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Changes in accumulation of organic matter and stable carbon and nitrogen isotopes in sediments of two Slovenian mountain lakes (Lake Ledvica and Lake Planina), induced by eutrophication changes

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Abstract

We measured accumulation rates of organic carbon (OC) and total nitrogen (TN) and stable carbon ($\delta^{13}C_{org}$) and nitrogen isotopes ($\delta^{15}N$) in the sediments of two mountain lakes (Lake Ledvica and Lake Planina, northwest Slovenia). Marked variations of these parameters were observed in both sediments. OC accumulation rates ranged from 4 to 23 g m^{-2} yr^{-1} in Lake Ledvica, whereas in Lake Planina they were one order of magnitude higher and increased substantially in the upper sediments. $\delta^{13}C_{org}$ and $\delta^{15}N$ decreased upward in Lake Ledvica, from −25.5‰ to −28.4‰, and from +2.1‰ to −3.4‰, respectively. In contrast, substantial variations in the $\delta^{13}C_{org}$ profile, ranging from −30.9‰ to −37.4‰, but a quite uniform $\delta^{15}N$ profile of approximately +1.8‰, was observed in Lake Planina. Elemental and isotopic changes of bulk sedimentary organic matter in the lakes were related to changes in the past trophic state of the lakes and their watersheds, inferred by natural development of the lake ecosystems, anthropogenic activities, as well as earthquakes and forest fires. Observed changes and differences in the $\delta^{13}C_{org}$ and $\delta^{15}N$ records could be attributed to global changes in isotopic composition of atmospheric CO_2 and nitrate in atmospheric deposition, as well as to differences in organic matter, dissolved inorganic carbon, and dissolved inorganic nitrogen sources at various altitudes. Although Lakes Ledvica and Planina are remote mountain lakes, the sediment records showed severe eutrophication in Lake Planina, whereas in Lake Ledvica eutrophication is still moderate, thus reflecting a high sensitivity of these ecosystems to any external inputs.

The organic matter accumulated in lake sediments constitutes an important fraction that provides crucial information in studies of the lacustrine paleoenvironment, the history of climate change, and the effects of man on local and regional ecosystems. It is introduced to lakes by multiple pathways (Meyers and Ishiwatari 1995; Herczeg et al. 2001). Terrigenous (allochthonous) organic matter, originating mostly from the catchment area via tributaries, consists mainly of degraded remains of leaves and grass and soil organic matter, as well as material that falls directly into the lake from the riparian zones. Additional contributions from more distant sources are delivered by precipitation and wind. On the other hand, biota (animals, plants, and bacteria) within the water column and the sediments contribute aquatic (autochthonous) organic matter. The different types of biota populating a lake and its watershed produce organic matter having a distinctive biochemical composition. Changes in the community structure of these biota create variations in the amounts and types of organic matter deposited at different times in the history of a lake (Meyers and Ishiwatari 1995). To distinguish between the different origins of sedimentary organic matter, atomic C : N ratios have often been used. It is well known that algae have C : N ratios between 5 and 8, whereas vascular plants have C : N ratios of ≥20 (Meyers 1994). In addition, stable carbon isotopic ratios are used to identify different sources of sedimentary organic matter (e.g., Deines 1980; Fry and Sherr 1984; Meyers 1994). Furthermore, the stable isotopes of nitrogen are also potentially useful indicators of both organic matter sources and algal productivity in lakes, but the dynamics of nitrogen biogeochemical cycling complicate interpretations of sedimentary $\delta^{15}N$ values (e.g., Lehmann et al. 2002 and references therein). Variations in the isotopic composition of carbon and nitrogen in sedimented organic matter integrate the overall changes in the relative abundance of these sources of particulate and dissolved C and N, as well as biogeochemical processes affecting these two essential nutrients in the water column of lakes (e.g., Schelske and Hodell 1991; Herczeg et al. 2001). Sources of dissolved C and N taken up by aquatic plants include atmospheric CO_2 and N_2, dissolved inorganic C (DIC), and dissolved NO_3^- and NH_4^+ derived from the catchment. The combined use of elemental abundances and isotopic ratios in sediments can, however, untangle and integrate the above apparent myriad of possible sources and transformations to reveal time histories of lake metabolism (Fogel and Cifuentes 1993; Meyers 1997).

Usually it is assumed that primary productivity is the principal control on carbon and nitrogen cycling, but in eutrophic systems the development of anoxic conditions in the water column and sediments may create conditions favorable

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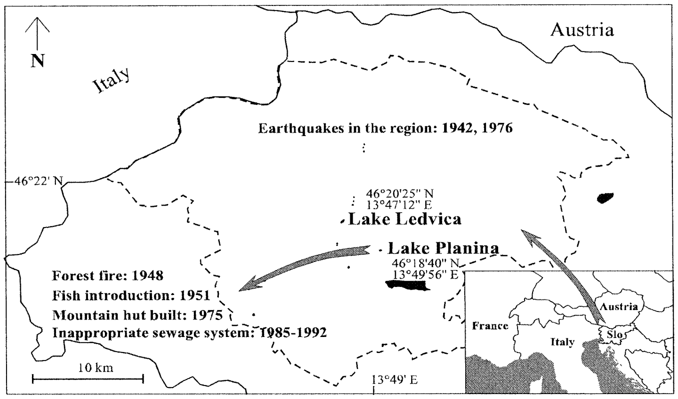

Fig. 1. Map of the Triglav National Park (dashed line) area in northwest Slovenia, showing the locations of Lake Ledvica and Lake Planina.

for expansion of microbially mediated carbon and nitrogen cycling processes, which can distinctly influence the isotopic signatures in the sediment (Hollander and Smith 2001; Lehmann et al. 2002).

Mountain lakes are considered an extreme environment. They are small and sensitive ecosystems with rapid flushing rates. Consequently, they respond quickly and markedly to any direct anthropogenic influence or change in the lakes themselves and their catchment areas. These effects and changes are potentially recorded in lake sediments and lake sediments can thus be used as a means of reconstructing past conditions of lakes over longer timescales (Battarbee et al. 2002).

In this study, organic carbon (OC) and total nitrogen (TN) accumulation rates and stable carbon ($\delta^{13}C_{org}$) and nitrogen isotopes ($\delta^{15}N$) were determined in sediments of two Slovenian mountain lakes (Lake Ledvica and Lake Planina) located at different altitudes. The former is still oligo/mesotrophic, whereas the latter is eutrophic. OC, TN, $\delta^{13}C_{org}$, and $\delta^{15}N$ were analyzed in the sediments and C:N ratios were calculated on an atomic basis. Sediments were dated radiometrically to follow temporal changes of the measured parameters. The results obtained in organic geochemical investigations in Lake Ledvica and Lake Planina are furthermore examined for the potential application of these techniques in studies of eutrophication processes in remote lakes.

Materials and methods

Site description—Two remote mountain lakes, Lake Ledvica and Lake Planina, were selected as study sites. The lakes are located in the Triglav National Park in the Julian Alps, in the northwestern part of Slovenia (Fig. 1). Their topographical characteristics and the physicochemical properties of the water column are summarized in Table 1. The lakes are relatively small (approx. 19,000 m^2), shallow (11 to 15 m), and of glacial origin. The catchment area of the lakes is mostly composed of limestones and dolomites. The annual precipitation rate averages 3,000 mm per year (Kastelec 1999). The lakes are located in the same air shed. Lake Ledvica is located at the level of the tree line and is surrounded by steep slopes where some bushes of *Pinus mughi* and *Larix decidua* grow (Brancelj et al. 2002). Its water column is saturated with oxygen throughout the year. It is still in a relatively pristine condition and the lake was rated as oligo/mesotrophic (Muri and Brancelj 2003). In contrast, Lake Planina is located below the tree line. It is surrounded by an extensive conifer forest to the north and a pasture used for grazing cattle to the south. The hypolimnion is dysoxic and occasionally anoxic during the stratified period (Vreca 2003). The lake was rated as eutrophic (Muri and Brancelj 2003). There were two strong earthquakes with epicenters in the area of the Julian Alps in 1942 and 1976 (M = 4.4 and M = 6, respectively), causing subsequent land or mudslides (or both) that increased sedimentation rates and also affected the trophic state of the lakes (Brancelj et al. 2000, 2002). The whole area of the Triglav National Park is protected by law, and consequently anthropogenic activities in the lakes' watersheds are relatively limited. Nevertheless, after 1975 a former cheese dairy situated just above Lake Planina has been used as a mountain hut (Vreca 2000). The old dairy was destroyed by fire in 1981 and on its ruins a new mountain hut was built in 1984. Although it operates only in the summer months, it represented a substantial burden of nutrients for the lake, mostly from inappropriate sewage system effluents during 1985–1992 (Brancelj et al. 2000). The anthropogenic effect on Lake Planina is thus stronger than on Lake Ledvica and this is also reflected in the deteriorated condition of the former.

Table 1. Topographic characteristics of the sampling sites (data from Brancelj 2002) and average physical and chemical properties of the water column.

	Lake Ledvica	Lake Planina
Altitude (m)	1830	1430
Surface area (m^2)	21,900	15,600
Max. depth (m)	15	11
Ice cover (months)	6.5	5.5
Secchi depth (m)	to the bottom	1.5
Cond (μS cm^{-1})	160	180/325†
O_2 (mg L^{-1})	9.0	8.5/1.0†
T (°C)‡	14.9/4.8†	19.5/6.5†
Alk (mmol L^{-1})	1.6	1.8/3.3†
Ca (mg L^{-1})	23	30/48†
Chl *a* (μg L^{-1})*	<1	13
pH	8.0	8.1/7.2†
TN (mg N L^{-1})	1.8	1.9/2.7†
NO_3^- (mg L^{-1})	1.5	0.2
TP (μg P L^{-1})	19	51/191†

Cond, conductivity; O_2, dissolved oxygen; T, temperature; Alk, the alkalinity; Chl *a*, chlorophyll *a*; Ca, calcium; TN, total nitrogen; TP, total phosphorus.

* Data from Brancelj 2002.

† Surface water/bottom water value.

‡ Maximal values.

Mineral composition was determined by X-ray diffraction only in the sediment from Lake Planina (Vreca 2000). The mineral component of the recent sediment was characterized as carbonate silt predominantly composed of calcite (69%) and clay minerals (23%). The other 8% represents quartz, dolomite, feldspars, micas, pyrite, and goethite. According to the similar lithology around Lake Ledvica, we assume that the sediment from this lake has a similar carbonate content to Lake Planina. In the sediment from both lakes no clear lamination was observed. In Lake Planina this could be explained by conditions that favor calcite dissolution during sinking of particles to the lake bottom and in the sediment (Vreca 2000, 2003). However, lake sediment represents an important sink of organic carbon, and in the sediment methanogenesis is an important process (Simcic et al. 2002; Vreca 2003; Muri and Simcic 2004).

Sediment sampling—Sediment samples were taken from the central part of the lakes using a modified Kajak gravity corer equipped with a plexiglass tube of inner diameter 6 cm and 70 cm long. Sediment cores collected from Lake Ledvica and Lake Planina were 28 and 25 cm long, respectively. The cores were immediately extruded in the field and sectioned into 1-cm intervals. Samples were freeze-dried, homogenized, and stored in polyethylene vials prior to analyses.

Dating—Sediments were dated radiometrically (Pennington et al. 1976; Appleby and Oldfield 1983). Details of sediment dating including vertical profiles of excess ^{210}Pb and ^{137}Cs activities are described in Appleby (2000) and Brancelj et al. (2002) for Lake Ledvica and in Brancelj et al. (2000) and Muri et al. (2002) for Lake Planina. Briefly, a γ-ray spectrometer equipped with a high-purity Ge well-type detector was used to measure the ^{210}Pb and ^{137}Cs activities in each sediment slice. Unsupported ^{210}Pb activity in the slice was calculated by subtracting ^{226}Ra activity from total ^{210}Pb. Sedimentation rates were obtained on the basis of unsupported ^{210}Pb activity levels using the constant rate of supply model. Maxima in the ^{137}Cs profile were used as reference points for the years 1986 and 1963 (fallout from the Chernobyl accident and maximum fallout from nuclear weapons, respectively) and confirmed the dating suggested by ^{210}Pb. Twenty-one cm of Lake Planina sediment represents a depositional time span of approximately 110 yr, whereas 17 cm of Lake Ledvica sediment represents approximately 150 yr.

Analyses of OC and TN—The OC and TN concentrations were measured after removal of inorganic carbon (carbonate). Dry sediment samples were acidified with HCl (Hedges and Stern 1984). The acid was added until effervescence upon acid addition ceased, indicating complete removal of inorganic carbon. Finally, the OC and TN concentrations were determined using a CHN elemental analyzer (Carlo Erba EA 1008) at a combustion temperature of 1,020°C. Blank samples and standard with known elemental composition (sulfanilamide) were used for quality assurance. The precision of the method, expressed in terms of standard deviation, was from 3% to 5%.

Accumulation rates of OC and TN—OC and TN accumulation rates were obtained by multiplying OC and TN concentrations and sediment accumulation rates at each sediment depth. The sediment cores were dated down to a depth of 17 cm in Lake Ledvica and 21 cm in Lake Planina. Thus, OC and TN accumulation rates were obtained only for these parts of the sediment cores.

Stable isotope analyses—Dry sediment samples were soaked in 1 mol L^{-1} HCl overnight to remove carbonates, and then filtered on quartz-fiber filters (Whatman GF/C), rinsed with deionized water, and redried. Subsamples of 2–4 mg were placed in tin capsules and the stable isotopic composition of organic carbon was determined on a continuous-flow Europa 20–20 isotope ratio mass spectrometer. All subsamples were analyzed in duplicate. Carbon isotopic ratios are expressed in standard delta notation ($\delta^{13}C_{org}$), which is the per mil (‰) deviation from the V-PDB standard. Analytical reproducibility was ±0.2‰, determined on repeated analysis of reference materials (IAEA-CH-7 and USGS24). Sample repeatability was ±0.1‰ or better. For determination of the isotopic composition of nitrogen approximately 5–30 mg of homogenized bulk sample material was placed into tin capsules and measured on a Europa 20–20 isotope ratio mass spectrometer. All subsamples were analyzed in duplicate. Nitrogen isotope ratios are reported as $\delta^{15}N$ with respect to atmospheric N_2 (air). Analytical reproducibility was ±0.3‰, determined on repeated analysis of reference materials (IAEA-N1 and IAEA-N2). Sample repeatability was ±0.1‰ or better.

Measured values for $\delta^{13}C_{org}$ are dependent on the historic isotopic signatures of DIC when organic carbon is produced photosynthetically (Schelske and Hodell 1995). To correct for the historic depletion of $\delta^{13}C$ in atmospheric CO_2 due to fossil fuel burning (Keeling et al. 1989), measured values for $\delta^{13}C_{org}$ in sediment samples were normalized to values recorded by fossil air trapped in ice cores (Friedli et al. 1986). According to Schelske and Hodell (1995) a $\delta^{13}C$ depletion of −1.4 has occurred since 1840 A.D. $\delta^{13}C_{org}$ values in this study were corrected for this effect using the following equation (from Schelske and Hodell 1995; the units for time are in years):

$$\delta^{13}C = -4{,}577.8 + 7.3430t - 3.9213 \times 10^{-3}t^2 + 6.9812 \times 10^{-7}t^3$$

The calculated time-dependent depletion in $\delta^{13}C$ since 1840 was subtracted from the measured $\delta^{13}C_{org}$ values for each dated sediment section.

Results

Organic carbon and total nitrogen concentrations—High OC concentrations were observed in Lake Ledvica and Lake Planina, exceeding 150 mg g^{-1} in surface sediments (Table 2). In Lake Ledvica, the OC concentration decreased gradually to 70 mg g^{-1} at the depth of 12–18 cm and increased to 105 mg g^{-1} at the bottom of the core. In contrast, the OC concentration in Lake Planina varied substantially down the core and amounted to 71 mg g^{-1} at the bottom of the core. The TN concentration was around 20 mg g^{-1} in the surface sediment of both lakes. In Lake Ledvica, the TN concentration decreased below 10 mg g^{-1} at the depth of 12–18 cm and increased to 24 mg g^{-1} at the bottom of the core. In

Table 2. Organic carbon (OC) and total nitrogen (TN) concentrations in Lake Ledvica and Lake Planina sediments.

Depth (cm)	Lake Ledvica		Lake Planina	
	OC (mg g^{-1})	TN (mg g^{-1})	OC (mg g^{-1})	TN (mg g^{-1})
0–1	156	19	169	21
1–2	136	16	149	17
2–3	126	15	152	17
3–4	116	12	139	14
4–5	109	11	113	12
5–6	115	12	122	13
6–7	109	11	112	13
7–8	109	11	94	10
8–9	107	11	105	11
9–10	96	10	153	16
10–11	101	11	142	15
11–12	85	9	95	10
12–13	66	9	120	13
13–14	66	8	144	15
14–15	69	8	118	12
15–16	69	8	119	13
16–17	70	9	117	12
17–18	67	10	113	12
18–19	82	10	102	10
19–20	84	11	85	9
20–21	80	12	126	13
21–22	112	17	132	13
22–23	60	12	105	11
23–24	103	18	67	7
24–25	97	19	71	7
25–26	95	20		
26–27	93	21		
27–28	105	24		

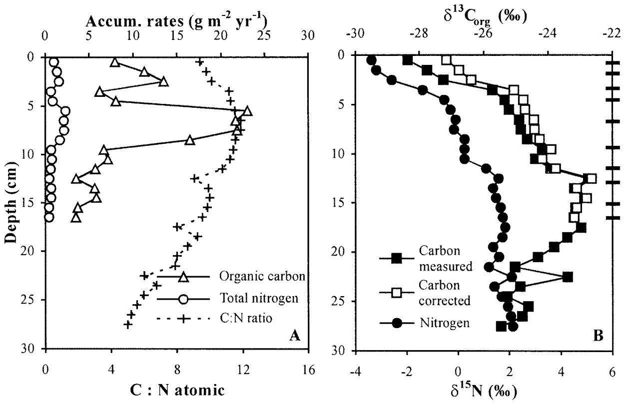

Fig. 2. Elemental and isotopic measurements of bulk sedimentary organic matter in Lake Ledvica, including (A) organic carbon and total nitrogen accumulation rate, atomic C : N ratio, and (B) measured and Suess effect-corrected carbon isotopic ratios and nitrogen isotopic ratios.

contrast, the TN concentration in Lake Planina decreased to less than10 mg g^{-1} at the bottom of the core.

Organic carbon accumulation rate—In Lake Ledvica, the OC accumulation rate in the surface sediment layer amounted to 8 g m^{-2} yr^{-1} and varied considerably in the upper 10 cm of the sediment (Fig. 2A). A subsurface peak value of the OC accumulation rate was observed at a depth of 6 cm, reaching 23 g m^{-2} yr^{-1}. In the deeper sediments (>10 cm), the OC accumulation rate dropped and was quite uniform, ranging from 3 to 7 g m^{-2} yr^{-1}. In contrast, the highest OC accumulation rate in Lake Planina was observed in the surface sediment layer (Fig. 3A). At 269 g m^{-2} yr^{-1} it was more than one order of magnitude higher than in Lake Ledvica. Nevertheless, a steep decrease of the OC accumulation rate was observed in the upper 5 cm of the sediment where it dropped to 29 g m^{-2} yr^{-1}. It averaged 31 g m^{-2} yr^{-1} in the deeper sediments.

Nitrogen accumulation rate—The TN accumulation rate closely followed the OC accumulation rates in both lakes (Figs. 2A, 3A). In Lake Ledvica, it amounted to 1 g m^{-2} yr^{-1} in the surface sediment layer. A peak value of 2.3 g m^{-2} yr^{-1} was again observed at a depth of 6 cm, whereas in the deeper sediments the flux dropped below 1 g m^{-2} yr^{-1}. The TN accumulation rate in Lake Planina was the highest in the surface sediment layer, reaching 33 g m^{-2} yr^{-1}. The accumulation rate decreased remarkably in the upper 5 cm of the sediment and dropped to less than 8 g m^{-2} yr^{-1} in the deeper sediment layers.

C : N ratio (atomic)—The surface sediment C : N ratio in Lake Ledvica was 9.5 (Fig. 2A). In the upper sediments, the C : N ratio increased from the surface, reaching a peak value of 11.9 at a depth of 7 cm. In the deeper sediments, the C : N ratio decreased gradually and dropped to 5.0 at the bottom of the core. In Lake Planina, the C : N ratio increased from 9.6 at the sediment surface to 12.1 in deeper sediment layers (Fig. 3A).

Carbon isotopes ($\delta^{13}C_{org}$)—In Lake Ledvica, δ^{13}C$_{org}$ values increased from −28.4‰ at the surface to −22.7‰ at the depth of 12 cm and then decreased to −25.5‰ at the bottom of the core (Fig. 2B). In contrast, in Lake Planina δ^{13}C$_{org}$ values decreased from −31.9‰ to −37.4‰ in the upper 5 cm of the sediment and then increased to about −32‰ with

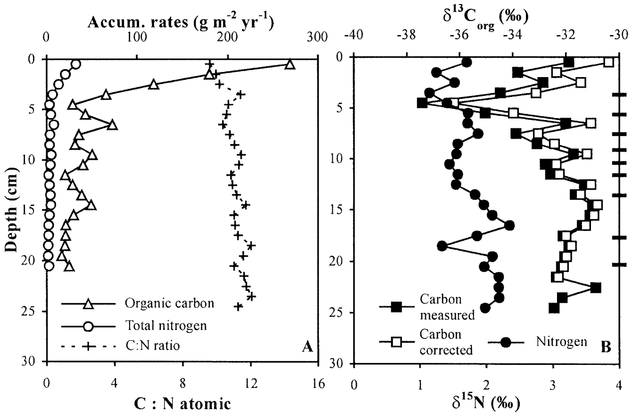

Fig. 3. Elemental and isotopic measurements of bulk sedimentary organic matter in Lake Planina, including (A) organic carbon and total nitrogen accumulation rate, atomic C : N ratio, and (B) measured and Suess effect-corrected carbon isotopic ratios and nitrogen isotopic ratios.

Table 3. Major elemental, isotopic, and biological trends in Lakes Ledvica and Planina. The most abundant diatom and cladoceran remains are reported in brackets.

Lake Ledvica	Lake Planina
Above 4 cm (after 1960) ● Decrease of OC and TN accumulation rates ● Decrease of C:N ratio ● Decrease of $\delta^{13}C_{org}$ and $\delta^{15}N$ ● Increase in abundance of diatom remains (*Denticula tenuis, Navicula cryptotenella*)* ● Decrease in abundance of Cladocera remains (*Chydrous sphaericus*)* ● Increase of CD and TC concentrations*	Above 4 cm (after 1987) ● Decrease of C:N ratios ● Increase of $\delta^{13}C_{org}$ ● Slight decrease of $\delta^{15}N$ ● Change in abundance of diatom taxa remains (*Stepanodiscus parvus, Fragilaria pinnata*)† ● Decrease of *Eubosmina longispina*†
12–4 cm (1910–1960) ● High OC and TN accumulation rates ● Increase of C:N ratio ● Decrease of $\delta^{13}C_{org}$ and $\delta^{15}N$ ● Increase in abundance of diatom remains (*Denticula tenuis, Navicula cryptotenella*)* ● Increase in abundance of Cladocera remains (*Chydrous sphaericus, Daphnia longispina*)* ● Increase of CD and TC concentrations*	15–4 cm (1910–1987) ● Increase of OC and TN accumulation rates ● Slight decrease of C:N ratios and $\delta^{15}N$ ● Decrease of $\delta^{13}C_{org}$ ● Change in abundance of diatom taxa remains (*Fragilaria pinnata, Stepanodiscus parvus*)† ● Change in abundance of Cladocera remains (*Eubosmina longispina, Daphnia longispina*)†
Below 12 cm (before 1910) ● Low OC and TN accumulation rates ● Increase of C:N ratio and $\delta^{13}C_{org}$ ● Slight decrease of $\delta^{15}N$ ● No significant change in abundance of diatom taxa (*Amphora libyca, Denticula tenuis*)* ● Decrease in abundance of Cladocera remains of *Chydrous sphaericus** ● Low CD and TC concentrations*	Below 15 cm (before 1910) ● Low OC and TN accumulation rates ● No significant change of C:N ratios, $\delta^{13}C_{org}$ and $\delta^{15}N$ ● Change in abundance of diatom taxa remains (*Stepanodiscus parvus, Fragilaria pinnata*)† ● Slight increase in abundance of Cladocera remains (*Daphnia longispina*)†

CD, chlorophyll derivatives; TC, total carotenoids.
* Data from Brancelj et al. 2002.
† Data from Brancelj et al. 2000.

sediment depth (Fig. 3B). As outlined above, trends in $\delta^{13}C_{org}$ values of the stratigraphic sequence deposited in historical times are overprinted by the addition of isotopically light fossil carbon into the terrestrial carbon dioxide pool (Schelske and Hodell 1995). The time-dependent correction for this anthropogenic effect is shown in Figs. 2B and 3B.

Nitrogen isotopes ($\delta^{15}N$)—Changes in the isotopic composition of sedimentary nitrogen are shown in Figs. 2B and 3B. In Lake Ledvica, $\delta^{15}N$ values increased progressively from $-3.4‰$ in the surface sediment to $+2.1‰$ at the bottom of the core, displaying an overall shift of 5.5‰. In contrast, in Lake Planina $\delta^{15}N$ values were relatively constant with a mean of $+1.8 \pm 0.3‰$.

Discussion

Environmental changes inferred from Lake Ledvica sediments—Three distinct periods of elemental and isotopic records are observed in contemporary Lake Ledvica sediments. In the sediment section below 12 cm that accumulated before circa 1910, no marked variations of both OC and TN accumulation rates were observed (Fig. 2A). The correlation between OC and TN accumulation rates is high ($r^2 = 0.99$, $p < 0.05$) throughout the sediment core. Increasing C:N ratios are also characteristic of these sediments. The C:N ratio increases from 5 at the bottom of the core to approximately 10 (Fig. 2A). The shift of C:N ratios suggests that sources of organic matter have changed. Autochthonous and allochthonous sources contributed organic matter but the former prevailed. The increasing trend of the C:N ratios could have also been caused by a relatively larger terrestrial contribution to the bulk organic matter. In this sediment section no significant change in the abundance of the diatom assemblage, represented predominantly by *Amphora libyca* and *Denticula tenuis*, was observed (Brancelj et al. 2002). In contrast, the concentration of Cladocera remains represented predominantly by *Chydrous sphaericus* decreased upward and concentrations of chlorophyll derivatives and total carotenoids slightly increased at the end of this period (Table 3). During this period, $\delta^{13}C_{org}$ values increased by 2.3‰, whereas $\delta^{15}N$ values remained rather uniform and average $+1.7 \pm 0.3‰$. The isotopic composition of organic carbon is significantly correlated with the C:N ratio ($r^2 = 0.52$, $p < 0.05$), whereas the isotopic composition of nitrogen is negatively correlated ($r^2 = 0.48$, $p < 0.05$). No statistically significant correlation was observed between the two isotopic parameters (Fig. 4).

An approximately fivefold increase of both the OC and TN accumulation rates is observed in the sediment section

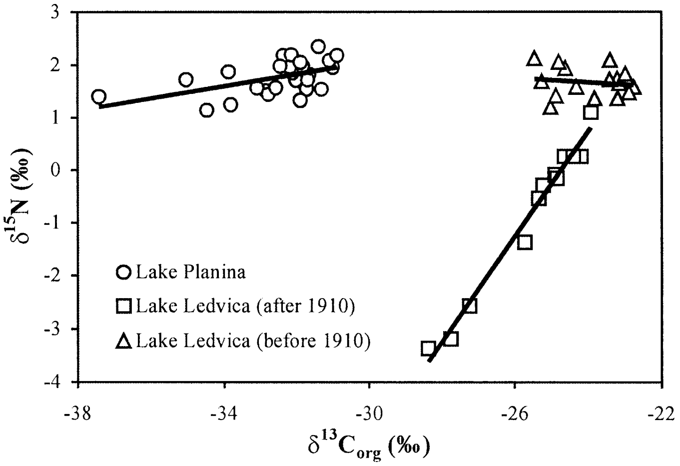

Fig. 4. Carbon vs. nitrogen isotopic ratios of sedimentary organic matter in Lake Ledvica (before 1910, after 1910) and in Lake Planina.

between 12 and 4 cm depth that accumulated from approximately 1910 to 1960 (Fig. 2A). In addition, the highest C:N ratios are also observed in this sediment section, ranging from 11 to 12 (Fig. 2A). Brancelj et al. (2002) found that concentrations of in-lake biota (diatoms and cladocera) also increased markedly in this sediment section. However, in this period abundance of *Denticula tenuis* and *Navicula cryptotenella* increased upward while *A. libyca* decreased. The concentration of Cladocera remains also increased. They were represented predominantly by *C. sphaericus* and *Daphnia longispina.* A sudden increase in concentrations of chlorophyll derivatives and total carotenoids followed changes in the diatom assemblage (Table 3). Brancelj et al. (2002) concluded that these increased concentrations resulted from a local slump event of around 1915 and a strong regional earthquake in 1942 that was followed by landslides, delivering terrestrial organic matter from the steep slopes around the lake to the lake itself and causing an increase in the trophic state of the lake. On the basis of the higher C:N ratios, a higher proportion of terrestrial organic matter could in fact have been contributed to the lake in this period. Because concentrations of nutrients in the Lake Ledvica water column are generally low (Table 1; for details see Muri and Brancelj 2003 and Muri 2004 for comparison with other Slovenian mountain lakes), an increased terrestrial contribution could be reflected in a temporary increase of primary producers in the lake. With higher nutrient concentrations in the water column, conditions for growth of primary producers would be more favorable. During this period, $\delta^{13}C_{org}$ and $\delta^{15}N$ values decreased by 3‰ (Fig. 2B). Changes in $\delta^{13}C_{org}$ values can be explained either by introduction of terrestrial organic matter with a lighter isotopic composition characteristic of C3 plants (Deines 1980), or by consumption of ^{13}C-depleted CO_2 formed during degradation of sinking organic matter by aquatic biota (Rau 1978; Hollander and Smith 2001) in combination with ^{13}C-depleted atmospheric CO_2 due to fossil fuel burning (Keeling et al. 1989). The extreme decrease in $\delta^{15}N$ values indicates that after 1910 sources of dissolved inorganic nitrogen (DIN) in Lake Ledvica have most probably changed (see Comparison of eutrophication effects on the lakes). In this section, $\delta^{13}C_{org}$ and $\delta^{15}N$ values are strongly correlated ($r^2 = 0.98$, $p < 0.05$), as shown in Fig. 4, and are also correlated with C:N ratios ($r^2 = 0.88$, $p < 0.05$).

The upper 4 cm of the sediment represent the section that was deposited after 1960. In this section the OC and TN accumulation rates dropped substantially (Fig. 2A). Nevertheless, a peak value is observed at a depth of 3 cm that accumulated around 1975. Increased accumulation rates in this sediment section could again be related to a strong earthquake in 1976, as also suggested by Brancelj et al. (2002). The C:N ratios decrease in the contemporary sediment (Fig. 2A). This decrease could be caused either by a lower terrestrial contribution in the most recent sediments or increased primary production in past decades. In a study of water chemistry in Lake Ledvica (Muri and Brancelj 2003; Muri 2004), it was found that nutrient concentrations occasionally exceeded the oligotrophic nature of the lake, suggesting a slight but possible chance of eutrophication in this lake. Furthermore, increasing concentrations of chlorophyll derivatives and total carotenoids in surface sediments also indicate nutrient enrichment in this lake. During this period, $\delta^{13}C_{org}$ and $\delta^{15}N$ values decreased by 2.7‰ and 2‰, respectively (Fig. 2B). The decrease of $\delta^{13}C_{org}$ and $\delta^{15}N$ values displays the progressive continuation of a process that started at the beginning of the 20th century, showing the influence of recycling of organic matter in the water column and an anthropogenic signal originating from atmospheric input. As reported by Brancelj et al. (2002) the concentration of the most abundant diatom species, *Denticula tenuis* and *N. cryptotenella*, was highly increased, whereas the concentration of Cladocera remains decreased in this section (Table 3). Recently, some N-fixing cyanobacteria (e.g., *Calothrix parietina*) were identified in the lake (Sisko pers. comm.) but no extreme expansion was observed.

Environmental changes inferred from Lake Planina sediments—The OC and TN accumulation rates in the sediment section below 15 cm that accumulated before circa 1910 are low (Fig. 3A) and almost of the same magnitude as those in Lake Ledvica (Fig. 2A). The correlation between the OC and TN accumulation rates is high ($r^2 = 0.99$, $p < 0.05$) throughout the sediment core. In contrast, a much lower correlation ($r^2 = 0.26$, $p < 0.05$) is observed between $\delta^{13}C_{org}$ and $\delta^{15}N$ values throughout the sediment core (Fig. 4). C:N ratios as well as $\delta^{13}C_{org}$ and $\delta^{15}N$ values are rather uniform in this sediment section and average 11.6 ± 0.4, -31.9 ± 0.4‰ and $+2.0 \pm 0.3$‰, respectively (Fig. 3B). During this period woodcutting and Alpine dairying were important activities in the surroundings of the lake and as a consequence erosion of hill slopes and input of nutrients was enhanced. C:N ratios show that allochthonous sources also contributed to sedimentary organic matter (Muri et al. 2004). In addition, the abundance of diatom taxa remains changed upward (Brancelj et al. 2000). According to Brancelj et al. (2000) the *Stepanodiscus parvus* population decreased in this section and that of *Fragilaria pinnata* started to increase around 1890. Furthermore, the abundance of Cladocera remains, predominantly represented by *Daphnia longispina,* increased in the sediment (Table 3).

In the sediment section from 15 cm to 4 cm, representing the time period from approximately 1910 to 1987, both OC and TN accumulation rates were higher and varied substantially (Fig. 3A). In this time period, some exceptional episodes occurred in the Lake Planina water column and its watershed, altering OC and TN distributions. In 1948, there was a forest fire in the watershed and around 1 km^2 of forests was destroyed (Muri et al. 2003). This event was also recorded in the sediment as a peak value in the OC and TN accumulation rates, due to increased erosion or input (or both) of terrestrial materials from the catchment. Furthermore, in 1951 fish were introduced into the lake, which was originally without fish. Several aspects could be related to fish introduction, as reported by Brancelj et al. (2000). The fish preyed upon large specimens of zooplankton (like *Daphnia longispina*) and the benthic fauna and reduced their numbers considerably. In parallel, the feeding on benthic animals resulted in increased resuspension of sediment and subsequent release of nutrients to the water column. As a consequence, the abundance of algae and small specimens of zooplankton (like *Eubosmina longispina*) increased, the latter probably due to reduced competition from the Daphnia population and increased algae concentrations (Brancelj et al. 2000). In contrast, diatom taxa abundance represented predominantly by *F. pinnata* and *S. parvus* decreased during this period (Table 3). A peak value of OC accumulation rates at a depth of 7 cm, corresponding to around 1975, could be explained by the 1976 earthquake that could have been followed by mudslides into the lake. In addition, a new mountain hut with an inappropriate sewage system was opened above the lake in 1984 (Brancelj et al. 2000). Thus, effluents rich in nutrients could have entered the lake until 1992 when a new sewage system was built. Although this period was short, the effluents affected this small ecosystem considerably. In contrast, the C : N ratios were quite uniform and averaged 11.0 ± 0.4 (Fig. 3A) but decreased slightly upward. The fairly low C : N ratios suggest that autochthonous organic matter prevails. However, allochthonous sources have also contributed organic matter in the past (Muri et al. 2004) and their contribution most probably has been rather constant, because no substantial variation of the C : N ratios in the deeper sediments is observed. At the end of this period, the lake was eutrophic and a dramatic change is recorded in the sediment by low $\delta^{13}C_{org}$ values (−37.4‰). A distinctive decrease in isotopic composition of organic carbon started above the depth of 7 cm (Fig. 3B). A possible explanation for such a change is recycling of organic carbon within the lake during moderate to severe eutrophication (Hollander and Smith 2001 and references therein). This model takes into account oxidation of ^{13}C-depleted biogenic methane released from the sediments to the water column that contributes isotopically depleted dissolved CO_2 to the DIC reservoir. If sufficient amounts of biogenic methane are oxidized, the $\delta^{13}C$ values of DIC are shifted to more negative values. The assimilation of this DIC would therefore result in the synthesis of ^{13}C-depleted biomass. Furthermore, ^{13}C-depleted biogenic methane formed in the sediment could enable expansion of methanotrophic organisms that produced a ^{13}C-depleted biomass in the anoxic part of the water column and in the sediment. It is also possible to attribute the negative shift in isotopic composition of carbon to the increased input of effluents from the mountain hut. Finally, as a consequence of several exceptional episodes during this period and multiple processes acting together, ^{13}C-depleted organic matter was sedimented at the bottom of the lake. However, likewise in the deeper sediment sequence, only a slight decrease is observed upward for $\delta^{15}N$ values and the average value of +1.8 ± 0.3‰ shows that variations are in the range of analytical reproducibility (Fig. 3B). The observed $\delta^{15}N$ values indicate compensation for the ^{15}N depletion of DIN from atmospheric sources by vegetation and soil cover (see Comparison of eutrophication effects on the lakes) and consequently the isotopic composition of sedimented nitrogen was not changed.

In the upper 4 cm of the sediment that accumulated after 1987, a rapid increase in OC and TN accumulation rates as well as $\delta^{13}C_{org}$ values was observed (Fig. 3A,B). The rapid increase in OC and TN accumulation rates is most probably related to the increased eutrophication. The C : N ratios averaged 10 and suggest that after 1990 autochthonous and allochthonous sources of organic matter were relatively important, since sedimentary atomic C : N ratios are higher than C : N ratios in plankton and bacteria, which usually range from 5 to 8 (Meyers 1994). However, the values are lower than C : N ratios in the most abundant terrestrial plants from the lake watershed that represent the allochthonous sources and have an average ratio of 26 (Cermelj unpubl.). Thus the C : N ratio in surface sediment indicates that the autochthonous contribution to sedimentary organic matter is more than 75% (Muri et al. 2004). The explanation for the increase in $\delta^{13}C_{org}$ values is more complicated. According to a model proposed by Hollander and Smith (2001), during severe eutrophication seasonal nutrification of surface waters is so intense that the flux of ^{13}C-enriched photoautotrophic biomass overwhelms all other biological sources contributing to the total sedimentary carbon pool. Furthermore, anaerobic decomposition of sedimentary organic carbon via methanogenesis can influence the isotopic signal considerably (Gu et al. 2004). As a consequence, the $\delta^{13}C_{org}$ values become more positive as the process intensifies. However, a much lighter isotopic composition (−35.1 ± 0.4‰) was observed in the upper 4 cm in some other sediment cores from this lake that were used for pore water extraction (Vreca 2003). In the sediment cores collected in 1998 no isotopic shift was observed in the surface sediment, whereas in the core collected in 1999 a positive shift was observed. The results reported in Vreca (2003) represent the mean isotopic composition of thicker segments but show a similar trend to lighter values in surface sediment. Yet a positive shift was also observed in the core collected in November 1999 when $\delta^{13}C_{org}$ value reached −33.1‰ in surface sediment (Vreca unpubl.). During this period the bottom concentration of O_2 reached 7.9 mg L^{-1} and high DIC production was observed (Vreca 2003). Thus, variations in isotopic composition of carbon in surface sediments of several cores collected in different years and different seasons of the year were observed and can be explained by a very dynamic and active sediment–water interface and the upper few centimeters of the sediment where sediment organic matter remineralization is limited by the supply of electron acceptors, e.g., O_2 (Vreca 2003) and the presence of methanotrophs with low isotopic composition (−68‰, Ogrinc pers. comm.), thus considerably influencing the $\delta^{13}C_{org}$ signal. In contrast, $\delta^{15}N$ values are slightly lower than in deeper sediments and again quite uniform, with an average value of +1.4 ± 0.2‰, which is similar to the average values observed by Vreca (2000) in sediment traps (+1.5 ± 0.4‰).

A study of lipids in Lake Planina sediments also confirmed that the lake has become more eutrophic recently (Muri et al. 2004). Shorter-chain lipids that are largely of autochthonous origin prevailed in surface sediments. *Anteiso*-C_{15} and C_{17} fatty acids were present in higher concentrations than their iso-homologues, which is characteristic of productive lakes (Cranwell 1973). The ratios of longer-chain to shorter-chain lipids that can be used to assess the relative contributions of allochthonous versus autochthonous components (Meyers 1997) were also the lowest in the upper 5 cm of the sediments. Although it is important to recognize that shorter-chain lipids are more susceptible to degradation than their longer-chain homologues, thus affecting their sedimentary distribution, it was nevertheless concluded that the high concentrations of shorter-chain lipids and the low ratios of longer- versus shorter-chain lipids in the upper sediment resulted from increased eutrophication that was the most intensive in the last ∼15 yr.

Comparison of eutrophication effects on the lakes—Small and shallow mountain Lakes Ledvica and Planina are both of glacial origin and are located in the same air shed with similar catchments lithology but at different altitudes. Progressive environmental changes in elemental and isotopic records of sedimentary organic matter together with major changes in diatom and cladoceran abundance are summarized in Table 3. The short core sediment records from both lakes can be divided into three periods showing moderate (Lake Ledvica) to severe eutrophication (Lake Planina) in recent sediments.

In the sediment of both lakes, similar OC and TN concentrations were observed (Table 2), but their accumulation rates were much lower in oligo/mesotrophic Lake Ledvica than in eutrophic Lake Planina (Figs. 2A, 3A). Rather high OC and TN concentrations in both lakes are generally characteristic of mountain lakes. This can be explained by generally low sedimentation rates and slower decomposition of organic matter in the water column, because mountain lakes are mainly small and shallow ecosystems with low water temperatures (Vreca 2003; Muri and Simcic 2004). As a consequence, sinking times of organic matter are short and a relatively higher proportion of organic matter can reach the sediment, accounting for higher OC and TN concentrations. Nevertheless, mass accumulation rates are better measures of changes in organic matter than concentrations because they compensate for changes in bulk sedimentation rates (Tenzer et al. 1999).

Furthermore, C : N ratios were low, indicating that inputs from terrestrial sources were relatively small in both lakes. However, lower C : N ratios were observed in Lake Ledvica than in Lake Planina. The latter lake is located below the tree line, resulting in a higher and relatively constant contribution of allochtonous organic matter compared to Lake Ledvica. In contrast, C : N ratios were more variable for Lake Ledvica, most likely due to several sudden inputs of terrestrial organic matter having higher C : N ratios (>20), and thus substantially changing the generally low Lake Ledvica C : N ratios. In addition, the progressive decrease in C : N ratio indicated that during the last century the autochthonous contribution increased in Lake Planina.

Distinct differences between the two lakes were also observed for the isotopic records (Fig. 4). The absolute range in sedimentary $\delta^{13}C_{org}$ values for each lake was approximately 6‰ and a difference of up to 9‰ was observed between the sediment cores (Figs. 2B, 3B, 4). $\delta^{13}C_{org}$ values are more positive in Lake Ledvica but decreased considerably during the last century and in surface sediment approach values observed in the deeper sediments from Lake Planina. The lowest values in Lake Planina were observed at the end of the period when natural events (i.e., forest fire, earthquake) and anthropogenic activities (i.e., introduction of fish, sewage effluents from the mountain hut) affected the carbon cycle within the lake. The differences in $\delta^{13}C_{org}$ values between the lakes could be related to (1) different sources of organic carbon as a consequence of altitude and vegetation cover differences, (2) different biogeochemical processes in oligo/mesotrophic and eutrophic lakes, (3) influence of ^{13}C-depleted atmospheric CO_2 due to fossil fuel burning, and (4) different sources of DIC in lakes at different altitudes. The $\delta^{13}C_{org}$ profiles show a similar trend to lighter values in sediments deposited until the mid 1980s but in the surface sediment from Lake Planina heavier values were observed. However, some other sediment cores from Lake Planina collected in the dysoxic/anoxic zone of the lake showed a trend to more depleted values also in surface sediment (Vreca 2003). These results are consistent with the possible influence of isotopically depleted atmospheric CO_2 on terrestrial and aquatic vegetation (Rau 1978) that is probably seen in Lake Ledvica. Despite known carbon sources, the seasonal and annual dynamics of carbon biogeochemical cycling (particularly at the sediment–water interface) complicates interpretation of $\delta^{13}C_{org}$ values in Lake Planina. Furthermore, comparison of all investigated short cores indicates that the sediments from this small lake are not homogeneous, resulting in variations of $\delta^{13}C_{org}$ values in different sediment cores but having similar overall trends.

Furthermore, DIC considerably influences the carbon cycle in carbonate environments. At high altitudes where vegetation cover is minor, ^{13}C-enriched DIC indicates that DIC is mainly produced through weathering of carbonates. As reported by Brencic (1998), springs from upper Triassic carbonate aquifers in Slovenia with a recharge area above the tree line have an isotopic composition of DIC between −4 and −6‰. The catchment area of Lake Ledvica is also composed of such carbonates; therefore we can assume that the groundwater inflow into the lake has a similar isotopic composition of DIC. In contrast, in springs with a recharge area below the tree line, lighter $\delta^{13}C_{DIC}$ values are consistent with production of DIC by carbonate weathering with CO_2 derived from the oxidation of soil organic matter. Therefore, $\delta^{13}C_{DIC}$ values between −12.6 and −14.4‰ were observed in the spring above Lake Planina (Vreca unpubl.). Thus a 6–10‰ difference in isotopic composition of DIC at various altitudes could explain the difference in isotopic composition of sedimentary organic carbon between the lakes.

Differences were also observed in the nitrogen isotopic records (Fig. 4). ^{15}N values were around $+2$‰ in sediments deposited before 1910. A 5‰ upward decrease in δ^{15}N values was observed in the sediment record from Lake Ledvica (Fig. 2B), whereas in Lake Planina no distinct change in nitrogen isotopic record was observed (Fig. 3B). The progressive ^{15}N depletion in Lake Ledvica shows that sources of DIN have probably changed during the last century. The main source of DIN in the lake situated at the level of the tree line is atmospheric deposition. Snow is present around Lake Ledvica for at least 7 months per year (Brancelj 2002). As reported by Pichlmayer et al. (1998), a characteristic decrease of δ^{15}N values in snow pack in the west to east direction was observed at high-elevation alpine sites in Austria. Furthermore, a much heavier isotopic composition of nitrogen was observed in ice older than 100 yr (δ^{15}N $= +4.8$‰) than in recent snow samples (-1.9 to -5.5‰). The marked decrease in isotopic composition is accompanied by a concurrent increase of nitrate concentrations in atmospheric deposition (Pichlmayer et al. 1998). If such isotopically lighter DIN had been available to primary producers during the last century, the δ^{15}N of the lake's biomass could have become more negative. In contrast, the nitrogen record in Lake Planina shows no distinct change during the last century. The runoff in the recharge area of Lake Planina is related to vegetation and soil cover around the lake and is thus different from that in the recharge area of Lake Ledvica (Brencic pers. comm.). Consequently, compensation of ^{15}N-depleted atmospheric DIN by vegetation and soil, as well as by increased primary production in the epilimnion, is possible.

The data obtained indicate that variations of carbon and nitrogen cycles in both lakes changed considerably in the last century because of natural and anthropogenic influences, and provide new insight into the interpretation of sedimentary records from mountain lakes. However, more detailed investigations are needed for complete understanding of carbon and nitrogen cycles in such lakes.

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