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ORIGINAL ARTICLE

Organic geochemical record of increased productivity in Lake Naukuchiyatal, Kumaun Himalayas, India

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Abstract Organic geochemical proxies have been studied in a 45-cm-long core retrieved from Lake Naukuchiyatal in Kumaun Himalayas, India. Increase in TOC, N, hydrocarbons and pigments concentration from bottom to surface sediments of the core indicates increase in the lake productivity. Stable isotopes (δ^{13}C and δ^{15}N), biomarkers (TAR, CPI and n-$\Sigma C_{15,17,19}$) and C/N atomic (between 9 and 12) suggest dominance of algal derived organic matter in these sediments. Decrease in organic δ^{13}C values (between −27 and −31‰) in surface sediments indicate influence of sewage and land runoff in shifting organic δ^{13}C values, whereas low (between −0.23 and 2.2‰) δ^{15}N values together with high pigment concentrations (zeaxanthin and echinenone) represent dominance of cyanobacteria in the lake.

Keywords Organic matter · Lake productivity · Stable isotopes · Hydrocarbons and pigments

Introduction

Lake sediments constitute an important link to study present and past in-lake processes and watershed activities. Sedimentary organic matter in the lake sediments are an important repository and hence they are used widely to study the paleoenvironmental conditions in lakes.

Pioneering studies by Meyers and Ishiwatari (1993), Meyers (1997), Brenner et al. 1999), Meyers and Lallier-Verges (1999), Routh et al. (2007) and many others led to significant research in the reconstruction of paleoenvironmental conditions based on the organic matter content in lake sediments. The useful deductions concerning the geochemical characteristics of organic matter with different origins and histories have been worked out from the geochemical signals in sediment records (Bernasconi et al. 1997; Meyers 2003). Periods of high sedimentation rates and primary productivity, preserve the evidence of short-term processes affecting organic matter delivery and burial in sediments (Urban et al. 1999; Routh et al. 2004).

The Kumaun Himalayan region is characterized by many sub-tropical lakes. During the past few decades, high tourist influx in the region has accelerated various anthropogenic activities (e.g., agriculture, construction, logging, etc.) which have impacted the ecology and water quality of these lakes (Chakrapani 2002; Das 2005; Choudhary et al. 2009a). Naukuchiyatal is one of the major lakes in the region, which has undergone such anthropogenic-related productivity changes in recent years. Several studies have highlighted the issue of eutrophication, heavy metal pollution and high sedimentation rates in some of the lakes in the area (Ali et al. 1999; Chakrapani 2002; Das 2005; Choudhary et al. 2009a). However, these studies do not elucidate the actual source of organic matter, nutrient dynamics or productivity estimates. Hence, the present study is focused on deciphering various organic geochemical proxies in the core sediments retrieved from Lake Naukuchiyatal. To the best of our information, the present study is one of its kind in Indian lake and hence, useful generalizations might be drawn in order to understand various biogeochemical processes in operation within the lakes.

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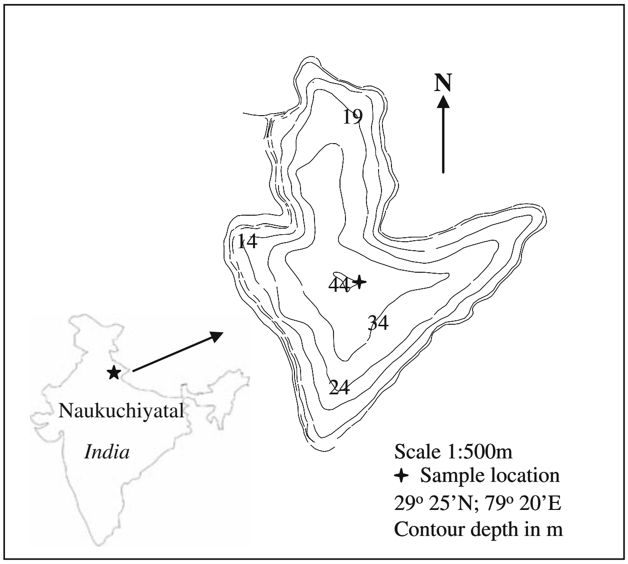


Fig. 1 Bathymetric map of Lake Naukuchiyatal showing the sample location (modified from Nainital Development Authority, India)

Study area

Naukuchiyatal Lake is situated at an altitude of 1,320 m above mean sea level in the Kumaun hills. The total surface and catchment area of the lake which lies between 29 45′ and 30 34′N and 78 45′ and 80 90′E are 0.375 km^2 and 3.2 km^2, respectively (Fig. 1). The basin of the lake is V shaped and has nine corners. The lake is 1,050 m long and 675 m in breadth with a maximum depth of about 42.2 m (Khanna and Jalal 1985). The lake receives an annual average rainfall of 2,420 mm and the temperature ranges from a maximum of 29°C to a minimum 1.8°C. The annual water inflow is of the order of 3.12 million cubic meters and retention time is 2.5 years. The major lithology around the lake area is constituted of metavolcanics and quartzites (Valdiya 1988).

Materials and methods

The sediment core was collected from the deepest part of the lake (Fig. 1) in December 2004. A gravity corer was used to obtain a relatively undisturbed core, 45 cm long and 55 mm in diameter. The core was sliced into 2-cm sections in the field and stored in polythene zip-lock bags under refrigerated condition. The samples were freeze-dried; water content and porosity were estimated in the different sediment intervals. The $C_{organic}$ and N_{total} isotopic composition of the acid-treated samples were analyzed using a continuous flow system consisting of a Carlo Erba elemental analyzer coupled to a Finnigan MAT Delta Plus mass spectrometer. Carbon (C%) and nitrogen (N%) contents in the samples showed standard deviation of <1%.

Isotope data were reported in the conventional delta (δ) notation versus Vienna PeeDee Belemnite V-PDB for C and atmospheric N_2 for total N. Reproducibility of duplicate analyses was 0.1‰. The total organic carbon (TOC) content and sedimentation rate was used to estimate the paleoproductivity (PP) in sediments by using the following equation (Ishiwatari et al. 2005),

$$\mathrm{PP} = (\%\mathrm{TOC} \times D)/\left(0.0030 \times S^{0.3}\right) \tag{1}$$

where D is the dry bulk density (mg cm^{-3}) of the sample and S is the sedimentation rate (cm kyr^{-1}) (Kumar et al. 2007). PP values derived from above equation do not indicate variations in terrigenous input and hence give integrated preserved total organic carbon information. In order to evaluate PP derived from autochthonous organic matter input. PP was also calculated based on the concentration of in-lake algal derived hydrocarbons (n-$\Sigma C_{15,17,19}$). Approximately 1–2 g of freeze-dried sediment was extracted with a mixture of dichloromethane and methanol (9:1 v/v) for the solvent extractable hydrocarbons. A Dionex Automated Solvent Extractor 300 was used (programmed for three extraction cycles at 1,000 psi and 100°C). The extracts were reduced using a Büchli rotovapor and injected in pulsed splitless mode into an Agilent 6890 gas chromatograph equipped with a Programmable Temperature Vaporising Agilent inlet and DB5-MS column (30 m × 0.25 mm i.d. × 0.25 μm film); the oven temperature was maintained at 35°C for 6 min, increased to 300°C at 5°C min^{-1} and held there for 20 min. The chromatograph was interfaced with an Agilent 5973 mass spectrometer operated at 70 eV in full-scan mode (m/z 50–500 amu). External and internal standards (S-4066 from CHIRON, Norway and deuterated perylene from Cambridge Laboratory, USA, respectively) were used for quantification. The hydrocarbons were used to calculate various hydrocarbon ratios in order to characterize the sedimentary organic matter (OM).

Carbon preference index (CPI; Allan and Douglas 1977) representing the predominance of odd over even n-alkanes:

$$\mathrm{CPI} = \frac{\sum (C_{23}-C_{31})_{odd} + \sum (C_{25}-C_{33})_{odd}}{2\sum (C_{24}-C_{32})_{even}}. \tag{2}$$

Terrigenous aquatic ratio (TAR; Bourbonniere and Meyers 1996) quantifying in situ algal versus terrestrial OM:

$$\mathrm{TAR} = \frac{(C_{27} + C_{29} + C_{31})}{(C_{15} + C_{17} + C_{19})}. \tag{3}$$

P_{aq} ratio (Ficken et al. 2000) quantifying the different plant types in the lake (e.g., submerged vs. emergent plant types):

$$P_{aq} = \frac{(C_{23} + C_{25})}{(C_{23} + C_{25} + C_{29} + C_{31})}. \quad (4)$$

Pigments were extracted for 2 min by ultra-sonication in HPLC-grade acetone (2 ml g^{-1} sediment) and stored overnight at 3°C (Bianchi et al. 2000, 2002). After filtration (with 0.02-µm filters; Gelman GHP Acrodisc 13), the samples were injected into an HPLC system consisting of a Waters 2690 separation module coupled to a Waters 996 photodiode array UV/VIS detector (set at 450 nm for absorbance). The injector was connected via a guard column to a RP-18 LiChroCART column (5 µm particle size, 250 × 4.6 mm internal diameter). The gradient (1 ml min^{-1}) program started with 100% mobile phase A (80:20 methanol:0.5 M ammonium acetate v/v ratio) after injection (50 µl; Westman et al. 2003). This was followed by 100% mobile phase B (90:10 acetonitrile:water v/v ratio) for 4 min, and to 25% B and 75% C (100% ethyl acetate) for 28 min. The program was changed to 100% B for 5 min with a final ramping to 100% A for 4 min. The pigment standards were purchased from DHI, Denmark.

Results

TOC contents in the sediments ranged between 3.3 and 6.0%, with a minimum value at 37 cm. Total N% varied between 0.33 and 0.63%, the maximum values (0.54–0.63%) occurred between 19 and 1 cm (Fig. 2). The TOC and N profiles indicated a decrease in values up to 37 cm, after that the values increased gradually towards the top of the core. The atomic C/N ratio measured in Naukuchiyatal sediment was between 9 and 12. The average value in the core was 11 (Fig. 2).

The PP estimates indicated a steady increase in productivity towards the top of the core. The minimum value (408 g C m^{-2} $year^{-1}$) was observed at 45 cm, whereas maximum value (1,149 g C m^{-2} $year^{-1}$) occurred at the depth of 17 cm (Fig. 3). PP calculated by using algal hydrocarbon (*n*-alkane $\Sigma C_{15,17,19}$) ranged between 0.4 and 15 g C m^{-2} $year^{-1}$.

Organic $\delta^{13}C$ values ranged from −27.1 to −31.6‰; the values gradually increased in younger sediments. The $\delta^{15}N$ values fluctuated between 2.1 and −0.23‰ and the maxi-

Fig. 2 Distribution of total organic carbon (TOC), total nitrogen, atomic $C_{organic}/N_{total}$, $\delta^{13}C_{organic}$, $\delta^{15}N_{total}$ and paleoproductivity in Lake Naukuchiyatal

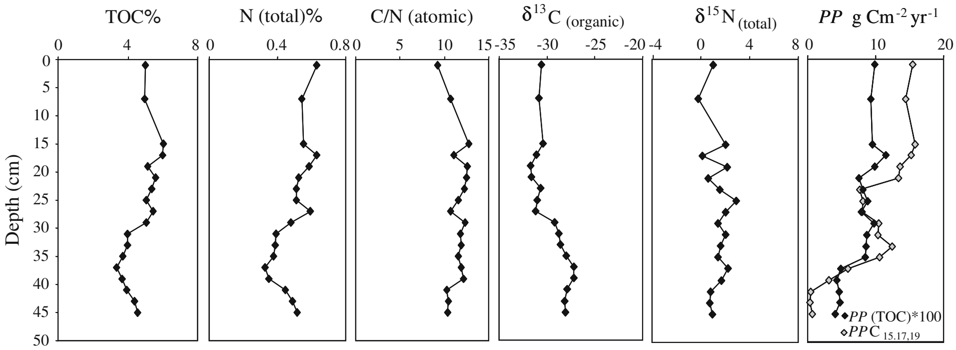
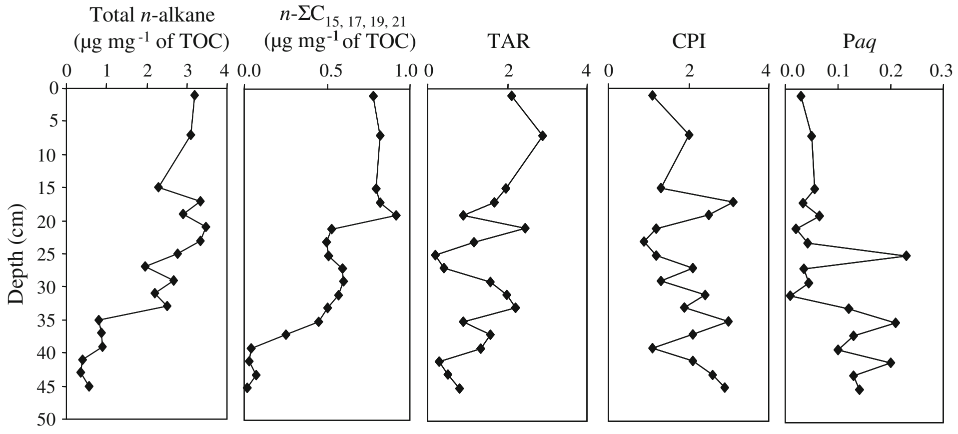


Fig. 3 Total hydrocarbon concentration and ratios (CPI, TAR, P_{aq},) in Lake Naukuchiyatal sediments



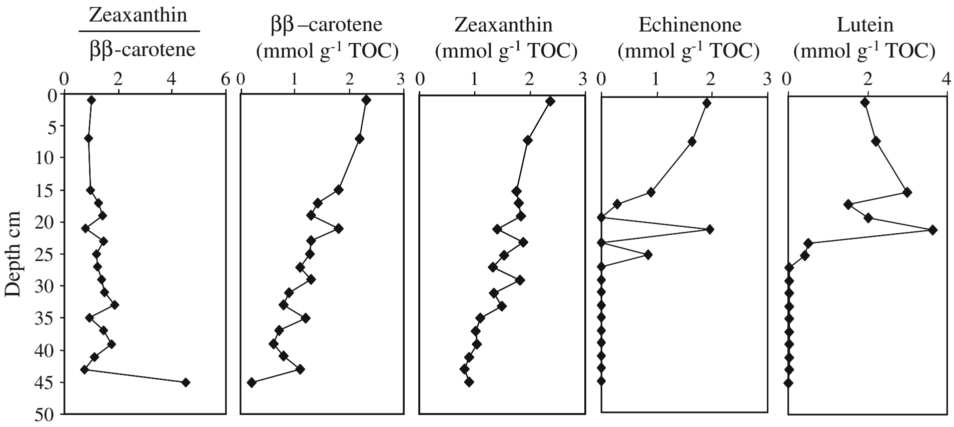


Fig. 4 Distribution of pigments ratios (zeaxanthin/β,β-carotene) and pigment concentrations (β,β-carotene, zeaxanthin, echinenone, and lutein) in Lake Naukuchiyatal

mum δ^{15}N value was measured at 19 cm (2.1‰), whereas minimum (0.1‰) value was recorded at 7 cm.

Total hydrocarbon concentrations varied between 0.05 and 3.3 µg mg^{-1} of TOC (Fig. 3). The $\Sigma C_{15,17,19}$ profile shows similar trend as the total hydrocarbon profile; the minimum (0.02 µg mg^{-1} of TOC) concentration is observed at 45 cm, whereas the maximum values (0.82 µg mg^{-1} of TOC) occurred at 17 and 7 cm, respectively. CPI values fluctuated between 0.9 and 3.1; high (3.1) CPI value was observed at 17 cm. TAR values in Naukuchiyatal sediments was between 0.3 and 2.9, where as *Paq* values varied between 0.02 and 0.23 and showed frequent variation with depth.

Pigment concentrations increased from the base of the core to the surface sediments (Fig. 4). $\beta\beta$-carotene and zeaxanthin were observed throughout the core. $\beta\beta$-carotene concentration varied between 0.2 and 2.3 mmol g^{-1} of TOC. Zeaxanthin varied between 1.0 and 2.4 mmol g^{-1} of TOC and is high in the surface sediments. The ratio between zeaxanthin and $\beta\beta$-carotene ranged between 0.74 and 4.5; the maximum value (4.5) was reported at 45 cm. Echinenone and luetin were absent below 25 cm, but the concentration increased above 25 cm and indicated values between 0.9 and 1.9 mmol g^{-1} of TOC and between 0.41 and 3.6 mmol g^{-1} of TOC, respectively.

Discussion

Elemental concentrations

TOC concentrations in the sediments represent the amount of organic matter which remained undegraded during progressive sedimentation. However, TOC values can be influenced by various factors, e.g., initial production of biomass, rate of sedimentation and post-depositional degradation (Schelske and Hodell 1991; Bourbonniere and Meyers 1996; Routh et al. 2004). TOC values in Naukuchiyatal sediments ranged between 3 and 6%. The values gradually decreased from 45 to 37 cm and co-vary with PP estimates based on both TOC and algal hydrocarbons (Fig. 2), which indicate that low TOC values in bottom sediments are related to low productivity. Consistent with this, these sediment layers indicate a decrease in N which suggests low nutrient flux and hence low productivity. However, post-depositional microbial degradation can also alter TOC values. Hodell and Schelske (1998) indicate limited organic matter degradation especially under anoxic conditions in the deeper sediments of Lake Ontario. TOC concentration together with PP values gradually increased above 37 cm and reached a maximum towards the surface sediments (Fig. 2). These values coincide with high N, which showed significant increase in recent years. Recent studies indicate increase in anthropogenic activities and considerable input of nutrients in the Kumaun Himalayan lakes (Chakrapani 2002; Das 2005; Choudhary et al. 2009a, b), implying that nutrient loading in recent years have impacted the productivity and consequently high TOC in the sediments. The increased pigment concentration in these sediments also provides additional evidence of increased productivity.

Atomic C_{org}/N_{total}

Atomic C_{org}/N_{total} ratio was determined from the elemental analyses of organic matter. The contribution of in situ aquatic organic matter compared to land plants in sedimentary organic matter can be distinguished by the C/N ratio (Meyers 1994; Routh et al. 2004, 2007). Land plants show atomic C/N ratio greater or equal to 20, whereas algae represent values between 4 and 10 (Meyers and Ishiwatari 1993; Meyers 1994; Talbot 2001). However, diagenetic processes can alter the C_{org}/N_{total} of sedimented organic matter (Brenner et al. 1999). Naukuchiyatal sediments represent atomic C_{org}/N_{total} ratio between 9 and 12 (Fig. 2). These values suggest that organic matter in Naukuchiyatal is primarily derived from in-lake algal production. Nonetheless, the presence of inorganic nitrogen can alter the C/N ratio and mislead organic matter source determination (Talbot 2001). The zero intercept of the regression line of the scatter plot between Ntotal versus Corganic show the absence of inorganic N. Hence, the use of atomic C_{org}/N_{total} in Naukuchiyatal sediments and source interpretation is acceptable.

Stable isotopes

Freshwater plankton and land plants which use common source of dissolved CO_2 (atmospheric) are isotopically indistinguishable from organic matter derived from the watershed (Meyers and Lallier-Verges 1999; Meyers and Teranes 2001). However, organic $\delta^{13}C$ values can be influenced by various factors, e.g., DIC sources in freshwater (dissolution of carbonate rocks from the catchment and land runoff), primary productivity and recycling of $\delta^{13}C$ depleted carbon from decay of organic matter in the water column and sediments (Gu and Schelske 1996; Hoefs 1997; Kendall et al. 2001).

Organic $\delta^{13}C$ values in Naukuchiyatal sediment range between −31‰ and −27‰ (Fig. 2). The values remain constant upto 37 cm from bottom upwards, and represent algal dominated organic matter. However, after 37 cm the $\delta^{13}C$ values decrease towards surface sediments, whereas PP values show steady increase in productivity. Generally, high productivity is correlated to increased $\delta^{13}C$ values in organic matter (Meyers 1997; Law et al. 1997, 1998, Teranes and Bernasconi 2000). However, the gradual decrease in $\delta^{13}C$ values in Naukuchiyatal sediments can be explained by various lines of evidence: (a) input of lighter DIC in the lake by land runoff and sewage input (Rosenmeier et al. 2004) and, (b) recycling of ^{13}C by methane production under eutrophic condition (Gu and Schelske 1996; Hoefs 1997).

This interpretation is consistent with recent studies by Choudhary et al. 2009a in the nearby lakes Nainital and Sattal. A similar phenomenon was also observed in Lake Petén Itzá (Guatemala), where sewage input in lake water resulted in $\delta^{13}C$ depleted organic matter (Rosenmeier et al. 2004). However, due to lack of evidence for methane production in Lake Naukuchiyatal, the recycling of ^{13}C in affecting organic $\delta^{13}C$ values remains speculative.

Dissolved nitrate represent $\delta^{15}N$ values between +7 and +10‰. Phytoplankton have $\delta^{15}N$ values of about +8‰, whereas terrestrial plants show values between 0 and +2‰ (Meyers 1997, 2003). However, the exception to this generalization can be visualized during eutrophic conditions, when organic matter produced by cyanobacteria represents isotopically lighter values ($\delta^{15}N$ values to −1 to +3‰; Brenner et al. 1999; Talbot and Laerdal 2000). $\delta^{15}N$ values in the sediments fluctuate between −0.2 and 2.2‰ (Fig. 2).

The $\delta^{15}N$ values in Lake Naukuchiyatal are low and represent an average value of 1.3‰. The low $\delta^{15}N$ values in Naukuchiyatal sediments characterize the range of organic matter produced by cyanobacteria (Brenner et al. 1999; Talbot and Laerdal 2000). Interestingly, cyanobacterial pigments measured in these sediments show a consistent increase in concentration in surface sediments. Hence, we infer the low $\delta^{15}N$ values in these sediments are most likely the results of the presence of isotopically light organic matter produced by cyanobacteria.

Hydrocarbons

Distinctive sources of biotic hydrocarbons in the sediments can be used to draw the changes associated with organic matter flux. Aquatic algae (both macro- and micro-algae) and photosynthetic bacteria are dominated by the $C_{15,17,19}$, whereas vascular plants contain large proportions of C_{27} C_{29}, and C_{31} *n*-alkanes (Cranwell et al. 1987; Tenzer et al. 1999). The abundance of these hydrocarbons over each other reflect different sources of organic matter and paleoproductivity (Rieley et al. 1991; Tenzer et al. 1999).

Total hydrocarbon concentrations in the sediments vary between 0.4 and 3.5 μg mg^{-1} of TOC, the values increase sharply towards the top sediments of the core (Fig. 3). The core represents a low total hydrocarbon concentration in the bottom layers, which indicates low productivity. In addition, the sediments represent low short chain *n*-alkanes ($\Sigma n\text{-}C_{15,17,19}$) concentrations, which signify algal sources of organic matter. The results are consistent with the PP and pigment data and indicate low productivity. After 37 cm, the total hydrocarbon and short chained *n*-alkane (Σn-$C_{15,17,19}$) show a steady increase in concentration towards the surface sediments. The trend is parallel to PP and pigment profile and indicates increased productivity in surface sediments.

Land plants containing *n*-alkanes (Σn-C_{25-31}) show a strong (>5) odd over even carbon predominance, also expressed as the carbon preference index (CPI). In contrast, algal derived organic matter represent low (<1) CPI values (Cranwell et al. 1987; Peters et al. 2005). CPI values in Naukuchiyatal sediments show variability between 0.9 and 3 (Fig. 3). The average value (1.9) of CPI indicates dominance of algal organic matter. Similarly TAR (terrestrial aquatic ratio) value, which is used to identify the *n*-alkane source to understand the changes in terrigenous versus aquatic hydrocarbons (Cranwell et al.1987; Meyers and Ishiwatari 1993) varies between 0.2 and 2.9, and indicates algal derived organic matter.

An *n*-alkane proxy (P_{aq}) was proposed by Ficken et al. (2000), to categorize the sources of submerged and emergent vegetation. P_{aq} values for emergent and submerged freshwater vegetation are generally within the range of 0.07–0.061 and 0.48–0.94, respectively (Ficken et al. 2000; Mead et al. 2005). P_{aq} values in Naukuchiyatal sediments range between 0.02 and 0.23 and indicate dominance of macrophytes. Singh and Gopal (1999) reported a dominance of submerged and floating macrophytes (*Nelumbium nucifera, Myriophyllum spicatum, Ceratophyllum demersum*) in the Naukuchiyatal lake.

Pigments

Algae and higher plants synthesize different kinds of pigments, and the numbers of the pigments are specific for different phytoplankton communities. For example, ubiquitous pigments such as $\beta\beta$-carotene are significant indicators of total algal abundance, whereas taxon-specific carotenoids allow distinction amongst different algal functional groups such as siliceous algae and dinoflagellates (fucoxanthin, peredinin), chlorophytes (lutein, Chl *b*, pheophytin *b*), cyanobacteria (echinenone, zeaxanthin and myxoxanthophyll) and various other prokaryotic groups (Leavitt and Hodgson 2001). However, the role of pigments in interpreting the source and preservation of the phytoplankton community needs to be established and should be handled with caution. Some pigments degrade faster than others, while some are chemically quite stable (Sanger and Crowl 1979; Leavitt 1993). Pigments extracted from Naukuchiyatal sediments indicate dominance of zeaxanthin and $\beta\beta$-carotene (Fig. 4). The zeaxanthin concentration in the sediment varies between 0.8 and 2.4 mmol g^{-1} of TOC and the values increase in surface sediments indicating the dominance of cyanobacteria (*Microcystis*, *Oscillatoria;* Paerl 1988; Leavitt 1993; Bianchi et al. 2000). Echinenone, which is considered as a specific marker for filamentous N_2-fixing cyanobacteria (e.g., *Anabaena*, *Anbaenopsis*; Bianchi et al. 2002) appear after 25 cm and increases towards surface sediments (Fig. 4). Echinenone is comparatively less stable than zeaxanthin and hence the absence of echinenone between 45 and 25 cm can be related to degradation of this pigment or may indicate absence of N_2-fixing cyanobacteria (Bianchi et al. 2002). An increase in cyanobacterial pigments in surface sediments may be associated with a nitrogen-limited and phosphorous-rich environment, as in Nainital Lake (Choudhary et al. 2009a), Norrviken lake (Routh et al. 2008), where increased phosphorous loading resulted in proliferation of cyanobacterial communities. $\beta\beta$-carotene concentration is low (0.20 mmol g^{-1} of TOC) in bottom sediments and show a steady increase (2.3 mmol g^{-1} of TOC) towards surface sediments (Fig. 4). Increase in $\beta\beta$-carotene concentration in surface sediments suggests augment input of green algal organic matter. However, the low ratios (0.73–4) between zeaedxanthin versus $\beta\beta$-carotene suggest dominance of cyanobacteria over other phytoplankton communities.

Lutein appeared after 25 cm and showed maximum values at 21 cm; the presence of this pigment is related to a possible increase in productivity and change in the phytoplankton community (Fig. 4). However, the concentration varied widely with depth suggesting frequent change in chlorophyte species. Furthermore, lutein is considered to be amongst the most labile pigments (Leavitt and Hodgson 2001), thus, the frequent change in concentration may also be related to degradation of this pigment.

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