become a severe issue in lakes within alpine terrain and the Arctic, where the nutrient availability is already limited. Oligotrophication in these ecosystems will further constrain diversity and the abundance of species populations, with the available nutrient and food supply altered. The main subject of the study was to see if the decreasing P is due to an increased P retention within the top sediment layers.

In order to understand the processes responsible for oligotrophication within the sub-Arctic areas of Sweden, two different lake populations were selected for comparison. The study was conducted by analysing water P concentration and sediment P concentration in seven sub-Arctic and eight lowland lakes.

The study shows that there is ongoing oligotrophication within the average sub-Arctic lake. Surprisingly the decreasing water P concentration did not correspond to the potentially releasable P retained in the top sediment layers. In which case it should have increased in the topmost sediment layer, if the potentially releasable P was responsible for the oligotrophication. On average, sub-Arctic lakes have a slightly elevated concentration of P bound to organic material in the top 4 cm sediment. This suggests a possible increased lake productivity together with a changed composition in the runoff water, possibly caused by a changed vegetation within the watersheds. Additionally, the study shows a high to medium risk of potential sediment P release within the sub-Arctic area. This could instead lead to increased P concentrations in the waters under conditions of low or no oxygen availability, potentially causing higher productivity than normal conditions in these lakes.

Documented changes in sub-Arctic ecosystems (greening, temperature, shift in microbial community), recognised to be caused by climate change, are believed to have a fundamental impact on the environment due to the effect they have on P concentrations within the watershed soil. It is therefore likely that these changes will impact the P fluxes within sub-Arctic lakes. However further studies are needed to determine if the origin of this oligotrophication is found within the effects of climate change.

Estimations of the sub-Arctic ecosystem responses to the varying potential outcomes of climate change consequences are difficult. It is however safe to say that further changes to these ecosystems will have a decisive effect on abiotic and biotic conditions, and that alterations in P-cycling and availability are likely to redraw the sub-Arctic landscape as we know it.

## Abstract

Oligotrophication is generally caused by a decreased nutrient availability, usually due to phosphorous limitation. This is a phenomenon impacting the entire food web and at worst causing cascading alterations within an ecosystem. Lakes of the Northern Hemisphere have lately been documented to be subjected to oligotrophication (e.g., Arvola *et al.* 2011, Eimers *et al.* 2009, Huser *et al.* 2018). The decreasing P concentration trends have previously been coupled with e.g., vegetation and pH shifts within ecosystems as well as changes in biogeochemical processes due to climate change and global warming. In Sweden, the greatest declines have formerly been reported within the northern, alpine lakes that are already oligotrophic (Huser *et al.* 2018).

The aim of this study is to elaborate the hitherto hypothesised pressures causing the changed P-cycling within sub-Arctic lakes, expanding on the possibility of global warming being the fundamental cause. Additionally, potentially increased P retention within sediment metal complexes was investigated. This possibility was evaluated by coupling water chemistry (1996-2021) and sediment subjected to P sequential extraction analysis. Seven sub-Arctic lakes and eight lowland lakes were investigated regarding the pressures of global warming to the P retention and release cycles.

The sediment concentrations and water chemical analyse showed no indications of a temporary increased mobile (potentially releasable) sediment P pool, contrary to expectations. The water chemical analysis, on the other hand, adds further evidence to former studies, agreeing with the previously detected oligotrophication. The magnitude of the TP declines, however, seem to have weakened compared to results from prior studies of the same area. Nevertheless, the PO<sub>4</sub>-P concentrations have in the intermittent years further decreased to less than analytical detection limits, lending additional evidence to the ongoing oligotrophication of lakes within the Northern Hemisphere and alpine area specifically. A relationship between sediment organic P concentrations and TP concentration in lake surface water was detected and hypothesised to be caused by the documented shift in terrestrial ecosystems within these watersheds. The correlations of this phenomenon, however, need further, quantitative study.

*Keywords: Phosphorous, P-cycling, Sediment-water exchange, P-fractionation, Lake sediment, Arctic, Climate change*

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## Abbreviations

Al	Aluminium
Al <sub>x</sub> (OH) <sub>y</sub> /AlOH	Aluminium hydroxides (various chemical structures)
Ca	Calcium
DO	Dissolved oxygen
DOC	Dissolved organic carbon
Fe	Iron
FeOH	Ferric hydroxides
Mn	Manganese
Mob-P	Mobile Phosphorus
OM	Organic matter
Org-P	Organic Phosphorus
P	Phosphorus
PO <sub>4</sub>	Phosphate/Orthophosphate
SD	Standard deviation
SLU	Swedish University of Agricultural Sciences
SWEDAC	Sweden's national accreditation body
TOC	Total organic carbon
TP	Total phosphorus
TP-frac	Sum of phosphor released during fractionation (i.e., the sum of P bound in porewater, Fe/Mn, Al, OM and Ca)

# 1. Introduction

## 1.1. Conditions of a lake

Oligotrophication, i.e., the issue of becoming less productive or more nutrient poor, has only within the last decades entered the research field. The comparative low number of studies looking at oligotrophication as an issue is probably due to the opposite condition, eutrophication, having a solid history of causing extensive problems. The general issue of eutrophication has long been subjected to assessment, monitoring, and remediation; and the problems caused have elicited both national and international concern (e.g., EEA 2018). In Sweden, the highly debated eutrophic condition of the Baltic Sea is merely an extension of the large-scale eutrophication of terrestrial ecosystems and subsequent run-off. Nevertheless, any change in nutrient availability will affect an ecosystem if it is a fundamental increase or reduction of essential and/or limiting nutrients. Either case will, in the long run alter the biodiversity and ecosystem services (Lento *et al.* 2019; Stockner *et al.* 2020).

Throughout the journey water takes from land to sea, there is an assimilation of nutrients, chemicals, and organic matter. The nutrient status of each watershed will determine the concentration of particles and chemicals assembled. Thus, it is the collection of pauses in lakes, rivers and ponds that will contribute to the final nutrient status in the ultimate waterbody – usually the sea.

All through the landscape, lakes are conditioned by their catchment as well as their morphology, oxygen levels, lake physiology and biology. All these variables will determine the lake nutrient status (see e.g., the review by Schindler 2006 and papers cited therein; various trophic status indexes such as The EU Water Framework Directive<sup>1</sup>; Carpenter *et al.* 1985; Fee 1979; Kolada 2014; Ohle 1980; Parker & Schindler 2006). Consequently, not all lakes stand the same risk of changed nutrient status in either direction (i.e., more or less nutrient availability).

In-lake nutrient availability is often strongly associated with stratification period(s). The period varies between lakes, mainly depending on lake

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<sup>1</sup> Available at: [https://ec.europa.eu/environment/water/water-framework/index\\_en.html](https://ec.europa.eu/environment/water/water-framework/index_en.html)

morphology. The effect of morphology on water column stability can be estimated using the relationship between lake mean depth and square root of the lake area (Osgood index, Osgood 1988). This, together with the local climate and the change of air temperature will induce either stratification or mixing of the water column. Depending on temperature oscillations within a given region, the water will mix between the epilimnion and hypolimnion. This works as a nutrient equalization event between the strata. When stratified, the water column is divided by the thermocline, controlling the nutrient input within the water gradient. The thermocline is the area where the water temperature is in transition, resulting in a broad variation of lake dependent thermocline depth. The seasonal temperature determines if either the epilimnion or the hypolimnion is the warmer/colder water agent. As the timing and multiplicity of these events are directly influenced by temperature change, as e.g., the length of ice cover is a direct result of the temperature of the surface water (Woolway & Merchant 2019). Higher temperatures are generally recognized to result in longer and stronger stratification periods, which may lead to lower in-lake nutrient cycling between bottom and surface waters during stratification period.

## 1.2. Phosphorus (P) in water and sediment

Phosphorus (P) is a crucial determinant of eutrophication due to its role as an essential nutrient for all living organisms, and is often the limiting nutrient for primary production in lakes (Schindler 1977). As a limiting nutrient, an increase in P generally leads directly to an increase in lake trophic status. Apart from the above-mentioned physio-chemical conditions, lake trophic status is determined by the amount of P loading and the in-lake cycling of P.

Trophic status is determined by the nutrient availability, ultimately defined by the P concentration. This has led to a classification of water bodies into categories defined by the P concentration: eutrophic (nutrient rich), mesotrophic (moderately nutrient rich) and oligotrophic (nutrient poor).

Changes to P-cycling can cause key equilibria shifts due to physical, chemical, or biological changes within the catchment. By such, alterations of either can change the entire ecology of a lake (Carlson 1977; Jeppesen *et al.* 2003; Schindler 2006). As the cycling of P determines the primary production, it will in extension also be crucial to the food web of any given ecosystem (Stockner *et al.* 2020).

Both lowland and mountain lakes are susceptible to nutrient concentration changes, depending on various environmental pressures. However, mountain lakes are commonly oligotrophic, whereas lowland lakes tend to have higher concentrations of nutrients due to a higher rate of sedimentary deposits (Ohle 1980). Lowland lakes also tend to be closer to nutrient saturation and thus have lower resilience against eutrophication (e.g., Carey & Rydin 2011). Because a

nutrient poor, oligotrophic lake usually has abundant of P binding sites within the sediment (commonly working as a nutrient sink), a further diminishing of nutrient input is crucial to these ecosystems. A decrease in P will thus negatively affect the food web, ranging from reduced primary production to fish abundance (Stockner *et al.* 2020).

P originates from erosion of soil and is subjected to transportation by the means of soil, chemicals, and biomaterial transported in runoff. Such particles entering the lake ultimately reach the lake sediment. The relation of continuous settling sediment imposes that sediment P content can be much greater than that in the overlying water column (Wetzel 2001). The relationship between sediment physiology, chemistry and biology combined with the water column condition dictate the in-lake P-cycling. Because these factors control the release and retention of P in sediment, alterations in either condition allow for variations in release/retention rates and in-lake P-cycling. The exchange of P between sediment and water is thus a fundamental function of P-cycling within water bodies.

### 1.3. Spatial distribution and abiotic effects

The global, natural distribution of P originates from bedrock erosion, as it has no gaseous stage like nitrogen. Generally, weathering processes emerge from either chemical or physical eroding of bedrock surfaces. This process can be viewed as a perpetual cycle because even small soil particles, such as clays are subjected to weathering. Nevertheless, weathering is not exclusively responsible for soil-P interactions; surface sorption processes (adsorption/desorption) also contribute to nutrient transport. Surface sorption is a continuous process of catch and release where nutrients as well as chemicals, salts *etc.* interact jointly with soil surfaces. The difference in P affinity varies depending on the soil particles, e.g., variations of clay mineral (Gérard 2016). So, this process varies depending on surface charge and overall particle composition, but is also affected by the surrounding abiotic conditions (e.g., Gustafsson *et al.* 2012). As a result, the nature of the soil and the surrounding environment will determine the possible reactions.

These biogeochemical processes have a considerable impact on P-cycling and transport since weathering, as well as sorption processes, greatly influence the potential transport of P (Gérard 2016). This, in turn, influences the availability of P within an ecosystem. Consequently, soil particles transported to lake sediment, affected by local physio/chemical conditions within a catchment, will determine lake productivity along with in-lake characteristics mentioned above.

A model of P-cycling within a lake can be simplified as a continuous flow between the bulk of bioavailable P (e.g., phosphate ( $\text{PO}_4$ )) and the pools of particulate P, soluble organic P, P bound in inorganic sediments (i.e., P bound in

metal complexes (e.g., Al, Ca, Fe)), organic matter and lastly weathering of mineral and bedrock (see e.g., Horne & Goldman 1994). The inflow of P with run-off as well as the P originating from wind/areal deposits play a large role in the P-cycling of lakes (e.g., Huser *et al.* 2018; Tipping *et al.* 2014). However, measuring of such is difficult, hence the complexity of atmospheric P deposition is not further covered within this study.

Another, equally important factor relating to in-lake P-cycling is the concentration of dissolved oxygen (DO), as stratified eutrophic lakes often have very little or no oxygen in deeper waters (Schindler 2006). These conditions affect both sediment and benthic organisms. DO limitations also affect the release rate of P from the sediment as these conditions will result in the release of P bound to e.g., ferric hydroxide (FeOH) complexes (Mortimer 1942). On the other hand, elevated DO will lead to uptake of P by the sediment, assuming P binding sites on iron (Fe) and manganese (Mn) based minerals are available.

Sediment release of P, Fe and Mn increases as the redox potential decreases with declining DO availability (Mortimer 1942). That is, the reduction of complexes and FeOH enables the diffusion of  $\text{Fe}^{+2}$  ions and P from the sediment into the water column. Aluminium hydroxides ( $\text{Al}_x(\text{OH})_y$ ) play a prominent role in P release/retention as well. They, like Fe and Mn, are sensitive to high pH but not to redox conditions. As such, they are used as a remediation measure to reduce eutrophication (e.g., Huser & Pilgrim 2014). The natural availability of both AlOH and FeOH will consequently, under certain conditions, control P concentrations in the water column by either binding it or release it from the sediment (Cooke *et al.* 2006; Mortimer 1942). The variation of P release/retention mechanisms result in a variety of availability to the in-lake cycling of P. Sediment P forms are thus subdivided into pools depending on affinity and binding substrate (see pools in Chapter 3.2 P-fractionation).

As particles migrate, the soil composition and bioactivity of an individual catchment will impact the environment it surround. Lakes are subsequently referred to as being *the mirror image of their surroundings*. This parable gives an immediate and graphic understanding of the capability of a lake to record physical, chemical, and biological changes in the surrounding watershed. The decrease in P concentrations reported within lakes throughout the Northern Hemisphere ought to accordingly have an explanation within the watershed of each individual lake, or viewing the issue holistically – through global changes to the environment affecting lake watersheds.

## 1.4. A new normal?

Due to climate change and global warming in particular, the Arctic and circumpolar areas have been subjected to irreversible changes in hydrology and



biodiversity (Lento *et al.* 2019; IPCC 2022). These changes will, as stated in the latest IPCC report, continue to share the pace of the uninterrupted increase in temperature. With the Arctic and circumpolar areas warming at twice the rate of the global average, understanding the resulting changes in these regions is essential. Even using the lowest projection for temperature increase, the Arctic will be severely impacted.

Increasing water temperatures are a direct result of climate change and have been shown to increase sediment P release rate, followed by a long equilibrium setting time (Wu *et al.* 2014). Wu *et al.* (2014) found the greatest increase in release rate (6.2 times) with a temperature rise from 10 to 30°C. Psenner (1984) also found increased P release rates due to higher temperature, already at an increase from 4 to 8°C, however Psenner stressed that this release rate is secondary to that caused by anaerobic conditions.

There are already reports of ecosystem change within the Arctic, such as vegetation encroachment into tundra and more elevated areas. These events have been positively accredited global warming (Tape *et al.* 2006; Truong *et al.* 2007).

Ecosystem change within any watershed, such as above mentioned greening, will also impact the waterbodies within the area, where a shift in trophic status caused by changed nutrient availability can have extensive effects. Here, the continuous monitoring and remediation programs of eutrophication comes to new light, as lakes have been noted to show a persistent decrease in P concentrations in lakes and streams of the Northern Hemisphere (e.g., Arvola *et al.* 2011; Eimers *et al.* 2009; Hu and Huser 2014; Huser *et al.* 2018; Stammmler *et al.* 2017). This trend deviates from that of Low and High Arctic lakes, as both these regions show an increase in P concentration. The main factor in these regions causing increased P was permafrost thaw and sloughing of thawed soil (Hobbie *et al.* 1999; Huser *et al.* 2020).

Huser *et al.* (2018) reported oligotrophication throughout the Swedish Boreal lake population, i.e., a broad latitudinal investigation showing large-scale declining TP trends (1988-2013). They found the steepest declining concentration trends in the already most oligotrophic lakes in the sub-Arctic area. These findings are, put in relation to the delicate environments in the circumpolar area, troubling, as a further decrease of nutrients will likely result in negative consequences for lake productivity and ecosystem services. The study and research of changes within pristine areas are important to understand the effects of global warming within other areas as well. Especially as Scandinavia and the sub-Arctic are subjected to a more rapid climate change compared to the global average.

In this study, the aim is to further elaborate the hitherto hypothesised pressures seemingly altering P-cycling and availability in lakes, with a hope to induce greater understanding of the ongoing oligotrophication in sub-Arctic lakes. This

was done by exploring water chemical and sediment data from seven sub-Arctic and eight lowland lakes in order to evaluate P concentration changes in the sub-Arctic lakes in relation to their lowland counterparts.

The formerly hypothesised cause of this system change has been largely accredited to climate change and global warming, e.g., such as the impact from greening (e.g., Huser *et al.* 2018; Lento *et al.* 2019). Special interest is therefore given to the possibility of such factors contributing to the oligotrophication in the Arctic.

The study hypothesis includes an increase in mobile P (i.e., an increased P binding to Fe) in the sediment in sub-Arctic lakes, accounting for a (temporary) retention. Potentially explaining, in part, the formerly recognized oligotrophication in the sub-Arctic study lakes and possibly elsewhere. Study of in-lake water chemical data is expected to confirm the already documented oligotrophication of water bodies within this region. The author also hypothesises that there will be clear indications of the environmental pressures triggering these changes, e.g., the impact from increasing temperature and greening of the watershed.

## 2. Background

### 2.1. Lake index and the landscape gradient

This study focuses on a comparison of P-cycling in oligotrophic/mesotrophic Swedish, pristine sub-Arctic lakes located in an alpine landscape and those in forest, lowland landscapes (Table 2.1 & Figure 2.1).

*Table 2.1.1 Geographic, morphological, and hydrological data for both lake populations but also former documented TP trends (data from Huser et al. 2018). Notation n/a meaning not applicable*

Lake	Population	Coordinates [SWEREF 99 TM]	Altitude [m.a.s.l]*	Mean depth [m]	Surface area [km <sup>2</sup> ]	Catchment area [km <sup>2</sup> ]	Residence time [yrs.]	TP trend %/yr.
Bysjön <sup>2</sup>	lowland	N6576747 E348540	124	n/a	1.00	n/a	n/a	n/a
Fiolen <sup>3</sup>	lowland	N6327724 E471494	226	n/a	1.60	5.46	n/a	n/a
Gipsjön	lowland	N6724037 E424887	376	3.8	0.75	10.75	0.44	n/a
Mäsen	lowland	N6654684 E536934	102	5.6	0.42	3.42	1.87	No Trend
Skärgölen	lowland	N6514333 E571374	88	4.1	0.18	0.74	4.20	-2.0
Stora Envätter	lowland	N6555227 E634745	62	3.2	0.38	1.43	3.80	-2.0
Ulvsjön	lowland	N6611064 E347304	212	4.5	0.55	3.98	1.16	-1.5
Älgsjön	lowland	N6551428 E578455	49	4.3	0.36	5.04	1.11	No trend
Abiskojaure	sub-Arctic	N758208 E161749	486	6.6	2.79	370.00	0.06	-4.6
Latnjajaure	sub-Arctic	N758677 E161050	976	11.3	0.74	10.55	1.59	-6.5
Louvvaajaure	sub-Arctic	N736804 E160569	456	4.9	0.83	4.06	1.87	-6.1
Stor-Björnsjön	sub-Arctic	N706083 E132287	565	7.0	0.43	22.92	0.15	-3.9
Stor-Tjulträsket	sub-Arctic	N731799 E151196	540	8.5	5.25	274.78	0.19	-3.9
Övre Fjätsjön	sub-Arctic	N690617 E134197	746	3.8	0.91	44.70	0.13	-3.0
Ö.Särnmannasjön <sup>4</sup>	sub-Arctic	N683337 E133785	949	n/a	0.41	n/a	n/a	n/a

\* <https://minkarta.lantmateriet.se>

<sup>2</sup> <https://viss.lansstyrelsen.se/Waters.aspx?waterMSCD=WA10190925> [2022-02-11]

<sup>3</sup> <https://viss.lansstyrelsen.se/Waters.aspx?waterMSCD=WA60899891> [2022-02-11]

<sup>4</sup> <https://viss.lansstyrelsen.se/Waters.aspx?waterMSCD=WA41997143> – km<sup>2</sup> for Särnmannsjöarna collectively.

The separation of the two populations occurs at  $\sim 61^{\circ}\text{N}$ , with the sub-Arctic lake population in a latitudinal gradient across the Swedish parts of the Scandinavian alpine region and the lowland lakes in the middle/southern lowland of Sweden (Figure 2.1.1). The sub-Arctic lake population ranges from ultra-oligotrophic to meso-oligotrophic whilst the lowland lake population range from meso-oligotrophic to mesotrophic (with one eutrophic lake) (see Appendix, Table A.1).

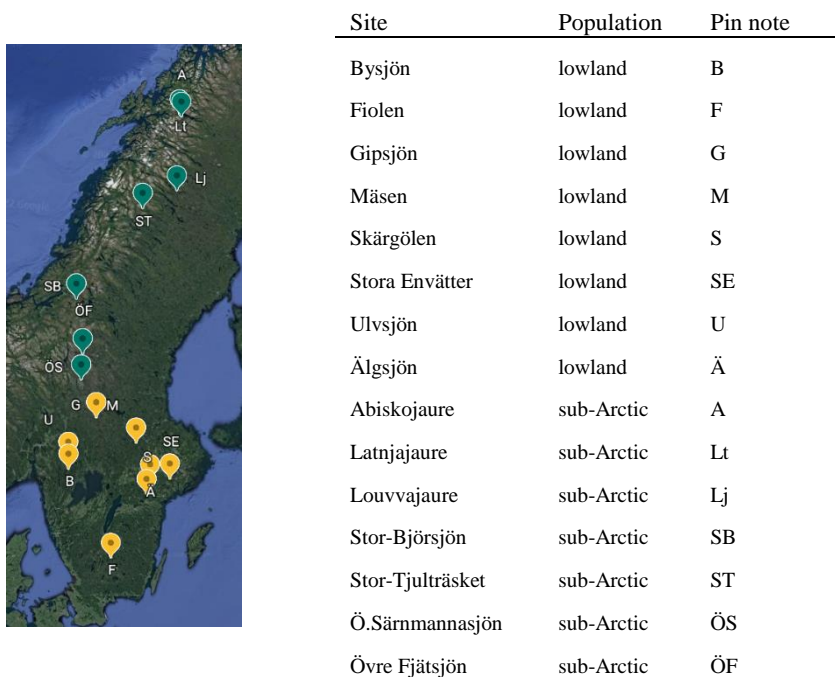
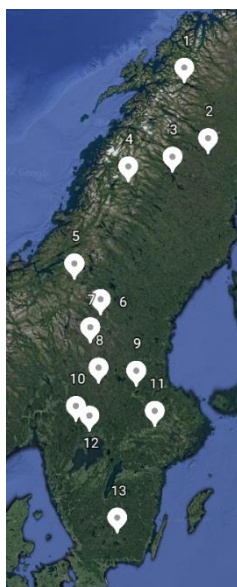


Figure 2.1.1. Lake location, green pin for sub-Arctic lakes and yellow pin for lowland lakes, pin notes in table to the right (Map generation; Google Earth project app)

In the north of Sweden and the alpine regions, the soil layer is mostly thin, and the bedrock lays close to the surface (Eriksson *et al.* 2011). In the middle to south of Sweden, the soil layer is deeper and soil combinations are more varied, with clays and silt as well as loam and sands being present.

Vegetation encroachment and increased vegetation growth has been noted in both higher latitudinal degrees and at higher altitude in Alaska, Greenland, and Scandinavia (e.g., Hollesen *et al.* 2015; Myers-Smith & Hik 2018; Tape *et al.* 2006; Truong *et al.* 2007; Wahren *et al.* 2005). Such evidence shows an already strong correlation to above-ground and terrestrial ecosystem change caused by global warming in these unique and threatened ecosystems.



Site (SMHI n.d. a)	Pin note
Abisko Aut	1
Jokkmokk Flygplats Mo	2
Arjeplog	3
Hemavan Flygplats	4
Särna	5
Hedeviken	6
Storlien-Storvallen	7
Malung	8
Falun-Lugnet	9
Arvika	10
Västerås	11
Karlstad Flygplats	12
Växjö	13

Figure 2.1.2 Weather stations position, pin notes in table to the right. With division of the populations, as sub-Arctic population north of pin 8 (Map generation; Google Earth project app)

Correlating the data in Table 2.1.2 and the weather station positions in Figure 2.1.2, compared to lake position in Figure 2.1.1, it is obvious that there is a change in meteorological seasonal length (longer summers and shorter winters, 1996-2021). There is also an increasing mean temperature of the year at close boarders to >42% of the sub-Arctic and 75% of the lowland lake populations.

Table 2.1.2 Climatic trend in seasonal changes in number of days\* in %/yr. and mean annual temperature °C (1996-2021). See pin note ref. in Figure 2.1.2, 1 indicating north, 13 south

Site	Pin note	Spring**	Summer**	Autumn**	Winter**	Mean annual temperature trend**
Abisko Aut	1	-0.37	0.72	-0.06	-0.10	n.s.
Jokkmokk Flygplats Mo	2	0.34	n.s.	0.13	-0.23	42.67
Arjeplog	3	-0.45	0.19	n.s.	-0.03	7.79
Hemavan Flygplats	4	-0.44	0.55	n.s.	-0.07	n.s.
Särna	5	0.17	0.41	n.s.	-0.35	n.s.
Hedeviken	6	0.58	0.35	-0.32	-0.31	3.73
Storlien-Storvallen	7	n.s.	0.84	n.s.	-0.42	n.s.
Malung	8	n.s.	0.42	-0.16	-0.28	n.s.
Falun-Lugnet	9	n.s.	0.38	n.s.	-0.33	0.84
Arvika	10	-0.15	0.19	n.s.	-0.26	0.85
Västerås	11	-0.38	0.29	0.27	-0.45	0.70
Karlstad Flygplats	12	n.s.	0.18	n.s.	-0.19	0.70
Växjö	13	0.44	0.09	0.34	-0.73	0.73

\*Based on the meteorological definitions of seasonal change; Spring: average temperature >0.0°C and <10.0°C for seven consecutive days. Summer: average temperature ≥10.0°C for five consecutive days. Autumn: average temperature <10.0 °C for five consecutive days. Winter: average temperature ≤0.0 °C for five consecutive days (SMHI n.d. b).

\*\*Analysed within this study with meteorology data from SMHI (n.d. b)

The meteorological data (collected from SMHI n.d. b) was subjected to a Mann-Kendal analysis, showing a great variation of change (Table 2.1.2). The change seems to correspond to the latest IPCC report (IPCC 2022), with most change to be found in the northernmost parts. However, the southern locations are not unaffected, lending evidence that the latitudinal gradient of Scandinavia is of special concern (IPCC 2022). The meteorological reasons behind the large variation of mean annual temperature (i.e., Jokkmokk Flygplats Mo) will not be discussed, as it is beyond the scope of this study.

## 2.2. Sedimentation rate & dating

For most of the lakes within the sub-Arctic lake population the sedimentation rates have been estimated based on their latitude and altitude (Brothers *et al.* 2008). Where more precise information was available from literature or alternative sources, these rates have been used instead (Table 2.2.1).

*Table 2.2.1 Sedimentation rates, estimated and modelled, sedimentation rates with values of both estimations and by other source are shown as a reference value between calculated and estimated values*

Lake	Population	Sedimentation rate [mm/yr.] **	Sedimentation rate other sources [mm/yr.]
Bysjön	lowland		3.1*
Fiolen	lowland		2.2*
Gipsjön	lowland		3.1*
Mäsen	lowland		6.1*
Skärgölen	lowland		3.6*
Stora Envätter	lowland		1.5*
Ulvsjön	lowland		3.5*
Älgsjön	lowland		1.5*
Abiskojaure	sub-Arctic	0.065	0.15 <sup>5</sup>
Latnjajaure	sub-Arctic	0.022	n/a
Louvvaure	sub-Arctic	0.092	n/a
Stor-Björnsjön	sub-Arctic	0.112	0.23 <sup>6</sup>
Stor-Tjulträsket	sub-Arctic	0.087	n/a
Övre Fjättsjön	sub-Arctic	0.112	n/a
Ö.Särnmannasjön	sub-Arctic	0.105	n/a

\* Huser unpublished data \*\* Estimated according to Brothers *et al.* (2008)

<sup>5</sup> Meyer-Jacob *et al.* (2017)

<sup>6</sup> Belle *et al.* (2021)

For the lowland lake population, the modelled estimation of sedimentation rates based on Brothers *et al.* (2008) deviate greatly from those calculated by  $^{210}\text{Pb}$  dating (Huser 2022, *unpublished data*). This might be explained by the land use variation which Brothers *et al.* (2008) found as an interfering factor to latitudinal precision regarding sedimentation in Europe where there is a great environmental heterogeneity. This factor should be less of concern for the barren landscape in northern Sweden where the sub-Arctic lake population is located. Hence, the estimated sedimentation rates based on Brothers *et al.* (2008) have been deemed reasonable for the sub-Arctic lake population (even among the rates based on more precise dating than estimates, the estimated rate has been kept for comparison). Whilst the  $^{210}\text{Pb}$  calculated values from Huser (2022, *unpublished data*) have been used for the lowland lakes.

## 3. Methods

### 3.1. P-fractionation

The sequential chemical extraction method used to assess P forms in sediment was developed by Psenner & Pucsko (1988), and Hupfer *et al.* (1995; 2009). It allows for an analysis of P fractions/pools in the sediment by four extraction steps giving five P forms: loosely bound and porewater P, Fe and Mn bound P, P bound to Al, P contained in organic matter (OM), and lastly P bound to Ca (calcium). An analysis of fractions by a spectrophotometer (880nm) with samples treated with the Ascorbic acid- ammonium molybdate method (Murphy & Riley 1962) gave absorbance, and P concentrations for each stratum of the sediment core were then calculated. For this study, only a portion of the total fractionation results are used as they are keys to explain the P-cycle. Henceforth the terms *Mob-P*, refers to loosely bound porewater P together with Fe bound P (i.e., mobile P), *Org-P*, equals P bound in organic matter (OM). Lastly *TP-frac* refers to the sum of all the extracted pools (porewater, Fe/Mn, Al, OM and Ca) which were released during fractionation (i.e., not total phosphorous (TP), which was unavailable for the sub-Arctic lake population). The P bound to Al and Ca is not studied as the mechanisms involved for their release/retention are not involved in the P-cycling processes herein studied.

The sediment cores for the lowland lake population were collected during 2014-2015, whereas the sub-Arctic lake population sediments were collected in the summer of 2021.

It should be noted that organic P was unavailable for the Lake Latnajaure core.

### 3.2. Data processing and statistical analysis

Water chemical analyse were conducted by the staff of the SWEDAC accredited Geochemistry laboratory at the Department of Aquatic Sciences and Assessment



at Swedish University of Agricultural and Environmental Science (SLU)<sup>7</sup>. For this study, the water chemistry data-set was retrieved from the SLU Environmental database<sup>8</sup> in its entirety, containing chemical analysis of epilimnetic samples (generally 0-2 m water depth) from 1996 to 2021. The chemical variables included in the study were aluminium (Al), iron (Fe), total organic carbon (TOC) and total P (TP). The water chemistry data set was initially cleaned and adjusted in Microsoft Excel 64 bit for Windows 10. Any less than detection limit values were divided by two, throughout the data set (i.e., if analysis showed <3; 1.5 was used). Due to multiple Al analytical methods used during analysis and duality between different lakes, [Al], [Al\_ICPAES], [Al\_NA] and [Al\_s] were used synonymously due to their chemical similarity. The risk of any errors due to differences in values from various analytical methods has been deemed negligible (Düker A., personal conversation<sup>9</sup>; SLU, Geochemical laboratory Accredited analytical method<sup>10</sup>).

The cleansed data-set was then analysed for trend using Mann-Kendal statistical analysis, with a confidence level of 95%. The analysis of multiple water samples per year allowed for determination of statistical trends both for the entire study period, i.e., annual changes of concentration, and seasonal changes of concentrations within specific months. All the significant trends were recalculated to trends in %/yr.

For Lake Ö. Särnmannasjön the collected water chemical data for relevant metals (Al and Fe) was produced partly by two different labs during 1996-2012<sup>11</sup>. When data from two labs were available on the same dates, an average of the two values was used. Also, for Lake Ö. Särnmannasjön, the analysis of dissolved metals was terminated in 2012 and neither Al nor Fe has been analysed since. Sampling of the lake was paused between 2012-09-21 to 2017-10-05 and resumed for the rest of the study period. Thus, in this study, total organic carbon (TOC) and total phosphorous (TP) data from Lake Ö. Särnmannasjön have been statistically analysed using 3 different scenarios: with and without the disrupted sampling period of 2012-2017 without any adjustment for missing values, as well as for short term trends during 2017-2021 (see Chapter 4.1).

For Lake Älgsjön the water chemical analysis for Al and Fe ended 2019, i.e., data for the two last years was lacking.

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<sup>7</sup> Detailed description can be found at <https://www.slu.se/en/departments/aquatic-sciences-assessment/laboratories/vattenlab2/>

<sup>8</sup> Available at <https://miljodata.slu.se/MVM/>

<sup>9</sup> Düker Anders [2021-12-09], Chemist at the Department of Aquatic Sciences and Assessment, SLU

<sup>10</sup> Detailed description can be found at <https://www.slu.se/en/departments/aquatic-sciences-assessment/laboratories/vattenlab2/>

<sup>11</sup> Additional analysis from Department of Environmental Science, Stockholm University

## 4. Results

### 4.1. Water chemical analysis

Long-term trends of Al water concentration for the lowland lake population showed no general trend. Only two of eight lakes had significant trends (Table 4.1.1). These trends (or lack thereof) were strikingly different from those of the sub-Arctic lake population, where all but one lake showed significant declining Al water concentrations (Table 4.1.1). The seasonal Mann-Kendal for Al in the sub-Arctic lake population showed a somewhat more continuous decreasing trend during the summer months than for the rest of the year. The sub-Arctic lake populations annual Al water concentration trend showed a significant monotonic, downward trend (Table 4.1.1).

*Table 4.1.1 All year water chemistry estimation trend for Lowland and sub-Arctic lake populations trend %/yr. for each parameter: aluminium, iron, total organic carbon and total P (1996-2021)*

Lake	Population	Al	significance	Fe	significance	TOC	significance	TP	significance
Bysjön	lowland			1.12	p<0.05				
Fiolen	lowland	-1.49	p<0.05			1.06	p<0.01		
Gipsjön	lowland								
Mäsen	lowland					0.65	p<0.05		
Skärgölen	lowland	-2.71	p<0.05						
Stora Envätter	lowland					1.05	p<0.001		
Ulvsjön	lowland								
Älgsjön	lowland					0.68	p<0.05	0.96	p<0.05
Abiskojaure	sub-Arctic	-3.16	p<0.001					-0.84	p<0.05
Latnjajaure	sub-Arctic	-1.24	p<0.01			-1.97	p<0.05	-5.09	p<0.001
Louvvaure	sub-Arctic	-1.05	p<0.05	-1.40	p<0.05			-1.97	p<0.01
Stor-Björnsjön	sub-Arctic	-1.57	p<0.01					-1.96	p<0.001
Stor-Tjulträsket	sub-Arctic	-2.16	p<0.01	-1.34	p<0.01			-3.54	p<0.0001
Övre Fjätsjön	sub-Arctic								
Ö. Särnmannasjön	sub-Arctic	-3.38	p<0.05						

The Mann-Kendall analysis of Fe concentrations in surface water showed no homogenous significant trend for either of the populations. Among the lowland

lakes, only one showed a positive significant trend (Lake Bysjön,  $p = 0.041$ ). The seasonal Mann-Kendall, showed variations in monthly Fe water concentration trends on an individual lake basis (not shown):

- Lake Byssjön and Lake Fiolen, had significant positive trends for August (both  $p = 0.05$ )
- Positive trends were also detected in Lake Stora Envättern for June ( $p = 0.04$ ), Lake Ulvsjön for April ( $p = 0.005$ ), Lake Älgsjön for April and October (both  $p = 0.04$ ).

The sub-Arctic lake population had two monotonic declining Fe concentration trends and three lakes showed monthly trends (not shown):

- Lake Louvvajaure and Lake Stor-Tjulträsket showed significant monotonic downward trajectories.
- Lake Latnjajaure had significant negative trends for July and September ( $p = 0.03$ ,  $p = 0.05$  respectively), Lake Louvvajaure for July and August ( $p = 0.03$ ,  $p = 0.04$  respectively) and Lake Stor-Tjulträsket for April ( $p = 0.001$ ) and August ( $p = 0.03$ ).

The TOC concentrations in the surface water had positive significant trends ( $p$  range 0.0003-0.0409) in 50% of the lowland lakes (Table 4.1.1). Only Lake Skärgölen and Lake Ulvsjön did not have a significant trend for any month according to the seasonal Mann-Kendall (not shown). The sub-Arctic lake population had no continuous, monotonic TOC trend with only one out of seven showing significance (Lake Latnajajaure,  $p = 0.032$ ). Seasonal Mann-Kendall analysis resulted in scattered monthly trends throughout the population. Three out of seven showed monthly trends (not shown):

- Lake Abiskojaure had significant negative and positive trends for April ( $p = 0.02$ ) and May ( $p = 0.04$ ), respectively.
- Lake Latnajajaure had negative trends for August ( $p = 0.04$ ).
- Lake Övre Fjätsjön had decreasing trends for March ( $p = 0.01$ ).

TP concentration in the surface water of the lowland lake population had only one significant trend, a positive trend for Lake Älgsjön. This deviated from earlier studies of these specific lakes where Huser *et al.* (2018) found half of the lakes within this population to have decreasing trends during 1988-2013, although 6% had a positive trend in that study.

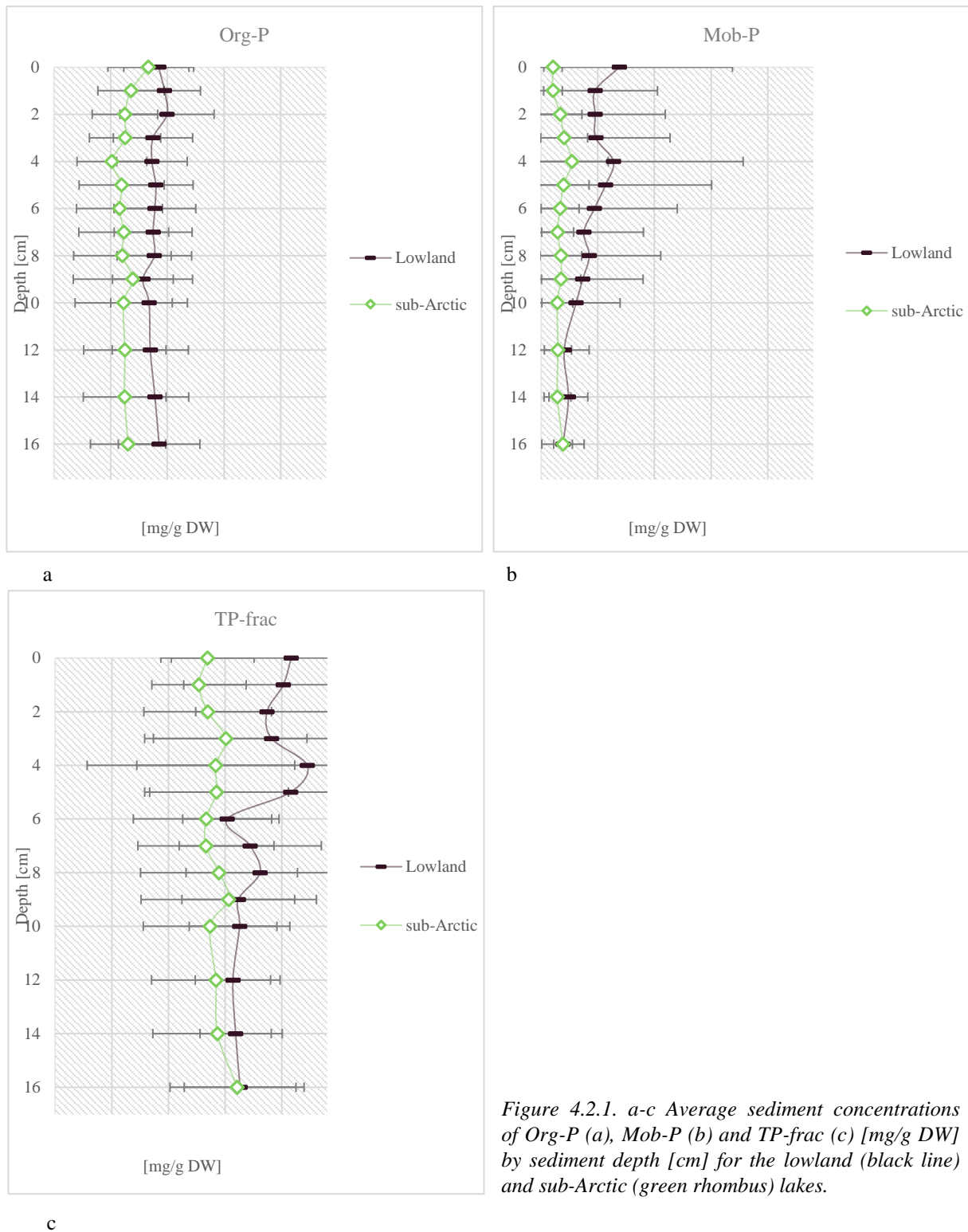
The sub-Arctic lake population had >70% significantly declining TP trends, with  $p$  ranging ~ 0.00007-0.045.

Due to a steep increase in the number of  $\text{PO}_4\text{-P}$  less than detection values (almost 100%) from 2013 and onward, trend analysis was not possible to conduct.

Statistical tests of TP and TOC concentration trends in Lake Ö. Särnmannasjön regarding the disrupted water sampling and analysis between 2012-2017 (as mentioned in Chapter 3.2 Data processing and statistical analysis) was tested for trends from 1996-2012, 1996-2021 (with no adjustment for missing values) and lastly, 2017-2021. Neither of these tested scenarios showed any significant trends.

## 4.2. Sediment analysis

Comparison of sediment by depth showed the fundamental differences between the lowland and the sub-Arctic lake populations. However, the standard deviation (SD) between both populations had large overlaps, indicative of large variability within each of the populations (Figure 4.2.1 a-c, compare water TP concentrations and trophic status in Appendix, Table A.1).



The core depth ranged from 16-18 cm to 42-44 cm as the deepest layers (lowland population). The comparison between the populations was standardized by the limiting depth (16-18 cm stratum). As the stabilization depth occurred within the core comparison depth (Org-P ~6 cm, Mob-P ~5-7 cm and TP-frac ~7-8 cm, see Figure 4.2.1 a-c) the difference in sampled core depth has most likely not affected the findings of this study (compare also the approximative sedimentation rates, see Table 2.2.1).

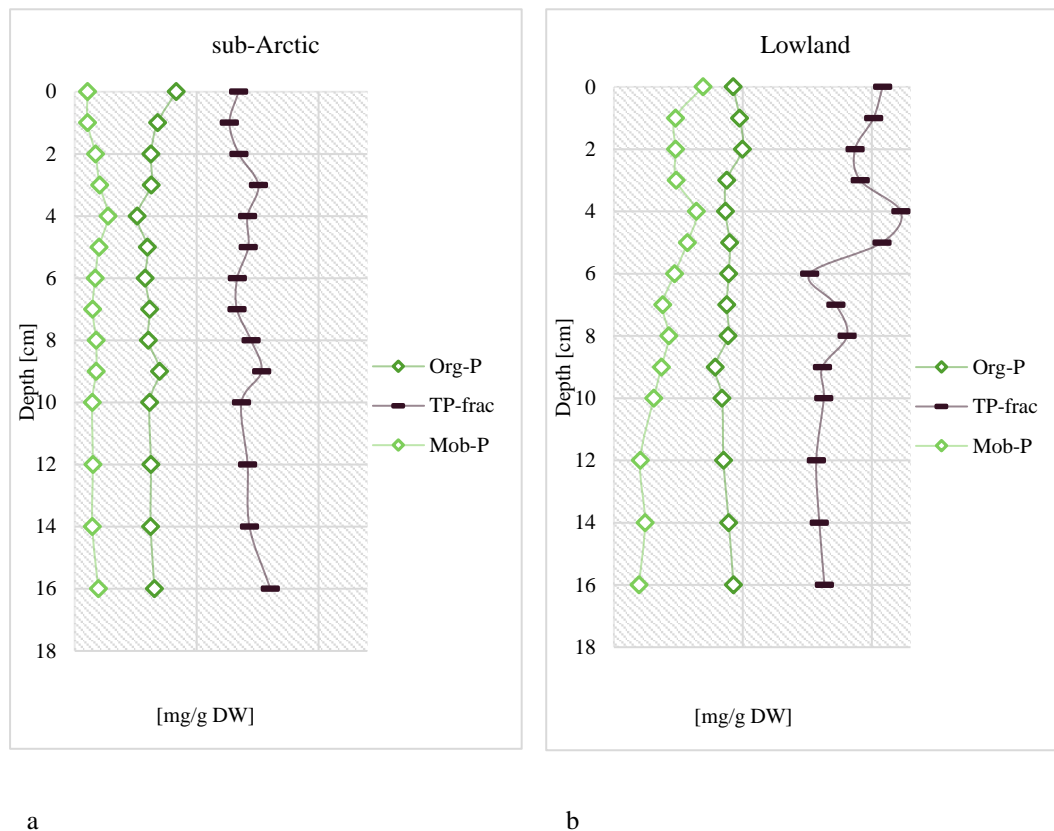


Figure 4.2.2. Average values of Org-P, Mob-P and TP-frac [mg/g DW] by sediment depth for sub-Arctic ( $n=6$  for Org-P,  $n=7$  for Mob-P and TP-frac) and lowland lakes ( $n=8$ ) respectively.

On an average there was a higher concentration of Org-P in the topmost sediment layer (compared to the lower layers) of the sub-Arctic lake population, levelling out from 1 cm (Figure 4.2.1 a). Whereas the lowland lake population showed more of a constant Org-P concentration throughout the sediment profile.

For Mob-P concentrations, the average lowland lake had elevated concentrations in the surficial sediment (0-6 cm), compared to deeper layers in the profile as well as the sub-Arctic lake average. Whereas the average sub-Arctic lake showed similarly low concentrations of Mob-P throughout the core profile.

TP-frac show a slightly heightened concentration in the top surface of both populations, as can be seen in Figure 4.2.2 a-b. This might reflect the Org-P concentration for the sub-Arctic lake average and the Mob-P in the average

lowland lake within the surface layer, or could also be explained by the remaining P fractions not studied herein.

The variability of P fractions in each population could be a consequence of the latitudinal alignment associated with the south-north gradient of landscape change. Although it is probably mostly due to the lowland lake population also being constituted of relatively nutrient-poor lakes, i.e., balancing between meso-oligotrophic to mesotrophic (detailed average and SDs in Appendix, Table A.2).

The average lowland lake, within the sediment depths 0-4 cm had Org-P and Mob-P ranges between 44.3-53.3% and 33.1-23.7%, respectively (out of the TP-fraction content). The sub-Arctic lake population averaged 41.6-61.9% for Org-P and Mob-P 7.8-13.5%, for the same depths (Figure 4.2.2 a-b).

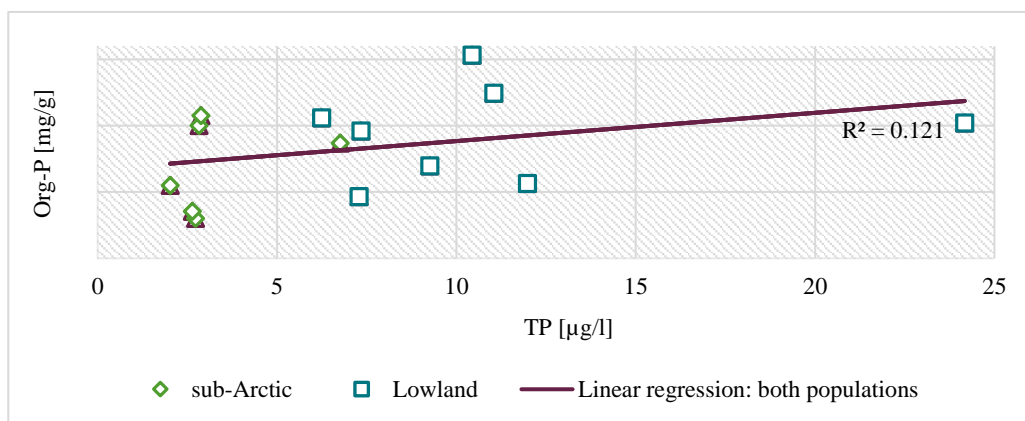


Figure 4.2.3 Regression of sediment concentration Org-P [mg/g DW] average top 4 cm on average TP water concentration [µg/l] the last 5 years of the series (2017-2021).

The regressions between TP water concentration and sediment Org-P concentration had a high variability and a weak relationship ( $r^2=0.12$ ) and was strongly influenced by the lake with the highest TP concentration in the surface water (Figure 4.2.3).

Table 4.2.2 shows the potential P release rate for each individual lake, based on the Mob-P concentrations of the top 4 cm of sediment (Pilgrim *et al.* 2007). Negative release rate indicates a potential net retention, whilst a high, potential P release rate,  $> 2$  and  $> 5$  mg/m<sup>2</sup>/day, indicate a great risk of degrading water status (due to increased P concentrations) in the pristine, sub-Arctic environments and lowland areas, respectively (Huser *et al.* 2022, *prelaminar report*).

*Table 4.2.2 Potential release rate, Li in [mg/m<sup>2</sup>/day]*

Lake	Population	Li (mg/m <sup>2</sup> /d)*
Bysjön	lowland	3.6
Fiolen	lowland	3.4
Gipsjön	lowland	5.3
Mäsen	lowland	35.2
Skärgölen	lowland	4.2
St Envättern	lowland	-0.2
Ulvsjön	lowland	4.2
Älgsjön	lowland	3.1
Abiskojaure	sub-Arctic	14.9
Latnjajaure	sub-Arctic	1.2
Louvvojaure	sub-Arctic	-0.5
Stor-Björnsjön	sub-Arctic	0.4
Stor-Tjulträsket	sub-Arctic	12.7
Övre Fjättsjön	sub-Arctic	1.6
Ö. Särnamannasjön	sub-Arctic	-0.5

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\* Estimated according to Pilgrim *et al.* (2007)

## 5. Discussion

The results for TP concentration trends in this study agree with former studies, i.e., that there is a long-term ongoing oligotrophication of lakes in the sub-Arctic region of Sweden (Huser *et al.* 2018). Nevertheless, the findings from the lowland lake population deviate from the former findings of Huser *et al.* (2018) as there was only one decreasing TP trend. Within the sub-Arctic population, the rate of decline (%/yr.) has decreased since this former study (compare TP trends in Table 2.1 and Table 4.1.1). As such, there are indications that these declines may cease or, alternatively, that there is a cause between 2013-2021 causing these developments. Such an event would have to have had a latitudinal impact as both populations are affected.

### 5.1. Sediment related effects

The primary process(es) causing the declines could not be determined from the comparison between water and sediment concentrations of sub-Arctic lakes and lowland lakes within this study. TP-f<sub>rac</sub>, if used as a proxy for sediment TP, showed no surprises, as the oligotrophic/mesotrophic lakes in both populations followed the patterns predicted by their water column trophic statuses (Figure 4.2.1 c; Appendix, Table A.1) (Carey & Rydin 2011). The hypothesised connection between water TP and sediment Mob-P, i.e., P retention in FeOH complexes, could not explain the general decreasing P concentrations within the sub-Arctic population. The overall constant sediment TP-f<sub>rac</sub> concentrations, used as a proxy for TP, indicate that there is still a general P-retention capacity within both lake populations (Hupfer *et al.* 2004; Wilson *et al.* 2010), even though the average lowland lake is not quite as uniform in this pattern (see Figure 4.2.1).

The average sub-Arctic lake sediment had a higher percentage accumulation of Org-P in the top 4 cm, although the actual concentration was lower compared to the average lowland lake (average 0-4 cm concentration was 0.69 and 0.94 mg/g DW for the average sub-Arctic and lowland lake, respectively). Both populations had a decreasing concentration by sediment depth. The surface sediment Org-P (0-1 cm) of the average sub-Arctic lake indicates a possible elevated sedimentation of allochthonous and/or autochthonous OM. The same sediment



layer for the average lowland lake is, as for the 0-4 cm average, smaller in relation to the TP-fraction (%), but again the actual concentration is greater. This relationship could be argued to somewhat contradict the trophic statuses of the two populations (Appendix, Table A.1), as the opposite relationship would have been expected according to previous work (Carey & Rydin 2011).

The elevated sediment Org-P concentration in the sub-Arctic population (compared to deeper sediment) is somewhat surprising. But considering the decrease in spring snow coverage coupled with increased temperatures (IPCC 2022; SMHI n.d. b) and a vegetation diversity shift (Tape *et al.* 2006; Truong *et al.* 2007), this suggests that there might be an increase of OM due to increased input from deciduous vegetation within these watersheds.

The main part of this study was to evaluate the sediment and water concentrations of P to induce greater insight to the importance of P-cycling of sub-Arctic lakes by examining patterns of variable concentration trends. Former studies have concluded that oligotrophic lakes accumulate P in the sediment, making it a permanent sink. Whilst mesotrophic lakes are hypothesised to accumulate deposited P yet are closer to saturation (Carey & Rydin 2011). The sediment concentration distribution of the sub-Arctic population (Figure 4.2.2) did not show the P retention patterns hypothesised or expected. Neither did the water concentration analysis show all previously documented patterns, e.g., previously shown increases in TOC concentrations (Gieseler *et al.* 2005). However, there was a non-hypothesised relationship between sediment Org-P and (epilimnetic) water TP concentrations (Figure 4.2.3). Such a relationship can have great bearing on these ecosystems, especially in consideration to climate change. Assuming that sediment Org-P could be used as a proxy for OM, oxygen levels could be decreasing in the interstitial water (due to increased mineralization of OM) and a subsequent release of Fe and associated P may occur (Mortimer 1942). Nevertheless, there are no such indications of this process in the water column TP trends as they indicate an ongoing oligotrophication or decline of P.

## 5.2. Lake chemistry related effects

Continued, declining P trends might indicate that oxygen concentrations have not yet decreased to such low values to induce the release of P bound in FeOH complexes (i.e., most of the Mob-P fraction). However, in the future, a succession to a new trophic status might occur if DO in these types of lakes start to decrease in bottom waters.

In lakes with periodically or constantly low oxygen in the hypolimnion, Mob-P controls sediment P release (Pilgrim *et al.* 2007). The correlation shown herein, between sediment Org-P concentration and TP concentrations in the epilimnion, however, suggests that the release of sediment P, or in-lake P cycling, is mainly

caused by decomposition of OM, and release of Org-P. Thus, an increase in OM input causes a P retention in these lakes, suggesting that lakes that go anoxic for shorter periods (if at all) have a general relationship between the sediment Org-P and the in-lake P.

### 5.3. Temperature

Analysis of changing temperatures and seasonal length (Table 2.2.1) showed that this area has been subjected to a general warming, even within the 26 years herein studied. A warmer climate will increase lake temperature and cause a greater temperature gradient to form throughout the water column (Pokrovsky *et al.* 2013). This in turn, will increase the stratification strength and length (i.e., the period between mixing events) and coupled with increased P concentration and lake productivity, could in turn lead to oxygen depletion in the hypolimnion (Hobbie *et al.* 1999). Nevertheless, stratification strength and length have been suggested to be extremely variable (Gerten & Adrian 2001; Woolway & Merchant 2019). Consequently, the responses of temperature induced P release rates (individual lake, increment of temperature and release rates) seem to be too complex to model, however former studies agree on an overall temperature impact. It is, however, unclear from this study how this type of change affects, or will affect, either of the lake populations.

Hypolimnetic water temperatures throughout the year vary greatly in different lakes, as do any response to increased air temperatures. However, it is not only the hypolimnetic water temperature that is affected by climate change. Shorter ice duration and increased evapotranspiration (i.e., warmer epilimnion/higher surface temperature) caused by global warming may decrease lake water level. As a consequence, lakes are hypothesised to depend even more on spring snow melt (Prowse *et al.* 2006). This causing a negative net result in overall water levels as the projected and increased precipitation will not be enough to neutralize the joined effect of evaporative losses and evapotranspiration. Subsequently, these developments are thought to lead to an increased in-lake productivity. These hypotheses are mainly based on studies of shallower lakes that are permanently ice covered. Consequently, the direct effect on deeper sub-Arctic lakes may not be as strong but cannot be entirely neglected.

### 5.4. Watershed changes

In the mountain areas where these lakes are located (sub-Arctic lake population), vegetation is limited (low woody plants and shrubs) and bare bedrock generally dominates the catchment. Thus, it seems there would be less chance for elevated

sedimentation of OM (Org-P). The majority of the lowland lakes, on the other hand, are located in watersheds rich in vegetation and often have forest dominated catchments. Here one would expect a higher percentage and amount of OM in the sediment. Earlier evidence of increased vegetation growth and/or vegetation encroachment in high elevated areas, with respect to both habitat and diversity, could indicate that there is an ecosystem shift at hand (e.g., Hollesen *et al.* 2015; Myers-Smith & Hik 2018; Tape *et al.* 2006; Truong *et al.* 2007; Wahren *et al.* 2005). The specific findings of this study do not show enough statistical evidence to either contradict or confirm such hypotheses, i.e., increased OM input to Arctic lakes due to vegetation. This can partially be explained by low sedimentation rates and changes within a catchment taking a long time to show within the sediment.

As sedimentation rates are so low (see Table 2.1.2 for reference), especially in the sub-Arctic area, this means that it might take decades or even longer before sediment cores can hold information on the recent accelerations of climate change. However, there is also the possibility that, to establish significant results regarding indications of an ecosystem shift caused by climate change, there is a need for a broader or larger study of sediment cores to determine if in-lake changes to nutrient cycling are indeed occurring.

Ecosystem reactions to climate change and increased temperatures are complex and, as of yet the holistic understanding of these transformations are not entirely known. The succession of vegetation encroachment in the circumpolar area is well documented. Although, there is an additional shift in the below ground community (i.e., microbes and fungi). Fungi symbiont spread go hand in hand with the deciduous vegetation spread, and an increased diversity in the vegetation will be echoed in the below ground community (Clemmensen *et al.* 2006; Deslippe *et al.* 2011). This could possibly lead to a higher retention of nutrients within the watershed, as one of these symbiont relationship's main advantages are to provide the vegetative community with P from the interstitial water in the root zone (Balestrini *et al.* 2015).

In former studies, decreasing TP concentrations have been coupled to increasing TOC/DOC (Huser *et al.* 2018). Thus, noted that there is a relationship between an increased TOC migration from the watershed and available P binding sites. Consequently, additional binding of P in AlOH complexes within the watershed soil, both due to increased binding sites and pH, is hypothesised to partially explain the decrease in TP water concentrations in this area (Huser *et al.* 2018; Gieseler *et al.* 2005). P bound in AlOH complexes does not desorb in the circumneutral pH range (Cooke *et al.* 2006), and this could point to a possible permanent retention of P within some of the studied lakes (see pH ranges in Appendix, Table A.1). Hence, if there is an increased precipitation of P and AlOH complexes in the watershed, this will subsequently limit the nutrient export to these lakes. Especially if the ratio of Al to P in watershed soil is elevated. Greater

Al:P increases the potential adsorption of P within the watershed. It has also been shown that lakes with a lower Al:P are less resilient (Spears *et al.* 2017). Consequently, such environments are more sensitive to the effect of climate change on P-cycling. Both above reviewed processes may have a sufficiently large effect on the P supply from the watershed. In view of these former findings, it is surprising that within the scope of this study there is no indication of any general changes in TOC concentrations, as it has been noted to increase in surface water in the circumpolar area in previous studies (Lento *et al.* 2019; Huser *et al.* 2018).

McLaren *et al.* (2017) indicated that vegetation encroachment, especially of deciduous shrubs will change the leaf litter turnover time. By such give rise to a decomposition shift within these areas. And in time it is hypothesised that the decomposition time will decrease after an initial increase. Such developments could possibly lead to a larger allochthonous deposit and thus further increase the risk of hypoxia.

Within the sediment, the degradation of OM will likely increase with increasing temperature (Gudaz *et al.* 2010). Increased production of Org-P and the subsequent settling of such on the sediment surface, provides a heightened risk of P input from the sediment to the hypolimnion, especially within well oxygenated lakes (Gudaz *et al.* 2010) as the sub-Arctic population. That is, even without an increased sediment Org-P, there is a risk of P release from OM due to increased temperatures. This could partially explain the seemingly flattened TP trend (comparing this study to the findings of Huser *et al.* 2018) as some of the watershed P declines may be offset by increased release of Org-P in the lake.

Further, a general induced microbial activity caused by higher temperatures from e.g., dormant microbes, could shift the equilibria of biological P within sediment (Haglund *et al.* 2003). This response is due to the subsequent increased microbial respiration, making P bioavailable. The increased microbial activity would further lead to lower redox potential, consequently followed by a higher production of enzymes (Pettersson 1998). Furthermore, oxygen depletion would generate anaerobic metabolism among microbes causing a lower immobilisation of P into bacterial cells. There is also, among certain microbes, an induced P release due to changed metabolism (than it would have during oxic conditions) (Boström *et al.* 1988). These additional mechanisms of P release would increase along with diffusion of Fe associated P due to the low redox potential.

## 5.5. A complex combination of factors

The multiple controlling factors to sediment P release/retention mechanisms are a complex web. The focus of this study has been the effect climate change has on

this cycle. In summary increasing temperatures, greening and climate change would generally lead to:

- more OM production in watershed leading to an increased accumulation in lakes
- more OM input will increase nutrient content of sediment
- more OM and higher temperatures may lead to more DO depletion
- more DO depletion will cause P to release from Fe/Mn minerals (i.e., diminish the sediment Mob-P pool and recirculate it to the water column)

Increasing temperatures will also lead to:

- stronger and longer stratification periods
- greater risk for oxygen depletion in the hypolimnion
- greater risk for elevated sediment P release, especially if there are greater amounts of OM in the sediment
- P accumulation in vegetation within the watershed

A general lake with a high potential P release rate ( $>5 \text{ mg/m}^2/\text{d}$ ) would, in case of temporary or permanent hypoxia, be at high risk of severe degrading water status. Within a northern/alpine lake the limit for such a risk is at a lower value than in lakes in general, preliminary data show that a potential P release rate  $>2.0 \text{ mg/m}^2/\text{d}$  indicates a high risk of internal loading (*unpublished report* Huser *et al.* 2022). To illustrate the predicaments of these developments a schematic categorization, with the studied lakes as an example, is suggested in Table 5.1.

As the risk of anoxia increases with higher OM in the settling sediment, lakes with a higher average sediment OM content stand an even greater risk of eutrophication. The risk of P release from OM would be particularly grave in an ecosystem where the binding sites related to the Mob-P pool are diminished. A temporary oxygen depletion in the hypolimnion e.g., caused by seasonally induced allochthonous deposits of semi-decomposed leaf litter, would thus be an abrupt transformation of these ecosystems. Hence, if the oligotrophication trend of the sub-Arctic lake population is indeed flattening, these ecosystems may be subjected to stark changes due to the increased potential for P release from the sediment.

Generally, this issue is greatest within ecosystems subjected to vegetation encroachment, such as the sub-Arctic lake population. This is due to a historically low production and allochthonous input of OM. The effect increased temperatures would have on OM composition, microbial activity, and the recirculation of P, would consequently lead to a higher primary production in the lake, potentially

further increasing the cycle of OM deposition and P release from the sediment. An equilibria shift concerning the entire food web could be argued as the ultimate consequence.

*Table 5.1 Risk intervals for a general lake <1 mg/m<sup>2</sup>/d low risk, 1 – 5 mg/m<sup>2</sup>/d medium risk and >5 mg/m<sup>2</sup>/d high risk, for northern/alpine lakes <0,5 mg/m<sup>2</sup>/d low, 0,5-2 medium and >2 high risk*

Lake	Population	Li (mg/m <sup>2</sup> /d)	Average Org-P [mg/g DW] 0-4 cm sediment	Risk of water status degradation in case of hypoxia
Bysjön	lowland	3.6	0.7	Medium
Fiolen	lowland	3.4	1.5	Medium
Gipsjön	lowland	5.3	1.2	High
Mäsen	lowland	35.2	0.6	High
Skärgölen	lowland	4.2	0.9	Medium
St Envättern	lowland	-0.2	0.5	Medium
Ulvsjön	lowland	4.2	1.1	Medium
Älgsjön	lowland	3.1	1.0	Medium
Abiskojaure	sub-Arctic	14.9	1.0	High
Latnjajaure	sub-Arctic	1.2	-*	Medium
Louvvaure	sub-Arctic	-0.5	0.6	Low
Stor-Björnsjön	sub-Arctic	0.4	0.3	Low
Stor-Tjulträsket	sub-Arctic	12.7	0.4	High
Övre Fjätsjön	sub-Arctic	1.6	0.9	Medium
Ö. Särnamannasjön	sub-Arctic	-0.5	1.1	Low

\*see chapter 3.2 Method

Nevertheless, if the results of climate change are either more or less productivity, it is herein hypothesised that these effects soon will be visible within sub-Arctic lakes. However, the lack of integrated understanding of anthropogenic effect on nutrient distribution combined with climate change makes it exceedingly difficult predicting these outcomes.

## 5.6. Recommendations for future work

Possible for error related to the findings of this study, which ought to be corrected in future studies include: collecting sediment during the same year (either spring or fall when lakes are well mixed). The lowland population sediment cores were 6 years younger than that of the sub-Arctic, but were collected in either the spring or fall periods. There is a risk that more recent sediment could have brought a different understanding to the differences between these populations.

A monitoring related issue that limited the analysis of water chemical data was the less than detection limitation for orthophosphate concentrations, varying

greatly between 2013 and 2021. The number of less than values and even the number of different detection limits was high within the data set and no reasonable way to handle the differences was found. The lack of reportable data is unfortunate because, even as  $\text{PO}_4\text{-P}$  is rapidly used in the environment, a study including this type of variable would have given information regarding the current biological conditions in the studied lakes. Improved detection limits might help, but in some cases  $\text{PO}_4\text{-P}$  might just not be present at levels that can be detected no matter the method.

Future studies should try and expand sediment sampling scale to see if the failure to establish statistical significance was simply due to low statistical power. In the future (at least 25 years) it might be possible to connect the sediment records to water chemistry and elaborate on the affect climate change has had on in-lake P-cycling in the sub-Arctic. The rapid climate change on one hand and the slow sedimentation rates on the other, makes it exceedingly difficult to shed necessary light on these processes. Extensive modelling of all known parameters affecting these ecosystems would take more time but might bring greater understanding to possible, future events.

A more detailed understanding of events recorded within the sediment could broaden the insight of these pristine ecosystems. Determination of sedimentation rate using  $^{210}\text{Pb}$ -dating joined with changes in P in relation to temperature and DNA analysis could potentially show changed microbial activity. This could help determine the impact climate change has had on P concentrations in lakes in the sub-Arctic area, both biotically and abiotically.

Further research on the potential relationship between sediment Org-P and epilimnetic water TP concentrations, elaborating if the relationship herein found was circumstantial or if there is more to the suggested explanation, is suggested. Due to the probable substantial difference between epilimnetic and hypolimnetic water concentrations these findings need a further in-depth, quantitative study including analysis of bottom water chemistry. Unfortunately these data are lacking for most long-term monitored lakes in Sweden.

Future studies could include the potential cocktail effect of the combined consequences of climate change to these ecosystems. For example, study of the riparian vegetation effects on sediment chemistry in relation to general vegetation encroachment/greening. This would be specially interesting if coupled with e.g., abiotic events such as the impact on P migration from intermittent droughts and heavy rainfall.

## 6. Conclusions

The findings of this study add to the already substantial evidence of an ongoing oligotrophication in lakes of the Northern Hemisphere, with an emphasis on the generally most oligotrophic lakes, especially in the sub-Arctic area. As the oligotrophication trend only recently entered the research field there is a knowledge gap regarding the underlying causes of the observed changes. However, to be able to qualitatively understand the in-lake effects on this oligotrophication trend and its relation to climate change, one might need another 20-40 years of settling sediment to have sufficient material to analyse. Nevertheless, there are indications of the sub-Arctic ecosystems being subjected to climate induced changes as discussed above (e.g., greening and temperature).

This study evaluated the possible climate change induced pressures that may have severe impacts on circumpolar ecosystems. 57% of the sub-Arctic lakes in this study have a medium or high risk of water quality degradation due to potential sediment P release rate herein identified. On average, the sub-Arctic lakes had a potential release rate of 4.27 mg/m<sup>2</sup>/d, whilst the lowland average had a potential release rate of 2.95 mg/m<sup>2</sup>/d without lake Mäsen (7.35 mg/m<sup>2</sup>/d with). The average release rate of the sub-Arctic lake population indicates a high risk of release of potential Fe associated P in the event of temporary or permanent hypoxia. As such, a further vegetation encroachment could be argued to have a substantial effect, not only on the terrestrial ecosystem, but also on in-lake P-cycling in Swedish alpine lakes.

This study failed to link the sub-Arctic oligotrophication trend to an increased retention within the Mob-P pool, herein determined by P bound in porewater and FeOH complexes. However, the relationship between sediment Org-P concentrations and TP water concentrations indicate that an increased primary production could indicate a P retention mechanism (within lakes that are permanently oxic or suffer hypoxia only for short periods). Although, the increased Org-P will likely lead to an elevated cycling of P between the sediment and water. Increased sediment OM might also be due to an increased allochthonous input caused by the terrestrial ecosystem shift called *greening*. The potential flattening of declining P trends is an indication that the above processes already may be at work (comparing the findings of this study and Huser *et al.* 2018). This may suggest that the elevated P-cycling between sediment and water



has already begun, though it is of yet so low that it does not show in the correlations between sediment and epilimnetic water chemistry in the study lakes. Due to the slow sedimentation rate, evidence of such a relationship might take many years to prove. If enhanced P-cycling processes have begun, it is possible that fluctuations are only detectable by examining interstitial water fluxes. In conclusion, this relationship needs considerably more research to be determined quantitatively.

The less than detection  $\text{PO}_4$  concentrations are crucial as it highlights the reduced easily accessible bioavailable P within these lakes and is a sign of these systems succession towards ultra-oligotrophic conditions. They also support previous work indicating potentially harmful declines in P in sub-Arctic lakes.

The consequence of climate change on lakes seem to be as multiple as there are differences between lakes. The similarities shared within each population are a useful tool to understand the general predicaments within other similar, pristine areas. Nevertheless, it cannot, from this study on its own, be concluded that the offset to P-cycling within these sub-Arctic lakes is caused by climate change. Though there are solid indications of irrevocable events in motion, and global warming seems to be at the centre of all herein examined, potential pressures.

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# Appendix

*Table A.1 Summary of lake water chemistry average by year average for the studied period (1996-2021) both populations*

Lake	Population	Al [ $\mu\text{g/l}$ ]	Fe [ $\mu\text{g/l}$ ]	TOC [ $\text{mg/l}$ ]	TP [ $\mu\text{g/l P}$ ]	pH
Bysjön	lowland	50.43	257.80	7.51	9.97	6.51
Fiolen	lowland	50.39	72.79	7.56	11.43	6.65
Gipsjön	lowland	197.25	1232.02	14.25	11.28	5.47
Mäsen	lowland	49.65	119.28	8.57	11.82	6.82
Skärgölen	lowland	33.82	101.06	7.79	8.03	6.78
Stora Envättern	lowland	62.35	95.28	10.42	7.45	6.53
Ulvsjön	lowland	124.48	255.55	8.86	6.90	6.08
Älgsjön	lowland	132.54*	605.39*	19.27	22.46	6.70
Abiskojaure	sub-Arctic	21.34	46.68	1.45	3.59	7.10
Latnjajaure	sub-Arctic	23.46	24.73	0.85	2.74	6.49
Louvvojaure	sub-Arctic	27.43	17.81	3.29	3.27	7.04
Stor-Björnsjön	sub-Arctic	37.37	100.74	4.67	3.94	6.96
Stor-Tjulträsket	sub-Arctic	14.81	35.04	2.22	3.95	7.29
Övre Fjätsjön	sub-Arctic	50.34	85.45	4.34	6.62	6.64
Ö. Särnamannasjön	sub-Arctic	49.70**	13.32**	1.51***	4.36***	5.61

\*1996-2019 \*\*1996-2012 \*\*\*no data 2012-2017

*Table A.2 Mean concentration, [mg/g DW]  $\pm$  standard deviation, for each sediment core by depth, [cm] for sub-Arctic (SA) and lowland (LL) lake populations correspondingly*

Depth	SA Org-P	SA TP-frac	SA Mob-P	LL Org-P	LL TP-frac	LL Mob-P
0-1	0.833 $\pm$ 0.358	1.344 $\pm$ 0.411	0.105 $\pm$ 0.081	0.924 $\pm$ 0.308	2.083 $\pm$ 1.058	0.690 $\pm$ 0.998
1-2	0.680 $\pm$ 0.293	1.269 $\pm$ 0.416	0.105 $\pm$ 0.082	0.975 $\pm$ 0.318	2.013 $\pm$ 0.878	0.477 $\pm$ 0.550
2-3	0.626 $\pm$ 0.289	1.346 $\pm$ 0.563	0.170 $\pm$ 0.191	0.997 $\pm$ 0.415	1.870 $\pm$ 0.629	0.478 $\pm$ 0.618
3-4	0.627 $\pm$ 0.315	1.506 $\pm$ 0.716	0.204 $\pm$ 0.207	0.874 $\pm$ 0.350	1.910 $\pm$ 1.043	0.483 $\pm$ 0.655
4-5	0.511 $\pm$ 0.308	1.417 $\pm$ 0.696	0.272 $\pm$ 0.320	0.864 $\pm$ 0.311	2.225 $\pm$ 1.942	0.638 $\pm$ 1.145
5-6	0.597 $\pm$ 0.375	1.424 $\pm$ 0.634	0.199 $\pm$ 0.225	0.898 $\pm$ 0.329	2.079 $\pm$ 1.245	0.569 $\pm$ 0.936
6-7	0.578 $\pm$ 0.379	1.333 $\pm$ 0.643	0.168 $\pm$ 0.167	0.890 $\pm$ 0.361	1.518 $\pm$ 0.393	0.471 $\pm$ 0.731
7-8	0.616 $\pm$ 0.396	1.330 $\pm$ 0.600	0.146 $\pm$ 0.140	0.875 $\pm$ 0.344	1.721 $\pm$ 0.627	0.378 $\pm$ 0.526
8-9	0.604 $\pm$ 0.430	1.446 $\pm$ 0.692	0.175 $\pm$ 0.184	0.886 $\pm$ 0.329	1.808 $\pm$ 0.653	0.425 $\pm$ 0.631
9-10	0.696 $\pm$ 0.526	1.532 $\pm$ 0.774	0.177 $\pm$ 0.173	0.785 $\pm$ 0.268	1.616 $\pm$ 0.497	0.368 $\pm$ 0.532
10-12	0.615 $\pm$ 0.429	1.367 $\pm$ 0.590	0.143 $\pm$ 0.136	0.839 $\pm$ 0.338	1.627 $\pm$ 0.444	0.309 $\pm$ 0.388
12-14	0.625 $\pm$ 0.363	1.418 $\pm$ 0.567	0.149 $\pm$ 0.120	0.850 $\pm$ 0.336	1.569 $\pm$ 0.332	0.204 $\pm$ 0.220
14-16	0.624 $\pm$ 0.365	1.433 $\pm$ 0.571	0.145 $\pm$ 0.118	0.891 $\pm$ 0.298	1.593 $\pm$ 0.314	0.241 $\pm$ 0.171
16-18	0.653 $\pm$ 0.332	1.605 $\pm$ 0.591	0.193 $\pm$ 0.187	0.927 $\pm$ 0.360	1.632 $\pm$ 0.493	0.194 $\pm$ 0.082