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C and N elemental and stable isotopic signatures in sedimentary organic matter from Lake Pamvotis (Greece) and Lake Bohinj (Slovenia)

C in N elementna in izotopska sestava sedimentirane organske snovi iz jezera Pamvotis (Grčija) in Bohinjskega jezera (Slovenija)

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Ključne besede: jezerski sedimenti, organska snov, ogljik, dušik, stabilni izotopi, Grčija, Slovenija

Abstract

Organic carbon (C_{org}) and total nitrogen (N) concentrations in combination with stable organic carbon ($\delta^{13}C_{org}$) and nitrogen ($\delta^{15}N$) isotopic composition were used to investigate the organic matter sources and influence of anthropogenic activities on recent sediments of eutrophic Lake Pamvotis (Greece) and oligotrophic Lake Bohinj (Slovenia). C_{org} and N concentrations increase upcore showing a progressive nutrient enrichment in both lakes. The C/N ratios vary with depth and indicate that allochthonous and autochthonous sources contributed to sedimentary organic matter. Nevertheless, an excursion to relatively low surface sediment C/N ratios suggests that recently the latter predominates. Finally, $\delta^{13}C_{org}$ and $\delta^{15}N$ records showed changes in carbon and nitrogen cycles related predominantly to local anthropogenic activities in Lake Pamvotis and to anthropogenic activities (i.e. woodcutting) and global atmospheric changes in Lake Bohinj.

Izvleček

Na osnovi določitev koncentracij organskega ogljika (C_{org}) in celotnega dušika (N) ter njune izotopske sestave ($\delta^{13}C_{org}$, $\delta^{15}N$) smo raziskali izvore organske snovi in vpliv antropogenih aktivnosti na recentne sedimente evtrofnega jezera Pamvotis (Grčija) in oligotrofnega Bohinjskega jezera (Slovenija). Koncentracije C_{org} in N, ki naraščajo proti površini sedimenta v obeh jezerih, nakazujejo postopno povečanje vnosa hranil. Razmerje C/N se z globino spreminja ter nakazuje spreminjanje deleža alohtone in avtohtone komponente v sedimentirani organski snovi, vendar relativno nizka razmerja C/N v površinskem sedimentu nakazujejo na povečan vpliv avtohtone komponente. Spreminjanje $\delta^{13}C_{org}$ in $\delta^{15}N$ z globino nakazuje spremembe v kroženju ogljika in dušika, ki so v jezeru Pamvotis predvsem posledica lokalnih antropogenih dejavnosti, v Bohinjskem jezeru pa jih lahko pripišemo antropogenim vplivom (sekanje gozdov) in globalnim vplivom preko atmosfere.

Introduction

Lakes represent very sensitive ecosystems and interesting sedimentation environments. The biggest concern related to lakes is induced eutrophication caused by anthropogenic activities. Despite 20th century advances in understanding eutrophication, it remains one of the foremost problems in protecting freshwater ecosystems (Schindler, 2006). It is now well known that anthropogenic nutrient loading of aquatic ecosystems from both point and nonpoint sources typically results in rapid a increase in the rate of biological production and a significant reduction in water column transparency, and can create a wide range of undesirable water quality changes in freshwater ecosystems (Smith et al., 2006 and references therein). Two of the most consistent eutrophication effects are an excursion in algal species composition and an increase in the frequency and intensity of nuisance algal blooms.

Furthermore, inflow of toxic elements and organic pollutants together with nutrient enrichment influences the ecosystem quality and typically results in significant alterations in biogeochemical cycling over space and time.

According to the EU Water Framework Directive (WFD 00/60/EC), the most important EU document in water management, lakes are classified as surface water bodies. For these, the achievement of good water status by 2015, including restoration of previous water status, the prevention of further deterioration, the reduction of polluting sources and finally the promotion of balanced and sustainable water use, is required. The rapid control of eutrophication by reducing point sources of phosphorus is one of the clearest cases where science has contributed to quickly solving a major management problem. However, loading of nutrients and other chemicals from nonpoint, diffusive sources like agriculture and atmospheric deposition, their accumulation in sediment and recycling between sediments (especially in the anoxic part of the sediments) and water remains a problem that has to be solved (Schindler, 2006). Therefore, the control of eutrophication remains one of the greatest challenges to limnologists and transfer of the knowledge on the complexity of the problem to policy makers the most important task in the future.

The organic matter accumulated in lake sediments represents 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 & 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. Additional contributions from more distant sources are delivered by precipitation and wind. On the other hand, biota within the water column and sediments contribute aquatic (autochthonous) organic matter. The different types of organic matter have a distinctive biochemical composition (Meyers & 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 and nitrogen isotopic ratios are used to identify different sources of sedimentary organic matter. 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 elements in the water column of lakes (e.g., Deines, 1980, Schelske & Hodell, 1991, Herczeg et al., 2001, Lehmann et al., 2002).

In this study, organic carbon (C_{org}) and total nitrogen (N) concentrations and stable organic carbon ($\delta^{13}C_{org}$) and nitrogen isotopes ($\delta^{15}N$) were determined in sediments of lakes Pamvotis and Bohinj located in the NW of Greece and NW Slovenia, respectively. The former is eutrophic, while the latter is still oligotrophic. In the catchment area of both lakes carbonate rocks prevail. Both lakes represent an important natural heritage and therefore, knowledge on past changes recorded in the sediment is needed. The main objectives were 1) to determine the sources of sedimentary organic matter, 2) to investigate their depositional history and 3) to attempt to identify anthropogenic imprints on the sedimentary records of these two different lakes.

Materials and methods

Study site

<u>Lake Pamvotis</u> is situated at 470 m a.s.l in north-western Greece. The biggest city in the vicinity is Ioannina (Figure 1) with approx. 150 000 inhabitants. The lake has a surface of 22.8 km^2 with a maximum depth of 8 m. The catchment of the lake consists of limestones, cherts and flysch. The fine (silty to silty clay) surface sediment consists of carbonates, quarz, feldspars and clay minerals. The size of the lake was dramaticaly reduced by agricultural drainage schemes, which begun around 1600 and culminated in the drilling of a tunnel to the west in 1944. There are no major fluvial inflows and outflows but only ephemeral streams on the surrounding hills with most precipitation percolating directly into the bedrock (Lawson et al., 2004 and references therein). The area recives on average 1200 mm of precipitation per year due to orographic uplift of moist warm air from the Adriatic (Lawson et al., 2004). The catchment has undergone substantial agricultural, industrial and urban development over the past 40 years which has resulted in lake eutrophication. A decrease in the external load of nutrients and pollutants occurred in the 1990s with the removal of the Ioannina sewage outfall

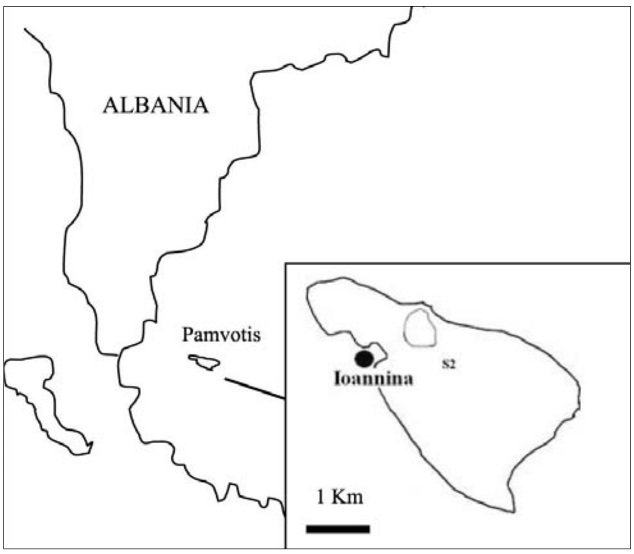

Figure 1. Location map of Ioannina and Lake Pamvotis

from the lake along with industrial discharges on the southern shoreline. Biological analyses in lake Pamvotis showed that chlorophytes and diatoms are the most abundant algae, while among zooplankton copepods, cladoserans and rotifers prevail (Romero et al., 2002, Kagalou et al., 2003). Heavy surface blooms of planctonic cyanobacteria were also observed (Kagalou et al., 2008). The lake is classified as eutrophic (Kagalou et al., 2003, Lawson et al., 2004 and references therein).

Lake Bohinj is situated at 526 m a.s.l. in the Triglav National Park in the Julian Alps in north-western Slovenia (Figure 2). Lake Bohinj, with a surface of 3.28 km^2 and a maximal depth of 45 m, is the largest natural lake in Slovenia, located in glacial valley surrounded by high mountains and is a typical subalpine ecosystem of great environmental importance. The catchment area is predominantly composed of carbonate rocks, i.e. limestones and dolomites. The silty surface sediment consists predominatly of carbonates, while quarz and feldspars represent a minor fraction (Molnar et al., 1978). Recharge of the lake is represented by surface waters with relatively stable inflow. However, an important part of lake hydrological balance is represented by underground water recharge from the karstified catchment area. Consequently, the lake has a relatively short retention time of water that is estimated to be approximately 0.3 years (ARSO, 2007). The area recives on average 3300 mm of precipitation per year. Past anthropogenic activities like ironworks until the mid 19th century and increasing tourism during the last forty years certainly influenced the lake ecosystem and are recorded in the sediment (Molnar et al., 1978). Biological analyses in the water column are also regularly performed during monitoring activities in Lake Bohinj. In general, diatoms (i.e., *Bacyllaryophyceae*) and green algae (i.e., Chlorophyta) are commonly the most abundant algae in spring, while in autumn dinoflagellates (i.e., Dinophyta) usually prevail (ARSO, 2007). According to the results of monitoring surface water quality performed since 1992 (Uhan & Bat, 2003) by the Environmental Agency of the Republic of Slovenia, Lake Bohinj is classified as oligotrophic but ocasionally shows some characteristics of a mesotrophic lake (e.g. in increased concentrations of inorganic nitrogen) (ARSO, 2007).

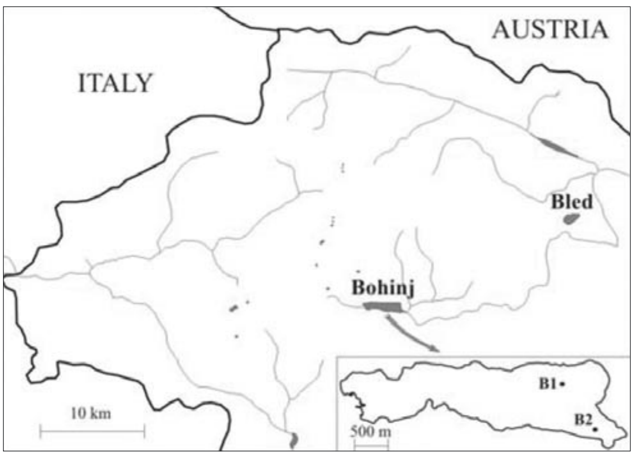

Figure 2. Location map of north-western Slovenia and Lake Bohinj

Sampling

Lake Pamvotis - Core samples were collected by gravity corer at sampling site in the deepest, SE part of the lake (S2) during the autumn overturn in November 2005 (Dascalou, 2007). One core was cut into 1 cm segments in the field, stored in plastic containers and frozen.

Lake Bohinj – Core samples were collected by scuba diver at two sampling sites in the easteren part of the lake (in Fužinski zaliv at the depth of 40 m – B1; and in the SE part at the depth of 15 m – B2) during the autumn overturn in October 2005. One core from each sampling site was cut into 1 cm segments in the field, stored in plastic containers and frozen. The sediment samples were lyophilized and stored until analysis.

Analyses

C_{org} and N concentrations were measured after removal of inorganic carbon (carbonate). Dry sediment samples were acidified with HCl (Hedges & Stern, 1984). The acid was added until effervescence upon acid addition ceased, indicating complete removal of inorganic carbon. Finally, the C_{org} and N concentrations were determined using a CHN elemental analyzer (Carlo Erba EA 1008) at the Marine Biology Station in Piran. The precision of the method, expressed in terms of standard deviation, was from 3 to 5%.

Furthermore, a portion of the dry sediment samples was soaked in 1 mol/L HCl overnight to remove carbonates, and then filtered on a quartz-fibre filter (Whatman GF/F), rinsed with deionized water, and redried. Subsamples of a few mg were placed in tin capsules and the stable isotopic composition of organic carbon was determined on a continuous-flow Europa Scientific 20-20 ANCA-SL isotope ratio mass spectrometer at the Jožef Stefan Institute. All subsamples were analysed 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 ± 0.1‰, determined by repeated analysis of reference materials (IAEA-CH-7). Sample repeatability was ± 0.1‰ or better. For determination of the isotopic composition of nitrogen, homogenized bulk sample material was placed into tin capsules and measured on a Finnigan DELTA^{plus} XP continuous flow stable isotope mass spectrometer with a Eurovektor CHNS Elemental Analyser at the Institute of Water Resources Management, Graz. 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 ± 0.1‰, determined by repeated analysis of reference materials (IAEA-N1, IAEA-N2 and USGS25). Sample repeatability was ± 0.1‰ or better.

Results and discussion

C_{org} in the sediment was found to decrease with the depth of the sediment as shown in Figure 3. C_{org} content varied between 37 and 94 mg/g in the sediment of Lake Pamvotis, while in the sediments of Lake Bohinj lower C_{org} contents were observed, i.e. from 5 to 58 mg/g at B1 and from 44 to 71 mg/g at B2. N content followed the changes in C_{org} content and varied between 3 to 7 mg/g in Lake Pamvotis. In Lake Bohinj, N content also ranged from 4 to 6 at B2, while at B1 lower concentrations were observed ranging from 1 to 6 (Figure 3). In Lake Bohinj, higher C_{org} and N contents are characteristic of sampling point B2 in comparison to B1. The differences could be attributed to the location of sampling point B2 which was closer to the shore and in the shallower SE part of the lake. In this area, a relatively higher proportion of allochthonous, more refractory, organic matter could be delivered to the lake as inferred by higher surface sediment C/N ratios at B2 (see discussion below), thus reflecting the higher surface sediment C_{org} content at B2. Furthermore, the influence of human activities (e.g. tourism, traffic) could have some influence on sediment at B2. On the other hand, a shorter sinking time of organic matter could also enable a higher proportion of organic matter to settle to the bottom of the lake at B2. In contrast, at B1 the deeper water column depth (i.e., 40 m) would favour a longer sinking time, and thus higher degradation of sinking organic matter. The increasing C_{org} and N content with time in both lakes is most probably a result of progressive nutrient enrichment and also the eutrophication process caused by changes in the past trophic state, inferred by natural development of the lake ecosystems (Vreča & Muri, 2006), as well as human activities that were intense, especially in the surroundings of Lake Pamvotis (Kagalou et al, 2003, Lawson et al., 2004).

The C/N ratios varied with sediment depth, i.e. from 10 to 16 in Lake Pamvotis, and from 13 to 16 and 10 to 20, at B2 and B1 in Lake Bohinj, respectively (Figure 3). The results obtained indicate that allochthonous and autochthonous sources contribute to sedimentary organic matter. Nevertheless, the relatively low surface sediment C/N ratios amounting to 12 in Lake Pamvotis, 13 at B1 and 14 at B2 in Lake Bohinj suggest that autochthonous sources could still predominate over allochthonous sources. Similar C/N values were observed in Lake Pamvotis (sampling point S2) and in Lake Bohinj at sampling point B2, while at sampling point B1 C/N ratios were markedly higher in the deeper sediments. The latter could be attributed to intense woodcutting (Molnar et al., 1978) that increased the contribution of allochthonous organic matter to the sediments during past operation of ironworks in this area. The excursions of C/N ratios suggest that sources of organic matter in both lakes have most probably changed in the time frames represented by the cores.

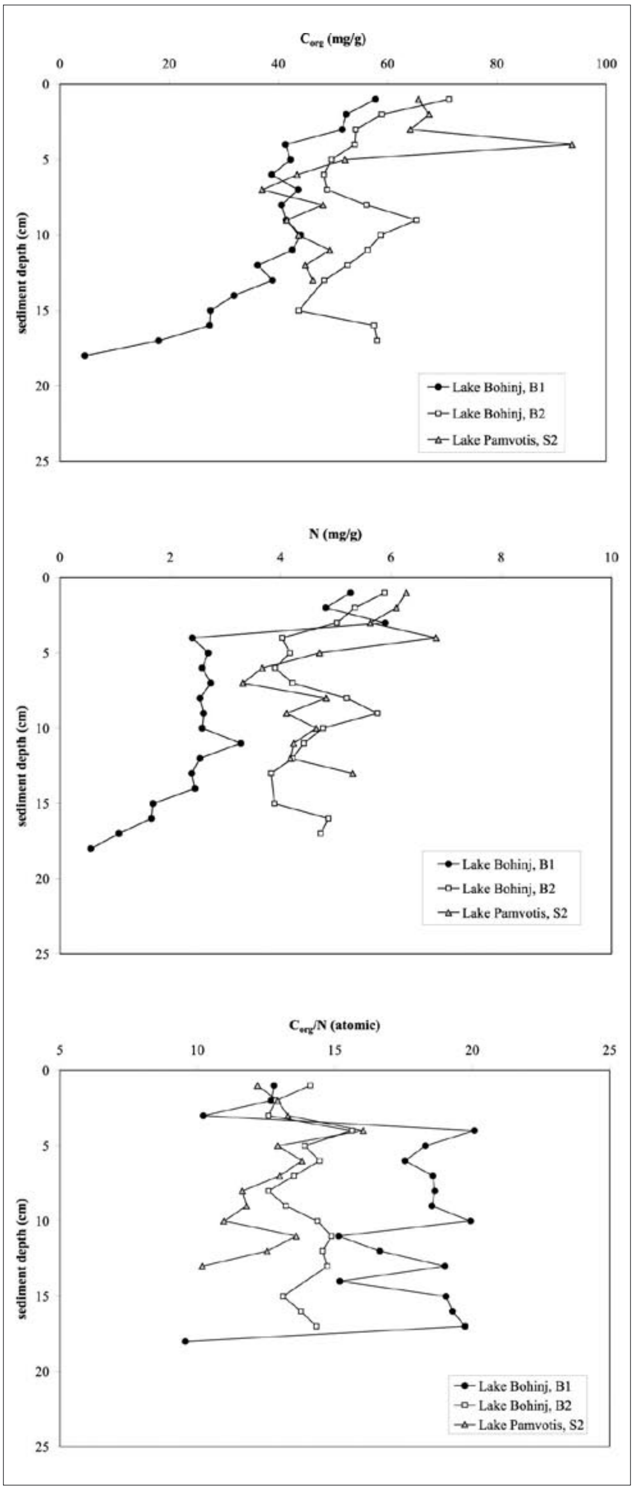

Figure 3. Changes in C_{org}, N and C/N ratio with depth

The isotopic composition of sedimentary organic carbon ($\delta^{13}C_{org}$) was found to increase with depth (Figure 4) from -27.3 to -22.7‰ at sampling point S2, from -26.5 to-27.7‰ at B1 and from -27.8 to -28.6‰ at B2. The most profound trend was observed in Lake Pamvotis. The explanation of the decrease of $\delta^{13}C_{org}$ values by 4.6‰ and 1‰ in Lakes Pamvotis and Bohinj is very complex. Changes could be explained either by introduction of terrestrial organic matter and soil with lighter isotopic composition, accompanied by consumption of ^{13}C-depleted CO_2 formed during

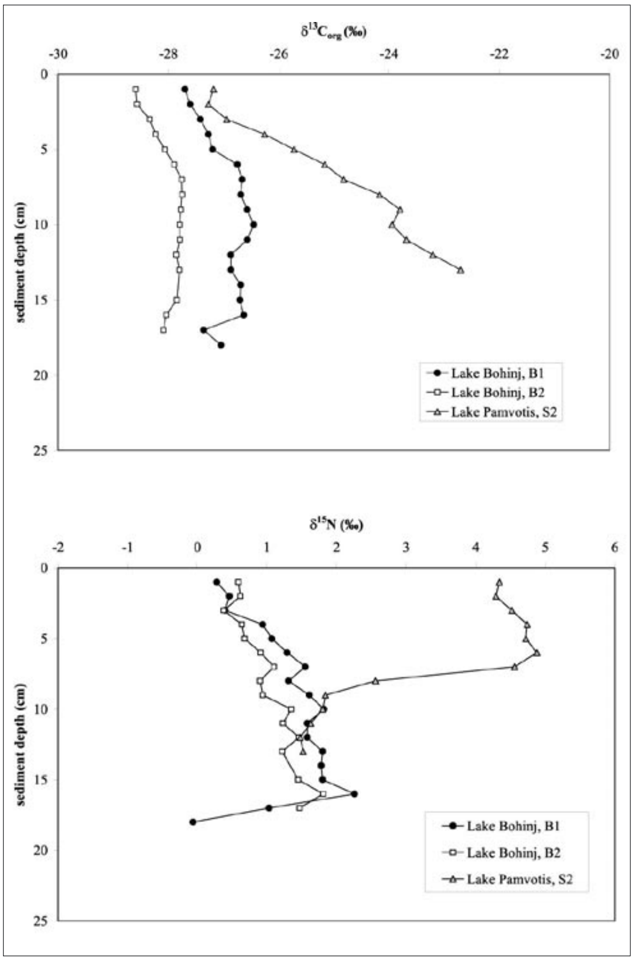

Figure 4. Changes in isotopic composition of organic carbon ($\delta^{13}C_{org}$) and nitrogen ($\delta^{15}N$) with depth

degradation of sinking organic matter by aquatic biota and excursion in isotopic composition of carbon in terrestrial and aquatic vegetation due to assimilation of isotopically depleted atmospheric CO_2 produced by fossil fuel burning (Rau, 1978, Hollander & Smith, 2001, Vreča & Muri, 2006). Similar negative trends were also observed in other high-mountain lakes in Slovenia (Vreča & Muri, 2002, Vreča & Muri, 2006). It is also possible to attribute the more negative $\delta^{13}C_{org}$ values in the surface sediments to the recycling of organic carbon within the lakes during moderate to severe eutrophication (Hollander & Smith, 2001, Vreča & Muri, 2006). It is assumed that oxidation of ^{13}C-depleted biogenic methane released from the anoxic part of the sediments to the water column contributes isotopically depleted dissolved CO_2 to the dissolved inorganic carbon (DIC) reservoir. If sufficient amounts of biogenic methane are oxidized, the $\delta^{13}C$ values of DIC are shifted to more negative values and assimilation of this DIC would result in the synthesis of ^{13}C-depleted biomass (Vreča & Muri, 2006). The trends typical for methane production in the sediments (Vreča, 2003 and references therein) were observed during pore water investigations from both lakes (Vreča et al., 2007) but more detailed experiments are needed. In shallow Lake Pamvotis the considerable excursion in $\delta^{13}C_{org}$ could also be attributed to intense degradation of organic matter during diagenesis, accompanied by increased input of effluents from the city of Ioannina (Kagalou et al., 2008).

In contrast to changes in $\delta^{13}C_{org}$, the isotopic composition of nitrogen shows different trends for Lakes Pamvotis and Bohinj (Figure 4). The isotopic composition of nitrogen varies between 1.2 and 1.8‰ in the sediments below 10 cm depth in both lakes and reflects the influence of soil nitrogen (allochthonous component) and lacustrine biomass. The mixing of both sources is also confirmed by the C/N ratios. However, in the upper sediments a distinctive increase is observed in Lake Pamvotis, while a slight decrease is recorded in both cores from Lake Bohinj. The changes in $\delta^{15}N$ values indicate that sources of dissolved inorganic nitrogen (DIN) changed during the last centuries in both lakes. The considerable increase in $\delta^{15}N$ values in Lake Pamvotis could be attributed to release of soil nitrogen (Routh et al., 2007), as well as agricultural runoff and sewage input from the city of Ioannina that stopped in the 1990s (Kagalou et al, 2003). In contrast, the decrease in $\delta^{15}N$ values in Lake Bohinj shows a similar trend as observed in other lakes in the area of Triglav National Park (e.g. Lake Ledvica; Vreča & Muri, 2006). The observed changes are believed to represent an ecological response to enhanced regional atmospheric deposition of fixed nitrogen from anthropogenic sources (Wolfe et al., 2001).

Conclusion

In this paper we present the first results on the elemental and isotopic composition of carbon and nitrogen determined in sediments of eutrophic Lake Pamvotis (Greece) and oligotrophic Lake Bohinj (Slovenia). The data obtained show considerable excursions in elemental and isotope sedimentary records that are related to changes in the lake catchments and the lakes themselves. According to the results considerable deterioration has occurred during the last centuries in Lake Pamvotis as a consequence of anthropogenic activities in the surroundings, whereas in Lake Bohinj eutrophication could be promoted by global atmospheric changes. The results obtained so far enabled a first comparison between the two lakes; however, more detailed investigations would be needed to understand the fate and cycling of nutrients in both lakes.

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