

The Sources of Carbon and Nitrogen in Mountain Lakes and the Role of Human Activity in Their Modification Determined by Tracking Stable Isotope Composition

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Abstract We studied the isotopic composition of organic matter in the sediments of eight mountain lakes located in the Tatra Mountains (Western Carpathians, Poland). The sediments of the lakes were fine and coarse detritus gyttja, mud, and sand. The total organic carbon content varied from 0.5 to 53 %. The C/N ratio indicated that in-lake primary production is the major source of the organic matter in the lakes located above the treeline, whereas terrestrial plant fragments are the major organic compounds in the sediments of dystrophic forest lakes. We also found that a clear trend of isotopic curves toward lower values of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ (both ~ 3 ‰) began in the 1960s. This trend is a sign of the deposition of greater amounts of NO_x from the combustion of fossil fuels, mainly by vehicle engines. The combustion of fossil fuels in electric plants and other factories had a smaller influence on the isotopic composition. This trend has been weaker since the 1990s. Animal and human wastes from pastures and tourism had a surprisingly minor effect on lake environments. These data are contrary to previous data regarding lake biota and suggest the high sensitivity of living organisms to organic pollution.

Keywords Stable isotopes · Acidification · Eutrophication · Alpine lakes · Fossil fuel combustion

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1 Introduction

Remote mountains and arctic lakes have been influenced by human activities for several decades. In some cases, the influence has been direct, i.e., waste input (Revenga et al. 2012), fish stocking (Brancelj et al. 2000; Pister 2001), deforestation of catchment areas (Schmidt et al. 2002; Zhang et al. 2010), etc. However, most of these lakes were impacted by regional and global factors: climate change and the deposition of pollution from the atmosphere, mainly products of the combustion of fossil fuels. The deposition of nitrogen and sulfur oxides, which leads to the acidification of lakes, has been described in several mountainous regions of the Arctic, America, and Europe (Brett 1989; Paterson 1994; Sienkiewicz et al. 2006). However, in many studies, the link between fossil fuel combustion and the acidification of lakes was based only on the time coincidence of both processes. Conversely, many lakes located in regions with significant deposition of nitrates and sulfur oxides showed only traces of acidification.

The C/N ratio, $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$ of organic matter in sediments are the result of several complex processes, including biosynthesis in the photic zone, organic matter degradation and bacterial growth in the water column and in the sediment, and the input from allochthonous sources (Brenner et al. 1999). The values of carbon isotopes and the C/N ratios depend on the source of carbon assimilated and the proportion of macrophytes to phytoplankton in the aquatic environment. The $\delta^{13}\text{C}$ values of aquatic plants and plankton are usually

between -30 and -25 ‰; however, the full range is -50 to -10 ‰. Similarly, global average value of $\delta^{13}\text{C}$ for terrestrial C3 plants was estimated to be -28.5 ‰ with a range from -20 to -37 ‰ (Kohn 2010). The C/N ratios are another indicator of changes in the source of organic matter. In general, the C/N ratios from aquatic plants (freshwater phytoplankton) are <10 . A higher C/N ratio (10 – 20) indicates a mix of aquatic and terrestrial organic material (Mackie et al. 2005; Zong et al. 2006). More precise C/N values for different types of plants fall within the following ranges: algae—ca. 5–8, C3 land plants—ca. 16+, and C4 land plants—ca. 35+ (Curtis et al. 2010).

A natural mixture of signals from terrestrial organic parts transported into the lake and organic matter due to the primary production inside the lake can be modified by extra inputs of C and N. Urban and farmland waste waters (Harrington et al. 1998), acidic precipitation (Wolfe et al. 2001, 2003), and modifications in the trophic web structure (Anderson and Cabana 2009; Rawcliffe et al. 2010) were identified as the most important sources of organic matter with specific isotopic signatures.

In this study, we present the results of the analysis of the isotopic composition of nitrogen and carbon in organic matter from the sediments of several mountain lakes in the Tatra Mountains (southern Poland). The major aim of this study was to identify the sources of these elements in organic matter deposited into the sediments of mountain lakes located in different altitudinal zones, from submontane to alpine. It helps to understand how these lakes were supplied with nitrogen and carbon and which factors can modify nitrogen and carbon cycles in these ecosystems. We also describe the modification of the isotopic composition of organic matter caused by natural and human-induced (i.e., pastures, tourism, and acid rain) processes. The records of carbon and nitrogen stable isotopes for the last five centuries are presented for the lakes of the Tatra region for the first time.

2 Material and Methods

The stable isotope analysis was performed on short sequences of sediments collected with a Kajak-type gravity corer from the deepest sites of eight lakes located in the Tatra Mountains (Fig. 1, Table 1): the Smreczyński Staw (SME), the Toporowy Staw Niżni (TSN), the Zielony Staw Gąsienicowy (ZSG), the Długi Staw (DLU), the

Czarny Staw Gąsienicowy (CSG), the Wielki Staw Polski (WSP), the Przedni Staw Polski (PSP), and the Morskie Oko (MOK). This material was the basis for paleoclimatic and paleoecological reconstructions for the last two millennia (Gąsiorowski and Sienkiewicz 2010a, b). These sediments were fine and coarse detritus gyttja or silty mud with sand lamination and contained variable proportions of total organic carbon (TOC), from <1 % in mud and sand from the oligotrophic lakes of the alpine zone to over 50 % in the gyttja and peat from the dystrophic lakes located in the forest zone (Fig. 2).

The methodology for coring, describing the lithology, and dating follows that reported by Gąsiorowski and Sienkiewicz (2010a, b). Sediment cores were collected with the Kajak-type gravity corer. The cores were described in the field and splitted in 0.5- or 1-cm-thick intervals. The samples were stored in plastic bags in cold condition. Sediment sequences were dated by lead ^{210}Pb (upper part of each sediment sequence) and the AMS radiocarbon method.

The samples for C_{org} and N_{org} content and stable isotope analysis were collected every 0.5, 1, or 2 cm. One cubic centimeter of sediment was dried at a temperature of 60 °C, and the sediment was grinded. The carbonate fraction was removed with hydrochloric acid. The stable isotope measurements were performed over a relatively long time period (2009–2011), but for every sample, the same pretreatment and analytical methods were applied to reduce the biases of the methods (Brodie et al. 2012). The organic nitrogen and carbon percentages and the isotopic composition were analyzed using a Thermo MAT 253 mass spectrometer with a Flash EA 1112 elemental analyzer, which was calibrated using an internal nicotinamide standard. The results of elemental analysis were reported as mass fraction (in percent), and the isotope analysis results are reported as per mill deviations versus atmospheric N_2 ($\delta^{15}\text{N}$) and Vienna Pee Bee Belemnite ($\delta^{13}\text{C}$). The analytical errors (1 SD) for the $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ measurements were 0.17 and 0.24 ‰, respectively. This analysis was performed in the Laboratory for Isotope Dating and Palaeoenvironmental Studies of the Institute of Geological Sciences of the Polish Academy of Sciences in Warsaw.

3 Results

In the sediments studied, the typical values of $\delta^{13}\text{C}$ were from -31 to -24 ‰, with a median of -27.42 ‰,

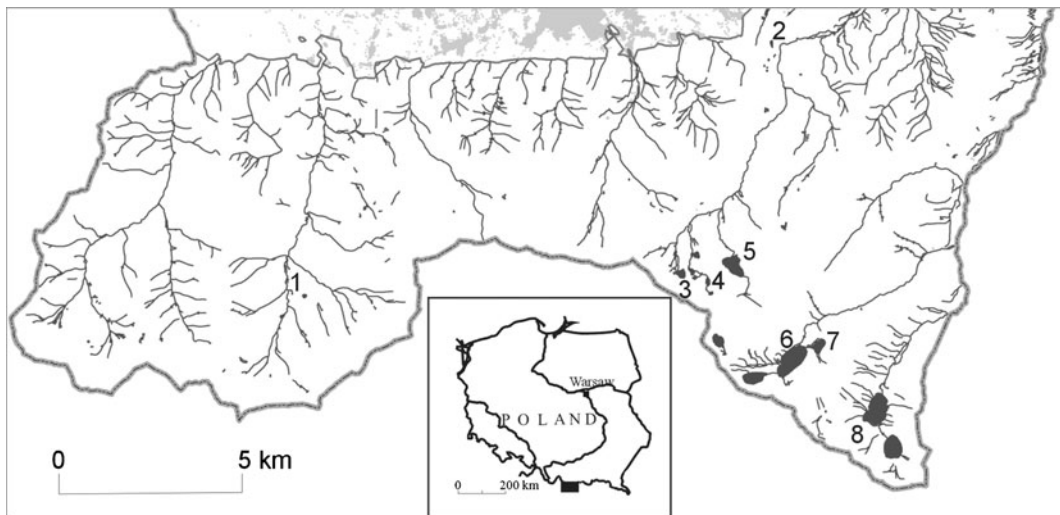


Fig. 1 Location of studied lakes: 1 Smreczyński Staw (SME), 2 Toporowy Staw Niżni (TSN), 3 Zielony Staw Gąsienicowy (ZSG), 4 Długi Staw (DLU), 5 Czarny Staw Gąsienicowy

(CSG), 6 Wielki Staw Polski (WSP), 7 Przedni Staw Polski (PSP), and 8 Morskie Oko (MOK)

and the values of $\delta^{15}\text{N}$ were from -2 to $+4$ ‰, with a median of $+0.85$ ‰ (Fig. 3). The $\delta^{13}\text{C}$ versus $\delta^{15}\text{N}$ relationship had a moderate coefficient of determination ($R^2=0.47$). The greatest variation in the $\delta^{13}\text{C}/\delta^{15}\text{N}$ ratio was detected in the PSP ($\delta^{13}\text{C}$ value was from -29.01 to -23.26 ‰ and $\delta^{15}\text{N}$ value was from -6.02 to 3.65 ‰), while the most stable isotopic composition was detected in the sediments from the DLU lake

($\delta^{13}\text{C}$ value was from -26.22 to -25.76 ‰ and $\delta^{15}\text{N}$ value was from 0.24 to 1.70 ‰).

The down-core analysis revealed relatively constant values of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in the preindustrial (pre-1850) period (Fig. 4) in five of the seven lakes studied (the DLU lake was excluded from this comparison because there was poor time control for this sediment sequence). The $\delta^{13}\text{C}$ values varied from -29.95 ‰ in the SME to

Table 1 Selected morphological and chemical parameters of studied lakes (after Kopáček et al. 2006)

| Name | Abbrev. | Location | Altitude (m a.s.l.) | Lake area (ha) | Max. depth (m) | pH | DOC ($\mu\text{mol L}^{-1}$) | TON ($\mu\text{mol L}^{-1}$) |
|--------------------------|---------|----------------------------|---------------------|----------------|----------------|------|--------------------------------|--------------------------------|
| Smreczyński Staw | SME | 49°13'21" N 19°51'52" E | 1,226 | 0.75 | 5.3 | 4.95 | 458 | 44.5 |
| Toporowy Staw Niżni | TSN | 49°17'00" N 20°01'52" E | 1,089 | 0.62 | 5.7 | 5.57 | 574 | 27.9 |
| Zielony Staw Gąsienicowy | ZSG | 49°13'44" N 19°59'59" E | 1,672 | 3.84 | 15.1 | 6.85 | 46 | 7.0 |
| Długi Staw Gąsienicowy | DLU | 49°13'38" N 20°00'33" E | 1,784 | 1.59 | 10.6 | 6.38 | 33 | 5.0 |
| Czarny Staw Gąsienicowy | CSG | 49°13'52" N 20°01'05" E | 1,620 | 17.94 | 51 | 6.49 | 41 | 5.6 |
| Wielki Staw Polski | WSP | 49°12'33" N 20°02'27" E | 1,655 | 34.35 | 79.3 | 7.03 | 39 | 8.1 |
| Przedni Staw Polski | PSP | 49°12'45" N 20°02'58" E | 1,668 | 7.71 | 34.6 | 7.23 | 47 | 10.8 |
| Morskie Oko | MOK | 49°11'49" N 20°04'12" E | 1,395 | 34.93 | 50.8 | 7.13 | 45 | 9.2 |

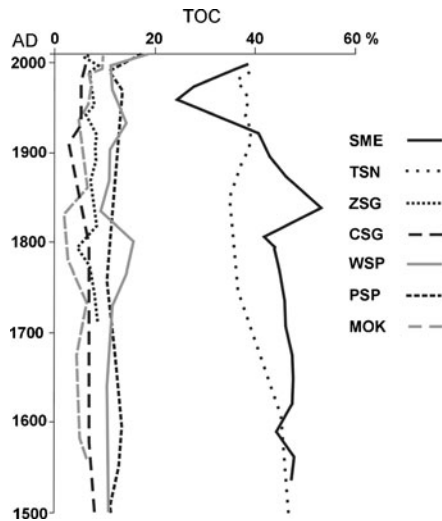


Fig. 2 Total organic carbon (TOC) in studied sediment sequences. Codes of lakes' names are similar to those in Fig. 1

-24.15 ‰ in the PSP sediments. The strongest trend toward lower $\delta^{13}\text{C}$ values was found for the ZSG sediments deposited at the end of the nineteenth century. The PSP showed a constant decline of $\delta^{13}\text{C}$ during the last two centuries. In other lakes, the carbon stable isotopic composition remained constant until the 1920s. After that, the changes varied between lakes. In the ZSG and the CSG, $\delta^{13}\text{C}$ remained constant. In the SME and the TSN, $\delta^{13}\text{C}$ declined during the second half of the twentieth century and reached -30.69 and -31.12 ‰, respectively. In both lakes, a slight trend toward the heavier C isotopes is observed since the 1980s. The twentieth century trends are toward lower values of $\delta^{13}\text{C}$ in the PSP, the WSP, and the MOK, and these trends are especially clear since the 1960s. The strongest trend toward low $\delta^{13}\text{C}$ values, over 3 ‰, is observed in

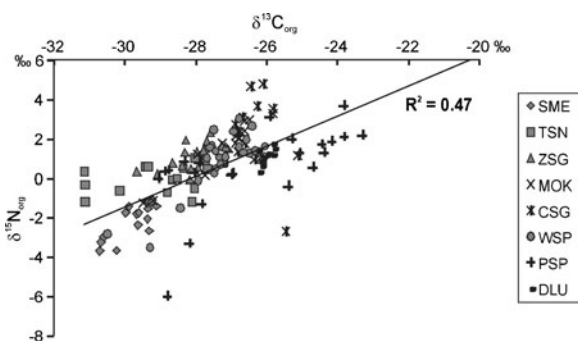


Fig. 3 $\delta^{13}\text{C}$ versus $\delta^{15}\text{N}$ in all studied sediment samples. Codes of lakes' names are similar to those in Fig. 1

the MOK, but the sediments that are most depleted in ^{13}C are in the TON (-31.12 ‰).

The $\delta^{15}\text{N}$ values during preindustrial (pre-1850) period varied from -2.40 ‰ in the SME to +2.72 ‰ in the CSG. In the latter lake, the isotopic composition of nitrogen underwent the smallest amount of change prior to the beginning of the twentieth century. The strongest shift in the $\delta^{15}\text{N}$ value, over 5 ‰, is observed in the PSP, and in the topmost sample, $\delta^{15}\text{N}$ reached -6 ‰. In contrast, the strongest enrichment in ^{15}N was recorded in the CSG (up to 4.7 ‰). In the SME, the TSN, and the ZSG, $\delta^{15}\text{N}$ remained nearly unchanged during the twentieth century.

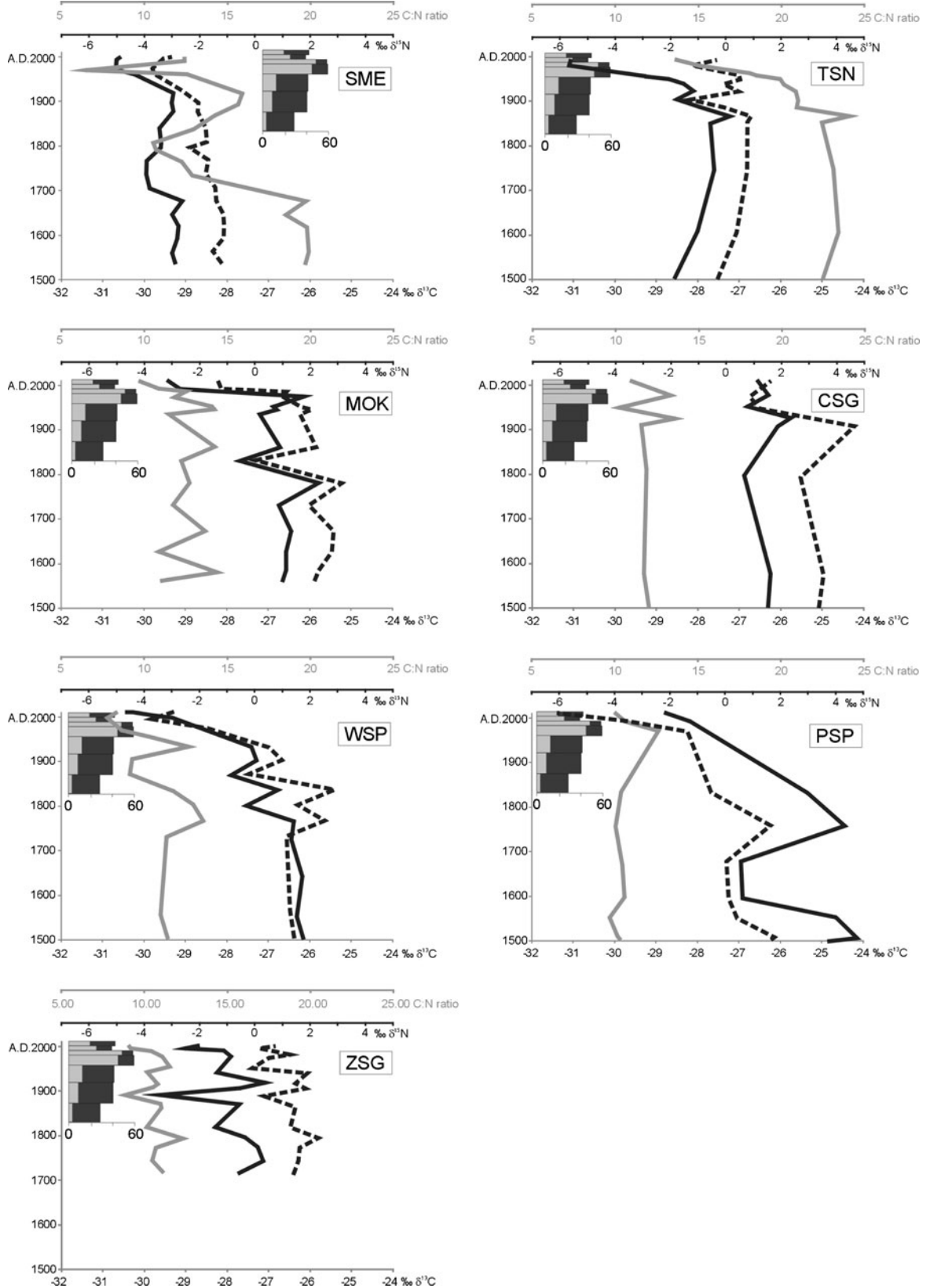
The C/N ratio in organic matter ranges between 9 and 15 in general. Only TSN sediments present significantly higher C/N values (13.6–23.2). However, a significant decrease was also observed during the past 120 years. Conversely, the sediments from the SME and the WSP lakes dated to the last three decades of the twentieth century show C/N values below 9. The C/N ratios of the sedimentary organic matter for the ZSG, the CSG, and the PSP lakes were the most stable, ranging only between 9 and 13.5.

4 Discussion

All of the lakes studied showed shifts in the sources of organic nitrogen and organic carbon deposited in sediments, as inferred from the C/N ratio, $\delta^{13}\text{C}$, and/or $\delta^{15}\text{N}$. However, when lakes of different origins and characters are included in the study, the values and magnitudes of the changes in the isotopic compositions of organic matter reveal spatial and temporal variations. The relationship between of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ shows only a moderate coefficient of determination (Fig. 3), suggesting that a different or complex process controls the isotopic composition of carbon and nitrogen in organic matter.

Relatively high C/N values clearly separate the TSN lake from the other lakes, indicating the significant

Fig. 4 Changes of $\delta^{13}\text{C}$ (solid black lines), $\delta^{15}\text{N}$ (dashed lines), and C/N ratio (solid gray lines) in organic matter from sediments of seven studied lakes. The long-term deposition of $\text{NO}_3\text{-N}$ and $\text{NH}_4\text{-N}$ (in milliequivalents per square meter per year) in the catchments of the Tatra lakes were presented as dark gray and pale gray bars, respectively (after Kopáček et al. 2003). The results for DLU were not reported due to lack of reliable chronology control for this sediment sequence. Codes of lakes' names are similar to those in Fig. 1



contribution of terrestrial plant tissues to the organic matter in this lake. The TSN is a dystrophic and colored lake located in a forest zone with many terrestrial organic parts deposited into sediments; intact leaves of spruce trees are a major component of these sediments. The specific environmental conditions (low pH and low nutrient concentration in the water) effectively limit macrophytes and algae productivity (Gąsiorowski and Sienkiewicz 2010a), and the isotopic signal is mainly due to terrestrial plant remains. A clear trend toward lower C/N values in the TSN suggests an increase of the in situ productivity of the lake (Thevenon et al. 2012) since 1950. However, there is no sign of an increase of the lake trophic state in the species composition of the diatom or in the TOC content (Fig. 2) over this time period, whereas the C/N ratio decrease is correlated with a depletion in $\delta^{13}\text{C}$ (Figs. 3 and 4), a decrease in the pH, and an increase of *Daphnia* biomass (Gąsiorowski and Sienkiewicz 2010a). The changes of the isotopic composition cannot be simply explained by the diagenetic decomposition of organic matter because this process causes depletion in ^{13}C and ^{15}N over time (Lehmann et al. 2002) and should produce lower $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values down the core. Hence, we instead associate this trend with a change in the food web in the TSN lake and a higher share of bacteria in the primary production of the lake (Bastviken et al. 2003; Eller et al. 2005) induced by lake acidification caused by NO_x deposition and possibly by an increase in the mean water temperature (Gąsiorowski and Sienkiewicz 2010a). The second dystrophic lake in the data set (the SME) also showed higher C/N ratios but only before the end of the seventeenth century. After that time, the C/N ratio decreased, which could be an effect of the limitation of the transport of terrestrial organic matter into the lake (Thevenon et al. 2012) by the peat bog zone surrounding the main basin (Skierski 1984). The steady decline of the C/N ratio in the SME during the eighteenth century changed into an increasing trend in the nineteenth century and again into a decline in the twentieth century (Fig. 4). These shifts may reflect changes in the spread of the peat bog along the shores of the lake and, consequently, in the transport of terrestrial plant remains into the basin (Gąsiorowski and Sienkiewicz 2010b). Furthermore, the SME sediments have a different isotopic composition than the other lakes. The organic matter is depleted in ^{13}C and ^{15}N throughout the entire sediment column, which indicates the relatively high contribution of methanogenic and N-fixing organisms to the primary

production (Dean 2006). The minima for the C/N ratio and $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in the SME occurred in the 1970s and 1980s. This was a period of maximal pollution and acidification in the region, as identified from the biota of acid-sensitive lakes in the Tatra Mountains (Stuchlik et al. 2002; Kopáček et al. 2004; Gąsiorowski and Sienkiewicz 2010a). In both lakes in the forest zone (the TSN and the SME), a return to the higher $\delta^{13}\text{C}$ values and a smaller increase in $\delta^{15}\text{N}$ are observed since the 1990s, indicating the chemical recovery of these ecosystems, while the recovery of the biota has not yet been observed (Gąsiorowski and Sienkiewicz 2010a, b).

In the other lakes, which are located above the treeline, the temporal changes in the isotopic composition varied between the sites, while the C/N ratio was similar (values from 7 to 14) and constant (Fig. 3). The ecosystems of these lakes were originally regulated by very low nutrient concentrations and low inputs of organic matter from their catchments. Additionally, from the end of the nineteenth century, the state of these lakes was determined also by locally diverse conditions and factors, i.e., the presence of fish (fish stocking) or the intensity of tourism (e.g., only two of the studied lakes have tourist shelters on their shores).

In the lakes located in the alpine zone, the strongest amplitudes in the isotopic composition of C (over 3 ‰) and N (also over 3 ‰) were recorded in the MOK, the PSP, and the WSP. The MOK and the PSP are the only lakes in the Polish part of the Tatra Mountains with tourist huts on their shores. The elevation of the WSP is a few meters lower than that of the PSP, and the WSP is fed by waters from the PSP and other lakes in the valley. Therefore, the changes in the isotopic composition of organic matter in this lake may be related to the changes and processes in the PSP. However, if human wastes were introduced into the lakes, positive shifts in $\delta^{15}\text{N}$ would occur (Bunting et al. 2007). This trend was not observed for any lake, which suggests that the activity associated with mountain huts in the Tatra Mountains may not have impacted significantly the lake environment. Human waste waters, if introduced into the lakes, had no influence on the water chemistry (Kurzyca et al. 2009) nor on the isotopic composition of sediments.

In this context, pasture land also seems to have had a limited impact on the lakes, though it produced an isotopic signal similar to that of human wastes. The limited impact is surprising because pasture activity in the Tatra Mountains began in the sixteenth century and was very intense between the seventeenth century and

the first half of the twentieth century (Radwańska-Paryska and Paryski 2004). At the peak of the pasture activity in the Tatra Mountains, approximately 1,000 sheep and cattle spent every summer in each valley. The traces of this process were recorded only in the CSG, where we observed a significant increase in $\delta^{15}\text{N}$ during the nineteenth century.

There is a question regarding the impact of air pollution on the isotopic compositions of C and N in the lakes of the Tatra Mountains. The trends toward negative values of $\delta^{15}\text{N}$ in the topmost sediments in the PSP and the WSP and, to a smaller extent, in the MOK and the CSG may be related to the deposition of NO_x from vehicle exhausts (Heaton 1990). In fact, the first symptoms of depletion in $\delta^{15}\text{N}$ were noted in the 1950s (Fig. 4) and are correlated with an increase in NO_x and NH_3 emissions in Central Europe, which peaked in the 1980s (Kopáček et al. 2002, 2004). After 1990, a slight decline in emissions and deposition rates was observed, and the NO_x and NH_3 depositions at Hala Gąsienicowa were 78 and 14 $\text{mmol m}^{-2} \text{ year}^{-1}$, respectively. The trends towards lower $\delta^{15}\text{N}$ values indicate that the pollution caused by the combustion of fossil fuels in electric plants played a smaller role in the modification of the composition of organic matter isotopes in the lakes studied. This fact can support the theory of the lower mobility of such types of pollution, which impacts the environment mainly within dozens of kilometers from the pollution source.

The significant depletion in ^{15}N in the second half of the twentieth century was not observed in the forest lakes or in the ZSG. The forest dystrophic lakes (the TSN and the SME) have water that is highly unsaturated with inorganic nitrogen, and these lakes are very small in area. Thus, the isotopic signal from precipitation was masked by the signal from terrestrial (allochthonous) organic compounds. The lack of significant changes in $\delta^{15}\text{N}$ in the ZSG was the effect of changes in the food web structure induced by the artificial introduction of charr. Fish were introduced into the ZSG in 1948, and the first major stocking was in 1951 (Gliwicz and Rowan 1984). Charr very effectively eliminate natural grazers, e.g., *Daphnia* and *Eurycercus* (Gąsiorowski and Sienkiewicz 2010a) and induce the development of phytoplankton. Increase of phytoplankton biomass produces higher values of $\delta^{15}\text{N}$ if there is a small contribution of ^{15}N -depleted, nitrogen-fixing cyanobacteria to the

sediment organic matter pool, and in this case, “phytoplankton effect” neutralized the effect of acid deposition.

All lakes included in this study and located in the alpine zone were artificially stocked with brook charr, with the exception of the WSP. The MOK also has a natural population of migratory fish. The introduction of fish effectively changes the food web structure in the lakes (Gliwicz and Rowan 1984). There is no strong signal of this process in the isotopic composition of sedimentary organic matter, except in the case of the ZSG. The ZSG is relatively small lake (Table 1) so even small population of fishes could rebuild food web structure, and this change can be reflected by stable isotope composition of sedimentary organic matter.

5 Conclusions

All of the data discussed above lead to the following conclusions:

1. Changes in the trophic web caused by acidification are clearly reflected by the $\delta^{13}\text{C}$ variation and the C/N ratio in dystrophic forest lakes because the acidification induced changes in the main primary producer group in these lakes.
2. Air pollution is reflected in the isotope composition of organic matter. The signal from the combustion of fossil fuels by vehicles is a strong decline in $\delta^{15}\text{N}$ for almost every sediment sequence.
3. The introduction of charr is not reflected in the composition of isotopes in most of the lakes studied, with the exception of the ZSG. In the ZSG, the charr very effectively eliminate planktonic and littoral grazers and thus cause a relatively substantial increase of the trophic state of the lake. An increase in $\delta^{15}\text{N}$ induced by the eutrophication process effectively masked the ^{15}N depletion caused by acid deposition.
4. The operation of mountain tourist huts on the shores of the MOK and the PSP has not had a significant impact on the isotopic composition of the organic matter in the lake sediments. The amount of nutrients introduced into the lakes from that source is small and most likely masked by the input of nutrients from other sources, mostly atmospheric deposition.

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