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Responses of Benthic Carbon Flows in Scandinavian Lakes to Atmospheric Sulphur and Nitrogen Depositions: Potential Influence of Nutrient Limitation

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Keywords: acidification | carbon stable isotope | Chironomidae | lake food webs | paleolimnology

ABSTRACT

1. Paleolimnological reconstructions of past changes in carbon stable isotopic composition of chironomid head capsules ($\delta^{13}C_{HC}$) were used to quantify the impacts of atmospheric sulphur and nitrogen depositions on carbon processing at the base of lake food webs.
2. We hypothesised that the impacts of atmospheric nitrogen deposition on carbon flows through food webs are influenced by nutrient limitation, as increasing nitrogen inputs are expected to drive higher primary producer biomass in nitrogen-limited lakes, thus increasing its availability for benthic consumers.
3. The $\delta^{13}C_{HC}$ values showed significant changes before/after 1960, corresponding to the time of early impacts of atmospheric deposition on water chemistry and biological communities in Scandinavian lakes. These results suggest that atmospheric deposition was a key driver of change in carbon flows through benthic food webs. Furthermore, our study showed that lakes responded differently to atmospheric deposition and that differences in $\delta^{13}C_{HC}$ values were largely explained by a combination of nutrient limitation, latitude and water transparency.
4. Overall, results support the hypothesis that the response of carbon flows through benthic food webs to changes in atmospheric nitrogen deposition differs partly depending on lake nutrient limitation. However, the observed strong collinearity between the explanatory variables complicates the interpretation of the relationships between the temporal changes in benthic carbon flows in response to atmospheric deposition and nutrient limitation.

1 | Introduction

Increased sulphur and nitrogen depositions due to anthropogenic atmospheric emissions triggered acidification (Mylona 1996) and affected the nutrient balance of many European and North American ecosystems during the last century (Galloway and Cowling 2002). Using a dataset of Swedish lakes, which included some as in the present study, Belle and Johnson (2024) reported large taxonomic shifts in chironomid assemblages observed at *ca*. 1960 corresponding to measures of acidification in Scandinavia. Yet, the concurrent, and potentially cumulative, impacts of sulphur and nitrogen depositions on lakes complicate the interpretation of the causal mechanisms of lake responses to atmospheric deposition, in particular, for

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the carbon flows through food webs. On the one hand, acidified freshwaters show decreases in pH and alkalinity (Fölster et al. 2007) and subsequently the gradual loss of acid-sensitive species and increase of acid-tolerant species (Schindler, Kasian, and Hesslein 1989). In lakes, acidification-induced changes in algal communities likely affect energy flows through food webs by shifting basal resources from high-quality food (such as diatoms) to lower quality and less edible taxonomic groups (filamentous green algae; cyanobacteria; Turner et al. 1995). On the other hand, increases in phytoplankton biovolumes in nitrogen-limited northern hemisphere lakes have been attributed to atmospheric nitrogen deposition, even in areas only moderately impacted by nitrogen deposition (Bergström, Blomqvist, and Jansson 2005; Bergström and Jansson 2006). The effects of atmospheric deposition on algal community composition and energy flows through lake food webs therefore likely depend on deposition intensity and on whether nitrogen or phosphorus limits algal growth. Whether biological changes driven by atmospheric deposition ultimately affect energy flows in lake food webs has, however, been largely overlooked.

Lake sediments constitute a natural archive allowing us to circumvent the lack of monitoring data (Walker 2001) and disentangle the effects of past environmental changes on freshwaters. Specifically, head capsules of Chironomidae larvae (Arthropoda; Diptera; Nematocera), nonbiting midges with larvae growing in superficial lake sediments, are chemically and morphologically well-preserved in sediments (Verbruggen et al. 2010). Stable isotope analysis of these head capsules, mainly of carbon ($\delta^{13}C$), provides quantitative and qualitative insights into carbon flows through lake food webs (Belle et al. 2017). Since differences in carbon isotopic fractionation by primary producers (i.e., aquatic vs. terrestrial producers) and bacteria (e.g., methanotrophic bacteria) lead to basal food resources that differ in their $\delta^{13}C$ values (Whiticar 1999; Karlsson et al. 2003; Vuorio, Meili, and Sarvala 2006), carbon stable isotopic composition of chironomid head capsules ($\delta^{13}C_{HC}$) can be used to reconstruct long-term changes in food resources incorporated into larval biomass (Frossard et al. 2013; Belle et al. 2017). As sediment-dwelling chironomid larvae form an important trophic link between primary producers and fish (Wagner, Volkmann, and Dettinger-Klemm 2012), carbon stable isotope analysis of chironomid head capsules can elucidate the impacts of atmospheric deposition on carbon flows through lake food webs.

In this study, we used centennial trends in $\delta^{13}C_{HC}$ values to infer past changes in basal resources for chironomid biomass in Swedish lakes. We anticipated to (i) quantify the impacts of acidification and atmospheric nitrogen deposition on carbon processing at the base of lake food webs and (ii) identify the mechanisms that underlie its observed temporal changes. We hypothesised that the effects of atmospheric nitrogen deposition on energy flow through lake food webs depend on whether nitrogen or phosphorus limit algal growth, as increasing nitrogen inputs are expected to drive higher primary producer biomass in nitrogen-limited lakes, and less so in phosphorus-limited lakes. More specifically, we predicted that among-lake variations in the TN/TP mass ratio would be associated with different responses to atmospheric deposition in the chironomid isotopic compositions in our study lakes.

2 | Material and Methods

2.1 | Study Sites

Nine lakes were selected to cover gradients in anthropogenic deposition, trophic status, and nutrient limitation (Figure 1). Modelled data for the period 1880–2020 available from the European Monitoring and Evaluation Program (EMEP) showed a sharp increase in deposition from 1940 followed by increasing trends in sulphur and nitrogen deposition between *ca*. 1960 and 1980 (Figure 2A,B). The onset and maximal imprint of acidification was also characterised by low temporal variability in temperature and precipitation (Figure 2C,D). After 1980, sulphur and nitrogen depositions followed a steady decrease to background levels around 2000. However, the postacidification period was also marked by a period of substantial climate warming starting in the 1980s and increasing precipitation after the mid-1970s.

The study lakes are relatively small (0.49–5.05 ha), deep (maximum water depth range 17.5–29.2 m) and situated in forested catchments on the Fennoscandian shield, where bedrock is predominated by siliceous granitoids, resulting in low conductivity, slightly acidic waters with poor acid neutralisation capacities (Table 1). Lakes were selected to be relatively deep to limit the reliance of profundal benthic communities on benthic primary production. Late summer water quality data were retrieved from the Swedish

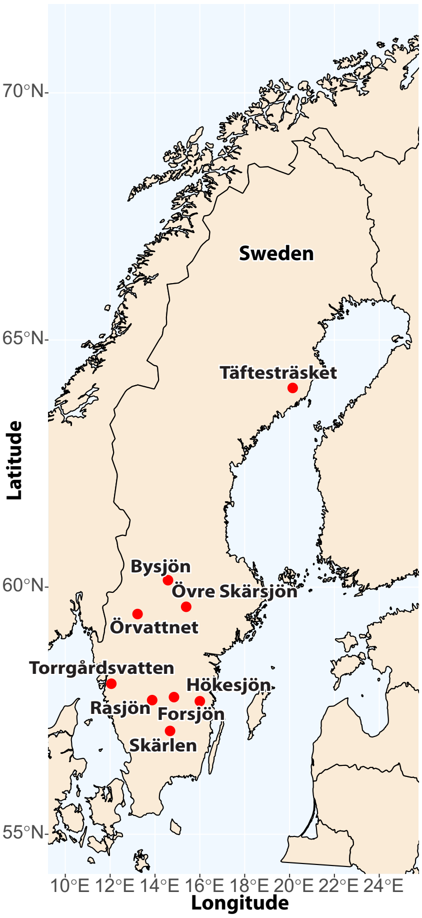

FIGURE 1 | Location map of the study region showing the sampling sites.

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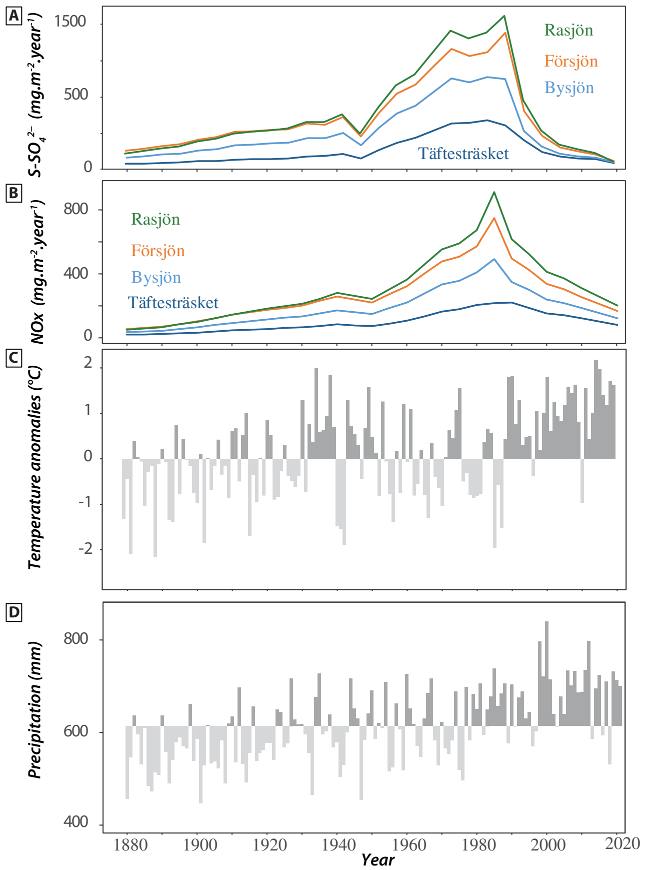

FIGURE 2 | Atmospheric sulphate sulphur (S-SO_4^{2-}; A) and oxidised nitrogen (NO*x*; B) deposition time series (1880–2020) in four of the study sites from the European Monitoring and Evaluation Program. (C) Mean annual precipitation in Sweden during the period 1860–2020. (D) Mean annual temperature in Sweden during the period 1860–2020. All data are shown as anomalies from the respective mean values over the entire time series (Sveriges Meteorologiska och Hydrologiska Institut; https://smhi.se).

National Monitoring Program database (https://miljodata.slu.se/mvm/): chlorophyll *a*, total organic carbon, oxygen and nutrients concentrations, as well as water conductivity, pH and Secchi depths (Table 1). The mass ratios between total nitrogen and total phosphorus concentrations (hereafter TN/TP ratios) were also calculated to reflect the degree of phytoplankton limitation, with TN/TP mass ratios >41 likely indicative of phosphorus-limited lakes and lower TN/TP ratios nitrogen-limited lakes (Bergström 2010). No local management actions (i.e., liming) have been taken to mitigate the effects of acidification (https://kalkdatabasen.lansstyrelsen.se), and it is thus reasonable to assume that all lakes follow a natural recovery after atmospheric deposition started to decline.

2.2 | Sediment Coring and Dating Techniques

In June–July 2020, surface sediment cores were retrieved from the deepest point of the lakes using a gravity corer (9 cm in diameter: UWITEC). All sediment cores were vertically split into two halves in the labortory, and subsamples of 1 cm thickness were collected and used for chronology. Sediment cores were dated by ^{210}Pb, ^{137}Cs and ^{241}Am at Liverpool University's Environmental Radioactivity Laboratory following Appleby et al. (1986) and Appleby and Oldfield (1992). Dates were calculated using the CRS model (see also Figure S1; Appleby and Oldfield 1978; Appleby 2002).

2.3 | Sediment and Chironomid Head Capsules Isotopic Analyses

One split half of each sediment core was continuously sectioned at 1-cm intervals, and sample numbers varied between 8 and 18 per core. The organic matter fraction of sediment samples was analysed for carbon stable isotopes ($\delta^{13}C_{OM}$) and used as an estimate of the isotopic baseline for chironomid larvae. Before analysis, potential carbonate was removed using small additions (50 μL) of HCl 1 M applied on 3 mg of dried and grounded sediments transferred in silver capsules. Carbonate-free sediment samples were then introduced in tin capsules and analysed for carbon stable isotopes using an Isotope Ratio Mass Spectrometer interfaced with an Elemental Analyser (EA-IRMS) at the Stable Isotope Laboratory, Swedish University of Agricultural Sciences. Results were expressed as the delta notation with Vienna Pee Dee Belemnite and atmospheric nitrogen as standards: $\delta^{13}C\ (‰) = [(R_{sample}/R_{standard}) - 1] \times 1000$; where $R = {}^{13}C/{}^{12}C$. Sample measurement replications from internal standards (wheat and maize flour) produced analytical errors (1σ) of ±0.15‰ for both $\delta^{13}C$ and $\delta^{15}N$ values ($n = 30$).

Carbon stable isotope analyses were performed on chironomid head capsules belonging to the fourth instar (following recommendations from Frossard et al. 2013) of morphotypes *Sergentia coracina*-type, *Chironomus plumosus*-type and *Chironomus anthracinus*-type identified using Brooks, Langdon, and Heiri (2007). These morphotypes typically inhabit the profundal zones of lakes, and they belong to the functional feeding group of the deposit feeders (Berg 1995). *Sergentia coracina*-type, *Chironomus plumosus*-type and *Chironomus anthracinus*-type also form an important link between basal resources and higher trophic levels in the food webs, thus being highly suited to unravel the effects of atmospheric deposition on the recycling of sedimentary organic matter and associated microbial communities. Finally, these taxa are usually found in high abundances in Swedish lakes and have large head capsules at the fourth instar, allowing us to obtain sufficient material for stable isotope analysis, and reliably sort them into morphotypes using a stereomicroscope. For the extraction of chironomid head capsules, sediment samples were deflocculated in NaOH (10%) solution, pretreated using washing with HCl (3.7%) solution and sieved through a 100-μm mesh size. The HCl rinse was used to remove potential carbonate contaminations and to allow for better comparisons with previously published paleolimnological studies. Head capsules were sorted under a stereomicroscope until a minimal mass of 30 μg was obtained. Carbon stable isotopic composition of chironomid head capsules ($\delta^{13}C_{HC}$) was analysed and expressed as the delta notation (see above). In this case, calculated analytical errors (1σ) were ±0.15‰ for analyses performed on low sample weights ($n = 14$). The potential shift in the isotopic baseline was taken into account by calculating the difference between the carbon stable isotopic composition of chironomid head capsules and

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TABLE 1 | Characteristics of the nine studied lakes.

Lake	CA	Depth	Area	Secchi	Chl	Cond25	pH	TOC	Tot_P	Tot_N	TN/TP
Bysjön (BYS)	38.9	20	5.05	3.4	2.7	3.4	6.7	9.3	7.7	250	32
Täftesträsket (TAF)	12.2	17.5	2.22	2.6	5.7	3	6.6	10.5	8.8	329	37
Örvattnet (OVR)	2.3	32	0.8	4.2	2	1.7	5.5	4.6	5.6	250	45
Övre Skärsjön (OSK)	8.77	32	1.7	2.1	2.3	2.2	5.8	10.2	6.6	321	49
Försjön (FOR)	10.37	21	1.56	4.1	3.3	4.9	6.8	10.4	8.8	482	55
Torrgårdsvatten (TOR)	57.3	29.2	0.49	2.4	0.9	4.7	5.5	1.8	4.5	271	60
Hökesjön (HOK)	40.32	22	0.51	6.7	2.9	4.61	6.9	5.1	3.6	261	72
Rasjön (RAS)	20.77	20	3.91	4.7	1.9	12.7	7.1	11.8	4	310	78
Skärlen (SKA)	13.14	28	3.18	8	2.7	4.1	6.6	4	4	450	113

Note: Abbreviations for variables include CA—catchment area (km^2); Depth—maximum lake depth (m); Area—lake area (km^2); Secchi—water transparency as Secchi disk depth (m); Chl—total chlorophyll concentration (mg/L); Cond25—specific conductivity at 25°C (μS/cm); TOC—total organic carbon concentration (mg/L); Tot_P—total phosphorus concentration (μg/L); Tot_N—total nitrogen concentration (μg/L); total nitrogen to total phosphorus mass ratio (TN/TP). Data are reported from https://miljodata.slu.se/mvm/. Lakes are ranked according to their TN/TP ratios.

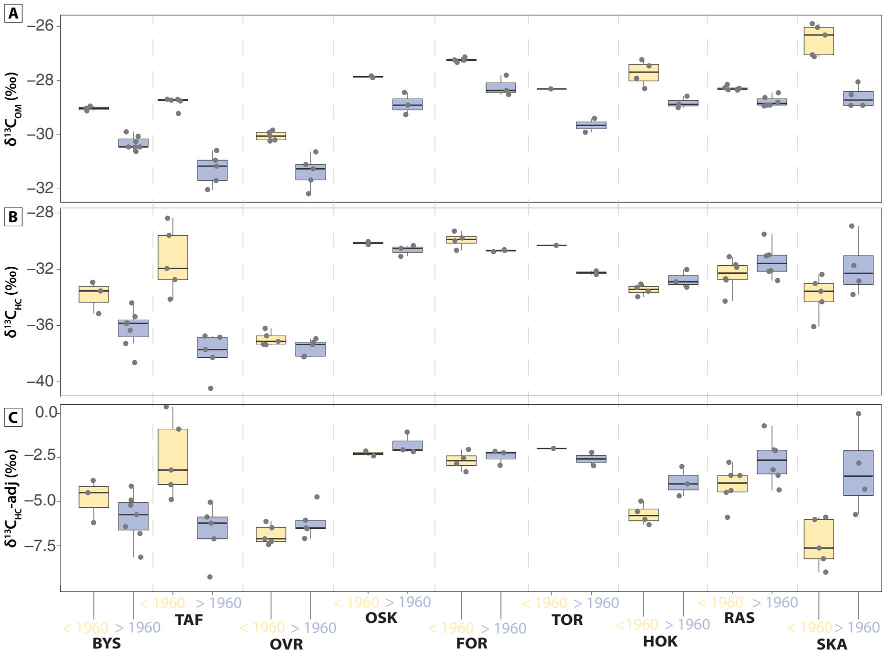

FIGURE 3 | Boxplots showing (A) carbon stable isotopic composition of organic matter ($\delta^{13}C_{OM}$; ‰), (B) carbon stable isotopic composition of chironomid head capsules ($\delta^{13}C_{HC}$; ‰), and (C) the difference between the carbon stable isotopic composition of chironomid head capsules and organic matter ($\delta^{13}C_{HC}$-adj $= \delta^{13}C_{HC} — \delta^{13}C_{OM}$) for the early 1900–1960 (in orange) and 1960–present-day (in dark blue) periods. Lakes are ranked according to their TN/TP ratios.

organic matter (i.e., $\delta^{13}C_{HC}$-adj $= \delta^{13}C_{HC} — \delta^{13}C_{OM}$). The values of $\delta^{13}C_{HC}$-adj have also been used as an indicator of selective feeding in previous studies (Belle et al. 2017).

2.4 | Data Analysis

Mean values of water chemistry data for the period covered by the first cm of each sediment core were calculated. To quantify the impacts of atmospheric deposition on carbon processing at the base of the lake food webs, time series of $\delta^{13}C_{HC}$ and $\delta^{13}C_{HC}$-adj values were divided into two periods: early 1900–1960 and 1960–present day. Differences between the average values of each period were calculated for each lake (e.g., [average $\delta^{13}C_{HC}$ values after 1960] − [average $\delta^{13}C_{HC}$ values before 1960] = Δ-$\delta^{13}C_{HC}$) and each variable (Δ-$\delta^{13}C_{HC}$ and Δ-$\delta^{13}C_{HC}$-adj).

Principal component (PC) analysis was applied to a standardised (mean of 0 and a standard deviation of 1) data set including lake water conditions (oxygen concentration, total nitrogen and phosphorus concentrations, TN/TP mass ratio), catchment and lake morphometric characteristics (catchment area, maximum water depth and latitude). The values of Δ-$\delta^{13}C_{HC}$ and Δ-$\delta^{13}C_{HC}$-adj were then added as passive variables to visually identify associations. Finally, statistical relationships between Δ-$\delta^{13}C_{HC}$ and Δ-$\delta^{13}C_{HC}$-adj values with the first PC axis were examined

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using linear regressions. We also tested for nonlinear relationships with GAM and since we did not find any, we continued with linear regression. All statistical analyses and plots were performed using the R 4.4.0 software (R Core Team 2024).

3 | Results

The $\delta^{13}C_{OM}$ values ranged from −32.2‰ to −25.9‰ and were consistently lower after 1960 (Figure 3A). Chironomid head capsules were found in sufficient numbers in 72 sediment samples (Table 2), and their $\delta^{13}C_{HC}$ values ranged from −40.5‰ to −28.4‰ (Figure 3B). Changes in $\delta^{13}C_{HC}$ values did not coincide with temporal changes in the morphotype used for isotopic analysis (Table 2 and Figure S2). Furthermore, $\delta^{13}C_{HC}$ values observed during the most recent period overlap only marginally with historical values (Figure 3B). The $\delta^{13}C_{HC}$-adj values, as the differences between $\delta^{13}C_{HC}$ and $\delta^{13}C_{OM}$ values, were almost consistently negative (ranging from −9.3‰ to 0.4‰) suggesting that $\delta^{13}C_{HC}$ values were generally lower than those of $\delta^{13}C_{OM}$ (Figure 3C). The Δ-$\delta^{13}C_{HC}$ values (e.g., [average $\delta^{13}C_{HC}$ values after 1960] − [average $\delta^{13}C_{HC}$ values before 1960] = Δ-$\delta^{13}C_{HC}$) ranged from −6.6‰ to 2‰ (Figure 4A) and were negative for six of nine of lakes (indicating lower $\delta^{13}C_{HC}$ values during the most recent period). Exceptions to these were Lakes Räsjön, Hökesjön, and Skärlen for which positive Δ-$\delta^{13}C_{HC}$ values were found (showing an increase in $\delta^{13}C_{HC}$ values after 1960). The Δ-$\delta^{13}C_{HC}$-adj values, as the differences between average $\delta^{13}C_{HC}$-adj values during [1960–present day] and [1900–1960] periods, ranged from −4.2‰ to 4.1‰ (Figure 4B). Negative Δ-$\delta^{13}C_{HC}$-adj values (such as in Lakes Bysjön and Täftesträsket) showed that the difference between $\delta^{13}C_{OM}$ and $\delta^{13}C_{HC}$ values increased during the most recent period, whereas positive Δ-$\delta^{13}C_{HC}$-adj values (see also Lakes Skärlen and Räsjön) showed a decrease in the difference between $\delta^{13}C_{HC}$ and $\delta^{13}C_{OM}$ during the 1960–present-day period.

TABLE 2 | Summary of the sample number, and morphotype analysed for carbon stable isotopic composition of subfossil chironomids during the early 1900–1960 (in orange) and 1960–present day (in dark blue) periods. Lakes are ranked according to their TN/TP ratios.

Lake	Period	Morphotype	*n*
Bysjön (BYS)	> 1960	*Chironomus anthracinus*-type	7
Bysjön (BYS)	< 1960	*Chironomus anthracinus*-type	2
Bysjön (BYS)	< 1960	*Sergentia coracina*-type	1
Täftesträsket (TAF)	> 1960	*Sergentia coracina*-type	1
Täftesträsket (TAF)	> 1960	*Chironomus anthracinus*-type	4
Täftesträsket (TAF)	< 1960	*Sergentia coracina*-type	1
Täftesträsket (TAF)	< 1960	*Chironomus anthracinus*-type	4
Örvattnet (OVR)	> 1960	*Sergentia coracina*-type	1
Örvattnet (OVR)	> 1960	*Chironomus anthracinus*-type	4
Örvattnet (OVR)	< 1960	*Chironomus anthracinus*-type	5
Övre Skärsjön (OSK)	> 1960	*Sergentia coracina*-type	3
Övre Skärsjön (OSK)	< 1960	*Sergentia coracina*-type	2
Försjön (FOR)	> 1960	*Sergentia coracina*-type	4
Försjön (FOR)	< 1960	*Sergentia coracina*-type	4
Torrgårdsvatten (TOR)	> 1960	*Sergentia coracina*-type	3
Torrgårdsvatten (TOR)	< 1960	*Sergentia coracina*-type	1
Hökesjön (HOK)	> 1960	*Sergentia coracina*-type	2
Hökesjön (HOK)	< 1960	*Sergentia coracina*-type	1
Rasjön (RAS)	> 1960	*Chironomus plumosus*-type	6
Rasjön (RAS)	< 1960	*Chironomus plumosus*-type	6
Rasjön (RAS)	< 1960	*Sergentia coracina*-type	1
Skärlen (SKA)	> 1960	*Sergentia coracina*-type	4
Skärlen (SKA)	< 1960	*Sergentia coracina*-type	5

Note: Lakes are ranked according to their TN/TP ratios.

The first two PC axes accounted for 45.5% and 24.5% of the total variance, respectively (Figure 5A). PC1 axis explained TN/TP ratios, latitude and Secchi depth (Figure 5B), with relative contributions of 22.4%, 20.8% and 17.3%, respectively. Negative values on the PC1 axis represented lakes at lower latitudes with high TN/TP ratios and high Secchi depth. The PC2 axis largely reflected the catchment size, maximum water depth and bottom oxygen concentration with relative contributions of 41.7%, 15.6% and 13.6%, respectively. The Δ-$\delta^{13}C_{HC}$ and Δ-$\delta^{13}C_{HC}$-adj variables were strongly correlated with PC1 axis (Figure 5B), and linear regressions showed that PC1 explained 80% and 92% of the overall variability of Δ-$\delta^{13}C_{HC}$ and Δ-$\delta^{13}C_{HC}$-adj, respectively. PC1 showed a negative relationship with Δ-$\delta^{13}C_{HC}$

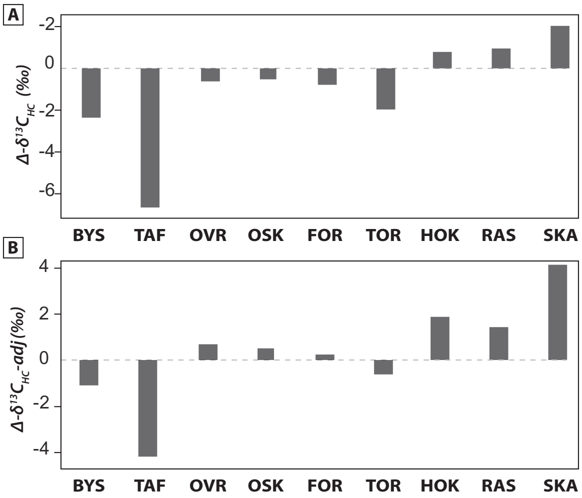

FIGURE 4 | (A) Barplots showing the differences in carbon stable isotopic composition of chironomid head capsules after/before 1960 periods (Δ-$\delta^{13}C_{HC}$ = [average $\delta^{13}C_{HC\ values}$ after 1960] − [average $\delta^{13}C_{HC}$ values before 1960]; ‰). (B) Barplots showing the differences in carbon stable isotopic composition of chironomid head capsules and sedimentary organic matter before/after 1960 periods (with Δ-$\delta^{13}C_{HC}$-adj = [average $\delta^{13}C_{HC}$-adj $_{values}$ after 1960] − [average $\delta^{13}C_{HC}$-adj values before 1960]; ‰). Lakes are ranked according to their TN/TP ratios.

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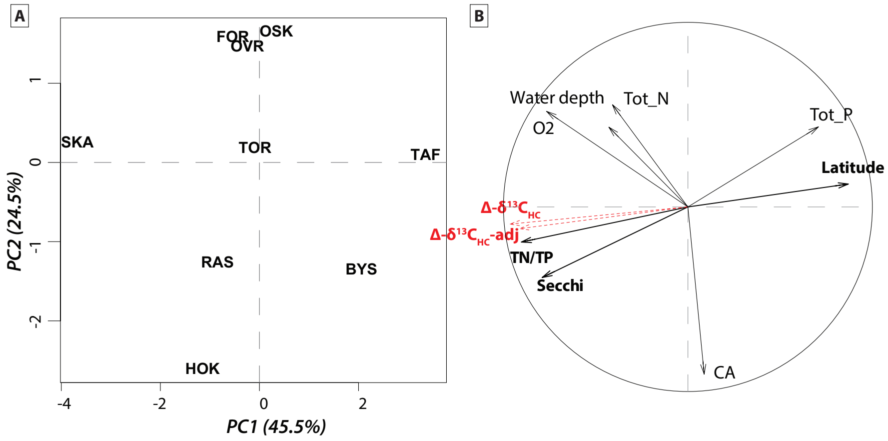

FIGURE 5 | (A) Principal component analysis factorial map, with the first two axes (PC1 vs. PC2) performed on a data set including lake water conditions (oxygen concentration, total nitrogen and phosphorus concentrations, TN/TP mass ratio), catchment and lake morphometric characteristics (catchment area, maximum water depth, latitude). Lake names are abbreviated as in Table 1. (B) Correlation circle representing variable contributions to the first two PC axes, in which Δ-$\delta^{13}C_{HC}$ and Δ-$\delta^{13}C_{HC}$-adj were added as passive variables.

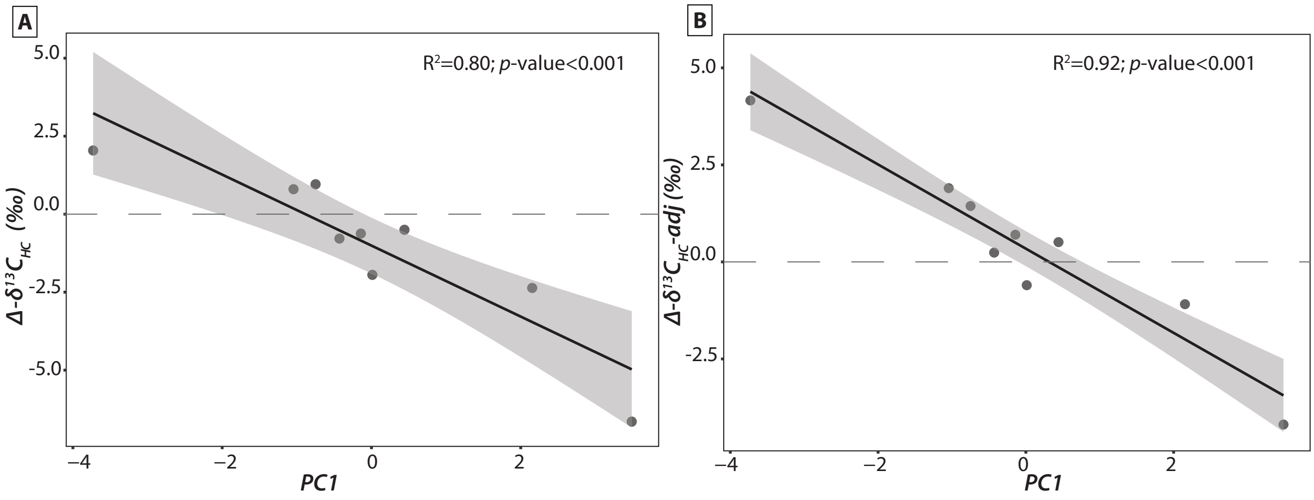

FIGURE 6 | Relationship tested using linear regressions between scores of the first PC axis and (A) differences in carbon stable isotopic composition of chironomid head capsules after/before 1960 periods (Δ-$\delta^{13}C_{HC}$ = [average $\delta^{13}C_{HC\ values}$ after 1960] – [average $\delta^{13}C_{HC}$ values before 1960]; ‰), and (B) differences in carbon stable isotopic composition of chironomid head capsules and sedimentary organic matter before/after 1960 periods (with Δ-$\delta^{13}C_{HC}$-adj = [average $\delta^{13}C_{HC}$-adj $_{values}$ after 1960] – [average $\delta^{13}C_{HC}$-adj values before 1960]; ‰). All relationships are statistically significant (p value < 0.001).

values (p value < 0.001; Figure 6A), suggesting that $\delta^{13}C_{HC}$ values increased after 1960 in low latitude lakes with high TN/TP ratios and high-water transparency. Conversely, higher latitude lakes with low TN/TP ratios and low water transparency showed a decrease in their $\delta^{13}C_{HC}$ values after 1960 (Figure 6A). Furthermore, PC1 scores showed a negative trend with Δ-$\delta^{13}C_{HC}$-adj values (p value < 0.001; Figure 6B), indicating that the difference between $\delta^{13}C_{HC}$ and $\delta^{13}C_{OM}$ values increased after 1960 in higher latitude lakes with low TN/TP ratios and low water transparency.

4 | Discussion

The present study was the first to use isotope-based paleolimnological reconstructions to quantify the impacts of atmospheric deposition on carbon processing by chironomid larvae at the base of lake food webs. Overall, our study supports the hypothesis that the response of carbon flows through benthic food webs to changes in atmospheric nitrogen deposition differs depending on whether lake phytoplankton is nitrogen- or phosphorus-limited. Temporal trends in $\delta^{13}C_{HC}$ values in our nine lakes showed significant changes before/after 1960, a year corresponding to early impacts of atmospheric deposition on water chemistry and biological communities in Scandinavian lakes (Schindler, Kasian, and Hesslein 1989; Cumming et al. 1992; Moiseenko 2005; Belle and Johnson 2024). Assuming only minor effects of temperature on $\delta^{13}C_{HC}$ values (i.e., trophic fractionation, Belle et al. 2020); these findings indicate that atmospheric deposition was likely a key driver of change in carbon flows in benthic food webs. Results also showed no evidence of recovery (i.e., shift back to historical values) of benthic carbon flows in response to decreasing sulphur and nitrogen depositions. This is because our $\delta^{13}C_{HC}$ values for the most

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recent period (i.e., after 1980) showed no large overlap with those found for the preacidification period, likely due to the growing influence of accelerating climatic changes after 1980 on aquatic communities of Scandinavian lakes (Johnson and Angeler 2010; Angeler and Johnson 2012; Svitok et al. 2021; Belle and Johnson 2024). Finally, our study clearly shows lake type-specific differences in $\delta^{13}C_{HC}$ values which are largely explained by a combination of variables, including nutrient limitation, latitude and water transparency. However, the strong collinearity among explanatory variables complicates the identification of the causal relationships between the observed temporal changes in $\delta^{13}C_{HC}$ values as a response to atmospheric deposition and nutrient limitation.

Our finding showing that $\delta^{13}C_{HC}$ values (ranging from −40.5‰ to −28.4‰) were lower than those of $\delta^{13}C_{OM}$ (ranging from −32.2‰ to −25.9‰) confirms the selective feeding habit of profundal chironomid larvae (Johnson 1985; Ravinet et al. 2010; Belle et al. 2017) and preferential incorporation of a lighter carbon source into their biomass. As pelagic algae have $\delta^{13}C$ values ranging from −38‰ to −28‰ (Vuorio, Meili, and Sarvala 2006) and are usually lighter (i.e., more negative $\delta^{13}C$ values) than other basal resources (e.g., terrestrial organic matter, Jones et al. 1999), results show that profundal benthic communities of the moderately nutrient-rich studied lakes are predominantly sustained by primary pelagic production. Nevertheless, the lake type-specific differences in $\delta^{13}C_{HC}$ values indicate that the limiting factor for primary productivity (i.e., nitrogen vs. phosphorus) arguably induces specific responses of lake food webs to atmospheric deposition. In our study, phosphorus-limited lakes (namely Lakes Hökesjön, Skärlen and Räsjön) showed that $\delta^{13}C_{HC}$ values did not follow the same trend as those of organic matter after the atmospheric deposition peak. Increasing $\delta^{13}C_{HC}$ values were reported in phosphorus-limited lakes located in areas heavily impacted by atmospheric deposition. After correcting for changes in isotopic baselines (i.e., $\delta^{13}C_{HC}\text{-adj} = \delta^{13}C_{HC} — \delta^{13}C_{OM}$), results still suggest preferential assimilation of the heavier carbon after 1960 in these lakes as $\delta^{13}C_{HC}$-adj values were less negative compared to the 1900–1960 period. Nitrogen deposition was not expected to affect algal growth in phosphorus-limited lakes (Bergström, Blomqvist, and Jansson 2005), but earlier works reported that algal communities of many lakes located at these latitudes shifted from diatom to filamentous green algae-dominated communities (Turner et al. 1995). We, therefore, conjecture that such algal shifts led to fewer edible species (or high-quality algae) available for benthic consumers in phosphorus-limited lakes, potentially increasing the contribution of terrestrial organic matter to the chironomid diet and leading to the observed increase in $\delta^{13}C_{HC}$ values.

In contrast, in lakes where nitrogen is more likely the limiting factor (e.g., Lakes Täftesträsket and Bysjön) decreasing $\delta^{13}C_{HC}$ values were found and changes in both $\delta^{13}C$ of organic matter and chironomids were positively correlated. Earlier studies have found increasing phytoplankton development in nitrogen-limited lakes in response to nitrogen deposition, even in areas moderately impacted by atmospheric deposition (Bergström, Blomqvist, and Jansson 2005; Bergström and Jansson 2006). Our results show the food web effects of increased algal production in response to nitrogen deposition on these lakes, driven by a higher use of pelagic algae by benthic consumers and consequently a lower contribution of the heavier carbon sources used before 1960 (most likely terrestrial organic matter with $\delta^{13}C$ values typically higher than those of phytoplankton). Furthermore, under specific conditions increasing phytoplankton development may also drive deep-water oxygen depletion and enhance CH_4 cycling. The very low $\delta^{13}C_{HC}$ values in the Lake Täftesträsket sediment core (i.e., $\delta^{13}C_{HC}$ values down to −40.5‰) indeed suggest a substantial contribution of methane-derived carbon (through reliance on CH_4-oxidising bacteria; Jones et al. 2008) to chironomid biomass. In anoxic lakes, biogenic CH_4 exhibits very low $\delta^{13}C$ values (from −110‰ to −40‰; Thottathil and Prairie 2021), and CH_4-oxidising bacteria can be used as a source of energy by consumers (Bastviken et al. 2003), thus leading to chironomids showing very low $\delta^{13}C$ values (Grey 2016). Only a few studies have, however, suggested that acidification and development of deep-water hypoxic/anoxic conditions may coincide (Henrikson, Olofsson, and Oscarson 1982), and the mechanistic relationship between pH and CH_4 production instead suggests that low pH inhibits CH_4 production (Ye et al. 2012; Bertolet et al. 2020). Combined analysis of sedimentary ancient DNA of CH_4-oxidising bacteria and stable isotopes of chironomid head capsules (see also Belle and Parent 2019; Belle et al. 2024) could help to unravel the underlying mechanisms of CH_4 cycling as a potential biological response to atmospheric deposition in nitrogen-limited lakes.

Author Contributions

S.B.: conceptualisation, conducting the research, data analysis, and preparation of figures and tables. S.B., J.F., W.G., R.K.J.: interpreting the data and writing.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data will be made available upon reasonable request.

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