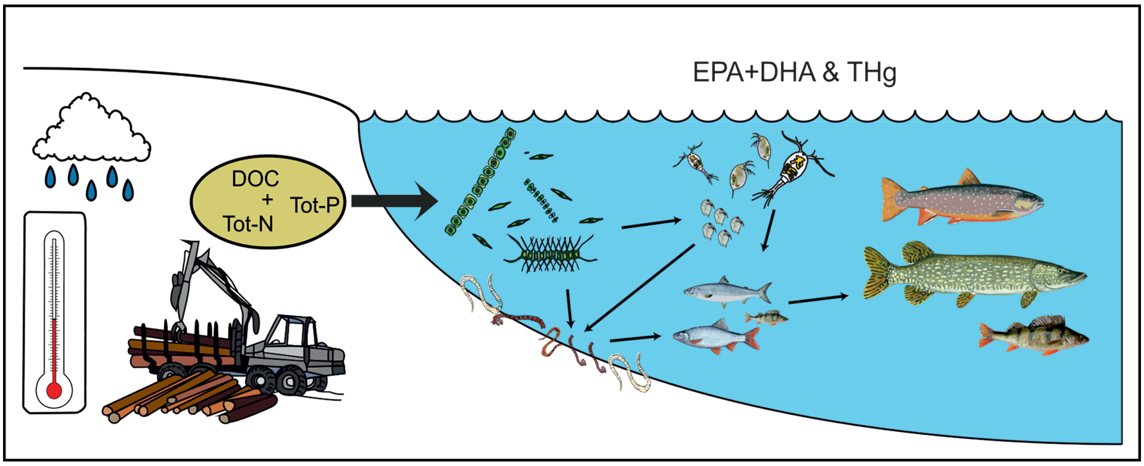
JYU DISSERTATIONS 525

Ossi Keva

Environmental Change Effects on Lake Food Web Structure and Nutritional Quality



UNIVERSITY OF JYVÄSKYLÄ
FACULTY OF MATHEMATICS
AND SCIENCE

JYU DISSERTATIONS 525

Ossi Keva

Environmental Change Effects on Lake Food Web Structure and Nutritional Quality

Esitetään Jyväskylän yliopiston matemaattis-luonnontieteellisen tiedekunnan suostumuksella
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ABSTRACT

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Climate change and intense land-use activities are promoting lake eutrophication and browning, affecting community structure and food web processes. In this thesis, space-for-time approach was used to study the environmental change impacts on food web structure, energy pathways, and organism nutritional quality (defined with fatty acids and mercury content) in subarctic and boreal regions. Only specific algal taxa can synthesize eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA), which are important polyunsaturated fatty acids (PUFA) for consumer growth and reproduction. Mercury is a toxic heavy metal that, bioaccumulates to organisms via diet. In subarctic lakes, increasing temperature and productivity negatively affected EPA+DHA content of seston and zooplankton. However, no such changes were observed from boreal regions. Seston and cladoceran PUFA contents were uncorrelated in the boreal zone where cladoceran preferred to feed on high-quality algae. European perch (*Perca fluviatilis* (L.)) showed slight decreasing trends in their muscle DHA content from more transparent lakes towards shallower and murkier ones in the boreal areas. In subarctic climate–productivity gradient, the decreasing prey item quality (zooplankton and profundal benthos) did not affect fish muscle EPA+DHA content at community level. Perch in boreal low pH lakes and highly forested catchments rely more on terrestrial energy sources than eutrophic lakes with neutral pH. This likely resulted in higher mercury and omega-6 PUFA content in perch muscle. Increasing temperature and productivity fundamentally alter subarctic lake communities' structure and function, resulting in an increasing share of cyanobacteria, smaller-bodied zooplankton, smaller benthos taxa, and warmer-water-adapted cyprinid fishes. Positive trends in biomass at each second trophic level (phytoplankton and invertivorous fish) were observed along with climate-productivity gradient shaping biomass pyramids. Food web processes and PUFA dynamics seem to differ between boreal and subarctic lakes. Future studies with harmonized methodology and wide lake gradients are needed to evaluate wheter methodology or ecology are driving these observed differences.

Keywords: Allochthony; browning; eutrophication; forestry; fish; mercury; nutritional quality.

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TIIVISTELMÄ

Keva, Ossi
Ympäristömuutoksen vaikutukset järviravintoverkkojen rakenteeseen ja ravitsemukselliseen laatuun
Jyväskylä: Jyväskylän yliopisto, 2022, 55 s.
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Ilmastonmuutos ja voimakas maankäyttö edistävät järvien rehevöitymistä ja tummumista vaikuttaen eliöiden yhteisörakenteeseen ja ravintoverkkoprosesseihin. Tässä väitöskirjassa tutkin ympäristömuutoksien vaikutuksia järvien ravintoverkkojen rakenteeseen, energiavirtoihin ja ravitsemukselliseen laatuun (eliöiden rasvahappo- ja elohopeapitoisuus) subarktisilla ja boreaalisilla vyöhykkeillä. Eikosapentaeeni- (EPA) ja dokosaheksaeenihappo (DHA) ovat muutamien leväryhmien tuottamia tärkeimpiä omega-3 monityydyttymättömiä rasvahappoja (PUFA) edistäen kuluttajien kasvua ja lisääntymistä. Elohopea on myrkyllinen raskasmetalli, joka kertyy eliöihin ravinnosta. Subarktisissa järvissä lämpeneminen ja tuottavuuden kasvu alensi kasvi- ja eläinplanktonyhteisöjen EPA+DHA-pitoisuutta. Boreaalisissa järvissä vastaavia muutoksia ei havaittu. Sestonin ja vesikirppujen PUFA-pitoisuudet eivät korreloineet boreaalisella vyöhykkeellä, jossa vesikirput valikoivat hyvälaatuisia leviä ravinnokseen. Boreaalisilla alueilla ahvenen (*Perca fluviatilis* (L.)) DHA-pitoisuus laski hieman kirkasvetisistä matalampiin tummavetisiin järviin. Subarktisella ilmasto-tuottavuus-jatkumolla kalayhteisön ravinnollinen laatu ei heikentynyt, vaikka niiden ravintokohteiden, eläinplanktonin ja syvänteen pohjaeläinten, EPA- ja DHA-pitoisuudet alenivat. Boreaalisissa happamissa metsäjärvissä ahventen energiasta suurempi osa oli peräisin maalta kuin järven sisäisestä tuotannosta, mikä oletettavasti nosti ahventen elohopea- ja omega-6 PUFA-pitoisuuksia. Subarktisissa järvissä lämpötilan ja tuottavuuden kasvu lisäsi sinibakteerien, pienten vesikirppujen, pienten pohjaeläinten ja lämpimiin vesiin sopeutuneiden särkikalojen osuutta eliöyhteisöissä. Samalla biomassa kasvoi erityisesti joka toisella trofiatasolla (kasviplankton ja selkärangattomia syövät kalat) muuttaen järvien biomassapyramidien muotoa. Ravintoverkkoprosessit ja rasvahappojen siirtyminen vaikuttavat eroavan eri lämpötilavyöhykkeillä. Jatkotutkimuksia yhtenäistetyin menetelmin tarvitaan selvittämään johtuvatko havaitut erot menetelmällisistä vai ekologisista tekijöistä.

Avainsanat: Alloktonia; elohopea; kala; tummuminen; metsätalous; ravinnollinen laatu; rehevöityminen.

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ORIGINAL PAPERS

LIST OF ORIGINAL PUBLICATIONS

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I Keva, O., Litmanen, J., Kahilainen, K.K., Strandberg, U., Kiljunen M., Hämäläinen, H., Taipale, S.J. Herbivorous cladoceran essential fatty acid content and diet across a phosphorous and DOC gradient of boreal lakes—importance of diet selection. Manuscript.

II Keva, O., Kiljunen, M., Hämäläinen H, Jones, R.I., Kankaala, P., Kahilainen, K.K., Laine, M.B., Schilder, J, Strandberg, U., Vesterinen J., Taipale, S.J. 2022. Allochthony, fatty acid and mercury trends in muscle of Eurasian perch (*Perca fluviatilis*) along boreal environmental gradients. *Science of the Total Environment* 838, 155982.

III Keva, O., Taipale, S.J., Hayden, B., Thomas, S.M., Vesterinen, J., Kankaala, P., Kahilainen, K.K. 2021. Increasing temperature and productivity change biomass, trophic pyramids and community level omega-3 fatty acid content in subarctic lake food webs. *Global Change Biology* 27: 282–296.

TABLE 1. Contributions of all authors in the original papers. Author abbreviations: AA = All authors, BH = Brian Hayden, HH = Heikki Hämäläinen, JL = Jaakko Litmanen, JS = Jos Schilder, JV = Jussi Vesterinen, KKK= Kimmo K. Kahilainen, MBL = Miika B. Laine, MK = Mikko Kiljunen, OK = Ossi Keva, PK = Paula Kankaala, RIJ = Roger I. Jones, SJT = Sami J. Taipale and SMT = Stephen M. Thomas.

	I	**II**	**III**
Original idea	SJT	PK, SJT, RIJ	KKK, SJT, OK
Planning	OK, SJT	OK, RIJ, HH, KKK, SJT, MK	OK, KKK, SJT
Field work	SJT, MK	MK, JS	KKK, BH, SMT, OK
Laboratory work	OK, SJT	MK, JS, OK	KKK, BH, SMT, OK
FA, THg and SI analyses	OK, SJT	OK, MK, KKK, MBL	OK, SJT, JV, SMT
Data cataloguing	OK	OK, MK	KKK, OK
Data analysis	OK, JL	OK	OK
First version of the manuscript	OK	OK	OK
Manuscript revisions	AA	AA	AA
Correspondence	OK	OK	OK

1 INTRODUCTION

1.1 Environmental challenges in boreal and subarctic lakes

In subarctic and boreal regions, lakes are facing environmental stressors such intense catchment area land use modification and climate change. These antropogenic-induced stressors affect lake chemistry with subsequent impacts on lake physio-chemical processes, community structures and lake food web processes (Creed *et al.* 2018). Lake browning is a phenomenon where lake water shifts toward browner color with increasing organic matter and iron concentration (Creed *et al.* 2018, Blanchet *et al.* 2022). Increasing nutrient content, especially phosphorous, has been shown to induce lake algal production (eutrophication) and modify the algal community (Schindler 1978, Kosten *et al.* 2012, Przytulska *et al.* 2017). Both lake browning and eutrophication have major impacts on the energy dynamics of lakes, lake productivity, and community structures at many trophic levels (Creed *et al.* 2018). Browning affects the light atmosphere of the lakes, narrowing the water layer available to photosynthesis and primary production (Ask *et al.* 2012, Seekel *et al.* 2015). Lake eutrophication has been shown to shift primary production balance from benthic algae production towards pelagic production (Vadeboncoeur *et al.* 2003, Vadeboncoeur *et al.* 2008, Hayden *et al.* 2019). Moreover, lake browning may impact the energy dynamics of the lake as the bacterial loop is able to transfer the humic substances to biomass further consumable by higher trophic levels (Creed *et al.* 2018, Blanchet *et al.* 2022). Anthropogenic intense land-use practises in lake catchment area such as, intensive forestry (e.g. ditching and clear cuttings) and agriculture, have been shown to increase organic matter flow and nutrient leaching from terrestrial environments to lake ecosystems often simultaneously (Kauppi 1985, Škerlep *et al.* 2020, Finer *et al.* 2021, Lepistö *et al.* 2021, Holopainen & Lehikoinen 2022). Also, recovery from acidification has been shown to contribute lake browning, where decreasing sulfate concentrations increase DOC coagulation and thus sedimentation and (Monteith *et al.* 2007, Lepistö *et al.* 2021). In addition to catchment area land-use modificitaion changes in climate has the potential to modify lake chemistry and further food we processes.

In subarctic and boreal Fennoscandia the mean annual air temperature has increased by approximately 2°C since 1850, being much higher compared to the global average ~1°C (Cohen *et al.* 2014, IPCC 2014, 2021). Global climate change has increased lake surface water temperatures by ca. 0.3 C per decade during past decades (Dokulil *et al.* 2021; O'Reilly *et al.* 2015; Woolway *et al.* 2019). Temperature and precipitation are predicted to increase in Fennoscandian regions by +2–7°C and +5–30%, respectively, by the end of the 21st century (IPCC 2014: RCP2.6—RCP8.5), ca. 1.5 times higher compared to the global average (Ruosteenoja *et al.* 2016). This predicted increase in air temperature likely also elevates the temperatures of surface waters affecting the lake food web structure by providing more suitable environments to warmer water adapted species but reducing the abundance of colder water adapted species (Kosten *et al.* 2012, Jeppesen *et al.* 2010). Higher precipitation has been linked to elevated nutrient, and organic matter flows from the catchment area to lakes (de Wit *et al.* 2016, Weyhenmeyer *et al.* 2016, Lepistö *et al.* 2021), which has documented impacts on food web communities.

1.2 Food web structures along eutrophication, browning and warming

Each species has their own fundamental niche, the range of different environmental parameters that they can survive and reproduce. These environmental parameters consist of temperature, nutrients, light atmosphere, and ecosystem size (Grinnell 1917). Hutchinson (1957) established a modern concept of the species niche, multidimensional hypervolume, where Grinnellian fundamental niche was reduced to a realized niche. This was based on the idea that species will occupy a position in the food web only where they are competitively superior, taking into account predation as well (Hutchinson 1957).

In lake food webs, nutrients, DOC, light regime, and temperature are probably the most critical factors governing the species dispersal and community structure of phytoplankton species. Total phosphorous (tot-P) has been shown to be an important factor defining phytoplankton productivity and community structure, where higher tot-P concentration in lakes promotes cyanobacterial dominance and lower tot-P value lakes have diatom dominance (Schindler 1978, Arvola *et al.* 2011, Przytulska *et al.* 2017). Also, in lakes with moderate tot-P concentration, increasing temperatures promote cyanobacterial dominance in the phytoplankton community (Kosten *et al.* 2012). Moreover, smaller phytoplankton taxa and cells have been found in warmer environments (Zohary *et al.* 2021). Browning favors mixotrophic phytoplankton taxa such as dinoflagellates and cryptomonads (Urrutia-Cordero *et al.* 2017, Senar *et al.* 2021). Lake primary production is unimodally related to DOC concentration of lake water, where the highest primary production rates are around 5mg l^{-1} of DOC. This is caused by the increased availability of nutrients below the threshold and decreased light availability above the threshold value (Seekell *et al.* 2015).

There are also differences in zooplankton's size range and community structure along environmental gradients. For example, large-sized copepods are more abundant in northernmost regions, whereas smaller-sized cladoceran is more typical in eutrophic and warm areas (Carter *et al.* 2017, Hayden *et al.* 2017). Zooplankton biomass in lakes has been shown to increase along with lake tot-P concentration (Hanson and Peters 1984); however, zooplankton biomass response to lake browning and nutrient enrichment does not always follow phytoplankton biomass (Bergström *et al.* 2021). This has been suggested to be a bottom-up mediated process by the decreased nutritional value of the phytoplankton to zooplankton (Bergström *et al.* 2021). However, standing biomass stock of zooplankton does not represent annual production, and top-down predation has been shown to decrease zooplankton biomass and size (Brooks and Dodson 1965, Amundsen *et al.* 2009). Moreover, warming and nutrient elevated primary production promotes the biomass of higher trophic levels (Jeppesen *et al.* 2000), where primary consumer invertebrates are a crucial energy transfer link between primary producers and fish.

Warming may result diversity loss in BMI community (Li *et al.* 2014). For example, ephemeropterans and plecopterans spatial distribution range may shrink (Bhowmik and Schäfer 2015). Browning impacts benthos community structure, e.g. reducing Plecoptera but increasing Chironomid contribution (Kesti *et al.* 2021). Benthos biomass has been shown to increase along lake total phosphorous content (Hanson and Peters 1984, Blumenshine *et al.* 1997). However, benthos size have been shown decreases with increasing predation (Blumenshine *et al.* 2000).

Smaller-sized perch (*Perca fluviatilis* L.) and roach (*Rutilus rutilus* L.) are predicted to increase their contribution in invertivorous fish community in European subarctic regions along eutrophication and warming (Jeppesen *et al.* 2012, Hayden *et al.* 2017). In even more eutrophic lakes (50—400µg l^{-1}), perch contribution declines (Jeppesen *et al.* 2000), and cyprinid biomass increases. Moreover, in Jeppesen *et al.* (2000) study, increment in predatory fish biomass along with eutrophication was less steep than invertivorous fish, resulting in proportional decrement of piscivorous fish. Predatory fish abundance in Finnish subarctic lakes has been shown to be lower in more oligotrophic lakes than in more eutrophic ones (Hayden *et al.* 2017, 2019). Piscivorous fish community structure also faces significant changes in the community structure along environmental gradients: Salmonid dominated systems shift towards pike and perch dominated (Hayden *et al.* 2019) and further to pikeperch (*Sander lucioperca* (L.)) dominated systems in temperate lakes (Jeppesen *et al.* 2000). 75% of cold water favoring Arctic charr (*Salvelinus alpinus* (L.)) dispersal (range of habitat) has been predicted to vanish by the end the century due to increased temperatures and invasions competitive predator pike (*Esox lucius* L.) (Byström *et al.* 2007, Hein *et al.* 2012). In addition to the nutrients, temperature have been shown to impact the size range of fish. In colder environments, fish species typically reproduce later in life, thus growing into a larger body size before sexual maturation (Heibo *et al.* 2005, Blanck and Lamouroux 2007, Estlander *et al.* 2017). Generalist fish species tend to have highest individual specialization at the most optimal temperature and nutrient circumstances (Sánchez-Hernández *et al.* 2021). Higher lake temperatures also induce fish omnivory (Jeppesen *et al.* 2010).

Food web theories such as "Green world hypothesis" have suggested that primary producers are bottom-up controlled by nutrient and light availability, herbivores are top down controlled by predators which are in turn resource limited (Hairston *et al.* 1960). The top-down control of herbivores is suggested to be the main reason why we see plants and other primary producers around the globe; if herbivores were only resource-limited, they would cyclically exploit the primary producers (Hairston *et al.* 1960). Terrestrial food chains are considered ca. one trophic level shorter than aquatic ones (Hairston and Hairston 1993). In lake ecosystems, primary producers are phytoplankton, benthic algae and macrophytes, primary consumers are zooplankton and benthos, predators are generally considered fish (Jeppesen *et al.* 2000, Carpenter *et al.* 2001). With these trophic dynamics, the standing stock biomass of each second trophic level (primary producers and predators) should be high (Paine 1980, Oksanen *et al.* 1981, Carpenter *et al.* 1985, Carpenter *et al.* 2001). Yet, the biomass does not directly indicate the production of different trophic levels. As there are differences in the longevity of organisms, biomass pyramids may be, in some cases, top-heavy (McCauley *et al.* 2018). However, as organism metabolism undeniably consumes energy, approximately 10% of produced biomass is converted to next trophic level biomass (Elton 1927, Brown *et al.* 2004). Thus, the year-round biomass production pyramids should take a regular pyramid shape.

1.3 Fatty acids in food webs

Fatty acids (FA) are fundamental biomolecules for cellular function. FAs are carbon chains attached to a carboxyl group with varying lengths and number and ordinations of carbon double bonds (Fig. 1). Fatty acids can be grouped based on the number of carbon double bonds in the backbone of the molecule: FAs without any double bonds in the termed saturated fatty acids (SFA), FAs with one double bond are monounsaturated fatty acids (MUFA), and fatty acids with two or more double bonds are termed as polyunsaturated fatty acids (PUFA). Furthermore, the place of the first double bond from the terminal methyl group is used to classify fatty acids into different omega groups (Fig. 1). Long-chain FAs here refer to fatty acids with ≥ 20 carbons in the backbone. Each fatty acid has a common name (e.g. docosahexaenoic acid), its abbreviation (DHA), and omega-name (22:6n-3) (Fig.1).

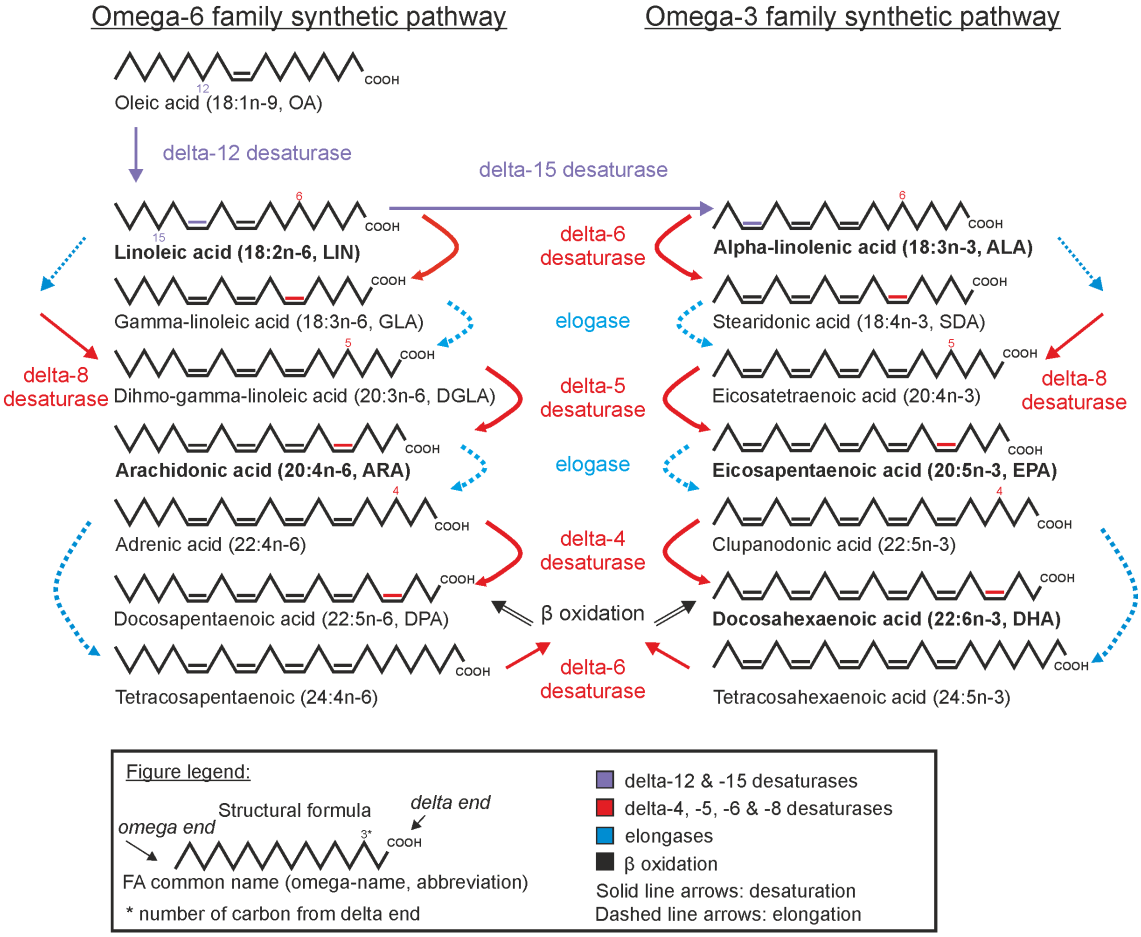

FIGURE 1 Simplified omega-6 and omega-3 fatty acid pathways (modified from Monroig *et al.* 2022). Structural formula, common names, omega names and abbreviation, are shown for each fatty acid. For simplicity, delta-8 pathway and Sprecher pathway (β oxidation) intermediate products are not shown. Desaturation processes are illustrated with solid line arrows (purple or red), elongation processes with dashed line arrows (blue) and β oxidation with black solid arrows. Purple colors indicate desaturase processes that are present only in low trophic level organisms. Colored numbers on top of the structural formula indicate the position from the delta end (carboxyl end) where an additional double bond will be added during desaturation. The newly added double bonds are highlighted with colored carbon to carbon double bonds in the end product. The fatty acids in the focus of this PhD thesis are bolded.

FAs are used, for example, as cellular membrane building material (phospholipids), cellular energy source (oxidation), energy storage (TAG), hormone precursors (e.g., eicosanoids), they are also impacting cellular signaling pathways (Arts and Kohler 2009). PUFAs, especially the long-chain ones, induce cell membrane fluidity, while shorter SFAs and MUFAs induce membrane stiffness (Guschina and Harwood 2006, Calder 2015). LIN and ALA serve as precursors for biologically more active long-chain PUFAs such as EPA, DHA, and ARA, which are used in the production of hormone and signaling molecules but are also crucial in adjusting the fluidity in cellular phospholipid membranes

(Arts and Kohler, 2009). FAs, especially EPA and DHA of the omega-3 PUFAs, have numerous beneficial health effects and are essential for neural and gonadal development (Connor 2000, Arts *et al.* 2001, Calder, 2015). These vital PUFAs are retained mainly from the consumer diet to maintain growth, neural function, and reproduction (Müller-Navarra *et al.* 2004, Brett *et al.* 2009, Peltomaa *et al.* 2017).

In lake ecosystems, phytoplankton and benthic algae are the main producers of PUFAs as animals lack the enzymes to convert SFA and MUFA to PUFAs (Henderson 1996, Cook and McMaster 2004, Bell and Tocher 2009, Galloway and Winder 2015). Thus, LIN and ALA are essential for consumers since they cannot biosynthesize these fatty acids (Arts *et al.* 2001). However, previous findings have shown that zooplankton and fish can biosynthesize long-chain PUFAs from their shorter counterparts (Geay *et al.* 2016, Ishikawa *et al.* 2019, Cheguaceda *et al.* 2020, Pilecky *et al.* 2022). However, this is thought to be energetically expensive and occurs mainly in situations where dietary long-chain PUFAs are not available in sufficient amounts to meet the physiological requirements of the consumer (Bell andTocher, 2009, Twinning *et al.* 2016, Scharnweber *et al.* 2021).

Phytoplankton taxa differ in their ability to synthesize different omega-3 and omega-6 PUFAs. Generally, phytoplankton groups can be classified non-EPA and non-DHA, EPA- and DHA-producers. For example, cryptomonads (Cryptophyceae), diatoms (Bacillariophyceae), dinoflagellates (Dinophyceae), and euglenoids (Euglenophyceae) have typically high EPA and DHA content (Taipale *et al.* 2013, Peltomaa *et al.* 2017). Green algae (Chlorophyceae, Conjugatophyceae, and Trebouxiophyceae) and cyanobacteria do not usually contain long-chain EPA and DHA (Taipale *et al.* 2016, Peltomaa *et al.* 2017), but can have relatively high amounts of ALA and SDA and saturated fatty acids. However, there are also taxa-specific variations in the biomolecule contents within the previously listed taxonomic groups (Ahlgren, Gustafsson, and Boberg 1992, Taipale *et al.* 2013, Peltomaa *et al.* 2017). As described earlier, environmental change can alter phytoplankton community structure and thus the transfer of PUFAs to higher trophic levels. Decreasing the EPA and DHA content of seston and zooplankton along eutrophication has been observed in natural lake systems and mesocosms studies (Müller-Navarra *et al.* 2004, Taipale *et al.* 2019, Trommer *et al.* 2019). Community structure at class/genus level has been suggested to be one of the main drivers for altering the FA content of the community (Kainz *et al.* 2004; Taipale *et al.* 2019).

Consumers do not modify their dietary fatty acids if not needed, thus they are suitable biomarker for ecologists (Dalsgaard *et al.* 2003). FA usage as biomarkers has the highest potential to model the diet of short-lived organisms as legacy FAs can confuse the interpretation in longer-lived organisms, such as fish (Galloway and Budge 2020, but see Jardine *et al.* 2020). Some FAs are not broken in animal digestion and metabolism and thus are suitable for biomarkers (Dalsgaard *et al.* 2003, Iverson *et al.* 2004, Iverson 2009). For example, branched fatty acids are synthesized by Actinobacteria and are transferred in the food web without modification (Taipale *et al.* 2012). Several 16C PUFAs are potentially good biomarkers for many different algal groups (Taipale *et al.* 2013). Generally, omega-3 PUFAs are thought to be mainly phytoplankton origin, and low omega-3:omega-6 FA indicates terrestrial derived energy sources (Lau *et al.* 2012, Hixson *et al.* 2015, Taipale *et al.* 2015). In addition to single usage of FAs as biomarkers, there is also fatty acid profile based on methods for estimating phytoplankton community structure and consumer's diet composition (Iverson *et al.* 2004, Galloway *et al.* 2014, Strandberg *et al.* 2015, Bromaghin 2017, Taipale *et al.* 2019, Litmanen *et al.* 2020, Jardine *et al.* 2020). These estimation tools use previously determined laboratory culture and feeding experiment-derived FA profile reference libraries to estimate the community or dietary structure (Litmanen *et al.* 2020).

Long-chain n3 FAs, especially EPA and DHA benefit fish growth and reproduction (Arts and Kohler 2009, Henrotte *et al.* 2010, Geay *et al.* 2016). These vital biomolecules are also beneficial for human neural development, neural and immune function, also decreasing e.g., cardiovascular disease risks (Swanson *et al.* 2012). Some studies have suggested that reducing PUFA trends in the primary producers, and primary consumers along environmental change would cascade to secondary and tertiary consumers, potentially impacting the quality for human consumed aquatic taxa as well (e.g., Taipale *et al.* 2016, Colombo *et al.* 2020).

1.4 Mercury in food webs

Mercury (Hg) is an easily volatile metal, and once entered to the atmosphere it is distributed globally via air currents (Pacyna *et al.* 2010). In northern regions, atmospheric mercury deposition is the primary mercury source in food webs as direct point mercury emissions are largely lacking (AMAP 2011, 2021, Stern 2012). Energy production (burning of coal), mining, metal, and construction industry are the primary anthropogenic mercury emission sources (Pacyna *et al.* 2010, Streets *et al.* 2019). Approximately 30 to 80% of the mercury in the atmosphere originate in anthropogenic emissions; however accurate estimations are challenging to establish as mercury has a long lifetime in the atmosphere and biosphere, and re-emissions of the historic mercury loads are hard to evaluate comprehensively (Downs *et al.* 1998, Pirrone *et al.* 2010, Outridge *et al.* 2018). Mercury emissions to the atmosphere have declined regionally in developed countries in the past decades, but globally the mercury emissions have increased (Outridge *et al.* 2018, Streets *et al.* 2019, Wang *et al.* 2019). Atmospheric mercury deposits to terrestrial and aquatic environments via dry and wet deposition (Pacyna *et al.* 2010). Historical deposition of mercury to lake catchment areas enhances the toxic heavy metal load to lakes as runoff from catchment is the main source of aquatic mercury (Grigal 2002), which is further promoted by land-use modification such as clear cuttings and peatland ditching (Porvari *et al.* 2003, Ahonen *et al.* 2018, Eklöf *et al.* 2018). After the industrial revolution, mercury levels in lake sediments have increased (Chételat *et al.* 2015). At a regional scale, point sources of mercury emissions in Finland have decreased in the past decades due to the banning of Hg usage in paper and pulp industries (Lodenius 1991); these actions have lowered the Hg levels in pike and perch (Braaten et al. 2019).

In lakes and catchment areas, inorganic mercury can be methylated to a more harmful compound, methyl mercury (MeHg), mainly by bacterial activity (Eckley *et al.* 2005, Bravo *et al.* 2017). Inorganic mercury and especially the more toxic organic methyl mercury (MeHg) bioaccumulate to organisms over time and biomagnifies in food webs towards higher trophic level organisms such as fish (Lavoie *et al.* 2013, Lescord *et al.* 2018). MeHg has a higher tendency to biomagnify in food webs, and thus a majority of fish mercury, especially in fish muscle, is MeHg (Lavoie *et al.* 2010; Lescord et al. 2018). Phytoplankton and other microbes passively uptake inorganic and methyl mercury from surrounding water (Morel *et al.* 1998, Benoit *et al.* 2001, Luengen and Flegal, 2009). Consumers receive the majority (ca. 90%) of their mercury from the diet (Hall *et al.* 1997, Tsui and Wang 2004), but ambient mercury levels have been shown to affect consumer mercury levels as well (Watras and Bloom 1992, Kelly *et al.* 2003; Porvari and Verta 2003).

Environmental change has been shown to impact aquatic food mercury content. DOC load from the catchment area has been shown to affect aqueous mercury and mercury levels in aquatic organisms (Driscoll *et al.* 1995, Watras *et al.* 1998; Ullrich *et al.* 2001). Lake browning and increasing food web reliance on allochthonous energy sources have shown lift mercury levels in primary producers, zooplankton, and fish (Watras *et al.* 1998, Strandberg *et al.* 2016, Poste *et al.* 2019, Wu *et al.* 2021). Contrary to DOC, increasing phosphorous content has been suggested to lower mercury content in organisms through biodilution (Sonesten 2003, Razavi *et al.* 2015, Finley *et al.* 2016, Kozak *et al.* 2021).

Mercury toxicity to aquatic invertebrates and fish has been evaluated, where higher mercury content has resulted in lower biomass of different organisms (Depew *et al.* 2013, Rodrigues *et al.* 2013). However, the health impact for humans is much better known, and mercury cause e.g. neurological disorders and fatalities in high doses and chronic exposures (Ratcliffe *et al.* 1996).

1.5 Allochthony of lake food webs

In lake food webs, allochthony refers to a ratio how much of food web energy or biomass originates from organic matter production from the catchment area (DOM, allochthonous production) and in-lake production (primary production, autochthonous production). In lakes, heterotrophic bacteria and protozoans can consume terrestrial organic matter that zooplankton is able to harvest (Hiltunen *et al.* 2017). Hydrogen isotopes can be used as a tool to trace the flow of autochthonous and allochthonous matter in food webs as there are marked differences between hydrogen stable isotope rations between the two sources (Doucett *et al.* 2007, Karlsson *et al.* 2012, Wilkinson *et al.* 2015). Terrestrial plants have approximately 100‰ higher hydrogen isotope ratios than aquatic algae, as the evaporation process from terrestrial plant stomata favors lighter hydrogen (Doucett *et al.* 2007). The differences δ^2H values between the two sources allow ecologists to estimate consumer hydrogen sources with mixing models (Karlsson *et al.* 2012). In addition to dietary hydrogen sources, a proportion of hydrogen from the surrounding environment is fixed to organism tissues (e.g., Vander Zanden *et al.* 2016). Thus, the trophic level difference between sources and consumer and hydrogen isotope values of surrounding water needs to be considered when modeling the proportion of consumer hydrogen sources (e.g., Vander Zanden *et al.* 2016).

1.6 Space-for-time substitution as a study method

Nature, biomes, and ecosystems tend to change over time in various ways. Anthropogenic impacts such as landscape modification and accelerating climate change has a myriad of effects on terrestrial and aquatic ecosystems. If the predicted environmental shift in time corresponded to the spatial gradient in environmental factors, space could be used to replace time to model future change (e.g., Pickett 1989). Space-for-time substitution is a widely used study method in ecological studies to bypass the problem with the lack of long-term datasets (Jeppesen *et al.* 2000, Taipale *et al.* 2016, Hayden *et al.* 2019, Laske *et al.* 2019; Senar *et al.* 2021). Long-term datasets, including relatively new techniques such as fatty acid content analysis, are still scarce (but see Lind *et al.* 2018) but would give presumably the most precise predictions when modeling future change in ecosystems, at least in a local scale (Blois *et al.* 2013).

Space-for-time substitution have caveats, for example: i) In the observed gradient some communities might have reached steady state whereas others are still developing (e.g. natural invasion of species) (Damgaard, 2019; Jeppesen *et al.* 2014) ii) Other confounding factors in the used spatial gradient impacting the observed phenomenon (Jeppesen *et al.* 2014). These confounding factors in freshwater ecosystems studies may include, for example, differences in lake morphology (area, depth, and shoreline shape) water resilience time. iii) Lack of causal evidence, providing only interferential, not mechanistic evidence of factors behind the observed phenomenon (Jeppesen *et al.* 2014). However, the mechanisms behind the observed correlations can be discussed and deduced based on, e.g., laboratory experiments. Moreover, space for time study approaches provides typically spatially high-resolution data from nature to future meta-analyses. Even with some caveats, future predictions are still an essential tool for mapping the possible changes in ecosystems and biota, giving the possibility to global and local cooperation organizations and alliances to react to the potential unwanted or harmful changes in nature.

Environmental gradients matching predicted climate change (**III**) and eutrophication (**I** & **III**) and browning (**I**-**III**) of boreal and sub-arctic lakes was used in all the studies of this PhD thesis. Even though we are not drawing any timely detailed predictions on how future global change will alter boreal and sub-arctic lake food webs, with these used space-for-time substitutions here, we can point the direction of the changes in food webs.

1.7 Main aims of the thesis

The main aim of this thesis is (Fig. 2): to reveal how environmental gradient affects the nutritional quality of seston, zooplankton, and fish in boreal and subarctic regions. Additionally, evaluating how climate and land use affect lake food web structures. More detailed hypotheses are described below.

In **I**, lake eutrophication (increasing tot-P) was expected to negatively impact the long-chain PUFA content of herbivorous due to decreased quality of phytoplankton (Müller-Navarra *et al.* 2004, Taipale *et al.* 2016). As the proportion of EPA and DHA synthesizing phytoplankton taxa is supposedly low in more eutrophic lakes, cladoceran diet preference of these taxa would increase with lake productivity to maintain viable cladoceran populations.

In **II, i**ncreasing lake DOC concentration and forest cover share in the catchment was hypothesized to correlate positively and lake productivity negatively with perch allochthony. In addition, small forest lakes with high DOC and allochthony, perch muscle THg and n6 PUFA contents was hypothesized to be higher and n3 PUFAs content lower compared to more productive and larger lakes (Hixson *et al.* 2015, Taipale *et al.* 2016, Rask *et al.* 2021) affecting the overall quality of perch muscle for human consumption.

In **III**, it was expected that lake communities shift towards warmer and more murky-water-adapted taxa, with a concurrent increase in food web biomass, and that increases in algae and fish biomass along with a environmental gradient would have effects on trophic pyramid shape (Jeppesen *et al.* 2010, Hayden *et al.* 2017, McCauley *et al.* 2018). Cyanobacteria form an increasing proportion of the phytoplankton communities in warmer and more productive lakes (Kosten *et al.* 2012, Przytulska *et al.* 2017), thus lowering the nutritional quality of primary producers (Müller-Navarra *et al.* 2004). I expected that the downgrading quality of primary producers and primar consumers would cascade to higher trophic levels (Müller-Navarra *et al.* 2000; Taipale *et al.* 2016).

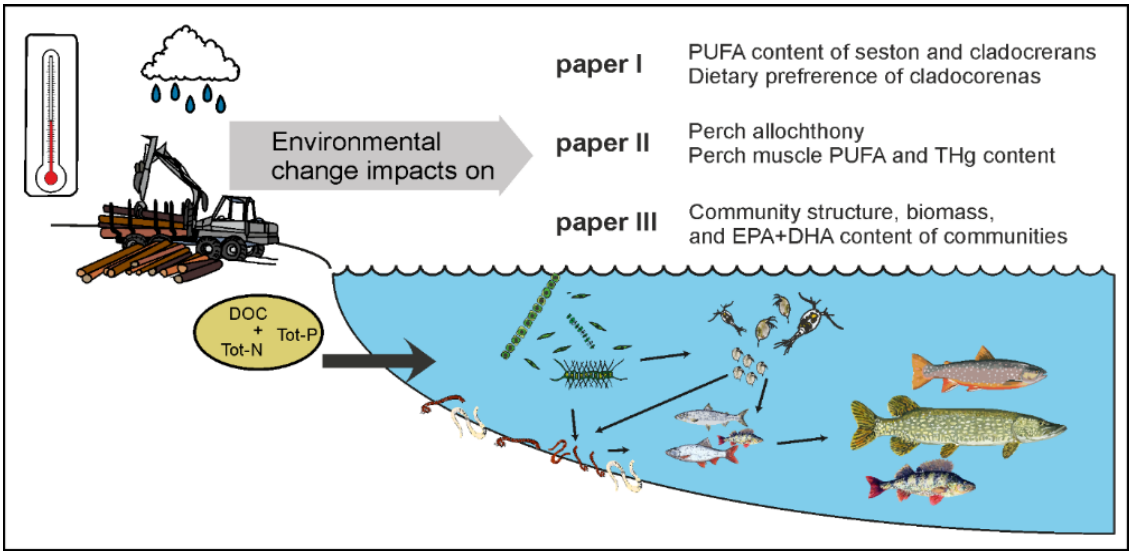

FIGURE 2 Graphical illustration of the main objectives of this PhD thesis.

2 MATERIALS AND METHODS

2.1 Study region

All the study lakes in this PhD thesis are located in Finland (Fig. 3). As the goal for each different study differed, also the type of lake gradients differed (Table 2). The study region of **I** and **II** consisted of Southern and Central Finland lakes (Fig. 3). And the study region of **III** covered lakes in Lapland. Sampling of the different lake gradients took place in late summer for all the studies between years 2010 and 2020. Please see more detailed descriptions of the study regions from the corresponding original papers **I–III**.

2.2 Data collection – Field work

2.2.1 Lake water chemistry, morphometrics, and land-use

In all the studies (**I-III**), lake chemistry such as total phosphorous (tot-P), total nitrogen (tot-N) dissolved organic carbon (DOC), chlorophyl-a was analysed from the sampled lakes (Table 2). In a case where samples were not taken, lake chemistry parameters were derived from Finnish Environment Institute (HERTTA-database). In **III,** long-term (1981) mean summertime air temperature and precipitation data were derived from nine Finnish Meteorological Institute weather stations along the study region (Table 2). For all the studies, lake morphological parameters such as size (lake area) and depth (mean and maximum) were derived from HERTTA and open terrain maps (National Land Survey of Finland). For **I** and **III**, land-use data and catchment area size data was retrieved with VALUE-tool (Finnish Environmental Institute), which uses CORINE (2012, level 3) land cover map. Here percent coverage of five major land cover types was categorized as: urban, agriculture, forest, wetland, and water areas (Table 2). For **II,** catchment area characteristics were derived from CORINE 2018 land cover (level 4) raster maps using geographical information system software (QGIS 3.16.13), where the resolution of different land-use types is higher compared to level 3 maps. The 46 different land-use types derived with level 4 raster maps were compiled to the previously mentioned five main categories plus peat-land forest cover (Table 2).

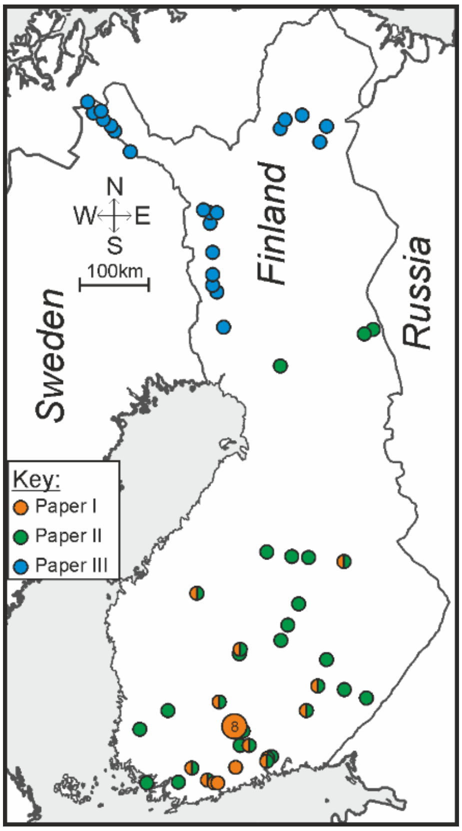

FIGURE 3 Map of the study area. The colors of the dots indicate lakes included in each original paper **I-III**. A detailed description of the studied lake gradients can be found in Table 2.

TABLE 2 Study region description of different PhD chapters. Article number and the number of lakes sampled are marked in the first column. The study lake range (min–max) of lake area (LA), lake mean depth (MD), total phosphorous (tot-P), total nitrogen (tot-N), chlorophyll a (Chl-a), catchment area (CA), forested area in the catchment (For.) and agriculture area in the catchment (Agr.) are listed in table columns.

Paper (lake n)	LA (km^2)	MD (m)	tot-P (µg l^{-1})	tot-N (µg l^{-1})	Chl-a (µg l^{-1})	DOC (mg l^{-1})	CA (km^2)	For. (%)	Agr. (%)
I – 21	0.004–107.5	1.2–21.1	4.0–149.0	300–2638	2.1–145.9	5.0–22.0	1.9–1936.8	39.8–99.1	0–33.2
II – 31	0.03–107.5	1.0–23.0	4.8–118.4	230–1470	1.8–72.2	5.0–24.3	1.9–1334.6	50.7–96.4	0–33.2
III – 20	0.3–1184.0	1.6–19.4	3.0–48.0	110–1100	1–24	1.9–14.9	20–11481	4.5–73.0	0–1.0

2.2.2 Sampling of seston, zooplankton, benthos and fish

For the seston samples in **I**, lake water was collected from epilimnion in the middle of the lake with a Limnos sampler and filtered through 50 μm mesh. The sieved water was filtered through a cellulose filter and freeze-dried. Zooplankton samples were collected from the same sites using a plankton net with vertical hauls. Live zooplankton samples were washed to a beaker with tap water, resulting in herbivorous cladoceran (Daphnia spp. and Bosmina spp.) to trap to the surface layer copepods to the bottom of the beaker. A glass plate was dipped into the beaker to pick cladocerans which after they were freeze-dried for FA analysis.

Quantitative phytoplankton taxa biomass data for **III** were extracted from the HERTTA database (Finnish Environment Institute) and from a previous study (Taipale *et al.* 2016). Shortly, phytoplankton community composition and biovolume were calculated with microscoping and taxa specific size to mass ratios, and dry mass per unit area was calculated (kg dw ha^{-1}) using lake-specific euphotic zone. Quantitative pelagic zooplankton samples for **III** were collected using a zooplankton net (diameter 25 cm, mesh size 50 μm) with three replicate vertical hauls from the deepest point of each lake (or in largest lakes, from the deepest point of the sampling area) and stored in a 5% formaldehyde solution (Hayden *et al.* 2017). Density and body size was determined by microscoping. The biomass of each identified zooplankton taxa was determined with microscoping and size to mass conversion factors (McCauley 1984). The biovolume of the zooplankton groups was transformed to areal biomass using lake-specific average depth.

Benthos was collected only from soft bottoms with an Ekman grab (area 272 cm^2) from a transect spanning from the littoral zone to the deepest point of lake or sampling area (**III**). Three separate replicates were taken from each depth, and animals were sorted into the lowest feasible taxonomic groups. Animals were counted, and the total wet biomass of each taxon was weighed. Samples were divided into littoral and profundal habitats based on water column light attenuation curves of the lakes. The habitat-specific benthos biomass estimations were done by weighing the taxa mass density with the littoral or profundal area contribution of each lake.

Fish samples were collected with gill net series for **II** and **III**. For **II,** only large-sized perch (Total length > 15cm) were selected for the subsequent analysis. For **III**, fish species were divided to insectivorous fish and piscivorous fish based on previous detailed diet analyses of fish communities (Thomas *et al.* 2017, Hayden *et al.* 2019). A piece of the dorsal muscle of fish (3-5 ind. taxa^{-1} lake^{-1}) was dissected and stored in plastic vials, freeze-dried, and stored in a deep freezer. In **III**, catch per unit effort (CPUE) was transformed to areal biomass estimations based echosounding and trawling by using regression formula (Malinen *et al.* 2014).

2.3 Laboratory analysis

2.3.1 Fatty acid analysis

Fatty acid analyses of seston filters or freeze-dried and powdered samples were conducted. Briefly, lipids of the samples were extracted and spiked with internal standard. the extracted fatty acids were methylated and analyzed with a gas chromatograph attached to a mass spectrometer. FAs were identified based on the ion spectra of the different compounds and quantified with external standards. The external calibration curves of FAME standard (566c, Nu Check Prep) were established for quantifying FAME concentrations. Only PUFA content of the samples is reported in **I** and **II** and EPA+DHA in **III**. For more detailed information about sample sizes and FA analyses, please see **I-III**.

2.3.2 Quantitative Fatty Acid Analysis (QFASA)

The basic idea in QFASA is to define a weighed mixture of prey item FA profiles that would resemble the consumer FA profile (Iverson 2009, Bromaghin *et al.* 2017). With species-specific long-term feeding experiments, consumer metabolism is taken into account, where discrimination factors for each fatty acid are produced (Iverson 2009). Prey mixtures closest to the discriminant factor weighed predator FA profile are estimated with statistical distance (e.g. Kullback–Leibler or $\chi 2$) (Iverson 2009). The measured FA profiles of seston and cladoceran were used to estimate seston and cladoceran diet compositions (**I**) with Quantitative FA Signature Analysis in R (QFASAR) (Iverson *et al.* 2004, Bromaghin 2017) with $\chi 2$ distance measure (Stewart *et al.* 2014). The method was applied with previously determined laboratory culture, and feeding experiment derived FA profile libraries (Galloway *et al.* 2014, Litmanen *et al.* 2020, Strandberg *et al.* 2015) that allowed to estimate seston composition and cladoceran diet (biomass %) of the following groups: diatoms, cryptomonads, chlorophytes, chrysophytes, euglenoids, dinoflagellates, cyanobacteria, raphidophytes, actinobacteria, methane-oxidizing bacteria (MOB), terrestrial organic matter (tPOM) and terrestrial organic matter consumed by microbes (mtPOM). The identified algal groups were divided to three quality groups (high, moderate, low) based on feeding experiments of *Daphnia* targeting on growth and reproduction responses on different diets (Peltomaa *et al.* 2017).

2.3.3 Total mercury analysis

Total mercury analyses were conducted with an atom absorption spectrometer by weighing freeze-dried perch muscle powder (25 ± 5 mg) to autosampler (**II**). Shortly, certified reference material (DORM-4, Research council of Canada) and blank samples were measured before and after each run. In addition, duplicates for each 5th sample were analyzed to test analytical error. As THg correlates linearly with total fish length and there were length differences in the sampled fish, the measured THg values were length corrected (Braaten *et al.* 2017):

$$THg_{weighed} = \frac{THg_{observed}}{TotL_{observed}} \times TotL_{mean}$$

, where $THg_{weighed}$ is the length adjusted THg values, $THg_{observed}$ indicates the analyzed THg values from the individual samples, $TotL_{observed}$ indicates measured individual total length and $TotL_{mean}$ indicates the mean of the total length of all sampled fish.

2.3.4 Stable isotope analyses

Stable isotope analyses were conducted for perch muscle, cladoceran, bulk zooplankton, inlet DOC, benthic algae, and lake water (**II**). Freeze-dried sample powder was weighed in tin cups (0.60 ± 0.05 mg) for stable nitrogen and in silver cups (0.350 ± 0.005 mg) for hydrogen isotope ratio analyses ($\delta^{15}N$, $\delta^{2}H$). $\delta^{15}N$ was analyzed with a continuous-flow isotope ratio mass spectrometer coupled to an elemental analyzer. $\delta^{2}H$ was analyzed with an isotope ratio spectrometer coupled to an elemental analyzer. Multiple samples of two reference materials were analyzed in each run. All stable isotope ratios are expressed as a ratio to the corresponding international standard (atmospheric nitrogen and VSMOW Standard Mean Ocean Water for nitrogen and hydrogen isotopes, respectively) using the delta notation.

2.3.5 Allochthony modelling

Consumer (large perch: TL > 15cm) allochthony was estimated based on hydrogen and nitrogen isotopes of the consumer and sources (**II**). The trophic level calculations were conducted with a simple one-source model (Post 2002):

$$TL_C = \frac{\delta^{15}N_C - \delta^{15}N_{base})}{\Delta_N} + \lambda$$

, where TL_C is the estimated trophic level of consumer. $\delta^{15}N_C$ and $\delta^{15}N_{base}$ are the measured nitrogen isotope values from consumer and base, respectively. Δ_N is the trophic fractionation of nitrogen isotopes between trophic levels and λ indicates the trophic level of the base, here zooplankton was used, and thus λ was 2. The allochthony calculations were done with slightly modified two-source mixing models according to Post (2002):

$$\text{allochthony index} = \frac{(\delta^2 H_C - \delta^2 H_{TL\,corrected\,base\ 2})}{(\delta^2 H_{TL\,corrected\,base\ 1} - \delta^2 H_{TL\,corrected\,base\ 2})}$$

, where allochthony index should vary between $0-1$ with higher values indicating predominant allochthonous hydrogen source. $\delta^2 H_C$ is the measured hydrogen isotope values from perch muscle. $\delta^2 H_{TL\,corrected\,base\ 1}$ and $\delta^2 H_{TL\,corrected\,base\ 2}$ indicate trophic level corrected allochthonous (inlet DOC) and autochthonous (phytoplankton) base hydrogen isotope values, respectively.

Please see the complete set of equations used for the allochthony index calculations in **II**.

2.3.6 Hazard quotient

There are obvious health benefits consuming long-chain omega-3 FAs and risks in mercury consumption for human health (Ratcliffe *et al.* 1996, Parrish 2009). Therefore, European Food Safety Authority (2012a, 2012b) has set daily minimum and maximum limits of n3 PUFA (250 mg EPA+DHA d^{-1}) and mercury (0.186 µg kg^{-1} body weight d^{-1}) consumption for humans. This provides a tool to combine these compounds' beneficial and adverse health impacts using a hazard quotient modeling (Gladyshev *et al.* 2009, Razavi *et al.* 2015, Strandberg *et al.* 2016). Hazard quotient (HQ) is a measure expressing the ratio of the harmful compound (here mercury) exceeding the limitations when fulfilling the daily requirement of beneficial compound (here EPA+DHA) (Gladyshev *et al.* 2009):

$$HQ = \frac{R_{EFA} \times C_{THg} \times 0.85}{C_{EFA} \times RfD \times AW}$$

, where R_{EFA} refers to daily recommended EPA+DHA supply for humans. C_{THg} is the THg content analyzed from perch muscle. The factor 0.85 was used to convert THg to methyl mercury content (e.g., Lescord et al., 2018). C_{EFA} is the measured EPA+DHA content of the perch muscle. RfD is the maximum tolerable daily intake of inorganic and methyl mercury, and AW is the average mass of an adult (here 70 kg was used). HQ > 1 represents a risk of adverse effects for human health when gaining all the required EPA and DHA from the fish, whereas HQ < 1 represents no risk (Gladyshev *et al.* 2009).

2.4 Statistical analyses

In **I**, Quantitative FA Signature Analysis (QFASA) was used to estimate seston composition and cladoceran diet composition. Correlation between lake chemistry-related parameters (tot-P and DOC) and seston community and cladoceran diet composition was tested with linear regression analysis. The relationship between lake chemistry and seston and cladoceran FA and cholesterol content was tested with linear regression analysis. Moreover, I tested if the predictive power of simple linear regression analysis on seston cladoceran biomolecule content could be enhanced with general linear models where lake chemistry and morphometrics were included. In **II** and **III**, Principal Component Analysis (PCA) was used to combine the studied environmental gradients (lake chemistry, morphometrics, and land-use data). In **II**, PC1 included forestry (−) and agriculture area (+) in the catchment, lake pH (+), and tot-P (+). In PC2, tot-P (+), DOC (+), average lake depth (−), and catchment area size (+) had the highest loadings for and thus was considered to represent a gradient from larger clear lakes toward smaller and more murky lakes. In **III,** PC1 combined open water season air temperature (+), tot-P (+), DOC (+), representing a climate-productive gradient. The formed PC components were subsequently used to model environmental change impacts on perch allochthony, muscle THg, FA content and hazard quotient (**II**), community-level biomass, and EPA+DHA content (**III**) with linear regression models. An alpha level of 0.05 was used in the statistical tests. All analyses were performed using R (R Core Team 2019), and results were visualized using ggplot2 -package (Wickham 2016).

3 RESULTS AND DISCUSSION

3.1 Cladoceran PUFA and diet across boreal tot-P and DOC gradients

In contrast to our predictions and previous findings (Müller-Navarra *et al.* 2004, Taipale *et al.* 2016), lake eutrophication (increasing tot-P) or browning (increasing DOC) was not found to downgrade boreal seston nor cladoceran PUFA quality (Fig. 4**, I**). Instead, sestonic LIN content increased, whereas sestonic ARA, ALA, EPA, and DHA contents were rather stable along tot-P and DOC and gradient in our dataset (Fig. 4, **I**). The DHA content of herbivorous cladoceran increased with tot-P, and LIN and ALA content increased along with lake DOC concentration (Fig. 4, **I**).

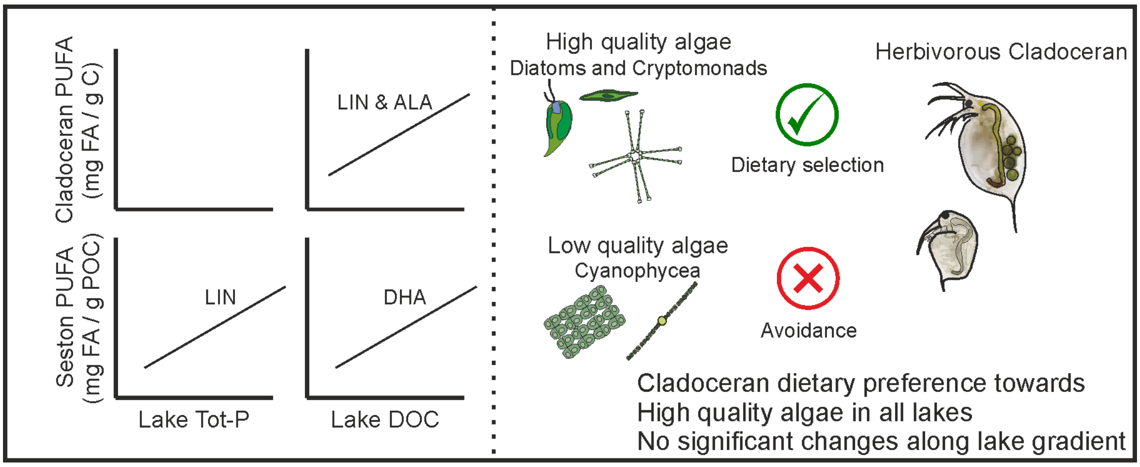

FIGURE 4 Graphical illustration of results and findings in **I**. PUFA= polyunsaturated fatty acids, POC = particulate organic carbon, LIN = linoleic acid, ALA = alpha linolenic acid, DHA = docosahexaenoic acid, Tot-P = total phosphorous and DOC = dissolved organic carbon concentration.

No relationship between sestonic and cladoceran PUFA contents was observed; indicating possible diet selection of cladoceran. However, this can be a also result of a time lag in the carbon turnover in *Daphnia*. Previously cladoceran diet shift has been shown to be visible in cladoceran FA composition in six-seven days (Taipale *et al.* 2009, Taipale *et al.* 2011). No clear trends in the community structure of seston along lake phosphorous gradient was found, but chlorophytes increased towards browner lakes (Fig. 5, 6, **I**). Interestingly, the seston composition results showed a very high contribution of cyanobacteria in the most oligotrophic lakes (Fig. 5), which presumably should include higher proportions of moderate and high-quality algae groups (Lepistö and Rosenström 1998, Taipale *et al.* 2016). However, there are bloom-forming cyanobacteria genera in oligotrophic lakes (Vuorio et al. 2020), and thus, phosphorous solely might not be the best predictor for phytoplankton community composition. Decreasing PUFA content in seston along the environmental gradients was not found, which could be explained by the single sampling from each of the lakes. Thus, occasional blooming and phytoplankton summertime succession could potentially have impacted the results, as decreasing PUFA content of seston has been studied in many other lake gradients (Müller-Navarra *et al.* 2004, Taipale *et al.* 2016, **III)**. Another alternative is that the space for time substitution here cannot trace all the differences between lakes. Thus, spatially distinct lakes with differing morphometry may react differently in nutrient supply, therefore not always showing similar phytoplankton community structure or quality trends.

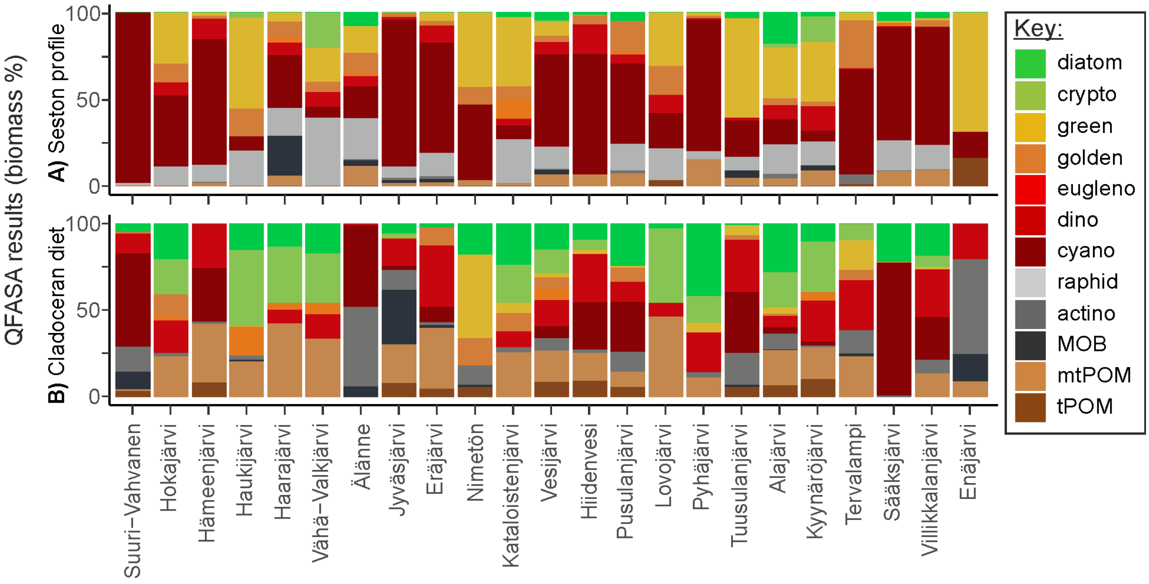

FIGURE 5 Seston algae biomass % composition (A) and herbivorous cladoceran diet biomass % composition (B) based on Bayesian posterior point estimate (median) results for each lake (A–B) (Modified from **I**). Lakes are sorted with increasing lake phosphorus content from left to right (A–B). Different groups are presented with different colors in the bar plots; the greener colors indicate better nutritional quality (A–B). Species abbreviations: diatom = diatoms, crypto = cryptophytes, golden = chrysophytes, green = chlorophytes, eugleno = euglenoids, dino = dinoflagellates, cyano = cyanobacteria, raphid = raphidophytes, actino = actinobacteria, MOB = methane-oxidizing bacteria, mtPOM = microbial terrestrial particulate matter, tPOM = terrestrial particulate organic matter.

In this study with 23 boreal lakes, herbivorous cladoceran preferred diatoms and cryptomonads in all lakes, whereas cyanobacteria were mostly avoided (Fig. 5, Fig. 6). These observations correspond with previous studies about cladoceran diet preferences (Lampert 1981, Gladyshev *et al.* 2000, Taipale *et al.* 2019) and laboratory experiment studies showing that diatoms and cryptomonads promote cladoceran growth (Peltomaa *et al.* 2017). As phytoplankton communities show spatial in-lake variability (Reynolds 2006), selective feeding of cladocerans could occur through attraction to algal patches with high-quality taxa as also suggested previously (Schatz and McCauley 2007, Taipale *et al.* 2019). Our results suggest that eutrophic and brown water lakes can provide feasible foraging environment for herbivorous cladoceran when high quality diet is present.

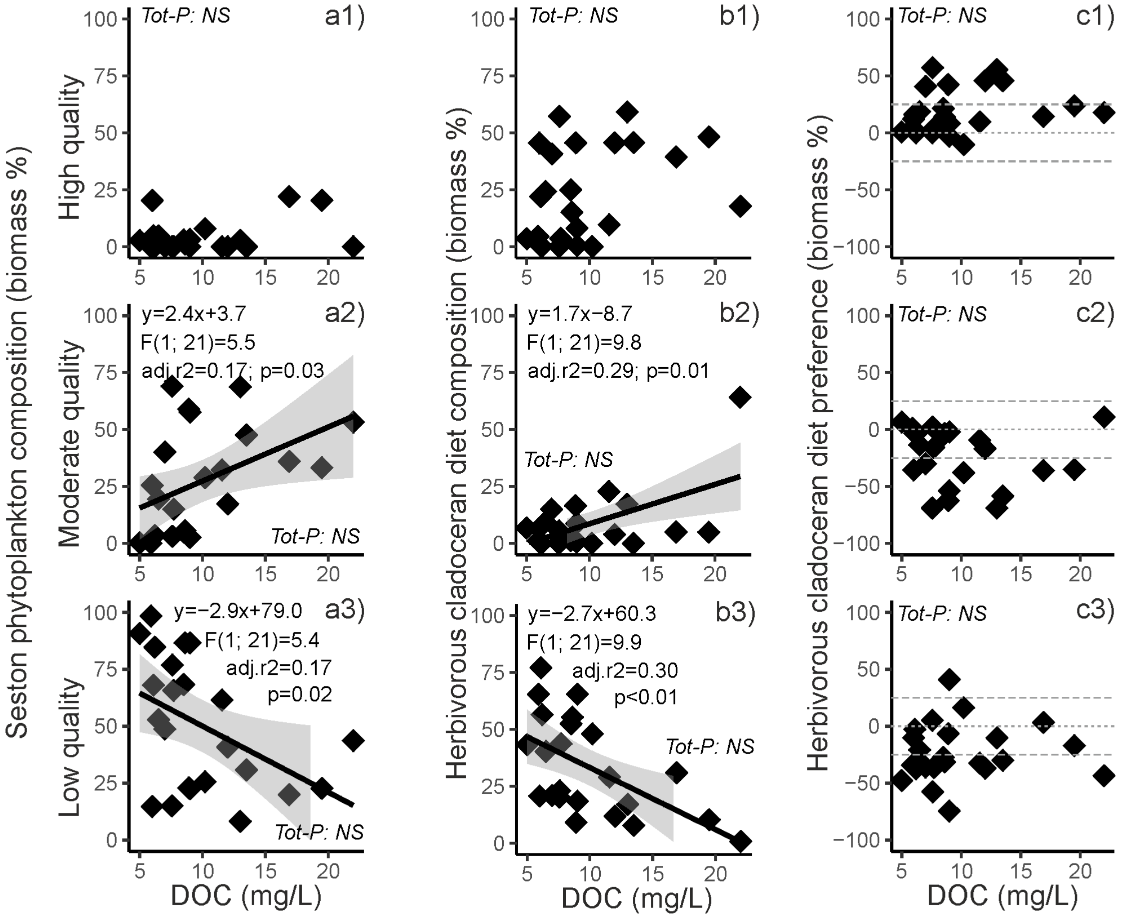

FIGURE 6 Linear regressions between lake DOC and estimated seston composition (a), cladoceran diet composition (b), and cladoceran diet preference (c) for the three diet quality groups (Modified from **I**). High-quality (diatoms and cryptomonads), moderate-quality (chlorophytes, euglenoids, cryptomonads), and low-quality (cyanobacteria and dinoflagellates) diet groups are presented in different rows of the figure. Diamonds represent values from different lakes. Linear regression lines with 95% confidence intervals (grey shaded areas) are drawn and regression equation with statistics (F, p, and adjusted r^2 [adj.r^2]) are presented if the model was statistically significant. Correlation between each dependent variable with lake tot-P is indicated with italics, where NS indicates a statistically non-significant correlation. Horizontal dashed lines in column c represent percent-point differences of 20%, 0%, and −20% from top to bottom, where values > 20 indicate a high preference, values < −20 high avoidance, and values between the lines low or no selection of diet items.

3.2 Perch allochthony, PUFA and THg across a boreal environmental gradient

Perch in lakes with low pH and high forested catchment areas relied more on terrestrial sources than perch in eutrophic lakes with higher pH (Fig. 7, 8). The observed clear positive correlation between perch allochthony and forest coverage (**II**) in the lake catchment corroborates previous findings (Tanentzap *et al.* 2014). However, a direct correlation between lake DOC and perch allochthony was not found (**II**). Therefore it is likely that lake DOC concentration does not alone result in high allochthonous dietary intake in fish.

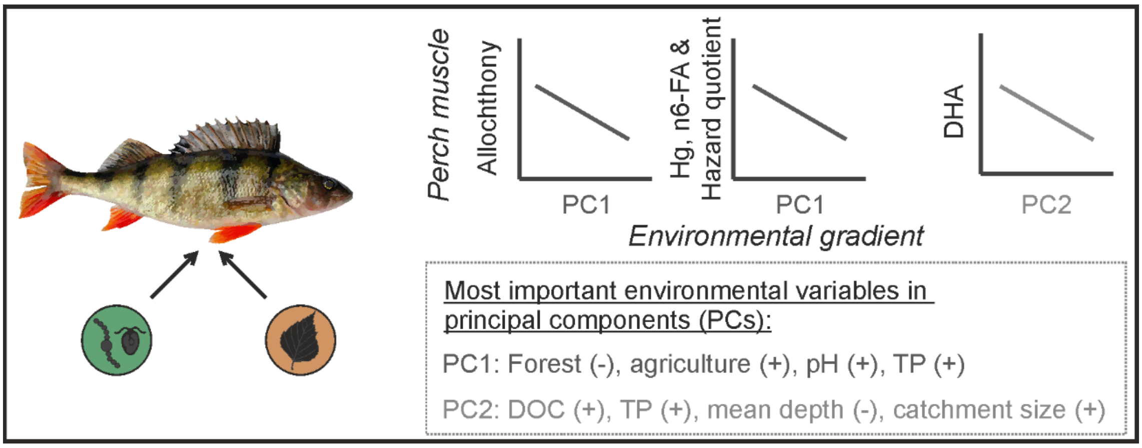

FIGURE 7 Graphical illustration of results and finindgs in **II** (Modified from **II**). Tot-P = lake total phosphorus and DOC = dissolved organic carbon concentration. PC1 combines catchment area forest and agricultural %, Tot-P and pH, where more acidic forest lakes are ranked to left side of the axis and more productive lakes to the right side. PC2 combines DOC, Tot-P, lake mean depth and catchment area size, where deeper clear water lakes are ranked to left side of the axis and shallow murky lakes to right side of the axis.

As lake DOC quantity and quality can be temporally variable, for example, in response to irregular rainfall events (Warner *et al.* 2020), thus our single snapshot measurement may not have adequately represented the longer-term organic matter flow or quality from the catchment. Therefore, it seems likely that catchment area land-use could provide a timely more integrated variable for predicting fish allochthony, and it could be beneficial to measure DOC quality in future studies. Interestingly, pH and allochthony showed a very high negative correlation (**II**). pH is measured with available protons in the water column, and hydrogen isotopes were used to model the fish allochthony. Thus, one could quickly think the strong link between deprotonation of humic carboxyl acid groups towards less acidic lakes could drive the observed trend. However, no clear links between lake water deuterium values and lake pH was found. Thus, the strong correlation between lake pH and perch allochthony is likely linked to the lake's catchment area characteristics and nutrient concentration, which were also correlated with lake pH (**II**).

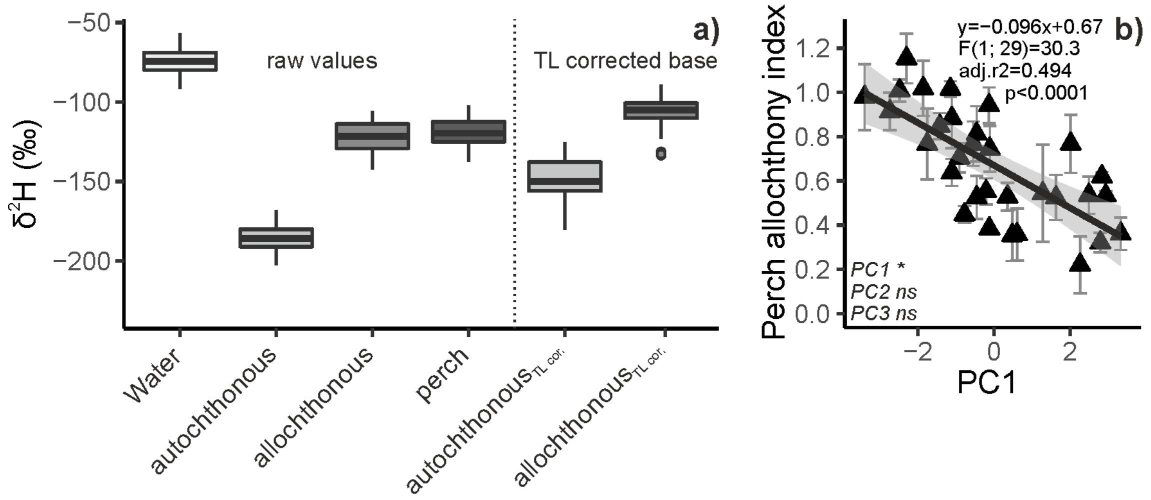

FIGURE 8 Boxplot chart showing the δ²H (‰) data used in the allochthony index calculations (all lakes pooled) (a) and the relationship between PC1 and perch allochthony (b) (Modified from **II**). See material and methods or Fig. 7 for PC-axis interpretations. In panel a, bold horizontal line indicates the median value, the boxes represent first and third quartiles, and the whiskers represent minimum and maximum values. Outliers (black circles) are presented if there are data points smaller or larger than 1.5 times the difference between the first and third quartiles (1.5*IQR). Water from epilimnion (lightest grey), autochthonous source (light grey), allochthonous source (grey), perch (dark grey). The dashed vertical line separates raw δ²H values (left) and trophic level (TL) adjusted base ($\text{autochthonous}_{\text{TL cor.}}$ and $\text{allochthonous}_{\text{TL cor}}$; right) δ²H values. In panel b, regression line (black) and 95% confidence intervals (grey shaded area) with regression equation and statistics (F, adjusted r^2 and p values) are shown, allochthony index correlation on other PCs are shown in bottom-left of panel b, where "ns" indicate statistically non-significant and "*" indicates significant correlation.

Perch muscle THg, ARA, and LIN content was highest in lakes with higher forest coverage than lakes with higher agricultural coverage, tot-P, and pH (Fig. 9). These are in line with previous studies suggesting that the mercury content of fish increases with lower lake pH and higher forested catchment coverage and decreasing tot-P (Ahonen *et al.* 2018, Braaten *et al.* 2018, Rask *et al.* 2021). Moreover, omega-6 PUFAs have been suggested to be terrestrial and omega-3 PUFAs autochthonous production biomarkers (Lau *et al.* 2012, Hixson *et al.* 2015, Taipale *et al.* 2015). Perch muscle DHA content was higher and ALA content lower in deeper clear water lakes than shallow, murky ones (Fig. 9). Here the decreasing perch muscle DHA content along the environmental gradient supports the previous findings (Taipale *et al.* 2016, Colombo *et al.* 2020) with the assumption that dietary sources along this gradient would be deleted with DHA along the same environmental gradient.

Perch allochthony was positively correlated with muscle n6 FAs and THg, but no correlation between allochthony and perch muscle n3 FAs was found (**III**). By combining the FA and THg results with hazard quotient calculations, perch quality was found to be lower in lakes with higher forest cover % in their catchment than lakes with higher agriculture cover %, tot-P and pH (Fig. 9), corroborating with previous finds (Strandberg *et al.* 2016). This observed trend was clearly driven by perch muscle THg values since there was a parallel correlation with perch THg content, hazard quotient, and the environmental gradient (Fig. 9). Thus, it is clear that the quality of perch muscle for human consumption decreases with increasing forest coverage and decreasing pH, and this is potentially mediated by increasing fish allochthony.

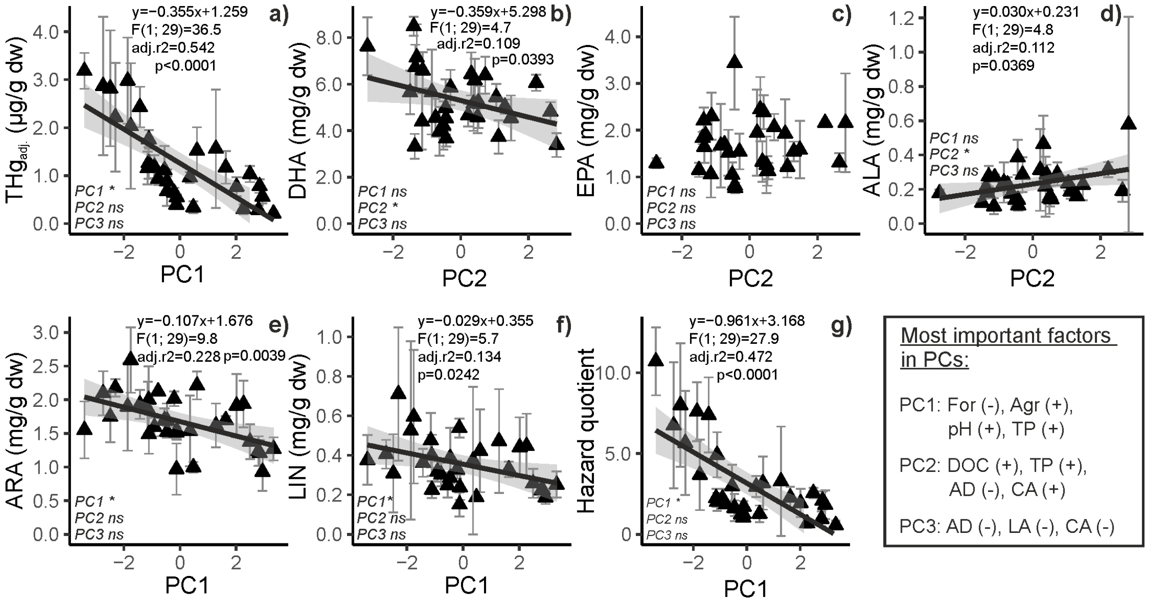

FIGURE 9 Relationship between environmental gradients and perch THg (a), DHA (b), EPA (c), ALA (d), ARA (e), LIN (f) content, and hazard quotient (g) (Modified from **II**). Note the different x-axis in panels a, e-g (PC1), and b-d (PC2). Lake mean values are expressed with black triangles and SD with grey whiskers. The regression lines (black) and 95% confidence intervals (grey shaded area) with regression equation and statistics (F, adjusted $r^{2,}$ and p values) are shown for each panel. The most important environmental factors impacting PCs (|factor loading| > 0.3) are shown in the bottom-right box, where parameter abbreviations correspond to Table 1. Compound correlations on other PCs are shown in italics in the bottom-left of each panel, where ns indicate statistically non-significant and * indicates significant correlation.

3.3 Food web structure and community quality in subarctic lakes

Along the Lapland lake gradient from north to south (increasing temperature and productivity), significant changes in lake community structures, biomass, and community EPA+DHA content was found (**III**). The phytoplankton community shifted from diatom to cyanobacteria-dominated, and the zooplankton community turned from large lipid-rich copepods towards smaller-sized cladoceran-dominated communities (Fig. 10). No clear shifts in the littoral benthos community along the studied environmental gradient was found. However, profundal benthos showed an increasing share of chironomids towards more warm and murky lakes (Fig. 10). Invertivorous fish communities shifted from salmonid-dominated towards percid and cyprinid-dominated communities (Fig. 10). Similar trends for the piscivorous fish community were observed: salmonid fishes such as Arctic charr and brown trout (*Salmo trutta* L.) disappeared along the gradient from north to south and were replaced with larger-sized perch and pike (Fig. 10). The omega-3 fatty acid content (mg EPA + DHA g^{-1} dw) of primary producers and primary consumers decreased along with the climate–productivity gradient. However, these downgrading quality of food sources did not cascade to longer living secondary and tertiary fish consumers (Fig. 10).

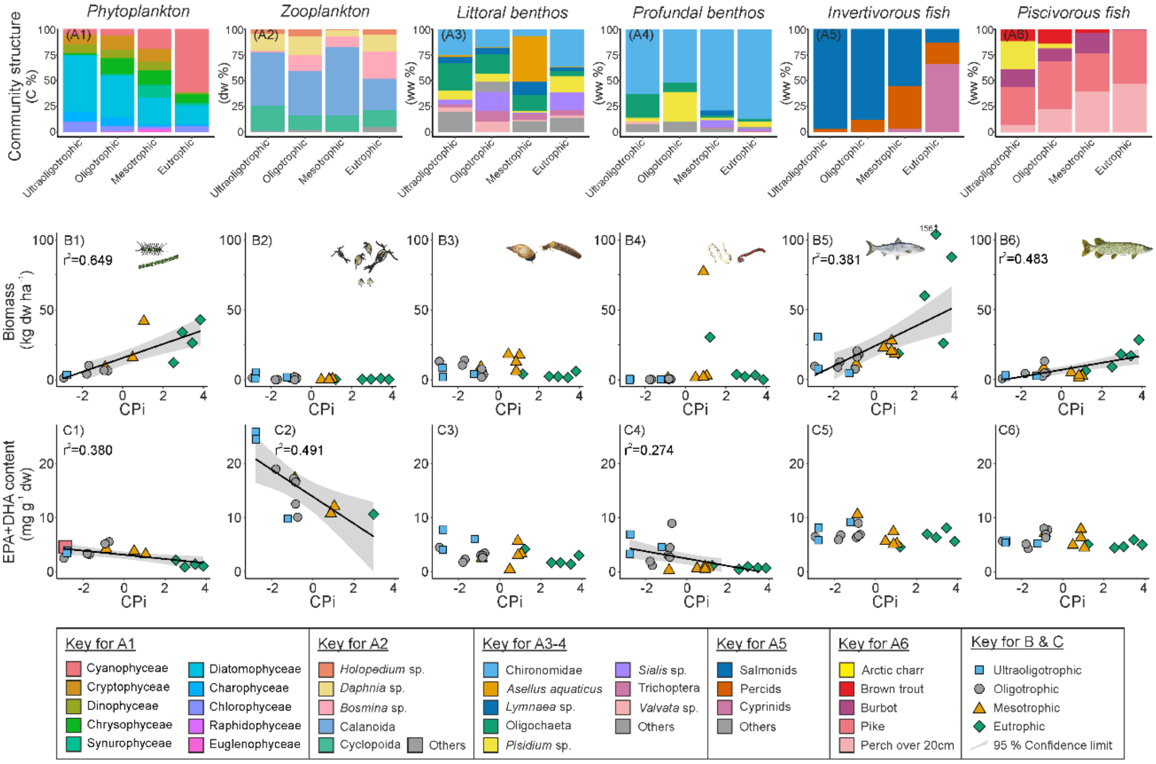

FIGURE 10 Community structure (A), biomass (B), and computed community quality (EPA+DHA content, C) across different lake types (A) and the climate productivity index (B-C) (Modified from **III**). Food web community compartments are shown in different columns of figure: phytoplankton (1), zooplankton (2), littoral benthos (3), profundal benthos (4), invertivorous fish (5), and piscivorous fish (6). The colored bars (a) indicate lake type normalized average of community composition (biomass %). Community biomass (kg dw ha^{-1}) and quality (mg EPA + DHA g^{-1} dw) against the climate-productivity index (CPi) are shown in rows A and B. The coloured shapes indicate different lake trophy: blue square = ultraoligotrophic, grey dot = oligotrophic, orange triangle = mesotrophic, blue diamond = eutrophic. Linear regressions fit (bolded line), 95% confidence limit (grey shaded areas), and adjusted coefficient of determination (r^2) are presented in the figures if the model was statistically significant ($p<0.05$). Detailed linear regression model descriptions can be found in **III**, Table S7.

Along the climate-productive gradient, the most profound positive trends in biomass in each second trophic level (i.e., phytoplankton and invertivorous fish) was observed (Fig. 10). In addition, increasing piscivorous fish biomass was observed along the lake gradient, but the slope compared to insectivorous fish biomass increment was clearly shallower (Fig. 10). Deviating trends in contrasting trophic levels had subsequent effects on shaping trophic pyramids, where only the warmest on most eutrophic lakes seem to settle into an hourglass shape (Fig. 10, 11). Previous food web level studies have found similar biomass ratio patterns (Jeppesen *et al.* 2000, Carpenter *et al.* 2001). As the insectivorous fish biomass increased towards warmer and more eutrophic lakes and at the same time invertebrate community biomass remained rather stable, it seems likely that the top-down control of invertebrate community is higher in more eutrophic and warmer lakes in the studied lake gradient. Increasing total biomass of food webs along with the climate–productivity gradient increased community EPA + DHA areal content. This was especially visible in invertivorous and piscivorous fish communities shaping the EPA, and DHA mass pyramids top-heavy in all the studied lakes (Fig. 11). As different organisms have different life spans, this observation is logical; fish are much longer lived than e.g., phytoplankton individuals. Thus, the biomass pyramids and PUFA pyramids may be top-heavy (Fig. 11).

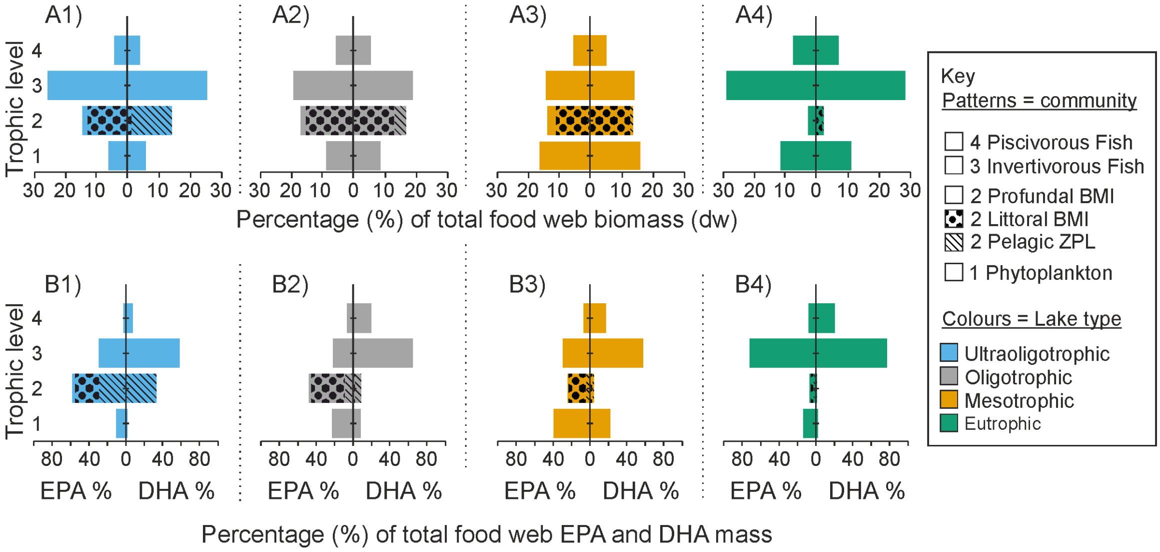

FIGURE 11 Biomass, EPARelative biomass (A) and FA stock (B) pyramids by food web compartments for the lake types (Modified from **III**). The steps of the pyramids illustrate lake type averages of relative biomass or EPA+DHA areal content and are ordered with trophic levels from bottom-up order: 1) phytoplankton, 2) invertebrates, 3) invertivorous fish, and 4) piscivorous fish. In the second trophic level (invertebrates) profundal benthos, littoral benthos, and zooplankton are separated with different patterns: clean, spots, stripes, respectively. Different lake types (ultraoligotrophic, oligotrophic, mesotrophic, and eutrophic) are drawn to different columns separated with dashed lines and additionally with colors blue, grey, orange, green, respectively. The lake-specific pyraminds are presented in Fig. S2 (**III**). Abbreviations in legend are BMI = benthos and ZPL = zooplankton.

3.4 Phytoplankton and zooplankton community structure and quality

Contrasting results of phytoplankton community structure and quality (EPA+DHA content) along environmental gradients between boreal and subarctic lake gradients was found (**I** & **III**). In **I,** no clear trends in phytoplankton community structure (based on fatty acid signature modelling) or EPA and DHA content was observed along with tot-P and DOC gradients. However, in **III,** where microscopy and laboratory determined FA contents were used in calculations, the trend was evident for both phytoplankton community structure and EPA+DHA content. Furthermore, based on microscopic results the phytoplankton community shifted from diatom to cyanobacteria dominated from cold clear water lakes towards warmer and murky lakes (**III**). These contrasting trends could be caused by the difference in used methods as there are obvious differences how the PUFA content of seston and community structure was derived in **I** and **III**. In **I,** seston PUFA content was directly analyzed from filtered lake water and weighted with lake particulate organic matter content. Yet, when PUFA content of seston was weighed with lake chlorophyll content (as an alternative proxy of algal biomass) still no decreasing quality was along with environmental gradients was found (**I**). Contrary to this, in **III,** lake phytoplankton community structure and previously determined laboratory grown algal monocultures were used to define the primary producer community EPA+DHA content. Thus, these results are not directly comparable and differences in methodolody is at least partly behind the observed differences in the seston quality and community structure trends along the environmental gradients. Additionally geographical and lake metric differences in the sampled lake gradients could have resulted the differences. In **I**, the sampled lakes were boreal and located in Central Finland approximately similar climatic zone, whereas the lake gradient from **III** was subarctic located in Northern Finland. Moreover, the sampled lake chemistry gradients were not equal between the two lake gradients (**I** & **III**). For example in **III,** subarctic lakes did not have the darkest lakes as DOC gradient spanned from 2 to 15 mg l^{-1}, but in **I** from 5 to 22 mg l^{-1} (Table 2). In addition **I,** did not any or large-sized ultraoligotrophic or oligotrophic lakes (LA range: 0.004–107.5 km^2, tot-P range: 4–149 µg l^{-1}), whereas in **III** the study lakes were mainly large and did not have any hypereutrophic lakes (tot-P range: 3–48 µg l^{-1}). In the lake gradient of **III**, there are multiple collinear environmental gradients (tot-P, temperature, precipitation, DOC), whereas environmental gradients in **I** were more scattered and did not include any climate variables. Therefore, it seems likely that lake gradients with multiple collinear environmental parameters including both water chemistry and climatic variables in space-for-time study setups may result in stronger observed shifts in community structures and biomolecular contents. One alternative for the observed differences in phytoplankton community and PUFA content in boreal and subarctic lake gradients could be that phytoplankton community response to environmental change may differ between the ecoregions. However, future studies with harmonized methods definening the PUFA content of seston is needed to verify if the there are true differences in seston quality and community structure response on environmental stressors between boreal and subarctic areas.

In the subarctic lake gradient, the zooplankton community shifted from copepod-dominated towards smaller cladoceran-dominated from clear oligotrophic lakes towards warmer and more murky lakes (Hayden et al. 2017, **III**). As no decreasing EPA+DHA content in zooplankton taxa (e.g., cladoceran) was observed, it is likely that the observed decreasing zooplankton community quality was mainly caused by the differences in the community structure where lipid rich calanoid *Eudiaptomus graciloides* was dominant copepod in most cold and ultraoligotrophic lakes (**III**). In **I,** no decreasing PUFA content in cladoceran samples from oligotrophic towards eutrophic lakes was found. The sample sizes for pure cladoceran samples in **III** were small (samples from 8 lakes) and did not cover the whole gradient. Thus comparing the cladoceran PUFA content between **I** and **III** directly could be misleading. However, we are providing more information that environmental stressors such as eutrophication or browning does not directly lead to decreased long-chain PUFA content of cladocerans as found also previously (Senar *et al.* 2019). This could be caused by cladoceran ability to rely on high PUFA content algae taxa such as diatoms and cryptomonads as shown in **I**.

3.5 Fish muscle long-chain PUFA content in subarctic and boreal lakes

In the subarctic region, no decreasing long-chain PUFA content of fish communities along with climate-productivity gradient was found (**III**). Moreover, when looking individual fish species, significant slightly decreasing quality of pike muscle was observed along the climate productivity gradient towards more eutrophic lakes, however, in large-sized perch (samples from 13 lakes) such trend was not found (**III**). Interestingly, in boreal regions, large-sized perch muscle (samples from 31 lakes) showed significant slightly decreasing DHA content along with the observed environmental gradient from deep clear water lakes towards shallow, murky lakes (**II**). In addition to sample sizes, there are apparent differences between the observed lake gradients, as described previously. Hence these differences could drive the observed differing fish muscle quality trends along environmental gradients. What makes these observations even more interesting is that decreasing quality of fish prey items (zooplankton and profundal benthos) along with the lake gradient was observed (**III**). However, despite this significant change in the prey community quality, fish muscle (combined value of several species at same trophic level, or large perch) PUFA content remained stable (**III**). It remains unsolved why in other lake gradients spanning from boreal to subarctic regions, the basal resources or diet of perch may result in cascading impacts on perch muscle quality (Taipale *et al.* 2016). And secondly, why downgrading quality of perch muscle here in boreal (**II**) and subarctic (**III**) regions, this mechanism seems to be less effective. As previous studies have shown, perch are able to compensate muscle PUFA content by prolonging shorter PUFAs to longer ones in environments where only low long-chain PUFA content prey are abundant (Geay *et al.* 2015, Ishikawa *et al.* 2019, Scharnweber *et al.* 2021, Taipale *et al.* 2022). This mechanism could be the driving factor for the observed relatively stable fish muscle PUFA content along the observed environmental gradients (**II** & **III**), emphasizing the conservative nature of fish muscle biomolecular structure.

3.6 Caveats and future perspectives

Differences in sampling and fatty acid calculations methods hinder the comparison of results of the articles in this thesis, but also from previous litterature. For example sampling of lake seston for FA analysis should be done with filttering subsample combined epilimnion water to pre-weighed filters. Biopsies from freeze-dried filter could be taken to carbon percentage measurement. Alternatively carbon percentage of the epilimnetic seston could be analysed from tangential filtered and freeze-drying concentrated particulate organic matter powder. FA content analyses derived using external FA standard (e.g. 566c, Nu Check Prep Inc.) calibration formulas and internal standard (e.g. 19:0 or 23:0) should be weighted with sample mass and carbon percentage. The used methods and sampled community or taxa is naturally dependent on the study questions in focus. However, FA content per sample carbon mass should be preferred in nutritional ecology studies to make different trophic level FA values and results from other studies more comparable. This would enchance the quality and reliability of drawed conclusions and mechanisms how environmental change impacts the fatty acid production and retention at different food web levels. Ultimately this would enable to produce more reliable and precise simulations how future change will affect nutritional quality of different organism.

In **I**, the seston composition and cladoceran diet composition were modeled based on the fatty acid profiles of the analyzed samples. Thus, not allowed to study the impacts of diet on cladoceran FA content as FA profile and content data are heavily correlated. However, I encourage to test with independent datasets how the diet of cladoceran impacts their PUFA content in the wild. For example, this could be done with other molecular tools such as eDNA which would provide even higher taxonomic resolution than FA dietary modeling.

In **II,** only land-use coverage data was used to together with lake chemistry and morphometry parameters to predict perch muscle biomolecular content. Thus, the results here do not indicate well the intensity of e.g., forestry activities such as ditching and clear-cuttings of study sites. However, in Finland, most of the forests outside protected areas are heavily managed. Yet, I encourage future research to compare directly fish allochthony and biomolecule content between lakes inside and outside of protected areas. In addition, the environmental parameters should include more accurate estimations of land-use activities e.g. catchment area ditch km ha^{-1}, and forest harvesting parameters such as clear-cutting history. With these approaches, future studies could answer how land-use activities specifically modify lake food web allochthony and consequently fish THg and PUFA content.

In **III,** not all the possible food web communities was studied. The biomass or FA data from benthic algae, macrophytes, bacteria, rotifer, and macrobenthos was lacking. Implementing the missing taxonomic groups to future data sets could provide more detailed knowledge on food web functioning and biomass distribution of lake food webs. Moreover, future studies should focus on year-round biomass and FA production of different communities and ultimately to produce biomass, carbon and FA production pyramids across lake gradients. Despite this would be extremely laborious and time-consuming, I believe that this would provide vital knowledge to ecologists of how ecosystems function, sparking new ideas and evolving old food web theories.

In subarctic lake food webs, warming and increasing nutrients downgrade primary producer and primary consumer PUFA content yet not cascading to fish communities. However, **II** provides more information that eutrophication and browning could slightly lower perch muscle PUFA content in the boreal region, which has also been observed previously (Taipale *et al.* 2016). It should be tested in future if differing PUFA trends in perch muscle along with environmental gradients could be found with harmonized methods and lake chemistry gradients from boreal and subarctic regions. Moving away from fish optimal niche could increase metabolic costs and hence lower the PUFA content of the fish muscle. Future studies could better assess what are the main ecological and physiological drivers behind this potential difference in fish muscle PUFA content respond on environmental stressors between the different ecoregions.

4 CONCLUSIONS

I did not observe clear seston community structure changes in boreal lakes along the lake tot-P gradient. Indicating that, especially with snapshot sampling, lake phosphorous may not be solely a good predictor for phytoplankton community structure nor quality. Thus lakes should be monitored throughout the season. Here, simultaneously collected seston PUFA content did not correlate with herbivorous cladoceran PUFA content. Moreover, in all the sampled 23 lakes, herbivorous cladoceran showed clear dietary preference towards high-quality algal groups (such as diatoms and cryptophytes), and at the same time, they seemed to avoid nutritionally low-quality algal groups (mainly cyanobacteria). These results corroborate with previous studies suggesting that cladoceran could be partially selective filter feeders (Hartman and Kunkel 1991), and the selectivity could occur through attraction to high-quality algal patches and restrain feeding when low-quality algae are present.

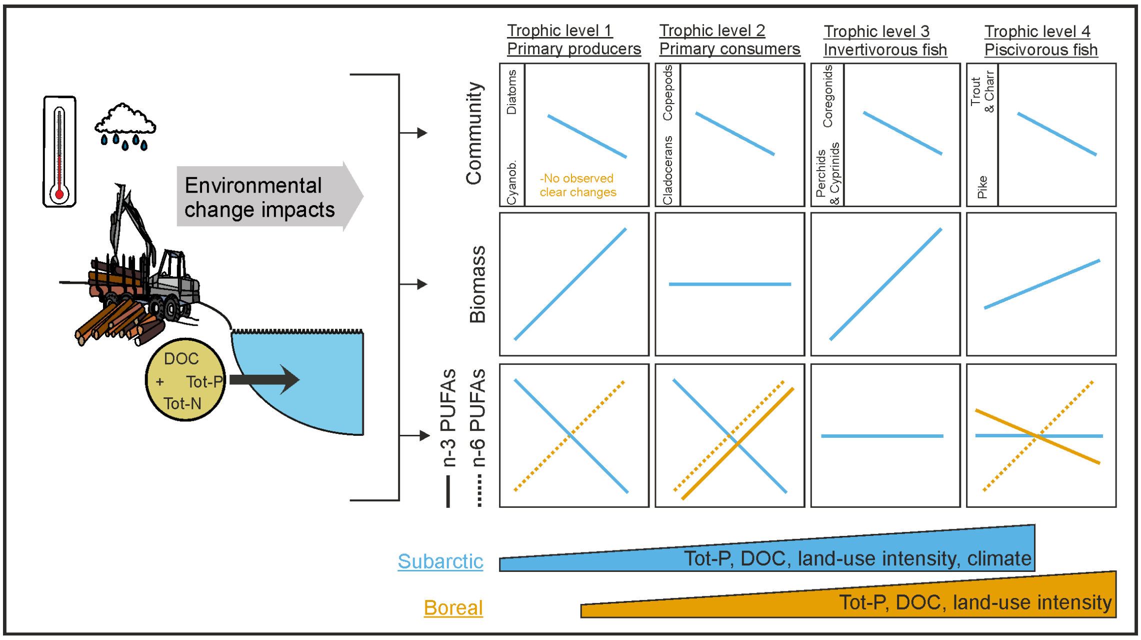

FIGURE 12 Main findings of the thesis – Environmantal change impacts on lake food web community structure, biomass and PUFA content.

Differences in lake catchment area land use have apparent impacts on lake food web energy sources, where lakes with higher forest coverage in the catchment perch rely more heavily on allochthonous production. This correlates with fish muscle mercury and omega-6 FA content but not omega-3 FA content. Collectively it seems that fish muscle quality for human consumption is lowest in lakes with low pH and high forest coverage in the catchment due to the higher mercury content of these fishes.

Subarctic lake food web communities become more murky water adapted species along with increasing temperature and productivity gradient. Phytoplankton species such as cyanophycean and invertivorous fish such as cyprinids and percids seem to benefit most from the warming environments. Moreover, due to the phytoplankton and zooplankton community changes, a profound decreasing PUFA content of these communities was found from cold, clear water lakes towards warmer and more murky lakes. However, this decreasing quality of fish prey items did not affect fish muscle quality, which could be related to fish homeostasis and the conservative nature of fish muscle structure. Yet, there seem to be differences in the food web processes between boreal and subarctic regions, as decreasing long-chain PUFA content of perch muscle along environmental gradients was observed only in boreal lakes. In boreal regions the studied lakes had wider tot-P gradient than in subarctic regions including hypereutrophic lakes, but lacking comprehensive representation of ultra-oligotrophic and large oligotrophic lakes. this could be one driving behind the observed trends in perch muscle long-chain PUFA content trends. More over, the subarctic lakes were generally much larger than the boreal lakes which likely enables predators a larger habitat space to forage in, thus preventing the prey item downgraded quality to cascade to fish muscle.

Even though I am not giving any timely related predictions on how future environmental change will alter the Finnish subarctic and boreal lake ecosystems, I can still point out the likely direction. It seems possible that future subarctic lakes will become more like boreal lakes as climate change and land-use intensity proceeds. This would result in a clear shift in community structure and lake food web functioning. For example, boreal fish species invasions towards the northernmost lakes in the Tornio-Muonionjoki tributary system are happening. Perch,ruffe (*Gymnocephalus cernua* L.) and roach have expanded their range towards the north in recent years. However, these populations are still relying on few year-classes and reproduction success (Hayden *et al.* 2013). Moreover, other space-for-time substitution studies have provided similar predictions (e.g., Jeppesen *et al.* 2000). Based on the previous studies (Braaten *et al.* 2019) it is likely that mercury content in fish muscle may continue to stabilize or decline in future. However, high intensity forestry activities might hider this trend and result even higher mercury content in fish muscle (Ahonen et al. 2018; Kozak et al. 2021). These activities likely increase also fish muscle omega-6 PUFA content. As future browning and eutrophication is projected to progress in future, this thesis provided data that in boreal regions perch muscle DHA content could decrease slightly in future. However, as lakes are unique, only long-term time series will show the actual impacts of climate change and increasing anthropogenic land use on lake communities and community quality.

Harmonizing sampling procedures, FA content calculations, studied lake gradient size and chemistry ranges, could enchance the comparability of ecological space-for-time studies focusing on nutritional quality of organisms. This would illuminate the complexity of lake food web processes potentially raising new ideas of FA retention dynamics and ultimatimately enabling accurate predictions how future environmental change will impact the nutritional quality of different organisms and communities.

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I am grateful to my supervisors, Docent Sami Taipale and Professor Kimmo Kahilainen, for giving me a chance to participate high standard research groups as a PhD researcher. I wish to express my gratitude for your persistent attitude in educating me as a scientist. I genuinely admire the enthusiasm and hardworking attitude towards science that both of you share. Sami, you have put a lot of effort into educating me in biochemistry and laboratory analytics and teaching me a great deal of independence early in my scientific career. Kimmo, you have had deep patience and trusted my visions, and provided courage to continue when needed during the whole project. Thank you for always having time to discuss even the smallest details when needed. I am deeply grateful that we met in the first year of my aquatic science bachelor studies at the University of Helsinki. You have truly inspired me towards a scientific career. Although the journey has not always been easy, I wish you both enjoy the time half as much I enjoyed conducting this project. I owe you both a great deal.

I wish to thank all the co-authors participating in the work. This thesis would not have been possible to conduct without the data and samples you have provided, your expertise, and constructive comments on the manuscripts. Even though there have been numerous co-authors in each of the papers, the revision rounds have been surprisingly smooth. Thank you for your extensive effort towards the manuscripts. Jaakko taught me a great deal about FA dietary modeling. I am grateful to Mikko for sharing his knowledge with stable isotopes and inspiring discussions about science. Without all your support, this thesis would have never been finished.

I am deeply grateful for the hardworking lab technicians, Mervi and Emma; you always helped when needed. You have helped a great deal in logistics and provided your expertise and help when I have been mingling with some problems in the lab. I wish to thank all the inspiring moments in the D2 coffee room. I wish the inspiring discussions at the coffee room would recover to their former glory when COVID time ends. Marco, you indeed provided me peer support when needed; I am grateful for the many eager and enthusiastic discussions during coffee breaks.

Special thanks to Salla for always supporting me when I crawled in the endless task lists and wailed before the frequent deadlines. Your support had truly carried me out, especially when my workload was almost unbearable. We indeed got some extra tasks when our beautiful firstborn, Uula was born. Your gratuitous tender care towards our child and endless support for me truly sparked to carry on. Thank you Salla for helping me with the graphical illustrations in the thesis cover. Finally, I wish to thank my siblings and parents who have supported me with my career choice.

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YHTEENVETO (RÉSUMÉ IN FINNISH)

Ympäristömuutoksen vaikutukset järviravintoverkkojen rakenteeseen ja ravitsemukselliseen laatuun

Ympäristömuutokset kuten lämpeneminen, rehevöityminen ja tummuminen muuttavat järvien perustuottajayhteisön rakennetta ja siten myös vaikuttaen tuotettujen rasvahappojen määriin. Järvien rehevöityminen lisää sinibakteerien osuutta leväyhteisössä, minkä on havaittu vähentävän merkittävästi pitkäketjuisten rasvahappojen tuotantoa vaikuttaen myös kuluttajien kudosten rasvahappopitoisuuksiin. Pii- ja nielulevät tuottavat entsyymiavusteisesti runsaasti pitkäketjuisia rasvahappoja kuten dokosaheksaeenihappoa (DHA, 22:6n3) ja eicosapantaeenihappoa (EPA, 20:5n3), toisaalta sinibakteerit eivät valmista näitä rasvahappoja. Toisenvaraiset eliöt tarvitsevat EPA:a ja DHA:ta kasvamiseen ja lisääntymiseen, mutta eivät voi itse valmistaa niitä tehokkaasti. Tämän takia tiettyjen levien tuottamat pitkäketjuiset rasvahapot ovat ravinnollisesti arvokkaita ja päätyvät levistä eläinplanktonin kautta kaloihin ja sitä kautta myös ihmisiin.

Elohopea (Hg) on terveydelle haitallinen raskasmetalli, joka kertyy eliöihin ja ravintoverkoihin. Ihminen on lisännyt teollisella aikakaudella ilmakehän elohopeapitoisuuksia esimerkiksi fossiilisia polttoaineita polttamalla. Ilmakehästä elohopeaa päätyy vesistöihin suoraan ilmalaskeumana ja valuma-alueelta. Voimakas maanmuokkaus järvien valuma-alueella, kuten kaivostoiminta, metsätalous ja metsäojitukset, lisää vesistöihin päätyvän orgaanisen aineksen ja siihen sitoutuneen elohopean määrää. Elohopea siirtyy passiivisesti vedestä leviin ja rikastuu niitä ravinnokseen käyttävissä kuluttajissa ja koko ravintoverkossa huippupetoihin saakka. Orgaanisen hiilen määrä korreloi usein veden ja eliöiden elohopean pitoisuuden kanssa. Rehevöitymisellä sen sijaan on yleensä päinvastainen laimentava vaikutus veden ja eliöiden elohopeapitoisuuksiin. Ihmisiin päätyvä elohopea on pääosin peräisin kalaravinnosta.

Muutokset järvikemiassa vaikuttavat koko ravintoverkon energialähteisiin. Esimerkiksi lisääntyvä valuma-alueelta peräisin olevan orgaanisen aineksen määrä lisää bakteerituotantoa, jota muut kuluttajat voivat hyödyntää ravinnokseen. Toisaalta rehevöityneissä järvissä suuri perustuotannon määrä on hallitseva energialähde toisenvaraisille eliöille. Järvien ravintoverkkojen sisäisen (perustuotanto) ja ulkoisen (maalta tulevan) energian määrää pystytään arvioimaan esimerkiksi pysyvillä isotoopeilla, mikäli em. energianlähteiden isotooppiarvot eroavat riittävästi toisistaan.

Tässä väitöskirjassa tutkin, miten ympäristönmuutos kuten lämpeneminen, rehevöityminen ja tummuminen vaikuttavat eri eliöyhteisöjen rasvahappopitoisuuksiin ja biomassaan. Lisäksi väitöskirjassa selvitin kuinka muutokset järvikemiassa vaikuttavat ensimmäisten asteen kuluttajien, vesikirppujen, ravinnon valintaan ja niiden rasvahappopitoisuuksiin. Tutkin myös, kuinka ympäristönmuutos vaikuttaa ravintoverkkojen energialähteisiin ja kalojen ravinnolliseen laatuun ihmiselle yhdistäen kalojen lihaksen rasvahappo- ja elohopeapitoisuuksia. Subarktisista ja boreaalisista järvistä otettiin näytteitä perustuottajista huippupetoihin ja näytteistä analysoitiin rasvahappo- ja elohopeapitoisuuksia sekä vedyn isotooppeja. Lisäksi subarktisista järvistä määritettiin eri trofiatasojen lajiston koostumukset, tiheydet ja biomassat yhteisörakenteiden vertailua varten.

Järvien rehevöityminen ja tummuminen ei alentanut sestonin (vedessä keijuva hiukasmainen aines) PUFA-pitoisuutta boreaalisilla järvillä. Sestonin ja sitä syövien vesikirppujen PUFA-pitoisuuksien välillä ei havaittu yhteyttä. Ravintomallinnuksiin perustuen vesikirput valikoivat ravinnokseen hyvälaatuisia leväryhmiä kuten pii- ja nieluleviä ja välttivät sinibakteereja boreaalisissa järvissä. Aikaisempien tutkimusten perusteella tiedetään, että järvissä perustuottajat eivät ole tasaisesti jakautuneita, vaan muodostavat lajikirjoltaan vaihtelevia laikkuja järvissä. Vesikirput siivilöivät leviä ravinnokseen, ja siksi vesikirput hakeutunevatkin laikkuihin, joissa on hyvälaatuisia leviä varmistaen tärkeiden rasvahappojen saannin.

Boreaalisilla alueilla ahvenen DHA-pitoisuudet laskivat hieman kirkasvetisistä järvistä matalampiin tummavetisiin järviin. Tämä johtunee leväyhteisön muuttumisesta rasvahappotuotannon kannalta epäedullisempaan suuntaan vaikuttaen siten myös kuluttajien rasvahappopitoisuuksiin kuten aikaisemmissa tutkimuksissakin on havaittu. Havaitsin myös, että happamissa metsäjärvissä ahventen energialähteistä suurempi osa on peräisin maalta kuin järven sisäisestä perustuotannosta. Lisäksi happamissa metsäjärvissä ahventen elohopea ja omega-6 PUFA-pitoisuudet olivat korkeampia kuin rehevissä järvissä. Happamien metsäjärvien ahventen korkea elohopeapitoisuus alensi ahvenen ravinnollista laatua ihmiselle. Nämä muutokset ahvenen laadussa johtunevat metsäjärvien suuremmasta maalta peräisin olevista energialähteistä ravintoverkkotasolla. Yhteyden kausaaliseen todentamiseen vaaditaan kuitenkin jatkotutkimuksia.

Lämpötilan ja tuottavuuden kasvu muuttaa eliöyhteisöjen ja ravintoverkkojen rakennetta subarktisissa järvissä: sinibakteerien osuus perustuottajista kasvaa, eläinplanktonin ja pohjaeläinten koko pienenee, särkikalojen osuus kalapopulaatiosta kasvaa. Perustuottajien ja selkärangattomia syövien kalojen biomassa kasvaa kohti lämpimämpiä ja rehevämpiä järviä muuttaen järvien biomassapyramidien muotoa tiimalasimaisemmaksi. Kalojen saalistuspaine selkärangattomiin kuluttajiin lienee voimakkaampaa lämpimämmissä ja rehevämmissä järvissä verrattuna kylmempiin kirkasvetisiin järviin. Lisäksi havaitsin, että subarktisissa järvissä perustuottajien ja selkärangattomien EPA+DHA-pitoisuudet laskivat kohti lämpimämpiä ja rehevimpiä järviä, mutta nämä muutokset eivät vaikuttaneet kalayhteisöjen PUFA-pitoisuuksiin. Sen sijaan koko kalayhteisöstä laskettujen kokoomanäytteiden EPA- ja DHA-pitoisuudet olvati vakaita subarktisissa järvissä huolimatta niiden verraten suurista eroista järvikemiassa. Tulevissa tutkimuksissa pitäisi selvittää johtuvatko havaitut erot boreaalisten ja subarktisten järvien välillä erilaisista menetelmistä vai todellisista muutoksista käyttäen kattavia aineistoja ja yhtenäistettyjä menetelmiä.

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ORIGINAL PAPERS

I

HERBIVOROUS CLADOCERAN ESSENTIAL FATTY ACID CONTENT AND DIET ACROSS A PHOSPHOROUS AND DOC GRADIENT OF BOREAL LAKES—IMPORTANCE OF DIET SELECTION

by

Ossi Keva, Jaakko J. Litmanen, Kimmo K. Kahilainen, Ursula Strandberg,
Mikko Kiljunen, Heikki Hämäläinen, Sami J. Taipale

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II

ALLOCHTHONY AND THE FATTY ACID AND MERCURY CONTENTS OF EURASIAN PERCH (*PERCA FLUVIATILIS*) ALONG BOREAL ENVIRONMENTAL GRADIENTS

by

Ossi Keva, Mikko Kiljunen, Heikki Hämäläinen, Roger I. Jones, Kimmo K. Kahilainen, Paula Kankaala, Miikka B. Laine, Jos Schilder, Ursula Strandberg, Jussi Vesterinen, Sami J. Taipale 2022

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Allochthony, fatty acid and mercury trends in muscle of Eurasian perch (*Perca fluviatilis*) along boreal environmental gradients

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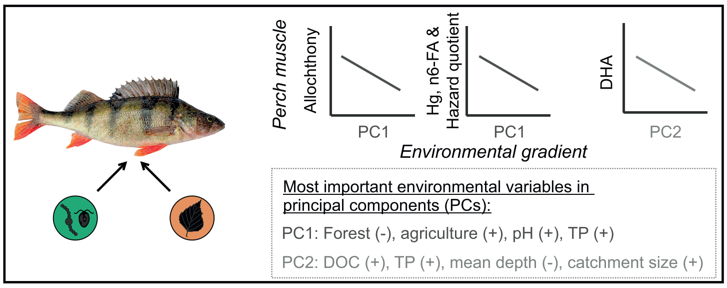
HIGHLIGHTS

- Perch allochthony, fatty acid (FA), and mercury (Hg) were studied in boreal lakes.
- Perch allochthony was highest in low pH lakes with highly forested catchment.
- Perch muscle Hg and omega-6 FA content showed a similar trend to allochthony.
- Perch muscle DHA content decreased towards shallower and murkier lakes.
- Hazard quotient (Hg/FA) elevated towards low pH lakes with forested catchments.

GRAPHICAL ABSTRACT



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ABSTRACT

Environmental change, including joint effects of increasing dissolved organic carbon (DOC) and total phosphorus (TP) in boreal northern lakes may affect food web energy sources and the biochemical composition of organisms. These environmental stressors are enhanced by anthropogenic land-use and can decrease the quality of polyunsaturated fatty acids (PUFAs) in seston and zooplankton, and therefore, possibly cascading up to fish. In contrast, the content of mercury in fish increases with lake browning potentially amplified by intensive forestry practises. However, there is little evidence on how these environmental stressors simultaneously impact beneficial omega-3 fatty acid (n3-FA) and total mercury (THg) content of fish muscle for human consumption. A space-for-time substitution study was conducted to assess whether environmental stressors affect Eurasian perch (*Perca fluviatilis*) allochthony and muscle nutritional quality [PUFA, THg, and their derivative, the hazard quotient (HQ)]. Perch samples were collected from 31 Finnish lakes along pronounced lake size (0.03–107.5 km^2), DOC (5.0–24.3 mg L^{-1}), TP (5–118 μg L^{-1}) and land-use gradients (forest: 50.7–96.4%, agriculture: 0–32.6%). These environmental gradients were combined using principal component analysis (PCA). Allochthony for individual perch was modelled using source and consumer δ^2H values. Perch allochthony increased with decreasing lake pH and increasing forest coverage (PC1), but no correlation between lake DOC and perch allochthony was found. Perch muscle THg and omega-6 fatty acid (n6-FA) content increased with PC1 parallel with allochthony. Perch muscle DHA (22:6n3) content decreased, and ALA (18:3n3) increased towards shallower murkier lakes (PC2). Perch allochthony was positively correlated with muscle THg and n6-FA content, but did not correlate with n3-FA content. Hence, the quality of perch muscle for human consumption decreases (increase in HQ) with increasing forest coverage and decreasing pH, potentially mediated by increasing fish allochthony.

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1. Introduction

Anthropogenic land-use practices such as intensive forestry, including clear-cutting, site preparation and peatland ditching (Škerlep et al., 2020; Nieminen et al., 2021; Finér et al., 2021), together with lake recovery from acidification (Vuorenmaa et al., 2006; Lepistö et al., 2021) and increasing precipitation (Larsen et al., 2011; de Wit et al., 2016) enhance organic matter load to boreal lakes. Increasing inputs of organic matter and iron shift lakes towards browner colour, a phenomenon termed browning (Creed et al., 2018; Blanchet et al., 2022). In addition to browning, land use practices may simultaneously elevate lake nutrient concentrations stimulating primary production (e.g. Hecky and Kilham, 1988; Palviainen et al., 2016; Finér et al., 2021) with subsequent impacts on higher trophic level biomass and molecular content (Hayden et al., 2019; Keva et al., 2021). Lake browning and changes in lake productivity can have major impacts on community structures, but also on the primary food sources of lake food webs (Creed et al., 2018; Keva et al., 2021). Browning could increase the contribution of allochthonous (exogenous, mainly terrestrial) primary food sources in the food webs, whereas in highly productive lakes autochthonous sources should be more dominant (Creed et al., 2018; Blanchet et al., 2022).

Phytoplankton use solar energy to synthesise energy-rich organic molecules such as carbohydrates that can be further modified, for example to polyunsaturated fatty acids (PUFAs). Terrestrial organic matter, especially the most labile compounds, mainly enters lake food webs via heterotrophic bacteria and protozoans, which in turn can be utilised by consumers such as zooplankton (Hiltunen et al., 2017). Primary consumers, benthos and pelagic zooplankton, can also directly ingest large terrestrial particles (Premke et al., 2010; Glaz et al., 2012; McMeans et al., 2019). Benthic algal growth on top of submerged leaves enhances the consumption of leaf litter by benthos (Guo et al., 2016). Terrestrial organic matter naturally entering lakes is generally deficient in essential biomolecules (sterols, amino acids, PUFAs), and thus, autochthonous production is essential for primary and secondary consumers (Karlsson et al., 2012; Taipale et al., 2016a; Wenzel et al., 2021). Any changes in lake food web pathways could potentially modify fish muscle composition and via this route influence the quality of human diet.

The balance between autochthonous and allochthonous primary food sources determines whether a lake food web is driven by the internal primary production or by organic matter inputs from the catchment area. Stable isotope ratio analyses provide a tool to trace the flow of autochthonous and allochthonous matter in food webs (Doucett et al., 2007; Karlsson et al., 2012; Vander Zanden et al., 2016). Since stable isotope ratios of hydrogen (δ^2H, expressed in ‰ versus a reference material) in terrestrial vs. aquatic primary producers are markedly different (ca. 100‰) (Doucett et al., 2007; Karlsson et al., 2012; Wilkinson et al., 2015), they enable tracing these initial energy sources in the aquatic foodwebs. Terrestrial plants have higher δ^2H values than aquatic algae, as terrestrial plants preferentially transpire isotopically lighter water (Doucett et al., 2007). Moreover, algae contain more lipids than senescent leaves, and lipids are depleted in the heavier stable hydrogen isotope, deuterium (Sessions et al., 1999). These differences in the relative amount of deuterium in the hydrogen in autochthonous and allochthonous matter provides a means of estimating consumer hydrogen sources with a two-end point mixing model. Tanentzap et al. (2014) estimated that greater forest cover in waterbody catchments increases the allochthnous subsidies in yellow perch (*Perca flavescens*) ranging from 30% to 60%. Similarly, Karlsson et al. (2012) found similar allochthony reliance (57%) in Eurasian perch (*Perca fluviatilis*) in boreal Sweden.

In lakes, phytoplankton and periphyton are the main source for physiologically important long-chain n3 PUFAs such as docosahexaenoic acid (22:6n3, DHA) and eicosapentaenoic acid (20:5n3, EPA) (Galloway and Winder, 2015; Taipale et al., 2015). Most consumers preferentially obtain EPA, DHA and arachidonic acid (20:4n6, ARA) from their diet to maintain growth, neural function, and reproduction (Brett et al., 2009; Parrish, 2009). These biomolecules are used in production of hormone and signalling molecules, but are also important in adjusting the fluidity in cellular phospholipid membranes (Arts and Kohler, 2009). Long-chain PUFAs are selectively retained in fish as the proportion of these molecules is often higher in the fish than its food sources. Thus, changes in phytoplankton community structure due to changes in lake chemistry (eutrophication and browning) are believed to impact directly on the PUFA content of organisms at higher trophic levels (Müller-Navarra et al., 2004; Strandberg et al., 2016; Taipale et al., 2016b; Taipale et al., 2018). It has been demonstrated that perch are capable of biosynthesizing ARA and DHA from α-linolenic (18:3n3, ALA) and linoleic acid (18:2n6, LIN), respectively (Geay et al., 2016). A recent study suggested that in lake ecosystems where phytoplankton produce less long-chain PUFAs, the biosynthesis of DHA from ALA in perch is intensified (Scharnweber et al., 2021). Terrestrial sources tend to be depleted especially with long-chain omega-3 (n3) FAs and enriched with omega-6 (n6) FAs, especially LIN (Hixson et al., 2015; Taipale et al., 2015; Mathieu-Resuge et al., 2021). Growth experiments have shown that invertebrates fed with allochthonous matter containing LIN will result high ARA content in their tissues (Goedkoop et al., 2007; Taipale et al., 2015). Moreover, fish lack the enzymes to convert saturated and mono-unsaturated fatty acids to PUFAs (Cook and McMaster, 2004). Thus, n3:n6 ratio can be used as a proxy for terrestrial and aquatic dietary sources, where higher values indicate more utilization of aquatic sources (Hixson et al., 2015). Perch do not store large quantities of lipids in the muscle tissue (Orban et al., 2007), yet different diets may result in differing lipid content and composition in muscle tissue (Jankowska et al., 2010).

Mercury (Hg) is a globally distributed harmful metal occurring in inorganic and organic forms that accumulate in organisms over time and magnifies in food webs through trophic linkages (e.g. Lavoie et al., 2013; Lescord et al., 2018a; Wang et al., 2019). Inorganic mercury compounds enter lakes via atmospheric deposition or leaching from the catchment area, and both of these pathways are intensified by anthropogenic activities such as energy production (e.g. coal and waste burning) and landscape modification (e.g. mining and intensive forestry practises) (Pacyna et al., 2010; Ahonen et al., 2018). Bacterial activity, especially in anaerobic conditions, has been shown to modify inorganic mercury compounds to an organic methylated form, MeHg (Eckley et al., 2005), and intensive forestry provide more favourable methylation conditions in catchment areas (Eklöf et al., 2018). Mercury enters fish mainly via dietary sources (Hall et al., 1997) and has a tendency to bind to the sulphur-containing amino acids in muscles and is therefore not excreted efficiently by fish (Thera et al., 2019). MeHg is s the dominant mercury form in fish muscle (Lescord et al., 2018a). Dissolved organic carbon (DOC) acts as a vector for transporting mercury from catchment areas to lakes, especially in low pH environments (Driscoll et al., 1995; Ullrich et al., 2001; Porvari and Verta, 2003). Lake sulphate concentration and thus lower pH enhances the bioavailability of mercury, and the methylation process occurs in the water column and sediment via the activity of sulphur-reducing bacteria and many other microbes (Winfrey and Rudd, 1990; Ponce and Bloom, 1991; Driscoll et al., 1995; Rask et al., 2007; Gilmour et al., 2013). Increasing organic matter in lakes (browning) and allochthony have been shown to be associated with increasing MeHg and total mercury (THg) content of zooplankton and fish (Strandberg et al., 2016; Braaten et al., 2018; Poste et al., 2019; Wu et al., 2021). However, mercury bioaccumulation factors in invertebrates and fish tend to decline with increasing DOC (after ca. 8–11 mg L^{-1}) and DOC aromaticity possibly through reduced bioavailability of mercury in the bottom of the food webs (French et al., 2014; Braaten et al., 2018; Lescord et al., 2018b). Increasing in-lake primary production has been suggested to result in lower mercury content in fish, possibly through biodilution in the food web and increased individual growth (Razavi et al., 2015; Finley et al., 2016; Keva et al., 2017; Kozak et al., 2021).

Long-chain n3 FAs, especially EPA and DHA, are beneficial for human neural development, neural and immune function, also decreasing e.g., cardiovascular disease risks (Parrish, 2009; Swanson et al., 2012). Freshwater fish are generally good sources of n3 fatty acids for humans (e.g. Gladyshev and Sushchik, 2019), but at the same time exposing consumers to mercury (e.g. Braaten et al., 2019). Mercury has been shown to cause neurological disorders and fatalities in high doses and chronic exposures (Ratcliffe et al., 1996). Daily minimum and maximum threshold limits of n3 PUFAs and mercury (e.g. European Food Safety Authority, 2012a, 2012b), respectively, enable combining the beneficial and the adverse health impacts of these compounds using a hazard quotient modelling (Gladyshev et al., 2009; Razavi et al., 2015; Strandberg et al., 2016). Despite the extensive literature on mercury and fatty acids, there are no previous studies directly measuring connections between fish allochthony and fish THg, FA content, and their derivative, hazard quotient.

In this study, a space-for-time substitution approach was used to test whether environmental change is likely to increase the dependence of a generalist consumer, Eurasian perch, on terrestrial organic matter sources in boreal lakes. And further, whether this shift in energy sources results in decreasing nutritional value (PUFA and THg) of fish for human consumption. Perch is one of the most common fish in Europe, showing a general ontogenetic dietary shift from invertivory (zooplankton and benthos feeding) to piscivory (fish feeding) from ca. 15 cm onwards in boreal lakes (e.g. Kottelat and Freyhof, 2007; Estlander et al., 2010; Sánchez-Hernández et al., 2021). In small humic lakes (characterized as higher DOC), perch supposedly rely more on terrestrial subsidies due to higher organic matter flow from the catchment entering food webs via bacterial activity. Thus, in this study it was hypothesized i) that perch allochthony is related positively with increasing DOC concentration and forest cover % in the catchment and negatively with lake productivity variables such as proportion of agricultural area of the catchment. Secondly, it was hypothesized ii) that in small forest lakes with high DOC concentration and allochthony, perch muscle THg content and n6 PUFAs are higher and n3 PUFAs content lower than in more productive and larger lakes. Finally, it was hypothesized iii) that these THg and PUFA trends would overall display decreasing quality (defined by hazard quotient calculation) of perch muscle quality for human consumption along the studied environmental gradients.

2. Materials and methods

2.1. Study region and field sampling

Thirty-one lakes were sampled in 2016–2018 (July–August) located in Southern and Central Finland across a DOC (5.0–24.3 mg L^{-1}) and total phosphorus (TP) gradient (4.8–118.4 μg L^{-1}) (Fig. 1 Table 1). Basic lake water chemistry analyses (DOC; total nitrogen [TN]; TP; chlorophyll-a [chl-*a*]; pH) were conducted for water samples collected from the epilimnion. Dissolved organic carbon and total nitrogen were analyzed from filtered (GF/F) water samples with a total organic carbon analyzer (TOC-L, Shimadzu, Japan) total nitrogen measuring unit (TONM-L, Shimadzu, Japan). Here calibration curves from potassium hydrogen phthalate ($C_8H_5KO_4$: 3, 6, 10, 15, 30 mg L^{-1}) and potassium nitrate (KNO_3: 0.15, 0.5, 0.75, 1.5 mg L^{-1}) were used to quantify the sample DOC and TN concentrations, respectively. Analytical accuracy (% error) and precision (RSD of multiple standards) for DOC were 5.8% and 1.1%, and for TN they were 1.9% and < 0.1%, respectively. TP, chl-*a* and pH values were derived from the Finnish Environment Institute HERTTA-database (www.syke.fi/avoindata).

Lake morphometry characteristics (average depth, max depth, surface area) were derived from an open database (Finnish Environmental Institute, OIVA). Lake catchment area characterises were derived from

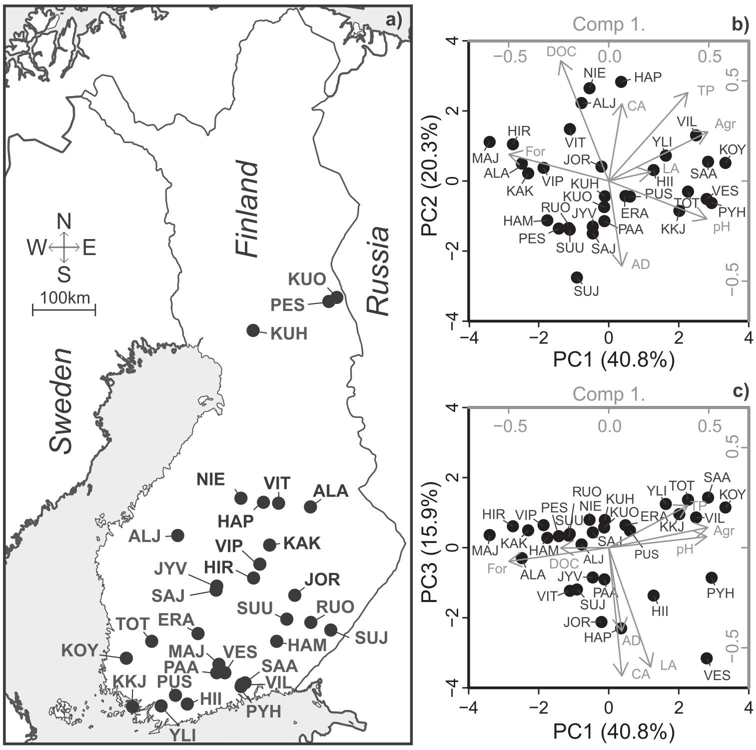

Fig. 1. Map of the study region (a) and principal component analysis orientation of the first three PCs (b and c) explaining most of the variation (total 76.9%) across the studied environmental gradients. PC1 express mainly forest cover %; For (−), agriculture cover %; Agr (+), pH (+) and total phosphorus; TP (−). PC2 indicate mainly lake average depth; AD (−), TP (+) and dissolved organic carbon; DOC (+). PC3 combine mainly lake morphometrics [AD (−), lake area; LA (−), catchment area; CA (−)]. The grey arrow lengths express the importance (loading) of an environmental variable for the PCs, loading scales (grey) are at the top and right sides of panels b and c. For lake name abbreviations and PC scores, see Table 1 and for variable loadings on the PCs, see Table S1.

Table 1

Study lakes listed in alphabetical order. Lake name, lake code, coordinates, lake morphometrics, chemistry, and catchment area characteristics. Lake area (LA), average depth (AD), maximum depth (MD), total phosphorus (TP), total nitrogen (TN), chlorophyll *a* (chl-*a*), dissolved organic carbon (DOC), pH, catchment area (CA). Catchment area characteristics are presented as cover % from the catchment area: urban area (Urb), agriculture area (Agr), forest in peatlands (For_p), forest in mineral soils (For_m), total forest area (For), wetland area (Wetl) and waterbodies (Wat). PC1-PC3 are the first three axes from the principal component analysis. Variable units are listed at the second header row in brackets, "NA" referes to not available.

Lake	Code	Coordinates	LA	AD	MD	TP	TN	chl-*a*	DOC	pH	CA	Urb	Agr	For_p	For_m	For	Wetl	Wat	PC1	PC2	PC3
		(°N, °E)	(km^2)	(m)	(m)	(μg L^{-1})	(μg L^{-1})	(μg L^{-1})	(mg L^{-1})		(km^2)	(%)	(%)	(%)	(%)	(%)	(%)	(%)			
Alajärvi	ALJ	63.0, 23.9	11.1	1.4	7.0	65.7	907	24.7	19.5	6.9	408.0	3.3	6.2	11.3	67.5	78.8	0.6	11.1	−0.78	2.23	0.09
Eräjärvi	ERA	61.6, 24.6	8.4	2.1	10.0	32.2	610	14.2	7.7	7.4	58.2	4.0	15.5	4.8	60.0	64.8	1.6	14.1	0.48	−0.43	0.64
Haapajärvi	HAP	63.6, 27.0	25.9	2.8	8.5	80.0	1013	39.4	14.4	7.0	1936.8	2.2	14.6	19.9	55.2	75.1	3.2	5.0	0.35	2.83	−2.30
Hiidenvesi	HII	60.3, 24.2	29.1	6.7	29.4	53.5	1018	15.3	8.6	7.8	925.9	6.8	16.0	6.4	61.0	67.4	0.8	9.0	1.28	0.31	−1.37
Hirvijärvi	HIR	62.4, 27.0	1.0	1.0	3.0	29.7	560	19.8	17.0	6.4	14.8	0.3	0.0	29.4	61.2	90.6	0.7	8.4	−2.74	1.05	0.61
Hämeenjärvi	HAM	61.3, 27.3	1.3	4.5	17.0	7.7	300	6.0	6.2	6.7	14.4	1.8	0.7	6.2	76.5	82.7	0.8	14.0	−1.75	−1.12	0.28
Joroisselkä	JOR	62.2, 27.9	13.0	9.1	55.0	25.3	485	16.0	10.4	7.2	1472.0	2.9	7.7	14.6	56.9	71.5	2.7	15.3	−0.21	0.41	−2.12
Jyväsjärvi	JYV	62.2, 25.7	3.3	7.0	25.0	23.8	561	9.5	8.6	7.1	366.9	15.3	5.7	6.2	65.1	71.2	0.7	7.1	−0.46	−1.30	−0.85
Kakkisenjärvi	KAK	62.7, 27.2	1.9	2.1	6.0	26.8	400	16.0	11.8	6.2	25.4	0.5	0.0	23.2	60.4	83.6	0.4	15.5	−2.30	0.22	0.50
Kakskerranjärvi	KKJ	60.3, 22.2	1.6	6.4	15.2	68.2	439	7.7	5.8	8.0	10.2	13.7	17.4	0.5	51.7	52.2	0.6	16.0	2.02	−0.85	0.96
Kuhajärvi	KUH	65.9, 26.7	3.1	2.1	6.3	34.4	620	27.6	9.3	7.0	16.0	3.1	5.3	18.3	39.1	57.4	5.5	28.7	−0.11	−0.44	0.79
Kuontijärvi	KUO	66.1, 29.0	6.0	1.7	12.9	29.3	540	12.0	6.8	7.5	89.1	3.7	5.2	17.2	52.1	69.3	6.2	15.5	−0.13	−0.74	0.57
Köyliönjärvi	KOY	61.1, 22.3	12.4	2.6	12.8	115.4	1327	72.2	7.2	8.4	145.2	8.5	27.7	8.6	45.0	53.6	2.0	8.2	3.33	0.52	1.15
Majajärvi	MAJ	61.2, 25.1	0.03	4.0	12.0	32.0	703	6.0	17.3	5.8	1.9	0.0	0.0	8.6	87.8	96.4	0.2	3.4	−3.40	1.12	0.36
Niemisjärvi	NIE	63.6, 26.5	4.2	1.6	5.0	63.6	984	36.9	24.3	7.0	181.1	2.5	19.6	18.1	55.1	73.2	1.9	2.9	−0.55	2.65	0.79
Pesosjärvi	PES	66.3, 29.5	0.4	4.7	11.9	5.9	230	1.8	6.2	7.4	5.9	0.1	0.0	16.2	70.1	86.3	3.9	9.7	−1.42	−1.36	0.33
Pusulanjärvi	PUS	60.5, 24.0	2.1	4.9	10.6	48.7	720	25.7	6.5	7.5	223.4	5.4	15.4	5.1	64.8	69.9	0.6	8.8	0.61	−0.45	0.49
Pyhäjärvi	PYH	60.7, 26.0	13.0	21.1	68.0	96.0	1350	20.8	7.6	8.0	457.7	4.2	30.2	2.4	56.7	59.1	0.5	5.9	2.94	−0.63	−0.86
Pääjärvi	PAA	61.1, 25.1	13.4	14.8	85.0	10.4	1350	5.5	9.8	7.3	223.8	3.0	15.3	12.2	61.4	73.5	0.6	7.6	−0.12	−1.15	−0.90
Ruokojärvi	RUO	61.6, 28.4	1.3	4.7	24.1	5.2	290	2.7	6.3	7.1	6.2	0.5	1.8	6.4	69.7	76.1	1.0	20.6	−1.15	−1.34	0.34
Suuri Jukajärvi	SUJ	61.5, 28.9	3.6	23.0	49.8	4.8	333	3.4	6.3	7.2	56.6	2.1	1.0	11.4	65.7	77.1	2.0	17.8	−0.91	−2.75	−1.19
Suuri-Vahvanen	SUU	61.7, 27.5	1.3	4.1	15.0	5.6	305	2.1	5.9	7.0	7.0	2.5	0.4	4.2	68.6	72.7	0.5	23.9	−1.11	−1.39	0.39
Sääksjärvi	SAJ	62.2, 25.7	0.6	5.6	16.1	14.7	290	4.7	6.3	7.1	6.1	30.2	0.0	1.8	58.0	59.8	0.3	9.7	−0.46	−1.50	0.43
Sääskjärvi	SAA	60.8, 26.0	5.1	2.4	5.0	107.2	938	28.8	6.1	7.7	65.1	3.8	33.2	0.8	54.3	55.1	0.2	7.7	2.84	0.55	1.43
Tottijärvi	TOT	61.4, 23.3	0.7	2.4	5.7	70.3	977	50.6	6.7	8.0	5.5	12.6	22.7	1.6	49.1	50.7	0.9	13.2	2.26	−0.30	1.37
Vesijärvi	VES	61.0, 25.6	107.5	6.1	40.0	43.1	443	5.0	5.0	7.6	510.1	8.6	18.0	2.1	49.0	51.1	1.7	20.6	2.80	−0.51	−3.16
Viitaanjärvi	VIT	63.6, 27.3	3.6	3.9	14.7	44.5	595	16.8	12.3	6.7	1334.6	1.8	8.0	25.9	56.3	82.2	3.5	4.3	−1.11	1.48	−1.23
Viipperonjärvi	VIP	62.5, 27.0	1.0	3.3	10.0	56.9	NA	10.0	11.1	6.5	10.2	0.6	0.1	13.9	74.3	88.2	0.3	10.9	−1.86	0.38	0.64
Villikkalanjärvi	VIL	60.8, 26.2	7.2	2.9	8.9	118.4	1470	22.6	8.5	7.6	411.4	4.1	30.8	2.6	59.1	61.7	0.3	3.0	2.49	1.30	0.86
Ylisjärvi	YLI	63.3, 23.3	1.8	2.1	4.3	95.6	998	30.8	8.4	7.5	129.1	5.6	24.5	4.8	60.2	65.0	0.7	4.2	1.63	0.72	1.25
Älänne	ALA	63.5, 28.1	10.0	3.2	15.4	32.8	480	11.0	10.2	5.5	357.6	0.2	0.5	32.6	33.7	66.3	1.4	31.7	−2.48	0.50	−0.30

CORINE 2018 land cover (level 4) raster maps (pixel size: 20 by 20 m; https://syke.fi/avointieto) using geographical information system software (QGIS 3.16.13). The following land coverage types were obtained: urban area, agriculture activities, peatland forests, mineral soil forests, total forests, wetlands, water bodies (Table 1). The sampled lakes varied in size (lake area range: 0.03–107.5 km^2, lake mean depth range: 1.0–23 m) and catchment area characteristics (forested area range: 50.7–96.4%, agricultural area range: 0–32.6%) (Table 1). In Southern and Central Finland most of the forested catchment areas are under heavy forestry practises (clear cutting, site preparation, peatland ditching), and only a small minority of forests are protected (Sallinen et al., 2019; Statistics Office Finland, 2021). For example, approximately 75% of the mires have been ditched to increase forest growth contributing more than one third of the total forest coverage in the region (Sallinen et al., 2019). Thus, the forest cover % in the catchment here is a good indicator of forestry activities.

For stable isotope analysis of DOC, 500 mL of water from lake inlet streams was filtered using Millipore tangential flow filtration (Pellicon P2GVPP05 cassette, Merck KGaA, Germany, pore size 0.22 μm) and filtrate was freeze-dried (Alpha 1–4 LD Plus, Martin Christ Gefriertrocknungsanlagen GmbH, Germany) at −55 °C until all the water was gone. The desiccated powder was transferred to a 1.5 mL class vial. Benthic algae were collected by scraping visually green patches of algae from hard substrate surfaces. Lake water (12 mL) was also collected for δ^2H measurements. Bulk zooplankton samples were collected from the deepest point of the lake with vertical hauls from thermocline to surface using a zooplankton net (mesh size: 250 μm, diameter: 25 cm). Cladocera were separated from the bulk zooplankton sample with a glass plate technique, whereby the bulk zooplankton sample is washed in a tall beaker with tap water resulting in trapping of cladocerans at the surface layer from where they were picked with a glass plate. Samples were checked under stereomicroscope to confirm they consist only cladocerans. Zooplankton samples were transferred to in 1.5 mL glass vials, freeze-dried and stored in a desiccator for stable isotope analyses.

Perch were sampled with Nordic multi-mesh gillnets (length: 30 m, height: 1.5 m, mesh sizes: 5; 6.25; 8; 10; 12,5; 15,5; 19,5; 24; 29; 35; 43 and 55 mm). Fish were euthanized immediately and transported in ice to the laboratory, where total length (cm) and mass (g) were measured. In this study perch >15 cm was considered as predominantly piscivorous, and were verified with stomach content analyses (mean fish contribution of stomach fullness: 65.8%, $n = 85$ stomachs with food). Total of 132 perch individuals (total length > 15 cm) were selected for this study, 2–5 ind. Lake^{-1} with an average (± SD) of 4.3 ± 0.8 ind. Lake^{-1}. A small piece of dorsal muscle was dissected from each individual, placed in a plastic vial, freeze-dried and stored at −80 °C prior to powdering the sample for fatty acid, mercury and stable isotope ratio analyses.

2.2. Stable isotope ratio, total mercury and fatty acid analyses

Stable isotope ratios of perch muscle were analyzed from 3 to 5 individual per lake. Stable isotope ratios of cladocera, bulk zooplankton, inlet DOC, benthic algae, and lake water were analyzed from pooled samples from three different parts of the lakes to obtain general average for each sample types. The pooling of the samples was done on site. Freeze-dried sample powder was weighed in tin cups (0.60 ± 0.05 mg) for stable nitrogen isotope ratio analyses (δ^{15}N). δ^{15}N was analyzed at the University of Jyväskylä, Finland using a continuous flow isotope ratio mass spectrometer (Thermo Finnigan DELTA^{plus}, Thermo Fisher Scientific Inc., US) coupled to elemental analyzer (FlashEA 1112, Thermo Fisher Scientific Inc., US). The primary international standard, atmospheric nitrogen, was used as the reference for nitrogen isotopes to calibrate lab-specific standard material (freeze-dried powder of pike (*Esox lucius*) muscle and birch leaves). Multiple lab-specific standards were added to each batch run. Analytical error for each run, derived from multiple laboratory standards, was below ~0.2‰ for δ^{15}N.

For hydrogen stable isotope (δ^2H) analyses, freeze-dried sample powder and reference materials were weighed in silver cups (0.350 ± 0.005 mg). Silver cups were stored open in laboratory atmosphere with laboratory standards for at least five days prior to folding the cups in order to allow interchange of exchangeable hydrogen between samples and laboratory air (Wassenaar and Hobson, 2003). Hydrogen stable isotope analyses were done at the University of Jyväskylä, Finland using an Isoprime 100 isotope ratio spectrometer (Isoprime Ltd., UK) coupled to an Elementar vario PYRO cube elemental analyzer (Elementar Analysensysteme GmbH, Germany). Multiple samples of two reference materials (caribou hoof [CBS], kudu horn [KHS]) were analyzed in each run expressed relative to VSMOW (Standard Mean Ocean Water) (Soto et al., 2017). Here, the standard deviations of replicate reference materials within each run were always below ~3‰. The lake water samples were stored in a refrigerator before δ^2H analyses that were performed at the University of Oulu, Finland with a CRDS laser instrument, Picarro L2120-i, (Picarro Inc., US). Here, the standard deviations between standards were below ~1‰. All stable isotope ratios are expressed as ratio to the corresponding international standard using the delta notation.

Total mercury (THg) analyses were conducted with an atom absorption spectrometer (Milestone DMA 80, Milestone Srl, Italy) by weighing freeze-dried perch muscle powder (25 ± 5 mg) to sample boats. At the beginning and at the end of each run of the THg analyzer (40 slot autosampler), certified reference material DORM-4 (National Research Council Canada, mean ± SD: 0.410 ± 0.055 μg THg g^{-1} dry weight) was measured along with blanks. The mean DORM-4 ($n = 10$) recovery was 102.0 ± 3.3% and the analyzed blanks were effectively zero: 0.0 ± 0.0 ng THg g^{-1}. Analyzed duplicates for each 5th sample had a mean RSD of 1.5 ± 1.9% ($n = 32$ pairs). As there were differences in the size distributions of the sampled perch among different lakes and THg correlates linearly with total length, the measured THg values were size-weighted with the ratio of whole study mean perch total length (20.1 cm, Table S1) to individual length. The average of length adjusted THg values of duplicate sample was used in the subsequent statistical analyses.

For fatty acid analysis, powdered perch muscle samples were weighed in tin cups (10 ± 1 mg) and the lipids from the sample were extracted in glass test tubes using chloroform-methanol solution (2:1 vol), sonicated and spiked with internal standard (Free c23:0). Duplicate analyses were made for every 10th sample. Distilled ultrapure water (0.75 mL) was added to test tubes to separate water-soluble compounds from lipids. Methylation reagent (1% H_2SO_4 in methanol) was added to samples and the methylation reaction was carried out during incubation in a water bath (50 °C for 20 h). The fatty acid methyl esters produced were diluted in hexane (1 mL) and analyzed with a gas chromatograph (GC) attached to a mass spectrometer (MS) (GC-2010 Plus and QP-2010 Ultra, Shimadzu, Japan) and using a Zebron DB-23 column (60 m × 0.25 mm × 0.2 μm). The injection temperature was 270 °C and the interface temperature 250 °C, and the linear velocity in the column was 36.3 cm s^{-1}. Samples were run with a splitless injection method.

FAs were identified based on ion spectra and quantified based on four-point standard mixture calibration curves (15 ng μL^{-1}, 50 ng μL^{-1}, 100 ng μL^{-1}, 250 ng μL^{-1}; GLC 566c, Nu-Chek Prep Inc., US) with GCMS solution software version 4.42 (Shimadzu, Japan). The calibration curves were analyzed before each run and coefficients of determination between peak area and standard FA concentrations were always higher than 0.995. Recovery % of the internal standard (c23:0) was 88.3 ± 12.0% ($n = 132$) and was used to adjust FA concentrations individually for each sample. An external EPA and DHA standard was analyzed in each batch to validate calibration curves, and the analyzed recovery percentages were 103.9 ± 1.7 and 81.5 ± 2.4 ($n = 7$) for EPA and DHA, respectively. Average sums of FA profile error and PUFA profile error between duplicate samples were 2.1 ± 1.7% (n = 13 pairs) and 1.0 ± 1.1% (n = 13 pairs), respectively. Only the content of PUFAs (DHA, EPA, ALA, ARA and LIN) are reported in this study.

2.3. Allochthony index modelling

Trophic level (TL_C) of each perch sample was calculated with Eq. (1) using a one-source model according to Post (2002):

$$TL_C = \frac{\delta^{15}N_C - \delta^{15}N_{base}}{\Delta_N} + \lambda, \tag{1}$$

where the base was pure cladocerans, or bulk ZPL in two cases (Hirvijärvi, Niemisjärvi) where insufficient cladoceran sample was obtained; thus, the estimated trophic level of the base (λ) was fixed to a value of 2. In the equation, $\delta^{15}N_{base}$ and $\delta^{15}N_C$ represent the nitrogen isotope ratio of the base and the consumer (perch), respectively. A trophic fractionation (Δ_N) of 3.4 in the trophic level estimations was used (Post, 2002).

Prior to allochthony index estimations, the percentage of environmental hydrogen in a consumer needs to be estimated. This was done with Eq. (2) according to Vander Zanden et al. (2016) as following:

$$\omega_{compound} = 1 - (1 - \omega)^{TL_C - 1}, \tag{2}$$

where $\omega_{compound}$ is the proportion of water δ^2H in consumer δ^2H values, ω is the estimated proportion of water δ^2H entering consumer (0.2 per each trophic level; Wilkinson et al., 2015). $TL_C - 1$ represents the difference between consumer and basal hydrogen sources in trophic levels, and TL_C was obtained from Eq. (1).

Consumer hydrogen isotope values were modelled with Eq. (3) from Vander Zanden et al. (2016):

$$\delta^2H_{C\ 100\%base\ 1\ or\ 2} = \left(\omega_{comp} \times \delta^2H_{water}\right) + \left(1 - \omega_{comp}\right) \times \delta^2H_{base\ 1\ or\ 2}, \tag{3}$$

where $\delta^2H_{C\ 100\%base\ 1\ or\ 2}$ are theoretical consumer δ^2H values for individuals receiving unexchangeable hydrogen 100% from base 1 or 2, respectively. This approach corresponds roughly to the idea of trophic level-corrected base δ^2H values. In the Eq. (3), δ^2H_{water} is the measured lake water δ^2H value, $\delta^2H_{base\ 1}$ equals to δ^2H value of inlet DOC (allochthonous source) and $\delta^2H_{base\ 2}$ is the hydrogen isotope ratio of phytoplankton (autochthonous source). The autochthonous source was modelled phytoplankton hydrogen isotope ratio with discrimination estimate of (−111.66‰ from water) derived from the mean difference between measured benthic algae δ^2H and pelagial water δ^2H values. This was done because for most lakes it was impossible to obtain sufficiently pure samples of phytoplankton for stable isotope analysis (SIA).

A perch allochthony index was calculated using a modified basic two-source mixing model (Post, 2002), where output values should be from 0 to 1, with low values indicating a predominantly autochthonous hydrogen source and high values a predominantly allochthonous hydrogen source:

$$\text{allochthony index} = \frac{\left(\delta^2H_C - \delta^2H_{C\ 100\%base\ 2}\right)}{\left(\delta^2H_{C\ 100\%base\ 1} - \delta^2H_{C\ 100\%base\ 2}\right)}, \tag{4}$$

where δ^2H_C measured δ^2H value of the consumer other parts from Eq. (3). Eqs. (1)–(4) was used to calculate the allochthony index to be able to visualize the relation of "Trophic level corrected base" δ^2H values to perch δ^2H values (Fig. 2). Eqs. (1)–(4) used in this study reduce to the equations in Karlsson et al. (2012) and provide exactly the same values, thus validating the introduced intermediate phase (Eq. (3)) of the allochthony index calculations.

2.4. Hazard quotient modelling

A human health risk-benefit ratio for fish consumption, the hazard quotient (HQ), was calculated according to Gladyshev et al. (2009):

$$HQ = \frac{R_{EFA} \times C_{THg} \times 0.85}{C_{EFA} \times R_{MeHg} \times AW}, \tag{5}$$

where HQ values express a ratio of mercury gain exceeding the health limitations when fulfilling the daily requirement of EPA + DHA by eating fish. R_{EFA}

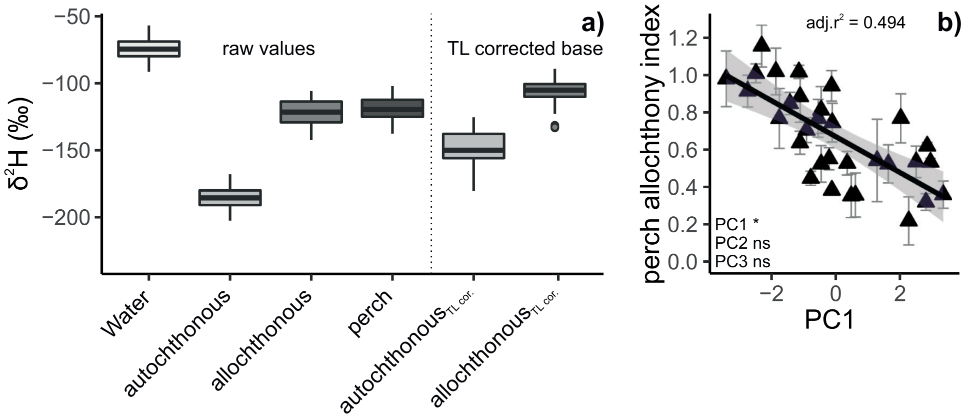

Fig. 2. Boxplot showing the δ^2H (‰) data used in the allochthony index calculations (all lakes pooled) (a) and relationship between PC1 (see Fig. 1) and perch allochthony (b). In panel a, bold horizontal line indicate the median value, the boxes represent first and third quartiles and the whiskers represent minimum and maximum values. Outliers (black circles) are presented if there are data points smaller or larger than 1.5 times the difference between first and third quartiles (1.5*IQR). Water from epilimnion (lightest grey), autochthonous source (light grey), allochthonous source (grey), perch (dark grey). Dashed vertical line separates raw δ^2H values (left) and trophic level (TL) adjusted terrestrial and aquatic base (autochthonous$_{\text{TL cor.}}$ and allochthonous$_{\text{TL cor}}$; right) δ^2H values. In panel b, regression line (black) and 95% confidence intervals (grey shaded area) with regression equation and adjusted r^2 value are shown. Allochthony index correlation on other PCs are shown in bottom-left of panel b, where "ns" indicate statistically non-significant and "*" indicates significant correlation. Model details including regression equation, F statistics and *p*-value can be found from Table S3.

refers to daily recommended EPA + DHA supply for humans (here 250 mg EPA + DHA d^{-1}, European Food Safety Authority, 2012a). C_{THg} is the THg concentration analyzed in the fish muscle (μg g^{-1} dry weight), the factor 0.85 was used to convert THg to methyl mercury content as >85% of total mercury in fish muscle is methyl mercury (e.g., Lescord et al., 2018a), C_{EFA} is EPA + DHA content of the fish muscle (mg g^{-1} dry weight), R_{MeHg} is the maximum tolerable daily intake of methyl mercury (0.186 μg kg^{-1} body weight d^{-1}: European Food Safety Authority, 2012b), and AW is the average weight of an adult. HQ > 1 represents a risk of adverse effect for human health when gaining all the required EPA and DHA from the fish, whereas HQ < 1 represents no risk (Gladyshev et al., 2009).

2.5. Statistical methods

The water chemistry variables (TP, DOC and pH), lake size related parameters (mean depth, lake area, catchment area size), and the most significant land-use activities in Finland (agriculture and forest coverage) were selected for principal component analysis (PCA) (Fig. S2). PCA using a correlation matrix was conducted to reduce the number of environmental dimensions (Fig. 1, Table 1). Linear regression analyses were used to test environmental impact (PC1–PC3) on perch allochthony (i), muscle FA and THg content (ii) and their derivative hazard quotient (iii). The variables used in the linear regression models were continuous, residuals in the models were approximately normally distributed (Fig. S3) and no obvious outliers were detected with Cook's distance. The connections between lake chemistry, morphometrics and catchment area characteristics were individually tested with Pearson's correlation and visualised using simple linear regression against perch allochthony, muscle FA content, THg content and perch muscle hazard quotient (Bonferroni correction implemented). These individual environmental variables were tested to represent the main environmental drivers behind the trends among principal components and perch muscle parameters. As an additional statistical approach, GLM models was created for perch allochthony, THg, PUFA and hazard quotient using the same independent variables than in the PCA approach. Here, the most parsimonious models were selected with both direction selection method based on AIC using MASS package (Venables and Ripley, 2002) in R. All statistical analyses were performed using R language (version 3.5.3; R Core Team, 2019) and the graphical illustrations were made with the ggplot2 package (Wickham, 2016).

3. Results

The cumulative proportion of variance explained in the lake environmental data exceeded 75% with the first three components, which were thus considered to be important for the subsequent analyses. PC1 explained 40.7%, PC2 20.3% and PC3 15.9% of the variance in the lake environmental data (Fig. 1, Table S1). Catchment area forest (+) and agriculture (−), TP (+) and pH (+) had the strongest loadings on PC1, which can thus be considered as a gradient of lake catchment area land-use and nutrient concentration, where low pH forest lakes are ranked at the left and nutrient rich agriculture lakes at the right end of the PC1 axis (Table S1). TP (+), DOC (+), lake average depth (−) and catchment area size (+) had the highest loadings for PC2 and thus can be considered to represent a gradient from larger clear water lakes towards smaller and murkier lakes (Table S1). PC3 mainly represents lake morphometrics, where lake average depth, lake area and catchment area size had negative loadings (Table S1).

The δ^2H values from the epilimnion water varied between −56.8 and −91.4‰ with a mean of −74.5 ± 8.8‰ (Fig. 2). The estimated autochthonous source (epiphytic algae) δ^2H values ranged from −167.9‰ to −202.6‰ with a mean of −185.6 ± 8.8‰ (Fig. 2). The allochthonous source (inlet DOC) δ^2H values ranged from −105.8‰ to −142.5‰ with a mean of −120.9 ± 9.5‰ (Fig. 2). Perch δ^2H values varied between −102.1 to −137.6‰ with a mean of −119.2 ± 8.3‰ (Fig. 2). Baseline δ^2H values corrected for trophic level ranged for allochthonous sources from −89.2 to −133.4‰ (mean ± SD: −105.4 ± 8.6‰) and for autochthonous sources from −125.4 to −180.4‰ (−148.4 ± 12.1‰) (Fig. 2). The mean value for the perch allochthony index was 0.67 ± 0.25, with a range of 0.22–1.15 (Fig. 2, Table S2). Lake PC1 scores correlated negatively with perch allochthony index (adjusted $r^2 = 0.542$, $p < 0.0001$) (Fig. 2). Perch allochthony index was negatively related to pH (adjusted $r^2 = 0.44$, $p < 0.0001$) and agricultural area (adjusted $r^2 = 0.43$, $p < 0.0001$) in the catchment area (Fig. S4). Allochthony index correlated positively (adjusted $r^2 = 0.337$, $p < 0.0001$) with forest coverage in the lake catchment area (Fig. S4).

Lake PC1 scores correlated negatively with perch muscle THg (adjusted $r^2 = 0.542$, $p < 0.0001$), ARA (adjusted $r^2 = 0.228$, $p = 0.0039$) and LIN (adjusted $r^2 = 0.134$, $p = 0.0242$) values (Fig. 3). Lake PC2 scores correlated negatively with perch muscle DHA (adjusted $r^2 = 0.109$, $p = 0.0393$) and positively with ALA (adjusted $r^2 = 0.112$, $p = 0.0369$) content (Fig. 3). Agriculture coverage correlated negatively with perch THg, ARA,

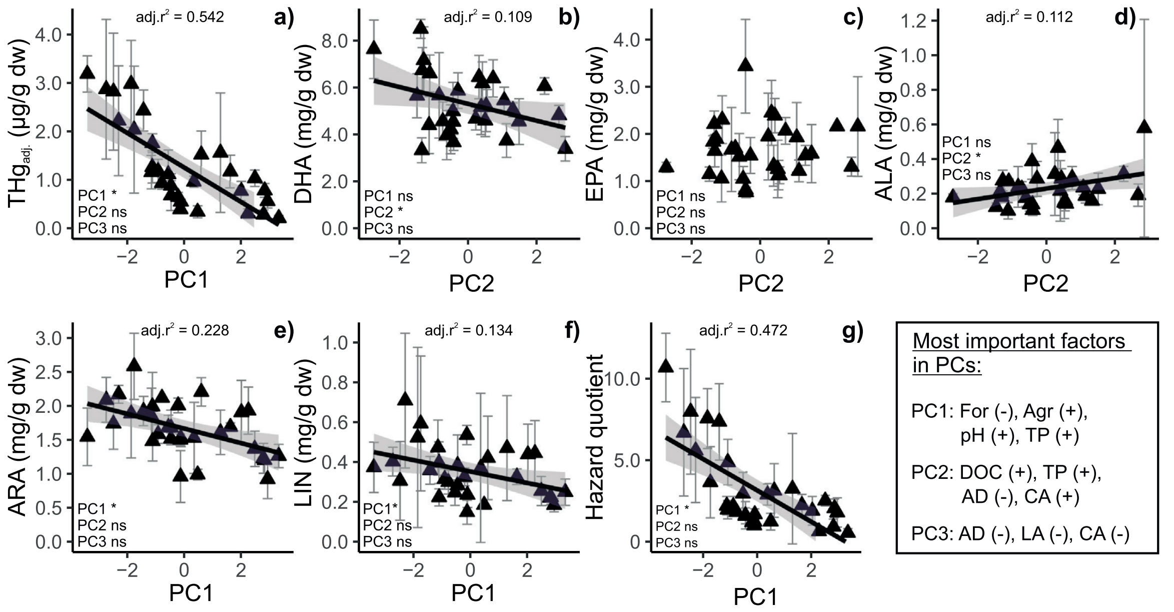

Fig. 3. Relationship between environmental gradients and perch THg (a), DHA (b), EPA (c), ALA (d), ARA (e), LIN (f) content and hazard quotient (g). Note the different x-axis in panels a, e-g (PC1) and b-d (PC2). Lake mean values are expressed with black triangles and SD with grey whiskers. The regression lines (black) and 95% confidence intervals (grey shaded area) with adjusted r^2 value are shown for each panel if the regression was statistically significant. Model details including regression equations, F statistics and *p*-values can be found from Table S3. Most important environmental factors impacting PCs (|factor loading| > 0.3) are shown in bottom-right box, where parameter abbreviations correspond to Table 1. Compound correlations on other PCs are shown in bottom-left of each scatterplot (also Fig. S9), where ns indicate statistically non-significant and * indicates significant correlation.

and LIN content and forest coverage positively with perch THg (Fig. S5). However, no correlation between lake chemistry, catchment area characteristics and perch n3 PUFAs was found (Fig. S5, Table S6). The generated general linear models for each dependent variable (Table S4–5) resembled a lot the PCA results and simple linear regression results. For example, agriculture coverage was selected to the most parsimonious models of LIN and ARA with negative signs, and pH was negatively associated with THg and LIN (Table S5).

The lowest hazard quotient (HQ) values were found from the eutrophic Lake Köyliönjärvi (0.6 ± 0.3) and the highest from the smallest and most humic Lake Majajärvi (10.7 ± 2.1); the total mean value for all lakes was 3.2 ± 2.8 (Table S1). Lake PC1 scores correlated negatively with perch muscle HQ (adjusted $r^2 = 0.472$, $p < 0.0001$) (Fig. S6). Lake forest coverage (adjusted $r^2 = 0.619$, $p < 0.0001$) in the catchment correlated positively with HQ (Fig. S6), while lake pH (adjusted $r^2 = 0.557$, $p < 0.0001$) and agricultural area (adjusted $r^2 = 0.233$, $p < 0.0035$) in the catchment correlated negatively with HQ (Fig. S6).

A positive linear correlation between perch allochthony and THg values (adjusted $r^2 = 0.339$, $p < 0.0001$) was found (Fig. S7). Perch allochthony correlated positively with perch ARA (adjusted $r^2 = 0.089$, $p = 0.0030$) and LIN content (adjusted $r^2 = 0.119$, $p < 0.0001$) (Fig. S7). No correlation was found between allochthony index and perch n3 content (Fig. S7). Allochthony and perch muscle HQ were positively correlated (adjusted $r^2 = 0.360$, $p < 0.0001$) (Fig. S7). Perch allochthony correlated positively with the sum of n6 FAs (adjusted $r^2 = 0.072$, $p = 0.0011$) and negatively with the n3:n6 ratio (adjusted $r^2 = 0.241$, $p < 0.0001$) (Fig. S8).

4. Discussion

4.1. Main results

In lakes with low pH and high forested catchment area perch relied more on terrestrial sources than in neutral and eutrophic lakes. However, no direct connection between lake DOC, morphometrics and perch allochthony was found. Perch muscle THg, ARA, and LIN content was elevated in lakes with high forest coverage and low pH. DHA content of perch muscle was higher and ALA content lower in deep clear water lakes compared to shallow murky ones. Perch allochthony was positively correlated with muscle n6 FAs and THg but did not have relationship to perch muscle n3 FAs. By combining the long-chain n-3 PUFA and THg results with hazard quotient modelling, the quality of perch muscle for human consumption decreases with increasing forest coverage and decreasing pH. This decreasing nutritional value for human consumption is clearly governed by the parallel trend with THg content and potentially mediated by increasing fish allochthony.

4.2. Environmental gradient and perch allochthony

The allochthony index of perch varied between 0.22 and 1.15 among the studied lakes. Generally, there seems to be less variation in allochthony values of perch individuals within a lake than between lakes. Theoretically, allochthony index values should vary between 0 and 1 (Post, 2002) indicating the proportion of allochthonous supply. In a few lakes (Eräjärvi, Ruokojärvi, Viitaanjärvi and Älänne) the value slightly exceeded one indicating some shortcomings in the modelling of allochthony or in the baseline sampling. For example, spatial and temporal differences in the source deuterium could lead to these higher allochthony index values. Although, in most cases the allochthony index did fall between 0 and 1, it should be noted that allochthony index was here used only as relative measure and not as an absolute indicator of derived allochthonous energy. Mean allochthony index value (0.63 ± 0.25) of current study was similar to a value observed from a boreal lake in Sweden (0.57) (Karlsson et al., 2012) and values from Canadian low-productivity lakes (0.6–0.7) (Solomon et al., 2011).

Perch allochthony increased with decreasing lake pH and increasing forest coverage in the catchment. Allochthony index was most strongly associated with lake water pH, with a negative correlation. Different to the initial expectations, there was no relationship between perch allochthony and lake DOC concentration. This indicates that browning or high DOC does not automatically lead to higher transfer of organic matter to food webs; instead, DOC quality (ie. molecular weight and structure) could be an important factor affecting community composition of heterotrophs responsible for the transfer. Furthermore, earlier research has indicated that increasing lake acidity reduces primary production and biomass, which has a negative impact on the nutritional quality of herbivorous zooplankton (Kwiatkowski and Foff, 1976; Locke and Sprules, 2000). This suggest that zooplankton and fish are forced to rely more on allochthonous carbon when phytoplankton is scarce (Taipale et al., 2016a). The observed clear positive correlation between perch allochthony and forest coverage in the lake catchment corroborates previous findings (Tanentzap et al., 2014). Lake DOC quantity and quality can be temporally variable, for example in response to variable rainfall events (Warner et al., 2020), and thus the used single snapshot measurements may not have adequately represented the longer-term organic matter flow or quality from the catchment. Therefore, catchment area land-use may provide a more integrated predictor for fish allochthony than single time point DOC concentration measurements. Interestingly, dividing catchment area forest coverage into soil types (peatland and mineral soils) did not increase the predictive power of catchment land-use for the perch allochthony index. Previously, it has been shown that peatland forest cover in lake catchments has a strong correlation with lake DOC (Kortelainen, 1993; Strandberg et al., 2016). However, in these studies a comparison between total forested area and peatland forest coverage in catchment was not made and the lake gradient of current study was geographically different and wider with potentially more confounding variables. Moreover, the allochthony index of the perch was low in the most eutrophic lakes such as Tottijärvi, Vesijärvi and Köyliönjärvi. The negative correlation with agricultural catchment area and perch terrestrial reliance likely reflects the higher within-lake primary production contributing to food web energy sources in the eutrophic lakes compared to more oligotrophic lakes.

4.3. Perch THg and FA content along environmental gradients

Perch muscle THg content were the highest in acidic, low productivity lakes with high forest cover % in the catchment. Lake acidity (pH) was clearly the best single environmental predictor explaining more than 50% of the variation in the mercury content of perch in different lakes. A similar negative relationship has been found for small boreal headwater lakes, which often have even more acidic water (Rask et al., 2021). Previous studies have found that more acidic lake water increases the direct accumulation of mercury from the environment to primary producers, bacteria, zooplankton and fish (Tsai et al., 1975; Watras and Bloom, 1992; Kelly et al., 2003; Porvari and Verta, 2003). However, direct accumulation of THg by respiration of fish represents only a minority (ca. 10–15%) of the total mercury burden (Hall et al., 1997), and is higher in low pH environments likely related to increased metabolic activities, decreased growth rate, and mercury bioavailability (Hall et al., 1997; Ponce and Bloom, 1991). Lower pH tends to increase the bioavailability of mercury through higher production of methyl mercury (Ponce and Bloom, 1991) and decrease the loss of volatile mercury from lake water (Winfrey and Rudd, 1990). Thus, ambient concentrations of reactive mercury could also be higher in the low pH lakes. Moreover, higher organic matter content in lakes has previously been linked to higher water column THg and MeHg concentrations (French et al., 2014; Braaten et al., 2018). Yet, the accumulation of mercury to organisms is highly dependent of the DOC quality as mercury bound to fulvic acids are not easily available for the bacteria and, consequently, for higher trophic level organisms (French et al., 2014; Lescord et al., 2018b). Unfortunately, mercury values from either water or lower trophic levels nor DOC quality estimates was available. Thus, in the low pH lakes the presumed higher accumulation of mercury to lower trophic levels will contribute significantly to THg accumulation in perch, even if the biomagnification factor was constant along the pH gradient (Ponce and Bloom, 1991). It should be also noted that in small headwater lakes, resources are often limited leading to slow growth rate which are related to higher mercury content in fish muscle (Sandheinrich and Drevnick, 2016; Rask et al., 2021).

Some studies have reported pH to be the most significant environmental factor explaining predator THg values (Porvari, 1998; Rask et al., 2007, 2021), whereas others have highlighted that lake DOC concentration or water colour could be the most important factor (Driscoll et al., 1995; Westcott and Kalff, 1996; Braaten et al., 2018). As DOC is typically mildly acidic and lowers lake pH, these interpretations of how DOC and pH impact fish tissue mercury content are not surprising (Thomas et al., 2020). Moreover, decreased bioavailability of mercury with increasing DOC concentration (after 8–11 mg L^{-1}) and DOC aromaticity lowers the bioaccumulation of mercury in highly humic lakes (French et al., 2014; Braaten et al., 2018). This has the potential to flatten organism mercury content towards more humic lakes complexing the relationship between DOC and organism mercury content (French et al., 2014). Here, no positive correlation between lake DOC concentrations and perch muscle THg content was found. Yet the highest mercury content in fish was observed from a humic lake (Majajärvi: 17 mg DOC L^{-1}). The negative correlation between forest cover and THg and the positive correlation between perch allochthony and THg could reflect a longer-term average organic matter flow and quality from the catchment area. Thus, catchment area land-use from forest coverage to agriculture, could be better predictors for perch muscle THg content than DOC concentration solely.

There was no correlation between perch muscle THg content and lake TP or chl-*a* concentration. However, a strong negative correlation with agricultural coverage in the catchment area and perch muscle THg was observed. The agricultural area in the catchment area is likely a seasonally more stable predictor of lake productivity than TP and chl-*a*. Many previous studies have suggested that mercury dilution within the larger amount of autochthonous matter in more productive lakes results in lower mercury content in fish (Håkanson, 1980; Pickhardt et al., 2002; Razavi et al., 2015; Kozak et al., 2021), as well as growth biodilution in fish where faster growing individuals contain less mercury (Karimi et al., 2016; Sandheinrich and Drevnick, 2016; Keva et al., 2017). Therefore, the observed negative correlation between agriculture coverage of lake catchment area and perch mercury content could be a result of biodilution process.

Perch muscle ARA and LIN content were higher in lakes with higher forest coverage and lower pH than in lakes with higher TP concentration and agricultural coverage in the catchment. Moreover, perch allochthony correlated positively with ARA and LIN. These likely resulted in the lower n3:n6 ratios in perch that had incorporated higher amounts of allochthonous OM. Growth experiments have shown that invertebrates fed with allochthonous matter include high amounts of ARA (Goedkoop et al., 2007; Taipale et al., 2015). Previously it has been suggested that a lower n3:n6 ratio indicates more utilization of terrestrial fatty acid sources in the organism as the pelagic primary production is typically richer in n3 fatty acids compared to terrestrial sources, while terrestrial sources are higher in n6 fatty acids. (Lau et al., 2012; Hixson et al., 2015; Taipale et al., 2015). Based on results, the n6 content and n3:n6 ratio of fish muscle reflect increased terrestrial organic matter contribution in the food web.

Simultaneous increment in lake TP and DOC concentration, and decreasing lake average depth slightly decreased perch muscle DHA and increased ALA content. However, when looking at single environmental variables, there was no correlation between lake chemistry, catchment characteristics, allochthony and perch muscle n3 fatty acid contents. The highest perch DHA content was found in a large oligotrophic lake (Suuri Jukajärvi: 8.5 ± 0.4 mg g dw $^{-1}$), but the lowest DHA values were also found from a small oligotrophic lake (Pesosjärvi: 3.3 ± 0.5 mg g dw $^{-1}$). Habitat availability and diversity of primary producer community is typically higher in deeper clear water lakes (Longhi and Beisner, 2010) where benthic algal community might form and important link for higher trophic level organisms (Seekell et al., 2015). Higher algal diversity induces the growth and FA diversity of zooplankton (Marzetez et al., 2017). Moreover, previous studies have suggested that in lakes or habitats with poor n3 fatty acid dietary supply, zooplankton and perch are producing more long-chain n3 fatty acid from ALA (Taipale et al., 2015; Geay et al., 2016; Pilecky et al., 2021; Scharnweber et al., 2021; Taipale et al., 2022). However, there are obviously differences in perch muscle DHA content among different lakes. Perch muscle DHA content was slightly higher in larger and clearer lakes than in shallower and murkier lakes, that was in accordance with a previous study (Strandberg et al., 2016). This could be related to differences in the primary producer community structure, where murkier lakes have less n3 PUFA synthesizing phytoplankton taxa (Taipale et al., 2016b; Taipale et al., 2018).

4.4. Environmental impact on human nutritional value of fish

The lowest EPA + DHA values in perch muscle were observed in a small humic lake (Majajärvi) and a mid-sized mesotrophic lake (Eräjärvi: ca. 5 mg g^{-1} dry weight) and the highest values were found in a mid-sized oligotrophic lake (Suuri Jukajärvi: ca 10 mg g^{-1} dry weight). The observed EPA + DHA range corresponds well with previously published data (e.g. Ahlgren et al., 1996; Strandberg et al., 2016; Keva et al., 2021). Fish muscle typically contains ca. 80% water (e.g. Ahlgren et al., 1996), so lowest and highest observed EPA + DHA values correspond to approximately 1 and 2 mg EPA + DHA g^{-1} wet weight. With these lowest and highest EPA + DHA content, daily consumption of 250–500 g and 125–250 g perch fillet should fulfil the daily recommended intake of EPA + DHA (250–500 mg d^{-1}, European Food Safety Authority, 2012a). The highest mercury content in perch muscle tissue, exceeding 3 μg g^{-1} dry weight, was measured in small humic lakes (Majajärvi, Viitaanjärvi) corresponding to ca. 0.6 μg g^{-1} wet weight that exceed the UN consumption limit. Perch muscle HQ values were almost all above one in the sampled lakes, meaning that fulfilling the daily recommended amount of EPA and DHA with perch fillet would in most cases simultaneously lead to an intake of Hg that exceeds the daily threshold. This highlights that, with the recommended daily intake of EPA + DHA (250–500 mg d^{-1}) and the maximum tolerable methyl mercury intake (0.186 μg kg^{-1} body weight d^{-1}, European Food Safety Authority, 2012b), freshwater lean piscivorous fish such as perch from boreal lakes might not be the best daily source of EPA + DHA for humans. Increased risk to gain guideline-exceeding mercury content when consuming lean piscivorous fish has been observed elsewhere as well (e.g. Loring et al., 2010; Strandberg et al., 2016). Because the DHA and EPA content in perch muscle did not correlate with the environmental gradient (PC1), it is clear that the decreasing HQ values along PC1 were driven by the mercury content of the fish rather than by n3 FAs. This is in accordance with previous studies from Canadian inland lakes and the Laurentian Great Lakes (Strandberg et al., 2017, 2020). Highest HQ values were found from low pH lakes with high forest coverage in the catchment. For example, in the smallest humic lake (Majajärvi) with high allochthony in this study, HQ values were higher than 10 (daily mercury threshold limit exceeded tenfold). A previous study from Eastern Finland also showed increasing HQ values along a gradient of lake colour (Strandberg et al., 2016). In the most eutrophic lakes, the perch HQ values were below or close to one, suggesting that eating perch from these lakes is likely a safer way to meet the daily EPA + DHA requirement than consuming perch from lakes with relatively small contributions of autochthonous primary productivity to the food web.

4.5. Conclusions

The increasing perch muscle THg and n6 FA content along reversed PC1 axis indicate that higher forest cover, lower TP and pH are the most important environmental stressors impacting these compounds. Perch allochthony had a parallel correlation to muscle THg and n6 FA content, thus allochthony could be at least partly the mechanism underlying the trend between perch THg and n6 FA content and environmental gradient (PC1). Perch in lakes with higher allochthony are likely to have elevated muscle THg content, but at the same time n3 fatty acid content in the fish muscle will likely remain unimpacted, since n3 FAs usually originate from autochthonous source and are preferentially retained by fish. Nutritional quality of perch for human consumption, defined with hazard quotient, was the lowest in low pH lakes with highly forested catchment area and this was clearly driven by the higher perch THg values in these lakes. Human consumption of perch from larger waterbodies with neutral pH should be favoured over fish from small headwater forest lakes with low pH.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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CRediT authorship contribution statement

OK: conceptualization, methodology, software, validation, formal analysis, investigation, data curation, writing original draft, review and editing, visualization. **MK**: conceptualization, methodology, validation, formal analysis, investigation, resources, data curation, review and editing. **HH**: conceptualization, review and editing, funding acquisition, project administration. **RIJ**: conceptualization, review and editing, funding acquisition, project administration. **KKK**: conceptualization, validation, resources, review and editing, supervision. **PK**: conceptualization, resources, review and editing, funding acquisition. **MBL**: formal analysis, investigation, review and editing. **JS**: conceptualization, formal analysis, investigation, resources, data curation, review and editing. **US**: resources, review and editing. **JV**: review and editing. **SJT**: conceptualization, validation, resources, review and editing, supervision, funding acquisition, project administration.

Data availability

Data are available from the authors upon reasonable request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.155982.

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III

INCREASING TEMPERATURE AND PRODUCTIVITY CHANGE BIOMASS, TROPHIC PYRAMIDS AND COMMUNITY LEVEL OMEGA-3 FATTY ACID CONTENT IN SUBARCTIC LAKE FOOD WEBS

by

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Increasing temperature and productivity change biomass, trophic pyramids and community-level omega-3 fatty acid content in subarctic lake food webs

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Abstract

Climate change in the Arctic is outpacing the global average and land-use is intensifying due to exploitation of previously inaccessible or unprofitable natural resources. A comprehensive understanding of how the joint effects of changing climate and productivity modify lake food web structure, biomass, trophic pyramid shape and abundance of physiologically essential biomolecules (omega-3 fatty acids) in the biotic community is lacking. We conducted a space-for-time study in 20 subarctic lakes spanning a climatic (+3.2°C and precipitation: +30%) and chemical (dissolved organic carbon: +10 mg/L, total phosphorus: +45 μg/L and total nitrogen: +1,000 μg/L) gradient to test how temperature and productivity jointly affect the structure, biomass and community fatty acid content (eicosapentaenoic acid [EPA] and docosahexaenoic acid [DHA]) of whole food webs. Increasing temperature and productivity shifted lake communities towards dominance of warmer, murky-water-adapted taxa, with a general increase in the biomass of primary producers, and secondary and tertiary consumers, while primary invertebrate consumers did not show equally clear trends. This process altered various trophic pyramid structures towards an hour glass shape in the warmest and most productive lakes. Increasing temperature and productivity had negative fatty acid content trends (mg EPA + DHA/g dry weight) in primary producers and primary consumers, but not in secondary nor tertiary fish consumers. The massive biomass increment of fish led to increasing areal fatty acid content (kg EPA + DHA/ha) towards increasingly warmer, more productive lakes, but there were no significant trends in other trophic levels. Increasing temperature and productivity are shifting subarctic lake communities towards systems characterized by increasing dominance of cyanobacteria and cyprinid fish, although decreasing quality in terms of EPA + DHA content was observed only in phytoplankton, zooplankton and profundal benthos.

KEYWORDS
DOC, food web structure, forestry, land-use, nutrients, omega-3 HUFA, trophic level, trophic pyramid

1 | INTRODUCTION

Temperature has increased much faster in Arctic regions than the global average, and rate of warming is expected to further accelerate in the future (Cohen et al., 2014). In subarctic Fennoscandia, mean annual air temperature has increased by approximately 2°C since 1850 and temperature and precipitation are further predicted to change by +2–5°C and +10%–30%, respectively, by the end of the 21st century (IPCC, 2014: RCP2.6–RCP8.5). Furthermore, precipitation in the region is more likely to fall as rain rather than snow (e.g. Dai & Song, 2020). Such climatic shifts will expand catchment vegetation towards higher altitudes and latitudes, promoting overall greening and facilitating changing land-use practices. In particular, forestry and agriculture are likely to replace traditional livelihoods such as reindeer herding, subsistence hunting and fishing (Huntington et al., 2007; Schindler & Smol, 2006). Expanding transport networks, settlements, oil drilling and mining activities are rapidly changing the hydrology and productivity of freshwaters (Cott et al., 2015; Denisov et al., 2020; Zubova et al., 2020). Joint effects of changes in climate and land-use in catchment area cause increased leaching of terrestrial dissolved organic carbon, nitrogen and phosphorous, with serious impacts on freshwater species richness, community structure and food web processes (Hayden et al., 2019; Jeppesen et al., 2012; Laske et al., 2019; Nieminen et al., 2015).

Globally, species richness tends to show a decreasing trend from the tropics towards the poles, with this linked to a gradient of decreasing temperature and productivity (for overview see e.g. Gaston, 2000; Hillebrand, 2004). The latitudinal gradient in species richness may be linked to multi-trophic level diversity and food chain length due to common underlying drivers of ecosystem size and productivity (Baiser et al., 2019; Gaston, 2000; Takimoto & Post, 2013). Productivity and ecosystem size are inherently linked, for example large lakes tend to be less productive but provide broader feeding or breeding areas for diverse communities, with subsequent effects on food chain length (Post et al., 2000; Takimoto et al., 2012; Ward & McCann, 2017). Food chain length determines the number of trophic levels and thus largely governs the strength of top-down and bottom-up processes. In systems with three or more trophic levels, primary production is suggested to be resource limited (bottom-up regulation), primary consumers top-down regulated and secondary consumers resource limited (Hairston et al., 1960; Hansson et al., 2013; Oksanen et al., 1981). Together with bottom-up regulation, top-down predator-mediated community-level trophic cascades have been suggested as a factor shaping food web structure driving opposing trends in the biomass of sequential trophic level (i.e. each second trophic level biomass is high; Carpenter et al., 1985, 2001; Paine, 1980). These processess govern the shape of trophic (Eltonian) pyramids, which may show different forms from a classic pyramidal shape where energetic loss in each trophic level leads to accumulation of max. circa 10% of produced biomass to next trophic level, and in certain cases may even show inverted pyramids (Brown et al., 2004; Elton, 1927; McCauley et al., 2018). Simple food webs in low diversity and productivity ecosystems may be prone to size-structured populations, trophic cascades and inverted trophic pyramids, but generally there is very little clear evidence of how food web biomass and trophic pyramid shape are influenced by temperature and productivity gradients.

Subarctic regions provide an excellent natural setting to study this phenomenon as they straddle boreal and Arctic biomes, providing clear diversity, climatic and productivity gradients with relatively simple food webs (Hayden et al., 2017). Furthermore, the subarctic landscape is scattered with lakes that provide abundant and well-defined ecosystems for food web diversity studies (Lau et al., 2020). Increasing temperature and productivity have been shown to shift subarctic lake communities towards more numerous, diverse, smaller-bodied, warmer-water-adapted taxa which are more reliant on pelagic energy sources (Hayden et al., 2017, 2019). However, we do not know how increasing temperature and productivity affect energy transfer efficiency and thus biomass distribution across different trophic levels. Aquatic food webs in many Arctic areas are based on production and transfer of lipids from primary producers to top consumers, where seasonal storage of lipids is important for survival and reproduction of long-lived organisms at higher trophic levels (e.g. Armstrong & Bond, 2013; Jørgensen et al., 1997). An important subgroup of lipids are polyunsaturated fatty acids (PUFAs), providing essential components for cell development and functioning; however, the distribution of lipids and fatty acids varies greately among taxa and trophic levels. While there are several PUFAs, eicosapentaenoic acid (20:5n-3, EPA) and docosahexaenoic acid (22:6n-3, DHA) are considered the most important since they are essential for growth, neural development and reproduction of consumers (Arts et al., 2001; Bou et al., 2017; Yeşilayer & Kaymak, 2020). Only certain algae groups, such as dinoflagellates, diatoms and cryptophytes, are able to produce EPA and DHA efficiently and are abundant in cold and nutrient-poor lakes (Taipale et al., 2013, 2016). However, warm and eutrophic lake primary producer communities, often dominated with cyanobacteria, are inefficient in EPA and DHA production (Müller-Navarra et al., 2000, 2004; Taipale et al., 2013, 2016). Some species can convert shorter-chain fatty acids to EPA and DHA (Geay et al., 2016; Ishikawa et al., 2019), but in many vertebrates this process is considered energetically expensive and thus direct sources from high-quality prey are often selected (Keva et al., 2019; Twining et al., 2016). This suggests that at a food web level, differences in primary producer communities induced by temperature and productivity may govern the quality of higher trophic levels (Colombo et al., 2020; Hixson & Arts, 2016).

Our main aim in this study was to test how increasing temperature and productivity change structure, biomass and nutritional quality of food web components from primary producers to top consumers using a gradient of Fennoscandian subarctic lakes ($n = 20$) spanning a climate and land-use gradient. Omega-3 fatty acids, EPA and DHA, were selected as proxies for quality of food web components, since they are physiologically important biomolecules for consumers. First, we hypothesized (H1) that communities shift towards warmer and more murky-water-adapted species, with a concurrent increase in food web biomass, and expected that increases in algae and fish biomass along the gradient would have effects on trophic pyramid shape (Hayden et al., 2017; Jeppesen et al., 2010; McCauley et al., 2018). Second, we hypothesized (H2) that cyanobacteria form an increasing proportion of the phytoplankton communities in warmer and more productive lakes (Przytulska et al., 2017), thus lowering nutritional quality of primary producers. As such, we expected that this difference would be passed through all higher trophic levels (Müller-Navarra et al., 2000; Taipale et al., 2016). Finally, we hypothesized (H3) that increasing temperature and productivity would increase total community biomass including an increase in physiologically important EPA and DHA of the total lake community (Hayden et al., 2019; Jeppesen et al., 2000).

2 | MATERIALS AND METHODS

2.1 | Environmental variables

We sampled 20 lakes, located in Finnish Lapland across the Tornio-Muoniojoki and Paatsjoki watercourses, spanning northern European climatic (average air temperature in June–September from 1980 to 2010: 8.4–11.6°C and precipitation 197–257 mm/open-water season) and water chemistry (dissolved organic carbon; DOC: 1.9–14.9 mg/L, total phosphorus; TotP: 3–48 μg/L and total nitrogen; TotN: 110–1,100 μg/L) gradients (Table S1; Figure 1). Here, all lakes are considered subarctic and located north of Arctic Circle (AMAP, 1998). Climatic differences originate from latitudinal and altitudinal gradients, with temperature and precipitation lowest in northernmost lakes and progressively increasing towards the south. Climatic data were derived from nine Finnish Meteorological Institute weather stations situated within the study region and refer to long-term (1981–2010) average open-water season June–September air temperature and annual precipitation as rain (Hayden et al., 2019). Differences in lake productivity originate predominantly from variation in vegetation and land-use practices within the catchment (Jussila et al., 2014). The northernmost lakes are situated near pristine areas with anthropogenic impacts limited to nature tourism and reindeer herding, and from the timberline southwards industrial forestry activities including large-scale clear-cut areas, site preparation and intensive ditching (Ahonen et al., 2018; Hayden et al., 2017; Jussila et al., 2014; Table S1). Catchment area characteristics and land-use variables were derived with the Finnish Environment Institute VALUE-tool combining catchment and CORINE-database and open map data (Ahonen et al., 2018; Hayden et al., 2017). Lake location (coordinates, altitude), morphometrical data (area, depth, mean depth, volume) and water physical-chemistry (nutrients, light) were derived from the Finnish Environment Institute (HERTTA-database) and National Survey of Finland, or from our own sampling (Table S1). We measured visible light in the water column (LI-COR, A-250, visible light 400–700 nm) at deepest point of lake or sampling area (largest lakes) to define 1% surface light level, that is, compensation depth needed to define proportions of the three major lake habitats littoral, profundal and pelagic (Table S1). Lake trophic states were categorized with TotP concentrations to ultraoligotrophic (<5 μg/L, $n = 3$), oligotrophic (5–9 μg/L, $n = 7$), mesotrophic (10–29 μg/L, $n = 5$) and eutrophic (≥30 μg/L, $n = 5$; Figure 1; Nürnberg, 1996). Phytoplankton, pelagic zooplankton, littoral and profundal benthic macroinvertebrates (hereafter benthos) and fish were sampled in the years 2009–2017. The sampling period was always in late summer (August–September).

2.2 | Community composition and biomass

Quantitative phytoplankton taxa samples for eight of the study lakes were extracted from the HERTTA database (Finnish Environment

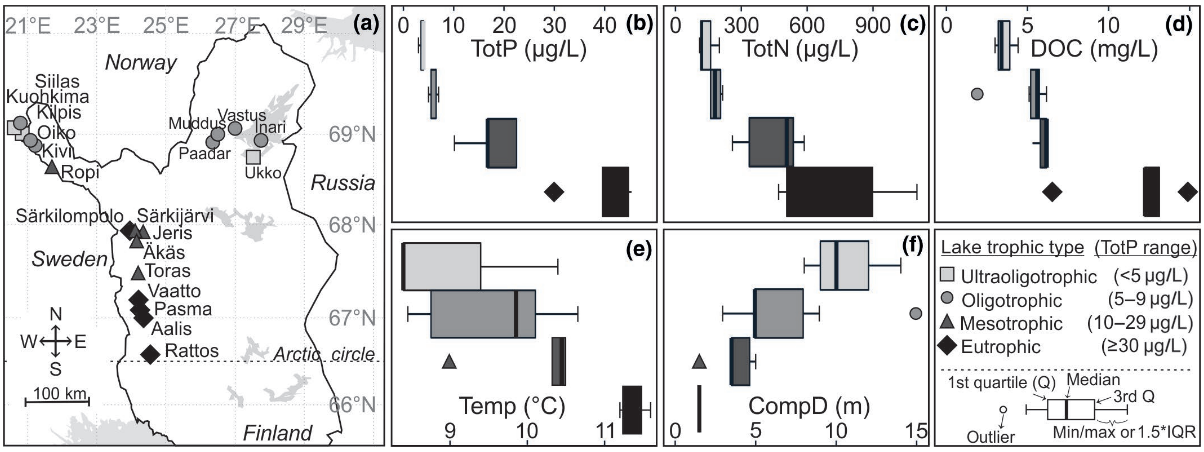

FIGURE 1 Map of the study region (a) in northern Fennoscandia and boxplot figures of the basic lake chemistry (b–d) and physics (e–f) of the sampled lakes. The study lakes are marked with grey shade shapes (a) or boxes (b–f) indicating different lake types (light grey = ultraoligotrophic, grey = oligotrophic, dark grey = mesotrophic, black = eutrophic). Abbreviations in *x*-axes labels are named as following: TotP = total phosphorous (b), TotN = total nitrogen (c), DOC = dissolved organic carbon (d), Temp = average air temperature in June–September 1981–2010 (e), CompD = compensation depth, that is, water depth where 1% surface light is left (f)

Institute) and from a previous study for an additional six lakes (Taipale et al., 2016). Samples were collected from the epilimnion (0–2 m) with a Limnos or corresponding water sampler in late summer (August–September) and stored in Lugol solution with added formaldehyde. Phytoplankton cell counts were calculated under a microscope using the Utermöhl technique (Utermöhl, 1958). Biomass estimations were done using taxa morphology-specific geometric formulas (Hillebrand et al., 1999) and adjusted with experimentally derived carbon-mass ratios (Menden-Deuer & Lessard, 2000). Phytoplankton community composition was calculated as class-specific percentage from carbon biovolume (g C/L), and dry mass (dw) per unit area was calculated as well (kg dw/ha, hereinafter biomass) using lake-specific euphotic zone.

Quantitative pelagic zooplankton samples were collected with a net (diameter 25 cm, mesh size 50 μm) using three replicate vertical hauls from the deepest point of each lake (or, in very large lakes, the deepest point of the sampling area) and immediately stored in a 5% formaldehyde solution (Hayden et al., 2017). We collected pelagic zooplankton for fatty acid analyses from the same point using repeated vertical hauls and these samples were condensed, frozen and freeze-dried in the laboratory. In quantitative samples, all crustaceans were identified to class level, and the body size of 30 first encountered individuals of each class was measured for density and biomass calculations. Biomass was calculated from identified individuals and length measured taxa using size-biomass (length to dry weight) conversion factors from McCauley (1984). The overall zooplankton community composition was calculated as taxon-specific percentage from dry mass biovolume (mg dw/L) and biomass was calculated (kg dw/ha) using lake-specific average depth.

Benthic macroinvertebrates were collected only from soft bottom with an Ekman grab (area 272 cm^2) from a transect (depths 1, 2, 3, 5, 10, 15, 20, 30 and 40 m) spanning from the littoral zone to the deepest point of lake or sampling area. Three separate replicates were taken from each depth and animals were stored in plastic buckets with lake water followed by sorting in laboratory. Animals were identified to lowest feasible level, sorted, counted and total wet biomass of each taxa were weighed (accuracy 0.1 mg) and areal biomass (g ww/m^2) was calculated for each sample using Ekman area to m^2 conversion factor 36.8. For the lake-specific biomass calculations, we transformed wet weights to dry weights using a conversion factor of 0.2 for all taxa (Smit et al., 1993). For each lake, we used water column light level to define compensation depth to define which sampling depths belong to littoral (≥1% light) or profundal (<1% light). Then whole lake bathymetry was used to calculate mean benthic macroinvertebrate biomass (kg dw/ha) in soft sediments in littoral and profundal regions.

Fish were sampled from three major lake habitats (littoral, profundal and pelagic) using a gill net series composing of eight 30 m long and 1.8 m high nets (mesh sizes 12–60 mm knot to knot) and one Nordic net with dimensions of 30 × 1.5 m (12 mesh sizes 5–55 mm). A minimum of three gill netting nights per major habitat was conducted and sampling amount was increased proportional to lake area (Hayden et al., 2017; Malinen et al., 2014). Fishing depth of gill nets was verified with echosounder and each fishing period length was recorded with accuracy of a minute. In most cases, gill nets were set overnight in the evening and collected during following morning (6–10 hr soaking time), but in the most eutrophic lakes we had to use shorter sampling time (<2 hr) to avoid oversaturation due to a massive increase in fish density in these lakes (Hayden et al., 2017). Fish were immediately killed during net lifting by cerebral concussion and chilled in ice. In the field laboratory, each fish was identified to species level, but whitefish (*Coregonus lavaretus*) were assigned to the morph (subspecies) level and whitefish × vendace (*Coregonus albula*) hybrids were separated based on morphology and colour, as well as head shape and gill raker count (Kahilainen et al., 2011, 2017). This subspecies level delineation was considered essential as many of the northern lakes in the study area are dominated by variable proportions of ecologically contrasting whitefish morphs (2–4 morphs in a lake) that play differential roles in food web structure, energy flows and fatty acid dynamics (Thomas et al., 2017, 2019).

Total length (accuracy 1 mm) and wet mass (0.1 g) were measured and the relative abundance of each species was calculated as catch per unit effort (CPUE), that is, number of individuals per species in gill net per hour (n/[net hr]). In addition to gill net data, we have previously collected both relative abundance (CPUE) and quantitative estimates of whitefish and vendace density in pelagic habitat (individuals/ha) using vertical echosounding and gill netting from subset of the lakes analysed in the current study (Malinen et al., 2014). We calculated a linear regression equation between CPUE and echosounding derived quantitative density to transform relative densities of whitefish and vendace to absolute densities. As one or both of these species are present in all sampled lakes, we used their calculated absolute density and their proportion of total catches to get fish density values for different species in each lake. These density values for each species and their mean individual mass in gill net catch were multiplied to get biomass (kg/ha). For consistency with other trophic levels, we transformed fish wet weight to dry weight with conversion factor of 0.2 (e.g. Ahlgren et al., 1996). In trophic level analyses, fish were classified as secondary consumers, that is, invertivorous (most species and morphs) and tertiary consumers, that is, piscivorous species (only pike, *Esox lucius*; burbot, *Lota lota*; Arctic charr, *Salvelinus alpinus*; brown trout, *Salmo trutta*; and >20 cm perch *Perca fluviatilis*) based on previous dietary and stable isotope studies of a subset of the studied lakes (Hayden et al., 2019; Kahilainen & Lehtonen, 2003; Thomas et al., 2017).

2.3 | Lipid extraction and fatty acid analysis

Homogenized freeze-dried (−50°C for 48 hr) invertebrate and fish dorsal muscle samples were weighed c. 0.5–1 mg and 3–4 mg, respectively, to Kimax glass test tubes. If material was not limited, duplicate samples were analysed. Fatty acids were analysed as methyl esters (e.g. Taipale et al., 2016), identified and quantified using a gas chromatograph attached to mass spectrometer. Samples were spiked with nonadecylic acid (PLFA c19:0). Lipids were extracted with 3 ml of chloroform–methanol solution (2:1 vol) and with sonicator (15 min). Ultra-pure water (0.75 ml of MilliQ) was add to Kimax tubes to separate water-soluble compounds from lipids. Lipid extract was evaporated and 3 ml of 1% H_2SO_4 add to Kimax tube which was incubated in +50°C for 20 hr to produce fatty acid methyl esters (FAMEs). The produced FAMEs were diluted with hexane and analysed using a gas chromatograph attached to a mass spectrometer (GC-2010 Plus and QP-2010 Ultra) with Zebron ZB-FAME column (30 m + 5 m guardian × 0.25 mm × 0.2 μm). At the begin of the each GCMS run, the column temperature was held in 50°C for 1 min, followed by raising with 10°C/min to 130°C, 7°C/min to 180°C, 2°C/min to 200°C and was held there for 3 min which after the colon temperature was raised with 10°C/min to 260°C. The injection temperature was 270°C and the interface temperature 250°C, total column flow being 27.5 ml/min and linear velocity 36.3 cm/s.

Fatty acids were identified based on ion spectrums and quantified based on four-point standard mixture calibration curves (15, 50, 100, 250 ng/μl; GLC 566c, Nu-Chek Prep) with GCMS solution software version 4.42. The calibration curves were ran before each run. The coefficient of determination between peak area and standard FA concentrations was always higher than 0.999. To validate the calibration curves, we run at least one external EPA and DHA standard (Sigma Aldrich CRM47571 & CRM47570) per batch, average recovery % ± RSD of the analysed external standards were 119.2 ± 21.4% ($n = 11$) and 104.9 ± 20.5% ($n = 11$) for EPA and DHA. The average sum of percent error between sample duplicate ($n = 47$) FA profiles and EPA + DHA contribution were 2.0 ± 1.3% and 0.6 ± 0.7%, respectively. The mean ± *SD* recovery of our internal standard nonadecylic acid in our samples was 90.7 ± 24.2% ($n = 503$) and the individual recovery % was used to adjust the sample FA concentrations. EPA and DHA were the only fatty acids used in the subsequent data analysis.

We combined a small subset of fatty acid data from previous studies having a different focus (Taipale et al., 2016; Vesterinen et al., 2020). Total dataset consisted of 868 samples (Table S2: phytoplankton = 32, zooplankton = 26, littoral benthos = 209, profundal benthos = 34, invertivorous fish = 377, piscivorous fish = 190). Community-specific lake average FA sample coverage of taxa-specific biomass contributions stands as follows: phytoplankton 93.2 ± 5.4%, littoral benthos 59.6 ± 24.3%, profundal benthos 71.9 ± 19.9%, invertivorous fish 92.9 ± 9.0%, piscivorous fish 89.7 ± 23.0% (Table S3). For zooplankton, only bulk samples were used in community quality calculations, thus assuming coverage being 100% (Table S3).

2.4 | Lake-specific community EPA + DHA content and areal content calculations

Computational phytoplankton EPA + DHA content (mg/g C) was calculated for study lakes using the class specific carbon mass and their corresponding FA production estimates from laboratory experiments (Taipale et al., 2016). The laboratory-culture experiment-derived FA production of phytoplankton taxa might be different from those found in nature, due to varying environmental conditions in natural environments. However, we think this estimation is relevant as microscopy data of phytoplankton community composition and biomass data was available for the study systems. For the consumer communities, EPA + DHA content (mg/g dw) was weighed with taxa-specific biomass contribution (%) determined for each lake and taxa, except for zooplankton which was sampled as a whole community. Lake-, habitat- and taxa-specific mean FA content values was used in the community EPA + DHA calculations where possible. If taxa specific fatty acid content was unavailable from a lake, we used measured taxa average FA content from different habitat (for benthos) or same type of lakes (e.g. oligotrophic), with this we supplemented our FA sample coverage near to 100% for each community (Table S3).

Biomass calculations (in Section 2.2) facilitated the calculation of trophic pyramids, where total food web biomass was divided among trophic levels. Here, we followed above classifications where the first trophic level are primary producers (i.e. phytoplankton), the second is primary consumers (zooplankton and benthic macroinvertebrates), the third is secondary consumers (invertivorous fish), while the fourth is tertiary consumers (piscivorous fish). To calculate community EPA + DHA mass per unit area (kg EPA + DHA/ha, herein after: areal content) for primary producers and different consumers, we multiplied community biomass (kg dw/ha) and EPA + DHA content. For fish community areal EPA + DHA content, we used only muscle tissue of fish instead of whole fish. Furthermore, we used community EPA + DHA areal content (kg FA/ha) to calculate trophic pyramids for these assimilated biomolecules through food webs. Our biomass calculations should be considered as minimum estimates, since we did not have any quantitative biomass and FA data from littoral primary producers (periphyton, macrophytes) nor small organisms such as bacteria, rotifers and microbenthos.

2.5 | Statistical methods

We calculated a climate–productivity index (CPi) that equals the principal component analysis axis one based on lake chemistry (TotP, TotN, DOC), and catchment properties (air temperature, precipitation) to reduce the number of environmental variables when testing H1 and H2 (Table S1; Hayden et al., 2019). Here, the PC1 axis (i.e. CPi) explained 81% of the variation in lake environmental data, and was the only PC used in subsequent analyses (Figure S1) due to inherent collinearity with land-use practices (Hayden et al., 2019). Within the final derived index, lakes with higher temperature and productivity receive higher CPi values. We built general linear models with forward and backward selection predicting each community biomass with CPi, prey item quality (EPA + DHA content) and predator biomass, where Akaike information criterion was used for selection of the best models. To summarize general linear model results, CPi was the main factor affecting community biomass (Tables S4 and S5). Therefore, we used a simple linear regression model to examine the relation between CPi (PC1 scores) and community total biomass (H1) and EPA + DHA areal content (H2). We also used linear regression analysis to test the dependence of community EPA + DHA areal content on lake total biomass (H3). An alpha level of 0.05 was used in each statistical analysis and tests were conducted using R through RStudio version 3.4.1 with the default base package (R Core Team, 2017). Figures were drawn using base graphics and ggplot2 package (Wickham, 2016).

3 | RESULTS

3.1 | Changes in community structure, food web biomass and trophic pyramid shape (H1)

Lake communities showed shifts from cold to warmer-water adapted taxa with increasing temperature and productivity (Figure 2). Phytoplankton changed from diatom-dominated communities in ultraoligotrophic lakes towards cyanobacteria dominance in eutrophic lakes (Figure 2a1). Copepods contributed 50%–80% of the total zooplankton community biomass along this gradient and calanoids had the highest biomass percentage in every lake type. However, copepod contribution was lowest in eutrophic lakes where cladocerans, especially *Bosmina* had a high biomass percentage (Figure 2a2). Littoral benthos communities were diverse throughout the lake gradient (Figure 2a3): *Asellus* sp. and *Lymnaea* sp. reached the highest proportional contribution in mesotrophic lakes, whereas oligochaetes had the highest biomass contribution in ultraoligotrophic lakes and lowest in eutrophic lakes. Profundal benthos was mainly dominated by chironomids and their biomass contribution increased from ultraoligotrophic lakes (c. 50%) towards eutrophic lakes (c. 90%; Figure 2a4). Invertivorous fish communities were dominated by salmonids (whitefish, vendace) in ultraoligotrophic lakes; by percids (perch and ruffe, *Gymnocephalus cernuus*) and salmonids in oligotrophic–mesotrophic lakes; and by cyprinids (roach *Rutilus rutilus*, bleak *Alburnus alburnus*) in eutrophic lakes (Figure 2a5). Piscivorous pike and perch increased their biomass contribution progressively from ultraoligotrophic lakes (c. 35% and 5%) towards eutrophic lakes (c. 50% for both species), while piscivorous Arctic charr and brown trout disappeared completely along the same gradient (Figure 2a6).

Phytoplankton mean mass increased more than 20-fold between ultraoligotrophic (0.08 ± 0.05; mean ± *SD* g dw/m^3) and eutrophic lakes 1.9 ± 0.8 g dw/m^3 (Figure 2b1), which was also evident for biomass along our lake gradient CPi (PC1) from north to south based on linear regression analysis (Figure 3a1). The lowest zooplankton biomass was observed in mesotrophic lakes (9.27 ± 5.96 mg dw/m^3; Figure 2b2); however, no clear trend in biomass was observed along the lake gradient (CPi) based on linear regression (Figure 3a2). The overall highest littoral benthos biomass was observed in mesotrophic lakes (Figure 2b3), but no clear trend in biomass along the continuous CPi axis (Figure 3a3). The profundal benthos biomass was low in ultraoligotrophic lakes (0.4 ± 0.3 kg dw/ha) and increased towards eutrophic lakes (8.2 ± 11.3 kg dw/ha). This trend was mainly caused by increasing chironomid biomass (Figure 2b4) and driven mainly by

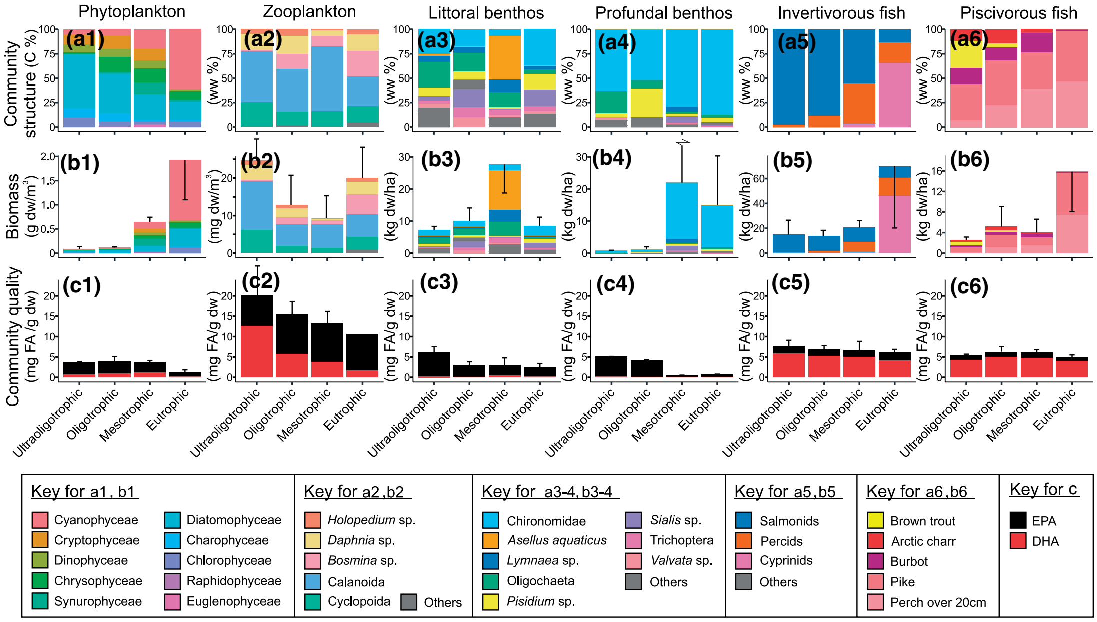

FIGURE 2 Community structure (a), biomass (b) and computed community quality (FA content, c) across different lake types (*x*-axis). Food web community compartments: phytoplankton (1), zooplankton (2), littoral benthos (3), profundal benthos (4), invertivorous fish (5) and piscivorous fish (6) are presented in different columns. The coloured bars indicate average taxa values of different lake types as following: normalized relative community composition (a: biomass %), lake type average biomass (b1: g dw/m^3, b2: mg dw/m^3, b3–b6: kg dw/ha) and lake type average community EPA and DHA content (c: mg FA/g dw). Lake-specific community bulk FA content is derived from the FA analysed bulk sample (c2: zooplankton) and calculated for other food web community compartments (c1, c3–c6) from taxa-specific biomass contribution and fatty acid content. Whiskers on top of the lake type average (b and c rows) represent ± *SD*

extremely high profundal benthos densities in two lakes with low oxygen (Figure 3a4). Generally, invertivorous fish biomass was circa five times higher in eutrophic lakes (69.7 ± 49.0 kg dw/ha) compared to other lake types (pooled average: 15.8 ± 12.9 kg dw/ha; Figures 2b5 and 3a5). The biomass of piscivorous fish was around four times higher in eutrophic lakes (15.9 ± 7.8 kg dw/ha) than other lake types (pooled average: 4.2 ± 3.2 kg dw/ha; Figure 2b6), and a clear linear trend along the CPi axis was observed (Figure 3a6). These massive increases in primary producer and secondary consumer biomass compared primary and tertiary consumers were evident in changes to the trophic pyramid shape (Figure 4; Figure S2). Ultraoligotrophic and oligotrophic lakes showed variable shapes from typical pyramids to inverted pyramids to across different lakes, while most mesotrophic lakes were generally typical pyramids and eutrophic lakes had hour glass-shaped biomass distributions (Figure 4; Figure S2).

3.2 | Changes in fatty acid content of food web components (H2)

Phytoplankton quality (EPA + DHA) was highest in oligotrophic and mesotrophic lakes, and clearly the lowest in eutrophic lakes (Table S6; Figure 2c1). Zooplankton quality was the highest in ultraoligotrophic lakes (20.1 ± 8.9 mg/g dw; mean ± *SD*) and progressively decreased towards eutrophic lakes (10.6 mg/g dw; Figure 2c2), but due to high variance and low sample size the EPA + DHA distributions did not differ statistically among the lake types (Table S6). Benthos quality was limited to the presence of EPA. Littoral and profundal benthos quality were the highest in ultraoligotrophic lakes and the lowest in eutrophic lakes (Table S6; Figure 2c3–4). There was no statistically significant differences in quality of invertivorous and piscivorous communities between lake types as EPA + DHA content remained stable (Table S6; Figure 2c5–6). EPA and DHA content of primary producers and consumers decreased along the CPi axis, but there was no statistically significant trend in secondary or tertiary consumers (Figure 3b1–6; Figure S3).

3.3 | Changes in community EPA + DHA areal content (H3)

The highest values of community EPA + DHA areal content came from invertivorous fish (pooled average ± *SD*: 0.188 ± 0.214 kg/ha), followed by piscivorous fish (0.050 ± 0.054 kg/ha), phytoplankton (0.036 ± 0.030 kg/ha), littoral benthos (0.023 ± 0.019 kg/ha),

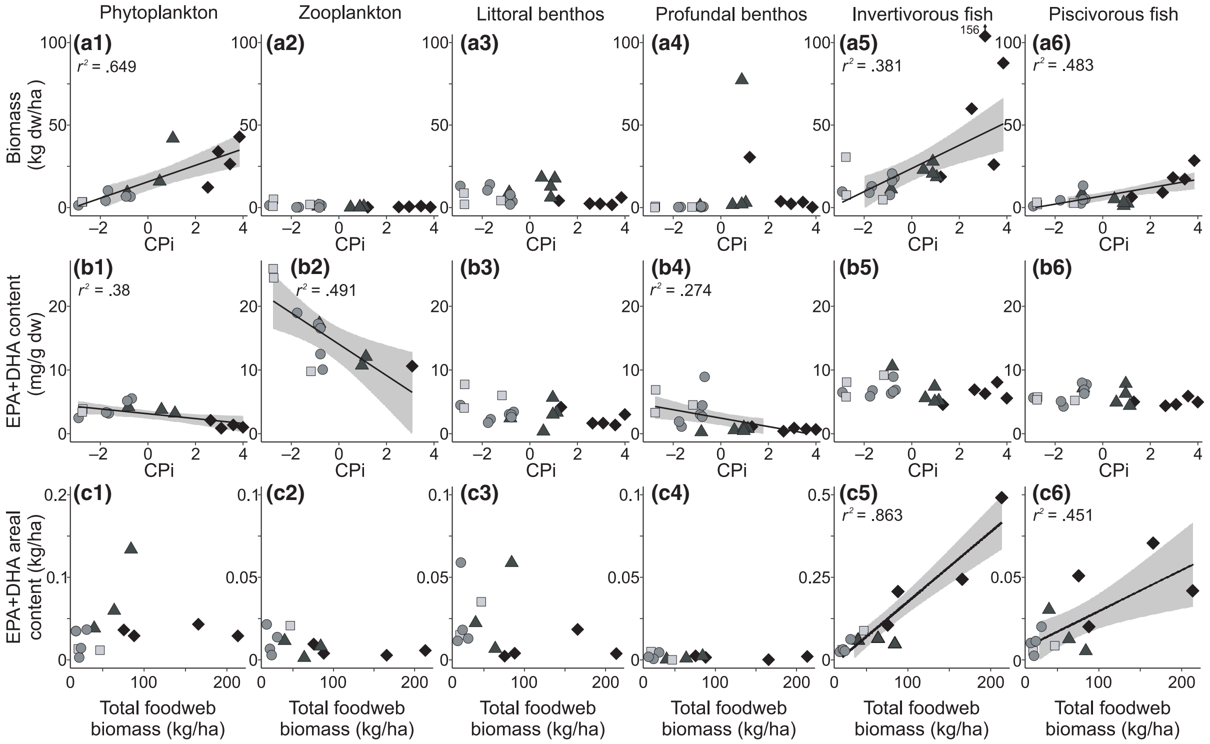

FIGURE 3 Linear regression models showing (a, b): Community biomass (kg dw/ha) and quality (mg EPA + DHA/g dw) against climate–productivity index (CPi) and (c): EPA + DHA areal content (kg EPA + DHA/ha) against total food web biomass (kg dw/ha). Food web community compartments: phytoplankton (1), zooplankton (2), littoral benthos (3), profundal benthos (4), invertivorous fish (5) and piscivorous fish (6) are presented in different columns. The grey-shaded shapes indicate different lake trophy corresponding to Figure 1: light grey square = ultraoligotrophic, grey dot = oligotrophic, dark grey triangle = mesotrophic, black diamond = eutrophic. Linear regressions fits (bolded line), 95% confidence limit (grey-shaded areas) and adjusted coefficient of determination (r^2) are presented in the figures if the model was statistically significant ($p < .05$). Linear regression equations, *F* statistics, adjusted r^2 and *p* values are presented for each model in Table S7

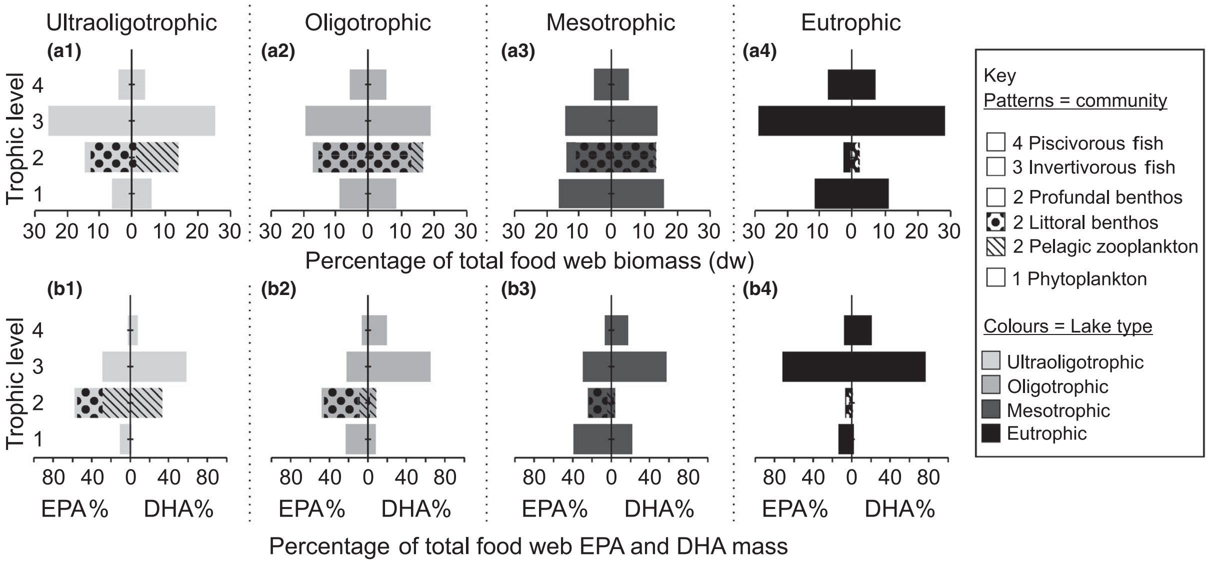

FIGURE 4 Relative biomass (a) and FA stock (b) pyramids by food web compartments for the lake types. The pyramid steps represent lake type averages of relative biomass or EPA + DHA areal content and are ordered with trophic level from bottom-up order: (1) phytoplankton, (2) invertebrates, (3) invertivorous fish and (4) piscivorous fish. The second trophic level (invertebrates) is divided among profundal benthos, littoral benthos and zooplankton with patterns: transparent, spots, stripes, respectively. Different lake types, ultraoligotrophic (a1, b1), oligotrophic (a2, b2), mesotrophic (a3, b3) and eutrophic (a4, b4), respectively, are separated into different columns delineated with dashed lines and additionally with light grey, grey, dark grey, black, respectively. The lake-specific data are presented in Figure S2

pelagic zooplankton (0.015 ± 0.026 kg/ha) and profundal benthos (0.007 ± 0.017 kg/ha), respectively. Trends in EPA + DHA areal content of phytoplankton, zooplankton and benthos communities with total food web biomass were not significant (Figure 3c1–4), though showing generally opposing patterns between trophic levels: EPA + DHA areal content of phytoplankton community was smallest in oligotrophic lakes but remained relatively stable in meso- and eutrophic lakes, while invertebrate taxa showed the opposite pattern. The areal contents of EPA + DHA of invertivorous (adj.r^2 = .86) and piscivorous (adj.r^2 = .45) fish communities were positively related to total biomass of food web (Figure 3c5–6; Table S7). Community EPA + DHA areal content showed a similar pattern as total biomass along the studied lake gradient, emphasizing the fundamental link between total community biomass and EPA + DHA areal content (Figure 3a,c; Table S7).

4 | DISCUSSION

4.1 | Major trends

Based on our findings, we suggest that increasing temperature and productivity fundamentally alter the structure and function of subarctic lake communities. Here, warmer, more nutrient-rich waters promote communities with an increasing share of cyanobacteria, smaller-bodied zooplankton, smaller benthic macroinvertebrate taxa and warmer-water-adapted cyprinid fishes. Moreover, we observed clear positive trends in biomass in each second trophic level (i.e. phytoplankton and invertivorous fish), which increased along the lake gradient. Deviating trends in contrasting trophic levels had subsequent effects on shaping trophic pyramids, where only eutrophic lakes seem to settle into a common hour glass shape. The omega-3 fatty acid content (mg EPA + DHA/g dw) of primary producers and consumers decreased along the climate–productivity gradient, but these trends were not observed in longer living secondary and tertiary consumers (i.e. fishes). Increasing total biomass of food webs along the climate–productivity gradient increased community EPA + DHA areal content, with this especially visible in invertivorous and piscivorous fish communities.

4.2 | Changes in community structure, biomass and trophic pyramids (H1)

Across our study lake gradient—from northern ultraoligotrophic lakes to eutrophic lakes—food webs shifted towards dominance of warmer and murky-water-adapted species. These changes were the clearest in primary producers, where communities moved from diatom dominance to cyanobacteria, and secondary consumers, where salmonids were replaced by cyprinids. While many of our biomass calculations follow the relative density results, obtained in a previous study conducted along a partially overlapping gradient (Hayden et al., 2017), there were clear differences in biomass trends among trophic levels. Here, primary producers and secondary consumer biomass increased most dramatically from cold ultraoligotrophic lakes towards warm eutrophic systems, but there were no clear trends in primary consumers and biomass increment of tertiary consumers was shallower. Past food web level studies have found similar patterns, whereby primary producers and invertivorous fish biomass tend to respond most strongly to increasing nutrient concentrations (Carpenter et al., 2001; Hanson & Leggett, 1982; Jeppesen et al., 2000). Moreover, joint effects of increasing temperature and productivity boost food web level change where oligotrophic subarctic lakes are mainly fuelled by benthic algae, but gradually shift towards phytoplankton-derived energy sources towards warmer and more productive lakes (Hayden et al., 2019; Vadeboncoeur et al., 2003). As key drivers of the biomass increment in our dataset for primary producers are cyanobacteria and for secondary consumers cyprinid fish those are both adapted to warm waters (Kosten et al., 2012; Rolls et al., 2017). Both are competitively superior in warmer and productive environment as many cyanobacterial taxa can fix nitrogen directly from air and form dense colonies to avoid predation, while cyprinids across the region have omnivorous diets and very high fecundity (Kosten et al., 2012; Lammens & Hoogenboezem, 1991; Lappalainen et al., 2008). Such uneven changes in community structure and biomass in different trophic levels were critical in shaping food webs and trophic pyramids.

Biomass patterns across trophic levels showed contrasting trends in response to increasing temperature and productivity. This corroborates the 'Green World Hypothesis', where primary producers (phytoplankton) are bottom-up controlled by nutrients, and primary consumers (zooplankton and benthos) are top-down controlled by invertivorous fish (Hairston et al., 1960). However, this model is a simplification of complex aquatic food webs such as the study lakes, as all additionally contain tertiary consumers (e.g. Arctic charr, pike; although their relative biomass and proportional contribution to trophic pyramids was variable). While top consumers biomass increment was shallower compared to invertivorous fish biomass with increasing temperature and productivity, the most diverse piscivore communities colonizing all lake habitats inhabited coldest and most nutrient-poor lakes only. Previous work from these cold and clear lakes suggest a strong top-down role of piscivorous fish on invertivorous fish density and biomass (Jensen et al., 2015; Kahilainen et al., 2019; Thomas et al., 2017). These cold and nutrient poor-lakes have generally low biodiversity that promotes both highly size structured fish populations, that is, strong year class dominance for 10–15 years, and potential for strong trophic cascades (Byström, 2006; Hayden et al., 2014a; Kahilainen et al., 2019). This likely explains the diverse shape of trophic pyramids we observed, especially in ultraoligotrophic and oligotrophic lakes. Increasing temperature and nutrients are fundamental controls that decrease piscivore top-down control and boost bottom-up processes, whereas trophic pyramids seem to stabilize into an hourglass shape in eutrophic lakes. Such trophic pyramid shape is obviously very persistent as common biomanipulation efforts of mass removal of invertivorous consumers (e.g. roach) tend to quickly return to original state (Carpenter et al., 1985; Hansson et al., 1998). Therefore, it is likely that especially in eutrophic lakes the piscivore predation on invertivorous fish is not limiting their population size, as observed elsewhere (e.g. Bartrons et al., 2020). Nutrient concentration is well-known driver of alternative stable states of lakes (Scheffer & Carpenter, 2003) and underlying mechanisms maintaining resilience of eutrophic state may include an hourglass shape of trophic pyramid driven by proportionally high biomass of primary producers and secondary consumers (Jeppesen et al., 2010). Yet, the nutrient level and reason for the possible tipping point of the lake food web communities should be further evaluated in future.

4.3 | Changes in EPA + DHA content of food web components (H2)

The nutritional quality (mg EPA + DHA/g dw) of primary producers and consumers with short lifecycles (i.e. invertebrates) showed a decreasing trend towards warmer and more productive lakes, while in secondary and tertiary vertebrate consumers (i.e. fishes) such trends in nutritional quality were not apparent. Along the environmental gradient, the phytoplankton community shifted towards more cyanobacteria-dominated communities, which are non-EPA and non-DHA synthesizing phytoplankton (e.g. Taipale et al., 2016). The decrease in zooplankton EPA + DHA content varied with an increasing abundance of small cladoceran and cyclopoid copepods. There are many differences in life-history strategies between different zooplankton taxa, where the calanoid *Eudiaptomus graciloides*, the dominant copepod in cold oligotrophic lakes, tends to accumulate lipids (including FAs) to survive winter, whereas short-lived cladocerans form resting stages and do not need high body fat reserves (Grosbois et al., 2017; Hiltunen et al., 2016). In a recent study, Senar et al. (2019) investigated a similar nutrient (total phosphorus 6–48 μg/L) and colour (dissolved organic carbon 2–10 mg/L) gradient in Canadian lakes, but did not record clear decrease in zooplankton taxa FA content. However, their study region was warmer and did not contain strong temperature gradients, which have clear influence on lipid rich copepod abundance in zooplankton communities and thus PUFA content (Gladyshev et al., 2011; Hiltunen et al., 2016; Senar et al., 2019).

Decreasing nutritional quality (mg EPA + DHA/g dw) in primary producers was predicted to result in similar trends in secondary and tertiary consumers (Ahlgren et al., 1996; Taipale et al., 2016). However, we did not find this to be the case and instead the EPA + DHA content of fish population was generally stable, in contrast to our second hypothesis. This could be related to diversity in lipid metabolism of different fish species during different development stages, since our study included many fish species at different developmental stages. A range of aquaculture and experimental studies indicate that EPA and DHA supplement increase fish individual growth and to some extent the muscle FA content of adult fish (Bou et al., 2017; Yeşilayer & Kaymak, 2020). However, similar evidence from the wild is very difficult to obtain due to limited food resources, slow growth rate of fish, long turnover time of muscle tissue, reproduction cycle and species-specific differences in lipid storages (Jørgensen et al., 1997; Keva et al., 2019; Thomas & Crowther, 2015). One alternative for stable fish EPA + DHA content could be that the ability for conversion of DHA from ALA varies greatly among fish species and development stage (Geay et al., 2016; Ishikawa et al., 2019; Kabeya et al., 2018). Since, benthic macroinvertebrates included only a minor amount of DHA as reported also previously, zooplankton is clearly the most likely dietary source of DHA for fish (e.g. Makhutova et al., 2016; Vesterinen et al., 2020). The EPA + DHA content of fish muscle did not differ among feeding habitats (zooplanktivore, benthivore, piscivore), possibly providing evidence for selective transfer or utilization of FAs in fish (Strandberg et al., 2015). Previous results from subarctic lakes indicated that the turnover rate of whitefish (*C. lavaretus*) muscle is slow, and thus stable isotope ratios and fatty acid content responded very slowly to any diet change (Hayden et al., 2014a; Keva et al., 2019; Thomas & Crowther, 2015; Thomas et al., 2019). Our results did not support the idea that prey quality is essential to supporting fish consumer biomass or quality. Together, these results suggest that lipid metabolism is variable and complex among different fish species and their life stages, where generalization of temperature and productivity induced responses to fatty acid content and even further to consumer biomass require more laboratory and field work with wild fish populations.

4.4 | Changes in community EPA + DHA areal content (H3)

The climate–productivity gradient of subarctic study lakes included both eutrophication and browning processes from cold and clear water lakes towards warm and murky waters (Hayden et al., 2019). While the EPA + DHA content of primary producers and consumers did show a clear decrease as suggested by earlier studies (Creed et al., 2018; Taipale et al., 2016), the EPA + DHA areal content at a systemic level did not show corresponding decrease due to massive increase of total biomass; similar patterns have previously been observed in mesocosm studies of benthic communities (Scharnweber et al., 2020). Surprisingly, EPA + DHA areal content in secondary and tertiary consumers indeed increased towards more murky lakes, indicating that total biomass clearly overrode the decline of quality. Changes in phytoplankton EPA + DHA areal content were highest in more eutrophic lakes, even if cyanobacteria contribution to this would be bypassed highlighting the importance of biomass to community EPA + DHA areal content and potentially to production as well. The total areal FA content does not exhibit a 1:1 relationship with the production of phytoplankton due to taxa-specific production rates (Taipale et al., 2020). However, even with a far lower areal content to production ratio in eutrophic lakes, the system-level production of EPA + DHA would still be higher in more eutrophic lakes due to massive phytoplankton biomass increment from oligotrophic towards eutrophic lakes. Therefore, we argue that in the studied climate and productivity gradient, the total production of fatty acids is higher in warm murky than cold and clear water lakes.

4.5 | Caveats

While our sampling protocol did not cover all the food web components, our results from sampled organisms are in general comparable to previous studies conducted from subarctic or boreal lakes. The biomass estimates of fish consumers were derived from simultaneously collected echosounding and gill net CPUE data of pelagic whitefish and vendace, which were collected from a subset of the same study lakes during optimal sampling window using standardized protocols suitable for these two species inhabiting all lakes in current study (Malinen et al., 2014). Our total fish biomass estimates of from ultraoligotrophic to mesotrophic lakes corroborates well with biomass estimates derived from subarctic Canada (Samarasin et al., 2015). In addition, results from our eutrophic lakes correspond to biomass estimates derived from mark–recapture, population analysis and echosounding methods of boreal eutrophic lakes (Olin et al., 2017; Rask et al., 2020). The comparison of biomass results of primary producers and primary consumer values to earlier studies from boreal lakes (Leppä et al., 2003; Nurminen et al., 2018) indicates broadly similar estimates, but clearly lower values than hypereutrophic and warmer lakes (Gyllström et al., 2005; Jeppesen et al., 2000). Future studies should include biomass estimates of benthic algae as they are very important sources of energy in oligotrophic subarctic lakes (Hayden et al., 2019; Vadeboncouer et al., 2003), but could be important in other types of lakes too (Vesterinen et al., 2016; Vadeboncouer & Power, 2017). Assuming a high benthic-to-pelagic production ratio (70%) in our ultraoligotrophic lakes and low ratio (0%) in shallow eutrophic lakes (Vadeboncoeur et al., 2008), the increment in primary producer biomass among the lake gradient would be less pronounced and this would affect also the biomass pyramids. However, whole food web primary production is known to increase along phosphorous gradients (Vadeboncoeur et al., 2003) and the biomass pyramids are more persistent to the changes in one trophic level biomass share due to multiple trophic levels accounting the total biomass. To make future fatty acid budget and production calculations more accurate, we suggest analysing whole fish or creating species-specific conversion factors from muscle tissue to whole body.

5 | CONCLUSIONS

While climate warming trends have been evident in the last decades in Finnish Lapland (Hayden et al., 2014b), increases in lake productivity via catchment greening and especially encroachment of the treeline will likely take much longer to become apparent. Historically, the treeline of Scots pine has been located in the highest latitude lakes of our study with circa 2.5°C higher July air temperatures, indicating the potential for alteration of catchment vegetation and lake productivity towards north (Kultti et al., 2006). While the lake productivity shifts apparent in our study systems originate from intensive forestry activities, there is increasing evidence of resource use related land-use in subarctic focusing on oil and gas drilling, and mining, as well as their associated infrastructure such as roads, pipelines and tailing ponds; activities that are simultaneously changing catchments and lake ecosystems with climate (e.g. Cott et al., 2015; Denisov et al., 2020; Schindler & Smol, 2006). Under current study settings, we show that increasing temperature and productivity have major impacts on lake ecosystems, shifting communities towards warm and murky-water-adapted taxa, where especially biomass of cyanobacteria and cyprinid fish are increasing. These have a major effect on lake food webs, where increasing biomass of primary producers and secondary consumers shift trophic pyramid shapes towards a persistent hourglass shape. Based on our findings, increasing temperature and productivity did reduce phytoplankton, zooplankton and profundal benthos community EPA + DHA content, but these changes in prey quality did not alter fish community EPA + DHA content. Instead, fish community areal EPA + DHA content did increase along the gradient due to a massive increment in fish biomass. While we are not proposing direct predictions of near future food web structure and quality, our results from a pronounced climate and productivity gradient clearly showed that warmer and murkier-water-adapted species will likely increase their biomass share, whereas a decrement in EPA + DHA content may only be observed in primary producers and consumers.

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AUTHOR CONTRIBUTION

K.K.K. provided the original idea for the paper. K.K.K., S.J.T. and O.K. contributed planning the manuscript. Field work was conducted mainly by K.K.K., S.M.T. and B.H. Fatty acid extraction and its analysis were performed mostly by O.K., with contributions from S.J.T., S.M.T., J.V. and P.K. O.K. compiled data, conducted statistical analyses and drafted the first version of the manuscript. All authors contributed to subsequent revisions of the manuscript.

DATA AVAILABILITY STATEMENT

More data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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