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Abstract

Over the last decades, the intensification of anthropogenic activities and associated disturbances on lake watersheds have led to major changes in lakes trophic functioning through accelerated eutrophication. In many lakes, these changes are characterized by an unprecedented increase in organic carbon fluxes, potentially leading to a shift in biogeochemical cycles and in the balance between carbon sequestration and greenhouse gas emissions. Understanding the response of the carbon cycle to natural and anthropogenic environmental changes is becoming a crucial challenge in the context of increasing global pressures. In this study, we reconstructed the changes in the trophic functioning of the benthic and pelagic food web, in response to accelerated eutrophication in four lakes over the last millenium. Changes in carbon pathways in food webs were assessed using stable carbon isotope analysis of chitinous subfossil remains of *Daphnia, Bosmina*, and Chironomini archived in sedimentary records. Changes in the trophic state were inferred from sedimentary geochemical analysis, including carbon accumulation rates, Chla accumulation rates and carbon-to-nitrogen ratios. Agro-pastoral activities were tracked by analysis of coprophilous ascospores. Results provided by this multi-proxy approach highlight recent disruptions in carbon sources and transfer pathways in lakes food webs. In particular, changes in the carbon isotopic signature of pelagic consumers suggest a recent increase in the use of ¹³C-depleted carbon sources such as methanogenic or respiration-derived carbon linked to recent changes in trophic state under the intensification of anthropogenic pressures.

Keywords

Bosmina, carbon isotopes, Chironomidae, Daphnia, eutrophication, lake, methane

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Introduction

Anthropogenic activities over the past few decades have caused disturbances in lake watersheds, resulting in significant changes in the trophic functioning of lakes. Accelerated eutrophication due to increased nutrient inputs has been observed as a direct consequence of these activities (Carpenter et al., 1998; Schindler, 2006; Smith et al., 1999). These changes in trophic status are likely to disrupt the carbon cycle by altering the carbon sources and pathways in lake food webs. Indeed, the increase in organic sedimentation can promote respiration-derived carbon production (Morales-Williams et al., 2021; Xiao et al., 2020), which can be transferred into lake food webs. Moreover, the intensity of methanogenesis (CH₄ production) and methanotrophy (CH₄ oxidation) is strongly dependent on sediment characteristics (organic matter quantity and C/N ratio; Duc et al., 2010; Praetzel et al., 2020) and oxygen conditions (Beaulieu et al., 2019; Rudd and Hamilton, 1978), both of which can be strongly affected by eutrophication. As lakes play a key role in the global carbon cycle (Bastviken et al., 2011; Tranvik et al., 2009), understanding the sources and pathways of carbon in lake food webs is an important challenge to predict the evolution of the functioning of the aquatic carbon cycle in the context of increasing human activities.

By using lake sediments as ecological archives, paleolimnological studies provide valuable reconstructions of changes in lake trophic functioning on decadal to millennial time scales, filling the gap in available instrumental data. The use of geochemical proxies, molecular techniques, and biological remains from limnological archives is a key approach to understand the response of lake ecosystems to disturbances (Saulnier-Talbot, 2016). For example, the concentrations of sedimentary pigments can be used as a proxy for past productivity and trophic state (Guilizzoni et al., 2011; Wolfe et al., 2006). The development of this approach in many lake sediment archives indicated that the intensification

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Table 1. Characteristics of the studied lakes.

Lake	Long	Lat	Alt.	Maximum depth	Lake area	Watershed area
Narlay	5.90 E	46.63 N	748	40	41	222
Remoray	6.26 E	46.77 N	850	28	85	2486
Abbaye	5.91 E	46.52 N	910	19	82	245
Brévent	6.86 E	45.90 N	2125	20	2.95	17

Geographical position are expressed in decimal degrees in WGS84 reference coordinate system, Alt indicates altitude (m.a.s.l.), Depth indicates maximal water depth (m), lake area and watershed area are expressed in ha.

of anthropogenic activities is associated with dramatic increases in autochthonous primary production (Jinglu et al., 2004; Lami et al., 1994; Schneider et al., 2018). These changes are also mirrored in sediment geochemical properties, with decreasing C_{org}/N ratios (Punning and Tõugu, 2000) and increasing organic matter accumulation rates (Anderson et al., 2014). Temporal changes in benthic consumer communities, and especially changes in chironomid assemblages, may also reflect the degradation of oxygen conditions in the hypolimnion (Belle et al., 2016b; Frossard et al., 2013). Finally, some paleolimnological studies have suggested that these changes in trophic state are associated with increased transfers of respiration-derived carbon (CO₂) and methanogenic carbon (CH₄) in lake food webs (Belle et al., 2016a, 2016b; Frossard et al., 2014; Schilder et al., 2017). However, the consequences of these changes in lake trophic state on the carbon sources and transfer pathways remain poorly understood, particularly for pelagic food webs. Understanding the lake trophic functioning, and in particular the long-term dynamics of carbon transfer in food webs in response to eutrophication, is therefore a major challenge in the context of ongoing human-induced changes.

Stable isotope analysis (SIA) of carbon is a widely used method for studying carbon flows in lakes (Frossard et al., 2014; Fry, 2006; Grey et al., 2001; Taipale et al., 2007). The carbon isotopic signature (δ^{13} C) of organisms closely reflects the isotopic signature of their diet (DeNiro and Epstein, 1978) and is therefore commonly used to trace carbon sources and pathways in food webs (del Giorgio and France, 1996). In lakes, the CO₂ fixed by photosynthetic organisms and the biogenic CH4 oxidized by methane-oxidizing bacteria (MOBs) are two common carbon sources that usually support the production of aquatic consumers. Phytoplankton δ^{13} C in lakes ranges from -25% to -40% (France, 1995; Masclaux et al., 2013; Peterson and Fry, 1987; Vuorio et al., 2006; Wang et al., 2013), depending on the intensity of isotopic fractionation and CO₂ origin (Bade et al., 2004, 2006; Gu et al., 2006). Indeed, respiration-derived CO₂ usually exhibits δ^{13} C values that are consistently lower than atmospheric-derived CO₂ as a result of photosynthetic and metabolic fractionation process occurring during carbon recycling (Degens, 1969; Fry, 2006; Keough et al., 1996; Raven, 1996). Moreover, biogenic methane is highly depleted in ¹³C (Jedrysek, 2005) and isotopic fractionation by MOBs during CH₄ oxidation leads to a further ¹³C depletion (Templeton et al., 2006). The incorporation of ¹³C-depleted carbon into consumer biomass is thus easily recognized by its δ^{13} C value, which is highly distinct from that of atmosphericderived carbon. In limnological studies, SIA has been widely used to track carbon transfers to pelagic (Lennon et al., 2006; Morlock et al., 2017) and benthic (Deines et al., 2007; Grey et al., 2004) consumers in lakes. SIA has also been successfully used in paleolimnological studies through the analysis of consumer remains preserved in sediments. These studies using the isotopic signature of subfossil remains of consumers preserved in sediments as an indicator of the past availability of methane and/or carbon from respiration are of great importance because they may indirectly provide records of variations in the intensity of greenhouse gas

fluxes over time. $\delta^{13}C$ analysis of chitinous remains of benthic consumers, such as chironomid head capsules retrieved from lake sediments, is increasingly used to assess past changes in carbon sources fueling the benthic food web and allows the reconstruction of shifts in carbon pathways associated with past environmental changes (Belle et al., 2016b, 2016c; Frossard et al., 2014; van Hardenbroek et al., 2010). Such changes can also be reconstructed for pelagic food webs using zooplankton remains, and more especially ephippia, which are resting eggs produced by cladocerans (Essert et al., 2023; Frossard et al., 2014; Schilder et al., 2015a, 2015b; van Hardenbroek et al., 2013, 2014; Wooller et al., 2012). The ephippia are well-preserved in sediments over time and are found in high abundance in most lacustrine records (Szeroczyńska and Sarmaja-Korjonen, 2007). However, δ¹³C values of Daphnia can vary considerably over the annual cycle of lakes due to seasonal variations in the availability of the different food sources for pelagic consumers (Essert et al., 2022a, 2023), and ephippia are generally produced by Daphnia within a very short time window (Caceres, 1998; Conde-Porcuna et al., 2014; Morlock et al., 2017; Perga, 2011). Therefore, potential changes in the timing of ephippia production relative to the seasonal availability of the different carbon sources may lead to misinterpretation of $\delta^{13}C_{ephippia}$ variations in sedimentary records. Nevertheless, such biases can be avoided by conducting parallel analysis with more time-integrated pelagic consumer remains, such as Daphnia claws or Bosmina head-shields (Essert et al., 2023). Despite the wide diversity of consumer remains that can be used to study past carbon transfers in lake food webs, there are still too few studies that simultaneously examine temporal changes in benthic and pelagic consumers in relation to geochemical signals of eutrophication (Frossard et al., 2014; Schilder et al., 2017).

The aims of this study are therefore (I) to investigate whether there have been major changes in the carbon sources fueling pelagic and benthic food webs of lakes during their recent history (i.e. the last millenium) and (II) to assess whether these shifts can be explained by changes in the anthropogenic activities in the lake watershed, and consequent changes in lake trophic states.

Paleolimnological reconstructions of carbon sources and pathways in lake food webs in four French mountain lakes were performed through the study of δ^{13} C values of pelagic consumers (*Daphnia* ephippia, *Daphnia* claws, and *Bosmina* head-shields), and also on benthic consumers (chironomid head-capsules). The design of our study includes the assessment of the accumulation rates of sedimentary organic matter, chlorophyll *a* accumulation rates, and C_{org}/N ratios as proxies for changes in lake trophic state. In addition, the intensity of agro-pastoral pressure on lake watersheds was inferred from previously published coprophilous fungi ascospore counts.

Material and methods

Study sites

Four dimictic lakes in eastern France were selected for this study due to their differences in morphological characteristics (water depth and lake area) as well as watershed characteristics (Table 1).

Table 2. Details of the studied proxies indicating core name, and references.

3

Proxy	Lake	Core	Reference
δ^{13} C Daphnia Ephippia	Brevent	Br12_P4	This study
	Narlay	Nar_19a	This study
	Remoray	Rem_19a	This study
	Abbaye	Abb_20	This study
δ^{I3} C Daphnia Claws	Brevent	Br12_P4	This study
	Narlay	Nar_19a	This study
	Remoray	Rem_19a	This study
	Abbaye	Unavailable	
$\delta^{I3}C$ Bosmina Head-Shield	Brevent	Br12_P4	This study
	Narlay	Nar_19a	This study
	Remoray	Rem_19a	This study
	Abbaye	unavailable	
δ^{I3} C Chironomid Head Capsule	Brevent	Br12_P4	Belle et al. (2016b)
	Narlay	Nar_19a	This study
	Remoray	Rem13_P1	Belle et al. (2015)
	Abbaye	Abb_20	This study
δ ^{I3} C OM	Brevent	Br12_P4	Belle et al. (2016b)
	Narlay	Nar10_P1	Belle et al. (2014)
	Remoray	Rem13_P1	Belle et al. (2015)
	Abbaye	Abb_20	This study
C _{org} accumulation rates	Brevent	Brel2_P4	Belle et al. (2016b)
0.8	Narlay	Nar13_P1	Belle et al. (2016c)
	Remoray	Rem I 3_PI	Belle et al. (2016b)
	Abbaye	Abb_20	This study
C _{ord} /N	Brevent	Brel2_P4	This study
5,8	Narlay	Nar13_P1	This study
	Remoray	Rem07_P1	This study
	Abbaye	Abb_20	This study
NIRS-inferred Chla + derivatives accumulation rates	Brevent	Brel3_PI	This study
	Narlay	Nar_19a	This study
	Remoray	Rem_19a	This study
	Abbaye	Abb_20	This study
Coprophilous ascospore accumulation rates	Brevent	Brel2_P4	Belle et al. (2016b)
	Narlay	Nar10_P1	Belle et al. (2016c)
	Remoray	Rem07_P1	Belle et al. (2016b)
	Abbaye	Abb_20	This study

Lakes Narlay, Remoray, and Abbaye are located at mid-altitude, ranging from 750 to 900 m within the Jura-Mountains (pre-Alps). These Jura lakes are of medium size, with areas of 41, 85, and 82 hectares, and maximum depths of 40, 28 and 19 m, respectively (Table 1). Owing to the calcareous bedrock prevalent in the region, the water in Jura lakes is notably mineralized, featuring elevated carbonate concentrations and high conductivity. The watershed for these three lakes encompasses 222, 2486, and 245 hectares, respectively, and is predominantly covered by agricultural activities and forests (Belle et al., 2016c; Essert et al., 2022b).

Lake Brévent is located in the French Alps at a high altitude, reaching 2125 m (Table 1). Lake Brévent is smaller in size compared to the other studied lakes, covering an area of only 2.9 hectares. The watershed of Lake Brévent is underlain by metamorphic rocks, resulting in relatively low mineralization of the lake's waters. The watershed is dominated by mineral cover, but also features alpine grasslands (Belle et al., 2016b).

All the lakes studied are thermally stratified in summer and are dimictic (Essert et al., 2022b; Réseau Lacs Sentinelles, 2020). The profundal zone of all four lakes always exhibits anoxic conditions at the end of the summer stratification period (Essert et al., 2022b; Réseau Lacs Sentinelles, 2020).

Sediment core and chronology

Sediment cores were collected from the deepest part of the lakes using a gravity corer (UWITEC, 90 mm diameter). Depending on the lake, samples analyzed in this study may come from different cores collected at the same location in the lake basins but at different years. Some proxies were from published sources. Information on the name of the core used for each proxy analyzed, and on the origin of the data are therefore summarized in Table 2.

Age/depth models for the cores Br12_P4 (from Lake Brévent), Nar13_P1, Nar10_P1 (from Lake Narlay), Rem13_P1 and Rem07_P1 (from Lake Remoray) were already available from Belle et al. (2015, 2016c, 2016b). For the present study, new age/ depth models were built for the cores Abb_20 (Lake Abbaye), Bre13_P1 (Lake Brévent), Nar_19a (Lake Narlay), and Rem_19a (Lake Remoray).

The chronology of the sediment core retrieved from Lake Abbaye in 2020 was constructed from radiometric methods (¹³⁷Cs, ²⁴¹Am, and ²¹⁰Pb analysis with gamma spectrometry) from the first 50 cm of the core Abb_20, combined with two radiocarbon dates from terrestrial remains (Poznan Radiocarbon Laboratory, Poland; Table 3 and Appendix 1).

For the new cores retrieved in 2019 from Lakes Narlay (Nar_19a) and Remoray (Rem_19a), ¹³⁷Cs and ²⁴¹Am analyses were performed on the first 20 cm of each core. Furthermore, these new cores were stratigraphically correlated with former sediment cores for which chronology was already available (Na 10_P1 and Rem13_P1) using magnetic susceptibility profiles measured with a multi-sensor core logger with an interval of 5 mm (GEOTEK, Appendix 2 and 3). The age/depth models of the 2019 cores from Lakes Narlay and Remoray relied (i) on the ages

 Table 3.
 Radiocarbon dates.

Lac code	Material	Core	yr BP	Cal AD (2σ range)
Poz-132028	Leaf - terrestrial	Abb_20	420 ± 30	1427–1619
Poz-131763	Leaf - terrestrial	Abb_20	730 ± 30	1227-1378
Poz-138173	Leaf - terrestrial	Rem_19a	755 ± 30	1223-1287
Poz-137476	Leaf - terrestrial	Nar_19a	890 ± 30	1044-1222
	Lac code Poz-132028 Poz-131763 Poz-138173 Poz-137476	Lac codeMaterialPoz-132028Leaf - terrestrialPoz-131763Leaf - terrestrialPoz-138173Leaf - terrestrialPoz-137476Leaf - terrestrial	Lac codeMaterialCorePoz-132028Leaf - terrestrialAbb_20Poz-131763Leaf - terrestrialAbb_20Poz-138173Leaf - terrestrialRem_19aPoz-137476Leaf - terrestrialNar_19a	Lac code Material Core yr BP Poz-132028 Leaf - terrestrial Abb_20 420 ± 30 Poz-131763 Leaf - terrestrial Abb_20 730 ± 30 Poz-138173 Leaf - terrestrial Rem_19a 755 ± 30 Poz-137476 Leaf - terrestrial Nar_19a 890 ± 30

Calibrated using IntCal20 calibration curve.



Figure 1. Age/depth models of the Abb_20 core based on radionuclide dating method (²¹⁰Pb, ¹³⁷Cs, and ²⁴¹Am) and two radiocarbon dates (a); age/depth models of the Rem_19a core based on cross-correlation with Rem13_P1 using magnetic susceptibility with 10 horizon markers represented by gray dots and verification of the obtained model with a radiocarbon date represented by the red bar (b); age/ depth models of the Nar_19a core based on cross-correlation with Nar10_P1 using magnetic susceptibility with nine horizon markers and verification of the obtained model with Nar10_P1 using magnetic susceptibility with nine horizon markers and verification of the obtained model with a radiocarbon date represented by the red bar (c); and age/depth models of the Bre13_P1 core based on radionuclide dating method (²¹⁰Pb, ¹³⁷Cs, and ²⁴¹Am) and cross-correlation with Bre12_P4 using magnetic susceptibility with seven horizon markers (d).

provided by the radio-nuclide analysis (depth of the 1986¹³⁷Cs peak and the 1963¹³⁷Cs and ²⁴¹Am peaks), and (ii) the ages of horizon markers in magnetic susceptibility profiles provided by the age/depth model of the old cores (Na10_P1 and Rem13_P1, Appendix 2 and 3). The age/depth models of the 2019 cores from Lakes Remoray and Narlay were further validated with new radiocarbon dates performed from terrestrial remains retrieved from the oldest part of each core (Poznan Radiocarbon Laboratory, Poland; Table 3, Figure 1).

For the core retrieved from Lake Brévent in 2013 (Bre13_P1), ²¹⁰Pb, ¹³⁷Cs and ²⁴¹Am analyses were performed on the first

10 cm of the core. Furthermore, this new core was stratigraphically correlated with the former sediment core for which a chronology was already available (Bre12_P4) using X-ray fluorescence profiles of lead measured with an Avaatech XrF Core Scanner (Edytem Laboratory, Appendix 4). The age/depth model of the Bre13_P1 core relied (i) on the ages provided by the radio-nuclides analysis (depth of the 1963¹³⁷Cs and ²⁴¹Am peak and chronology derived from the ²¹⁰Pb flux dating model), and (ii) the ages of 7 horizon markers in the magnetic susceptibility profiles provided by the age/depth model of the old core (Bre12_ P4, Appendix 4). The age/depth modeling for the different cores (Figure 1) was performed using a simple linear interpolation (Clam package for R; (Blaauw, 2010)).

$\delta^{I3}C$ analysis

Isotopic carbon composition was analyzed on bulk organic matter ($\delta^{13}C_{OM}$) and five types of chitinous subfossil remain: *Daphnia* ephippia ($\delta^{13}C_{ephippia}$), *Daphnia* claws ($\delta^{13}C_{claws}$), *Bosmina* head-shields $(\delta^{13}C_{Bosmina} + s)$ and chironomid head capsules of the Chironomini tribe $(\delta^{13}C_{Chironomini}; of the genus Chironomus$ in Abbaye and Brévent, and genus Sergentia in Remoray and Narlay records). Sediment samples were sieved at 60 and 120 µm and Daphnia claws, Daphnia ephippia, Bosmina headshields, and chironomid head capsules were carefully handpicked using fine forceps under a stereomicroscope. The Daphnia claws were recognized according to the description provided by Szeroczyńska and Sarmaja-Korjonen (2007). For each sample, a minimum of 0.05 mg dry weight of material required for isotopic analysis was picked up. Unfortunately, the abundance of Bosmina head-shields and Daphnia claws in the sedimentary records of Lake Abbaye was too low to allow isotopic analyses. All the subfossil samples were exposed to a 3.7% HCl solution for carbonate removal, rinsed three times with demineralized water, and put into ultra-clean tin capsules. The samples were then dried at 60°C in an oven for 2 days. Stable carbon isotope analyses were performed using an isotope ratio mass spectrometer interfaced with an elemental analyzer (EA-IRMS) at INRAE Nancy (Silvatech platform, Champenoux, France). The isotope ratios were expressed in the delta notation with Vienna Pee Dee Belemnite as the standard: $\delta^{13}C$ (‰) = $(R_{sample}/R_{standard} -1) \times 1000$, where R= ¹³C /¹²C. Replication of sample measured on an internal standard gave analytical errors of ± 0.3 ‰ for all δ^{13} C measurements.

Sediment elemental analysis

Sediment samples were dried and acidified with a solution of HCl (3.7%) to remove carbonate. Elemental analysis of sedimentary organic carbon was performed using a VARIO TOC CUBE analyzer (Elementar). C_{org} concentrations were converted to accumulation rates (C_{org} AR, g/100cm²/year) from sediment dry bulk density and sedimentation rate provided by the age/ depth model. Elemental analysis of total nitrogen was performed using a VARIO MAX CNS. The carbon-to-nitrogen ratio (C_{org}/N) was calculated from the C_{org} concentration and total nitrogen concentration measurements performed with a VARIO MAX CNS.

Coprophilous ascospores analysis

Sediment samples were first prepared by chemical treatment (HCl, NaOH, HF, HCl, and acetolysis) and by removing any material larger than 100 µm by sieving following the standard procedure of Faegri and Iversen (1964). Spores of coprophilous fungi were counted following Etienne and Jouffroy-Bapicot (2014) and were identified using mycological and palynological literature (van Geel et al., 2003). The identification of strict coprophilous fungal ascospores (Sporormiella-type and Podosporatype) in lake sediments reflects the presence of herbivores in the watershed and can be used as a proxy for local grazing pressure (Etienne et al., 2013). Coprophilous ascospores concentrations were converted to accumulation rates (coprophilous fungi sensu stricto accumulation rates, nb/cm²/year) from the dry bulk density of the sediment and sediment accumulation rate provided by the age/depth model. Coprophilous ascospores results were expressed as accumulation rate (nb/cm²/years).

NIRS-inferred chlorophyll a analysis

Concentrations of chlorophyll *a* and related derivatives, including its degradation product, were determined by near-infrared reflectance spectroscopy (NIRS) according to the method published by Wolfe et al. (2006). The NIRS spectra were measured every 0.5 cm on each core analyzed using a multi-sensor core logger (GEOTEK). NIRS-inferred chlorophyll *a* + derivative concentrations were then calculated from the reflectance spectra using the model developed by Wolfe et al. (2006). NIRS-inferred chlorophyll *a* + derivative concentrations were then converted to accumulation rates (Chl*a* + derivatives AR, μ g/cm²/year) by using the dry bulk density of the sediment and the sediment accumulation rate provided by the age/depth model.

Data analyses

To identify the main phases of lake trophic functioning, zonation in the geochemical data (Corg AR, Corg/N, and Chla AR) of each lake was performed by a constrained hierarchical cluster analysis using Euclidean distance on standardized data and CONISS linkage method (Grimm, 1987). The significance of the phases was assessed using the broken stick model (Bennett, 1996). As C_{org} AR, C_{org}/N , and Chla + derivatives AR data were not always derived from the same core for each lake, the values of the different variables for each stratigraphic point are derived from linear interpolation for the ages determined by the variables with the fewest observations (C_{org}/N or C_{org} AR depending on lakes). Differences in the accumulation rate of coprophilous ascospores, δ^{13} C of the *Daphnia* ephippia, and δ^{13} C of the chironomid headcapsules between the different phases of each lake were then assessed using pairwise Wilcoxon rank-sum tests with p-value correction for multiple testing (Benjamini and Hochberg, 1995). The significance level was set at $\alpha = 0.05$. All statistical analyses and figures were performed using R 3.5.1 statistical software (R Core Team, 2019) and additional R packages (Dunnington et al., 2022; Juggins, 2022; Wickham, 2009).

Results

Organic carbon, Chla accumulation rates, and C/N ratio

The four sediment records are all characterized by an increase in the rate of organic carbon accumulation (hereafter referred to as C_{org} AR) from older to younger layers, resulting in an unprecedented high Corg AR during the last century with respect to the last millenium. This increase in C_{org} AR is accompanied by an increase in Chla accumulation rate (Chla AR) and, in most cases, by a decrease in the carbon-to-nitrogen ratio. However, despite these common features, the temporal dynamics of these changes over the last millenium vary from lake to lake. Cluster analysis performed on the geochemical characteristics of sediment organic matter indicates significantly different periods that correspond to the main phases in the functioning history of the four studied lakes (Figure 2). Four distinct phases have been identified in the sedimentary record of Lakes Abbaye and Brévent, and three phases for Lakes Remoray and Narlay.

The oldest phase (i.e. phase I) for Lake Abbaye (from 990 to 1680 A.D.), Remoray (from 1090 to 1770 A.D.), Narlay (from 1180 to 1380 A.D.) and Brévent (from 630 to 1100 A.D.) shows the lowest C_{org} AR compared to the subsequent phases of each lake studied (Figure 2). It also presents the lowest Chla AR, except for Lake Remoray. A significant increase in the Chla AR (pairwise Wilcoxon rank test *p*-value < 0.005) occurred in phase II of the lakes Abbaye (from 1680 to 1870 A.D.), Narlay (from 1380 to 1910 A.D.) and Brévent (from 1100 to 1270 A.D.). This increase in Chla AR is associated with a significant decrease in



Figure 2. Trends in organic carbon accumulation rate, Chlorophyll *a* accumulation rate and C_{org} /N for sediment records of lake Abbaye (a), Remoray (b), Narlay (c) and Brévent (d). Dendrogram is based on these three proxies previously standardized and constructed by hierarchical clustering analysis (Euclidean distances, CONISS linkage method). Significance of the zonation was assessed using broken-stick model and significant zone were represented by dashed lines and Roman number. C_{org} AR data for lake Remoray, Narlay and Brévent were previously published in Belle et al. (2016c, 2016b).

 C_{org}/N ratios for Narlay (*p*-value < 0.001) and a significant increase in C_{org}/N ratios for Abbaye (*p*-value < 0.001) and Brévent (*p*-value < 0.005). In addition, these changes are also followed by an increase in the C_{org} AR for Lake Brévent (*p*-value < 0.001). For Lake Remoray, we observe a significant increase in C_{org} AR (*p*-value < 0.001) associated with a significant decrease in Chl*a* AR (*p*-value = 0.001) during this second phase. The phase III of the sedimentary records of Lake Abbaye (from 1870 to 1945 A.D.) and Brévent (from 1270 to 1840 A.D.) is again characterized by a significant increase in Chl*a* AR (*p*-value < 0.001) together with a significant decrease in C_{org}/N ratios (*p*-value < 0.001). These changes are followed by a significant increase in C_{org} AR for Lake Abbaye (*p*-value < 0.001), while these accumulation rates remain comparable to the previous phase for Lake Brévent.

The most recent phase identified from sediment geochemistry (i.e. phase IV for Abbaye and Brévent since 1945 and 1840 A.D. respectively, and phase III for Remoray and Narlay since 1940 and 1910 A.D. respectively) exhibits the highest C_{org} AR with the exception of Lake Abbaye, where accumulation rates remain comparable (Figure 2). Indeed, the C_{org} AR increase significantly (*p*-value < 0.005) to reach unprecedented levels for the three lakes of Remoray, Narlay, and Brévent during this latest phase, and is followed by a significant increase in Chla AR for Lake Remoray (*p*-value=0.001). Although Chla AR increase to reach

unprecedented levels during this most recent phase for Lake Narlay and Brévent, they are not significantly different from those of the previous phase. These recent changes are also accompanied by a significant decrease in C_{org}/N ratios to values around 10 for Lakes Remoray (p-value=0.02) and Narlay (p-value<0.001), leading to the lowest values in the records during the most recent phase. On the other hand, for Lake Brévent, the C_{org}/N ratios are not significantly different from those of the previous phase. For Lake Abbaye, C_{org} AR values are not significantly different from the previous phase and seem comparable (p-value=0.2). Moreover, in contrast to the other three lakes, the latest phase does not show a further increase in Chla accumulation rates, but rather a slightly significant decrease with respect to the previous phase (p-value < 0.001). However, the accumulation rates of Chla during the most recent phase of Lake Abbaye remain very high compared to the two first and oldest phases. Moreover, as with Lake Remoray and Narlay, a significant decrease in the Core/N ratios is observed during this most recent phase (*p*-value < 0.001), which shows the lowest values of the Lake Abbaye chronology.

Accumulation rates of ascospores of coprophilous fungi

The accumulation rates of strictly coprophilous ascospores (hereafter referred to as Cop AR) are characterized by high



Figure 3. Distribution of coprophilous fungi accumulation rate according to the lake phases inferred by CONISS analysis of geochemical sedimentary data for lake Abbaye (a), Remoray (b), Narlay (c) and Brévent (d). Means sharing the same letter are not significantly different according to Wilcoxon rank test with corrections for multiple testing (α = 0.05). Data are not presented for phase II of lake Brévent due to the limited number of observations during this phase. Data for lake Remoray, Narlay and Brévent were previously published in Belle et al. (2016c, 2016b).

between-lake differences and strong temporal variations along the records of each of the four lakes (Figure 3). Indeed, Cop AR range from 14 to 440 spores/cm²/year for Lake Abbaye, 5-455 spores/ cm²/year for Lake Remoray, 1 to 119 spores/cm²/year for Lake Narlay and from 0 to 13 spores/cm²/year for Lake Brévent. The lowest Cop AR are always found in the oldest phase of the sedimentary record for all the lakes studied (Figure 3). A significant increase in coprophilous ascospore fluxes is observed in the second phase of Remoray and Narlay records (*p*-value= 2.6×10^{-5} and 0.03, respectively) and in the third phase of Abbaye and Brévent records (p-value=0.04 and 0.009) (Figure 3). In the latest phase of Lake Remoray record, the Cop AR values increase again significantly compared to the previous phase (p-value=0.02), while in Lake Narlay and Brévent, they remain at a relatively high level. In Lake Abbaye, in contrast to the other lakes studied, the Cop AR decrease significantly during the most recent phase (p-value=0.04) (Figure 3).

$\delta^{I3}C$ of consumers and bulk organic matter

In the four lakes, the most recent phases (phases III and IV for Lake Abbaye, III for Lake Remoray and Narlay, and phase IV for Lake Brévent) are characterized by a strong decrease in $\delta^{13}C_{ephippia}$ (Figures 4 and 5). The $\delta^{13}C_{ephippia}$ of these recent phases are significantly lower than those of the preceding phase (Figure 5), and are the most negative values (near or below $-40 \ \%$) for the whole records, except at Lake Brévent. Indeed, in this lake, despite a strong decrease compared to the preceding phase, the recent $\delta^{13}C_{ephippia}$ remain close to those of the beginning of the record (phase I, mean = $-35.2 \pm 0.6 \ \%$, *p*-value = 0.1) (Figure 5). Moreover, the $\delta^{13}C_{ephippia}$ in Lake Brévent record are higher than in the three other lakes (mean = $-34.1 \pm 1 \ \%$).

In Lake Abbaye, the abundances of Bosmina head-shields and Daphnia claws were too low to allow carbon isotope analyses. In the other three lakes, however, the $\delta^{13}C_{claws}$ were always higher than $\delta^{13}C_{ephippia}$ (Figure 4). $\delta^{13}C_{claws}$ are also significantly correlated with $\delta^{13}C_{ephippia}$ for Lake Narlay (Spearman test *p*-value=0.006, rho=0.92) and Lake Brévent (Spearman test p-value < 0.001, rho = 1). They follow the same temporal trends, characterized by a decrease during the most recent phase. At Remoray, $\delta^{13}C_{claws}$ follows the decrease in $\delta^{13}C_{ephippia}$ during the most recent phase (i.e. Phase III), but the temporal trends in δ^{13} C differ between ephippia and claws for the oldest samples. As a result, we did not observe a significant relationship between $\delta^{13}C_{claws}$ and $\delta^{13}C_{ephippia}$ when considering the whole Remoray dataset (Spearman test, p-value=0.3). The temporal changes in $\delta^{13}C_{\textit{Bosmina}\ h-s}$ seem similar to $\delta^{13}C_{\textit{claws}}$ in the most recent phase in Remoray, Narlay, and Brévent records. However, the two proxies were not significantly correlated over the whole sediment records according to Spearman's test.



Figure 4. Temporal evolution of (1) $\delta^{13}C_{ephippia}$, (2) $\delta^{13}CBosmina_{h-s}$ and $\delta^{13}C_{claws}$, (3) $\delta^{13}C$ of bulk OM and $\delta^{13}C_{Chironomini}$, and (4) $\delta^{13}C$ difference between bulk OM and Chironomini for lakes Abbaye (a), Remoray (b), Narlay (c) and Brévent (d). The white and gray zone sequences together with the Roman numerals indicate the lake phases inferred by CONISS analysis of geochemical sedimentary data. $\delta^{13}C_{Chironomini}$ data for lake Remoray and Brévent were previously published in (Belle et al., 2015, 2016b).

There is no clear temporal trend in $\delta^{13}C_{Chironomini}$ along the four sediment records (Figure 4). No significant differences in chironomid isotopic signatures were found between the different phases of the trophic history of the lakes (Figure 5). Furthermore, the near disappearance of head capsules in the uppermost sediment samples for Lakes Remoray and Narlay precludes analysis of the carbon isotopic signature of chironomid remains after the 1970s. Regarding the $\delta^{13}C_{OM}$ values, they decrease significantly during the latest phases of lakes Abbaye, Remoray, and Narlay

(*p*-value < 0.05), although we have only three measurements for the most recent phase of Lake Narlay. In contrast, no significant differences in the $\delta^{13}C_{OM}$ are observed between the different phases of Lake Brévent, except for the oldest phase, which has significantly lower $\delta^{13}C_{OM}$ values (*p*-value < 0.005). In the whole dataset, $\delta^{13}C_{Chironomini}$ are always lower than $\delta^{13}C_{OM}$ (Figure 4), and no significant differences in $\Delta\delta^{13}C_{OM-Chironomini}$ are observed between the different phases (*p*-value > 0.1), except for the Lake Remoray record. In this lake, $\Delta\delta^{13}C_{OM-Chironomini}$ show a decrease



Figure 5. Distribution of δ^{13} C value of *Daphnia* ephippia and chironomids head-capsules according to the lake phase inferred by CONISS analysis on sedimentary geochemical data for lake Abbaye (a), Remoray (b), Narlay (c) and Brévent (d). Means sharing the same letter are not significantly different according to Wilcoxon rank test with corrections for multiple testing ($\alpha = 0.05$). Data are not presented for phase II of lake Abbaye and Brévent and for phase I for Chironomini of lake Narlay due to the limited number of observations during these phases.

in the early 20th century, followed by a further increase in the latest phase. As a result, the mean $\Delta\delta^{13}C_{OM-Chironomini}$ during the second phase of Lake Remoray is significantly lower than during the first phase (*p*-value=0.01). For Lake Brevent, $\Delta\delta^{13}C_{OM-Chironomini}$ is significantly lower compared to the other lakes, with a mean of 4.9 ± 1.9 ‰ compared to 7 ± 2.1 ‰ for Abbaye, 10.7 ± 1.8 ‰ for Remoray and 7.1 ± 3.3 ‰ for Narlay.

Discussion

Evidence of the recent eutrophication of lakes

In the three Jura lakes (Remoray, Narlay, and Abbaye), the temporal dynamics of organic carbon and algal pigment accumulation rates and carbon-to-nitrogen ratio undoubtedly indicate that the limnological conditions are currently in an unprecedented state with respect to a millenium-long record. The trophic history of these three lakes is characterized by a sharp increase in the organic sedimentation rate over the last two centuries, paralleled by a decrease in the C/N ratio of organic matter to values around 10. While diagenetic processes may have contributed to a part of the observed trends in C/N ratios, they do not appear to be the primary explanation for the changes over the past two centuries. Although diagenetic processes affect the C/N ratio of sediments with aging due to different rates of carbon and nitrogen degradation, the ratio stabilizes after 5 years due to extremely low degradation rates (Gälman et al., 2008). However, in our study, the observed reductions occurred earlier in the record, suggesting that these processes play a minor role in the declining C/N ratio of organic matter. Phytoplanktonic organic matter has commonly low C/N ratio ranging from 5 to 10, whereas terrestrial organic matter (e.g. leaf and wood of vascular plants from the watersheds) is rich in cellulose and therefore has a higher C/N ratio (20 or more; Meyers and Lallier-vergés, 1999). The recent decrease in C/N ratios then suggests a change in the nature and origin of the sediment organic matter, corresponding to either an increase in phytoplankton production, a decrease in inputs of terrestrial detritus from the watersheds, or both. However, the increase in organic matter accumulation rates combined with the decrease in C/N ratios indicates that these changes are most likely the result of an increase in autochthonous production. This hypothesis is further supported by the increase in the accumulation rates of chlorophyll a and its degradation products for the most recent phases, which suggests a recent increase in primary production (Wolfe et al., 2006). Similar recent shifts, characterized by an increasing

organic sedimentation combined with an increasing primary production, were seen in many lakes located in the Jura Mountains (Belle et al., 2016a). These quantitative and qualitative changes in sediment organic matter are likely to be the result of accelerated eutrophication of water bodies caused by excessive nutrient inputs related to human activities in the catchments (Carpenter et al., 1998; Knoll et al., 2003).

The high accumulation rates of coprophilous fungi during the recent phases of Lake Abbaye and Remoray history reinforce this assumption since they are efficient proxies of agro-pastoral pressure intensity on the watersheds (Etienne et al., 2013; van Geel et al., 2003). These findings suggest that the recent changes in the trophic status of these two lakes could be linked, at least in part, to the intensification of pastoralism in their watersheds. The potential impact of these agro-pastoral practices on the trophic status of lakes has already been shown in different geographical contexts by several authors (Belle et al., 2016a; Hausmann et al., 2002; Tiberti et al., 2014).

Our results suggest that the history of Lake Narlay is characterized by two successive phases of accelerated eutrophication. During phase II (1400 to 1900 A.D.), the highest accumulation rates of coprophilous ascospores are combined with an increase in primary production, as indicated by high accumulation rates of chlorophyll a and its degradation products and lower C/N values. Diffuse nutrient inputs linked to the development of pastoralism therefore appear to be at the origin of this first stage of eutrophication. The further increase in the accumulation rate of organic carbon and phytoplankton production indicates that a second stage of eutrophication occurred during the 20th century. This more recent and stronger eutrophication of the lake does not seem to be related to an intensification of pastoralism in the lake catchment, since the accumulation rates of coprophilous ascospores remain stable. According to historical data, Lake Narlay suffered from point source nutrient inputs from wastewater from cheese factories and pig farms during the 20th century (Belle et al., 2016b, 2016c).

The alpine Lake Brévent has also undergone an increase in organic carbon sedimentation rates, which have more than doubled since the mid-19th century. However, we do not observe any significant changes in the C/N ratio for this recent period. This finding suggests that the increase in organic accumulation rate is probably not related to a change in the relative contribution of autochthonous versus allochthonous material. For the most recent phase, the C/N ratios (from 10.5 to 13) indicate a contribution of allochthonous inputs to the organic matter in the sediment (Meyers and Lallier-vergés, 1999). The recent increase in organic sedimentation in Lake Brévent seems to be less dependent on primary production than in Lakes Abbaye, Remoray, and Narlay, and probably results from a simultaneous increase in autochthonous production and inputs of allochthonous organic particles. Phase I of Lake Brévent is characterized by low organic sedimentation, low primary production, and an almost complete absence of coprophilous ascospore. On the contrary, during phase III, the accumulation rate of coprophilous ascospores as well as Corr and Chla AR increases significantly. These first changes in the trophic state of Lake Brévent could be linked to the beginning of pastoralism on the alpine watershed during the medieval era (Schmidt et al., 2002). The significant increase in organic sedimentation during the most recent phase (phase IV) is not followed by a significant increase in the flux of coprophilous ascospores compared to phase III. The flux remains nevertheless at the highest levels. Belle et al. (2016b) showed a significant increase in Rumex alpinus coverage over the watershed, probably linked to an increase in soil nutrient concentrations. These changes could be the result of alterations in the biological functioning of alpine soils as a consequence of climate warming, which could lead to an increase in nutrient availability (Chersich et al., 2015). They may also be the result of a

new intensification of agro-pastoral practices on the watershed. Nutrient inputs (Hausmann et al., 2002) and watershed soil erosion (Arnaud et al., 2016) linked to this intensification may therefore be partly responsible for the recent and unprecedented increase in organic sedimentation.

Impact of eutrophication on the origin of the carbon fueling lake food webs

Benthic food webs sustained by methane-derived carbon. $\delta^{13}C_{\mbox{\it Chironomus}}$ and $\delta^{13}C_{\mbox{\it Sergentia}}$ of Lake Abbaye, Remoray, Narlay, and Brévent sediment records do not differ significantly between the different phases of their respective trophic history. However, in the three Jura lakes (Abbaye, Remoray, and Narlay), the isotopic signatures of chironomid head-capsules decrease slightly during the most recent phases that correspond to the accelerated eutrophication stage. This decrease could be explained by an increase in the relative contribution of C-CH₄ to the chironomid diet via MOBs consumption (van Hardenbroek et al., 2010), as MOBs are highly depleted in ¹³C (Jedrysek, 2005; Templeton et al., 2006) compared to the carbon fixed by phototrophic organisms (Vuorio et al., 2006; Wang et al., 2013). The high accumulation rates of easily degradable autochthonous OM that characterized these most recent phases promote the establishment of favorable conditions for methane production in the sediments (Beaulieu et al., 2019; Duc et al., 2010; Naeher et al., 2014; Praetzel et al., 2020). Moreover, the genera Sergentia and Chironomus are known to construct tubes allowing them to stimulate the growth of MOBs by circulating oxygen-rich water (Jones and Grey, 2011), and previous studies have highlighted that the Chironomini tribe has a high affinity for MOBs as a food source (van Hardenbroek et al., 2012). Finally, Chironomus and Sergentia head-capsules were strongly ¹³C-depleted relative to the bulk organic matter for most samples of these three lakes, especially in Remoral sediment records. The $\delta^{13}C$ of bulk organic matter is generally considered an integrated estimate of the $\delta^{13}C$ of autochthonous and allochthonous organic matter available to benthic consumers at the time of its sedimentation, assuming little influence from diagenetic processes (Meyers, 1994). Large differences between the 813C of bulk OM and subfossil remains of Chironomidae can then be interpreted as a significant contribution of MOBs to their diet (Belle et al., 2014; van Hardenbroek et al., 2013), and indirectly as a high methanogenic activity within the sedimentary compartment. At Brévent, these differences were less pronounced, suggesting a less sustained transfer of C-CH₄ to benthic consumers than in the three Jura lakes. Moreover, the near disappearance during the 20th century of these chironomid head capsules in the sediment records of the three lakes of Remoray, Narlay, and Brévent indicates a significant degradation of physico-chemical conditions in the benthic compartment (Belle et al., 2016b) that prevented the benthic metazoan fauna populations to grow and thrive. The recent increase in primary production and organic sedimentation is most likely responsible for an intensification of heterotrophic processes in benthic compartment of these stratified lakes, leading to the appearance and development of anoxia in the hypolimnion (Foley et al., 2012; Matzinger et al., 2010), which in turn may promote the methanogenesis (Beaulieu et al., 2019; Rudd and Hamilton, 1978).

Combining studied remains to assess the trend in $\delta^{13}C$ of pelagic consumers. In the four lakes, $\delta^{13}C_{ephippia}$ decreases significantly during the recent period, which is also characterized by an increase in organic accumulation rates, combined, in the case of Lake Abbaye, Remoray, and Narlay, with a rise in phytoplanktonic pigments accumulation rates and a decrease in the C/N ratio. Indeed, the most ¹³C-depleted isotopic signatures of Daphnia

resting eggs were always found in the uppermost samples corresponding to the accelerated eutrophication period. $\delta^{13}C_{ephippia}$ reaches values around or below -40 % for the Lakes Abbaye, Remoray, and Narlay, while $\delta^{13}C_{ephippia}$ values from Lake Brévent are less ¹³C-depleted with minimum values of -35.9 %.

These decreasing trends in ephippia δ^{13} C suggest changes in the δ^{13} C of carbon fueling pelagic food webs over time. However, shifts in the seasonal timing of ephippia production by Daphnia may induce variations in $\delta^{13}C_{ephippia}$ trends in the sediment records independently of changes in carbon sources and transfer pathway (Essert et al., 2023). Indeed, $\delta^{13}C$ of ephippia reflects the $\delta^{13}C$ of Daphnia at the time of ephippia production (Perga, 2011; Schilder et al., 2015b). However, ephippia are known to be usually produced by Daphnia within a relatively short time window (Caceres, 1998; Conde-Porcuna et al., 2014; Morlock et al., 2017; Perga, 2011), and δ^{13} C values of *Daphnia* can vary considerably over the annual cycle of lakes due to seasonal variations in the availability of the different food sources under the effect of stratification and turnover mechanisms (Essert et al., 2022a, 2023). Consequently, the information provided by $\delta^{13}C_{ephippia}$ mainly depends on the timing of ephippia production with respect to the timing of lake autumnal turnover. On the other hand, $\delta^{13}C$ of Daphnia claws and Bosmina head shields reflect a time-averaged image of source population δ^{13} C of pelagic consumers (Davidson et al., 2007; Perga, 2010). In our study, the decreasing trend in $\delta^{13}C_{ephippia}$ was also evidenced for $\delta^{13}C$ of other pelagic consumer remains (Daphnia claws and Bosmina head shields). This finding then supports that the decrease in $\delta^{13}C_{ephippia}$ reflects changes in the $\delta^{13}C$ of carbon fueling pelagic food webs over time rather than changes in the seasonal timing of ephippia production by Daph*nia* (Essert et al., 2023). Moreover, the $\delta^{13}C_{ephippia}$ were always well below the $\delta^{13}C_{claws}$ and $\delta^{13}C_{Bosmina h-s}$ for the four lakes. This suggests that ephippia are mostly produced by Daphnia during periods of high availability of ¹³C-depleted carbon sources relative to the rest of the annual cycle (Essert et al., 2022a, 2023). Ephippia retrieved from the sediment records of the four lakes are then most likely produced by Daphnia after autumnal turnover and during winter, when the water column is no longer stratified and C-CH₄ particularly available for pelagic consumers (Essert et al., 2022a; Taipale et al., 2008, 2009). Indeed, the reduction in photoperiod and the probable decrease in availability of other food sources for pelagic consumers during this period is likely to triggers ephippia production by Daphnia (Essert et al., 2023; Kleiven et al., 1992; Stross and Hill, 1965). On the contrary, the higher δ^{13} C values of *Daphnia* claws and *Bosmina* head shields rather reflect an annual average of pelagic carbon transfers (Davidson et al., 2007; Perga, 2010).

Accelerated eutrophication leads changes in the carbon sources fueling pelagic food web. According to the existing literature, three main carbon sources are generally recognized for *Daphnia* in lakes (Perga et al., 2008; Taipale et al., 2007): (i) phytoplankton, with an isotopic signature ranging from -25 to -40 ‰, depending on the origin of the carbon used and the intensity of isotopic fractionation during photosynthesis (de Kluijver et al., 2014; France, 1995; Masclaux et al., 2013; Peterson and Fry, 1987; Vuorio et al., 2006; Wang et al., 2013), (ii) bacteria and heterotrophic microorganisms, including MOBs for which δ^{13} C is between -90 ‰ and -60 ‰ (Jedrysek, 2005; Templeton et al., 2006), and (iii) allochthonous detritus with δ^{13} C between -25 and -29 ‰ (France, 1995; Grey et al., 2001; O'Leary, 1988; Peterson and Fry, 1987). The recent ¹³C-depletion of *Daphnia* ephippia may therefore have two explanations.

First, the recent decrease in $\delta^{13}C_{ephippia}$ could reflect a decrease in phytoplankton $\delta^{13}C$ as a result of the increasing use of respiration-derived CO, for photosynthesis, as this carbon is particularly depleted in ¹³C compared to other dissolved carbon sources available for photosynthesis (Fry, 2006; Parker et al., 2010). The sharp increase in organic accumulation in the sediment that characterized the recent phase of accelerated eutrophication certainly led to an intensification of heterotrophic activities in the hypolimnion (Foley et al., 2012). This assumption is supported by the disappearance of the benthic metazoan fauna, suggesting the prevalence of harsh oxygen conditions due to increased respiration by heterotrophic organisms. This probably led to the increased availability of respiration-derived CO₂ for photosynthesis (Morales-Williams et al., 2021; Xiao et al., 2020).

The second hypothesis is that the decrease in $\delta^{13}C_{ephippia}$ may be the result of changes in the relative contribution of the three components described above to the Daphnia diet. Indeed, most of the $\delta^{13}C_{ephippia}$ in recent samples were in the lower range or outside the range of phytoplankton δ^{13} C according to the literature. This recent decline in $\delta^{13}C_{ephippia}$ could therefore also be the result at least in part of increased C-CH₄ transfers into pelagic food webs, as MOBs are highly depleted in 13C (Jedrysek, 2005; Templeton et al., 2006) compared to the carbon fixed by phototrophic organisms (Vuorio et al., 2006; Wang et al., 2013). This hypothesis is strengthened by the observed disparity between $\delta^{13}C$ of ephippia and the other pelagic consumers remains, suggesting that ephippia are produced by Daphnia during periods of high availability of ¹³C-depleted carbon sources, likely after autumnal turnover and during winter when C-CH₄ is particularly available for pelagic consumers. Many studies have highlighted that CH₄ production in lake sediments is promoted by (i) a strong organic sedimentation rate combined with low C/N ratio of lake sediments (Duc et al., 2010; Praetzel et al., 2020), (ii) high primary production (Beaulieu et al., 2019; Essert et al., 2022a; Kortelainen et al., 2000), and (iii) oxygen depletion (Beaulieu et al., 2019; Rudd and Hamilton, 1978). The recent changes in trophic states and associated physico-chemical conditions in the hypolimnion of the four studied lakes, as evidenced by the shifts in both the quantity and quality of organic matter and the disappearance of chironomids at depth in three of the four lakes, are likely to have established a favorable environment for the initiation and/or enhancement of methanogenesis. This, in turn, may have led to an increased assimilation of C-CH₄ into the pelagic food webs.

Alternatively, some authors suggest that these recent decreases in isotopic signature in paleolimnological studies may be due to the Suess effect (Verburg, 2007). Indeed, the δ^{13} C of atmospheric CO₂ has decreased over the last 200 years due to fossil fuels burning, known as the Suess effect. This may have contributed to the recent decrease in δ^{13} C values of primary producers, and consequently in the δ^{13} C baseline of food webs. However, according to the literature, the decrease in atmospheric CO₂ signature caused by the Suess effect is only 1.5 ‰ since 1770 (Körtzinger et al., 2003), which is lower than the observed decreases in $\delta^{13}C_{ephippia}$ in the four lakes studied during this period (ranging from 3 to $10 \ \%$ depending on lakes). Although the Suess effect may have played a role in the recent decline of consumer δ^{13} C, it alone cannot fully explain this phenomenon. A significant portion of the decrease in δ^{13} C is likely a result of changes in carbon sources and transfer pathways due to eutrophication, specifically for the lakes of Remoray and Narlay, which have experienced the most significant declines of 8‰-10‰ over this period, indicating a shift in the carbon cycle.

In summary, the recent $\delta^{13}C_{ephippia}$ depletion may be due to an increase in the transfer of different ¹³C-depleted carbon sources linked to the development of heterotrophic activities: either carbon from respiration, methane-derived carbon, or both, under the influence of accelerated eutrophication, as already suggested by Frossard et al. (2014), Schilder et al. (2017) and Rinta et al. (2016). These results also indirectly suggest that greenhouse gas

emissions into atmosphere, especially methane, a significant contributor to global warming, may have significantly increased during the last century in response to accelerated eutrophication.

Conclusion

Through the use of a novel approach, which combines the study of sediment geochemistry with the carbon isotopic signature of different consumer subfossil remains, we have been able to demonstrate a shift in carbon sources and transfer pathways in lake food webs in response to anthropogenic disturbances. The recent accelerated eutrophication of the water, linked to the intensification of human activities in the watersheds, seems to be the cause of the shift in the carbon cycle in these lakes. Indeed, the isotopic signature of pelagic consumers has decreased significantly during the last century, reflecting an increasing use of ¹³C-depleted carbon sources such as methanogenic and/or respiration-derived carbon in pelagic food webs during the recent accelerated eutrophication of these systems. These results suggest an increase in the production and availability of ¹³C-depleted carbon sources, resulting from the intensification of heterotrophic and methanotrophic processes in response to accelerated eutrophication. More broadly, these results indirectly suggest that greenhouse gas emissions from these lakes into the atmosphere, especially methane, a significant contributor to global warming, may have increased during the last century. In contrast to pelagic consumers, temporal variations in carbon isotopic signatures were not very pronounced for the benthic consumer remains. Nevertheless, the profundal chironomid community disappeared in three of the four lakes studied during the 20th century due to the degradation of oxygen conditions in the hypolimnion. In this context of loss of the benthic fauna under ongoing global changes, the response of pelagic consumers to the disappearance of benthic consumers in terms of methanogenic carbon transfers seems important to consider. Indeed, their disappearance in the benthic compartment can potentially affect the flux and availability of methane in the pelagic compartment for these systems. Unfortunately, the sampling resolution implemented in this study does not allow us to assess the impact of the recent disappearance of methanogenic carbon transfers in the benthic compartment on pelagic carbon transfers. These interactions between benthic and pelagic food webs should therefore be considered in future studies.

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Supplemental material

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