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Reconstruction of Paleoclimate and Environmental Fluctuations Since the Early Holocene Period Using Organic Matter and C:N Proxy Records: A Review

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ABSTRACT

In this review, the shifts in organic matter (OM) accumulation and C:N ratios in lake sediments to reconstruct paleoclimate and paleo-environmental changes since the early Holocene period are presented. The C:N proxy data of total OM reflect wet climatic conditions during early Holocene (10 to 8.2 kyrs BP) due to enhanced southwest monsoon. This was followed by intermittent arid conditions during the mid and late Holocene period (8.2 to 2.8 kyr BP). Enhanced values of C:N ratio during middle to late Holocene (7.8-2.3 kyrs B.P) indicate periods with lower lake levels and minimum precipitation, while decreased C:N ratio point to stronger SW monsoon and expansion of the lakes. Further, C:N and δ^{13}C results from the lake sediments reveal a detailed and continuous paleo-environmental changes in the relative sources of OM (allochthonous vs autochthonous). Proxy records using such natural archives have also been utilized to reconstruct past extreme events and environmental changes around the lake systems, such as causes for lake desiccation, hydrographic changes, alternations between C_3 and C_4 vegetation and historical disturbances in the catchment area since the early-late Holocene period coupled with the Indian summer monsoon.

INTRODUCTION

Organic rich sediments accumulating at the bottom of suitable climate-sensitive lakes provide continuous and high resolution (inter-annual to decadal scale) records of past climate variability, paleo-environmental changes of the associated catchments (Verschuren, 2003; Baresic et al., 2011). Mineralogical, organic and isotopic composition of lacustrine sediments also provide important information of palaeo-environmental changes, human-induced impacts and help in predicting future trends in environmental evolution following the global and the local/regional factors that influence them.

Production of OM in soils and sediments is directly linked to the intensity of the southwest monsoon (SWM) and is crucial for agricultural productivity, sustainable civilization, land use and land pattern dynamics in India (Staubwasser et al., 2003; Gupta, 2004; Nagasundaram et al., 2014). Early farming and agricultural practices were dependent on the amount and intensity of monsoon rains (Bellwood, 2005; Ruddiman et al., 2008). One of the prime factors for the rise and decline of river valley civilization such as the Harappan was due to the climatic fluctuation, particularly due to the overall decrease in the SW monsoon rains since the middle Holocene period (roughly 7 to 5 kyr BP) (Crawford, 2006). We have compiled from literature, the data on fluctuating OM from early to late Holocene period; data collected on OM and C:N are from six sites (five lakes and one river delta) (Fig.1) that are located in the Indian region receiving rain dominantly from the SWM. Out of these five lakes, two lakes are located on the fringes of the Thar Desert where the climate is mainly controlled by SWM and westerly winds.

The C:N ratio remains constant during periods of higher productivity, but shows variation during periods of lower lake productivity due to the changes in the internal productivity of OM and isotopic changes of carbon and nitrogen in lakes (Routh et al., 2004). Determining the source and amount of OM deposited is useful because it provides a better understanding of the biochemical processes within the lake and its surroundings. The flux and ratio of elements like carbon (C) and nitrogen (N) in an ecosystem are also dependent on the anthropogenic intervention, e.g., land use, management intensity coupled with other environmental factors like precipitation and climatic gradient (Ghaley et al., 2015).

The nature, magnitude and type of organic substances reflected by C:N ratio and δ^{13}C in the lake sediments contribute to a better understanding of paleoclimate and paleoenvironmental records (Meyers, 1994; Meyers and Lallier-Verges, 1999). Since many of a lake's specific nutrient dynamics represent the catchment landscape processes and changes in these processes may serve as indicators of terrestrial processes that are otherwise difficult to detect. OM and C:N ratios in lakes are likely to be altered as a consequence of changes in terrestrial export of nutrients related to climatic fluctuations. Similarly, the change in isotope ratios probably indicates a change in watershed vegetation from C_4 to C_3 plants. Thus, monitoring and understanding the effects of natural and anthropogenic changes in C:N and OM nutrient dynamics is very important because of the multitude of processes operating within and outside the ecosystem. Despite the extensive work on paleolimnological processes, the significance of OM and C:N ratio as climate proxy sources for paleoclimatic reconstruction appears to have remained uncovered so far.

Based on high-resolution geochemical proxy records (C:N, δ^{13}C and OM%) of sediment cores retrieved from lakes, it is aimed to establish the stages of environmental shifts and identify the initial time frame and pattern in which past environment(s) influenced the human activities. Whilst both natural climate change and human-influenced environmental changes are likely to be recorded in the Holocene lake sediments, it is critical to evaluate the influence of human activities on the reliability of sedimentary proxies in inferring the past climate changes (Crawford, 2006; Lu et al., 2009; Li et al., 2012).

ORIGIN AND ACCUMULATION OF OM IN LAKE SEDIMENTS

C:N ratio and δ^{13}C provide information whether OM is derived from terrestrial or lacustrine sources (Meyers and Teranes, 2001; Osleger et al., 2008; Chaudhary et al., 2013). Further, the amount of OM underlying in the sub-surface sediments is a small fraction of that originally ingressed into the surface waters (Ishiwatari and Uzaki, 1987;

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Mahapatra et al., 2011). OM being an important constituent of lake sediment participates in a variety of biochemical and geochemical processes (Meyers and Ishiwatari, 1993; Castaned and Scheouten, 2011) such as microbial reprocessing, sediment-pore water interaction, digenesis along with other redox reactions, in which oxidized inorganic species become reduced by interaction with OM. OM preserved in lake sediments provides key information on lake nutrient dynamics and helps in elucidating the idea of past accumulating sources (terrestrial vs. aquatic) of OM (Meyers, 2003; Das et al., 2007). The concluding result of the multiple processes occurring within the lake system is that the OM in sediments has been often markedly different from that produced by the biota in and around the lakes (Meyers and Lallier-Verges, 1999; Lamb et al., 2007).

C:N RECORDS FROM INDIAN REGION

Changes in the nature of OM accumulation in lake sediments discussed in this review highlights its resolution to serve as a proxy indicator for paleoclimate reconstruction. OM records from the Indian lakes, although a very few, are very detailed (Prasad et al., 1997; Enzel et al., 1999; Chakraborty et al., 2006; Menzel et al., 2014) to assess the paleo-climate changes within Indian subcontinent, particularly in relation to the fluctuations in the intensity of the SWM. The selected lakes of the Indian region fall in the pathway of SWM (Fig.1a). A couple of them such as the Nal Sarovar and the Lunkaransar lie along the arid margins of the Indian Thar Desert and receive less annual average precipitation of the SWM and were thus also selected to investigate the effect of changing monsoon precipitation pattern on the accumulation of OM. From the available published data, it is revealed that the climate during the Holocene fluctuated from dry, arid and wet phases as a result of changing monsoon pattern. However, till date no paleoclimate reconstruction based on published C:N ratio, $\delta^{13}C$ and OM content records from these lakes since the early Holocene has been compiled and presented and hence this work.

C:N and OM Records from Other Regional Lake Sites

High resolution records of regional and global paleoclimate data based on C:N ratio, $\delta^{13}C$ and OM content from lake sediments are now available with a comprehensive integration for reconstruction of past environmental and paleoclimate shifts. A compilation of few paleoclimate data sets generated from both the high and low altitudinal lakes from India and sites elsewhere are presented in Table 1 for reference.

The sudden increase in the C:N ratio of the Pleasant lake sediments (Kaushal and Binford, 1999) around 1780 A.D is attributed to the large-scale watershed deforestation and thus enhancing the terrestrial input of OM into the lake. The C:N ratios from the Shawnigan and Elk lakes reflected increased nitrogen loading from catchments, due to watershed disturbances in the post-disturbance period as also indicated by the source isotope signatures. The results confirmed that the C:N ratio in lake sediments can be used to elucidate a generalized idea of historical accumulating sources of sedimentary OM (Das et al. 2007).

Multi-proxy record of paleoclimate change from varved sediments in lake Xiaolongwan, north-eastern China reveal distinct stratigraphical patterns in organic carbon, total nitrogen, organic carbon isotope, nitrogen isotope, clastic content and dinocyst concentration over the

Table 1. A compilation of C:N ratios and $\delta^{13}C$ (‰) of different lake sediments and biota reported in the available literatures

S No.	Lake	Location	Source	C/N ratio	$\delta^{13}C$ (‰)	Reference
1	Balgoda Lake	Srilanka	Core sediments	24.3	-	Eriksson & Olsson (2015)
2	Bohai Bay	China	Surface Sediments	21.3±6	-	Gao et al. (2012)
3	Coburn Pond	Maine	Surface Sediments	12	-28.4	Meyers & Ishiwatari (1993)
4	Daihai Lake	China	Surface Sediments	8.2-12.1	-	Hou et al. (2013)
5	Eastern Lake	Florida	Aquatic plants	39±17.1	-13.5 to -30.8	Das et al. (2013)
6	Elk Lake	Canada	Core sediments	15-25	-26 to -29.7	Das et al. (2007)
7	Galapagos Lake	S America	Core sediments	10-50	-	Conroy et al. (2008)
8	Hudson Bay	Canada	Core sediments	9.77	-	Haberzettl et al. (2010)
9	Knudsenheia Lake	China	Core sediments	18.2	-	Jiang et al. (2011)
10	Lake Baikal	Russia	Surface sediments	11	-29.9	Meyers & Ishiwatari (1993)
11	Lake Bhimtal	India	Core sediments	9.4-11	-25 to -30	Choudhary et al. (2013)
12	Lake Biwa	Japan	Surface Sediments	6	-25.3	Meyers & Ishiwatari (1993)
13	Lake Bosumtwi	Ghana	Palm fronds	91	-25.5	Meyers & Ishiwatari (1993)
14	Lake Brunnsviken	Sweden	Core sediments	10-102	-28.5 to -26.2	Routh et al. (2004)
15	Lake Kariba	Zimbabwe	Core sediments	9.1	-28.8	Kunz et al. (2011)
16	Lake Kazjak	Croatia	Core sediments	11.6	-31.7	Baresic et al. (2011)
17	Lake Michigan	N America	Surface Sediments	9	-26.3	Meyers & Ishiwatari (1993)
18	Lake Ohrid	Macedonia	Surface Sediments	3-14.5	-	Vogel et al. (2010)
19	Lake Tahoe	California	CoreLT-99-4	13	-26.7	Osleger et al. (2008)
20	Lake Ximencou	China	Core sediments	11	-23.6	Pu et al. (2013)
21	Lake Xingyun	China	Core sediments	18.71	-18.37	Zhang et al. (2014)
22	Manasbal Lake	India	Core sediments	6-13	-	Kusumgar et al. (1992)
23	Mangrove Lake	Bermuda	Surface Sediments	13	-	Meyers & Ishiwatari (1993)
24	Meli Lake	Alaska	Core sediments	11-14	-27.5 to -28.5	Anderson et al. (1999)
25	Nakaumi Lagoon	Japan	Core sediments	8-18	-	Sampei & Matsumoto (2001)
26	Naples Harbour	Italy	Surface Sediments	0.39-68.83	-23.8 to -28.9	Rumolo et al. (2011)
27	Pyramid Lake	Nevada	Mixed plankton	09	-28.3	Meyers & Ishiwatari (1993)
28	Sangla Lake	India	Trench sediments	9-17	-25 to -23	Chakraborty et al. (2006)
29	Shownigam Lake	Canada	Core sediments	20	-24.8 to -27.2	Das et al. (2007)
30	Svalbard Archipel.	China	Core sediments	18.2	-19 to -25	Jiang et al. (2011)
31	Tamshui Basin	Taiwan	Surface Sediments	6.1	-	Ku et al. (2007)
32	Tangledup Lake	Alaska	Core sediments	6-15	-25 to -30.5	Anderson et al. (2001)
33	Upper Lochnan	Scotland	Core sediments	9-28	-16 to -30	Mackie et al. (2005)
34	Varthur Lake	India	Surface Sediments	23-33	-	Mahapatra et al. (2011)
35	Walker Lake	Nevada	Surface Sediments	8	-24.2	Meyers & Ishiwatari (1993)
36	Welwich Marsh	England	Surface Sediments	14.7	-21 to -28	Lamb et al. (2007)
37	Xialongwan Lake	China	Core sediments	17	-29.6	Chu et al. (2009)
38	Yellow Sea	East China	Core sediments	6.15-12.57	-21.6 to -25.84	Badejo et al. (2014)

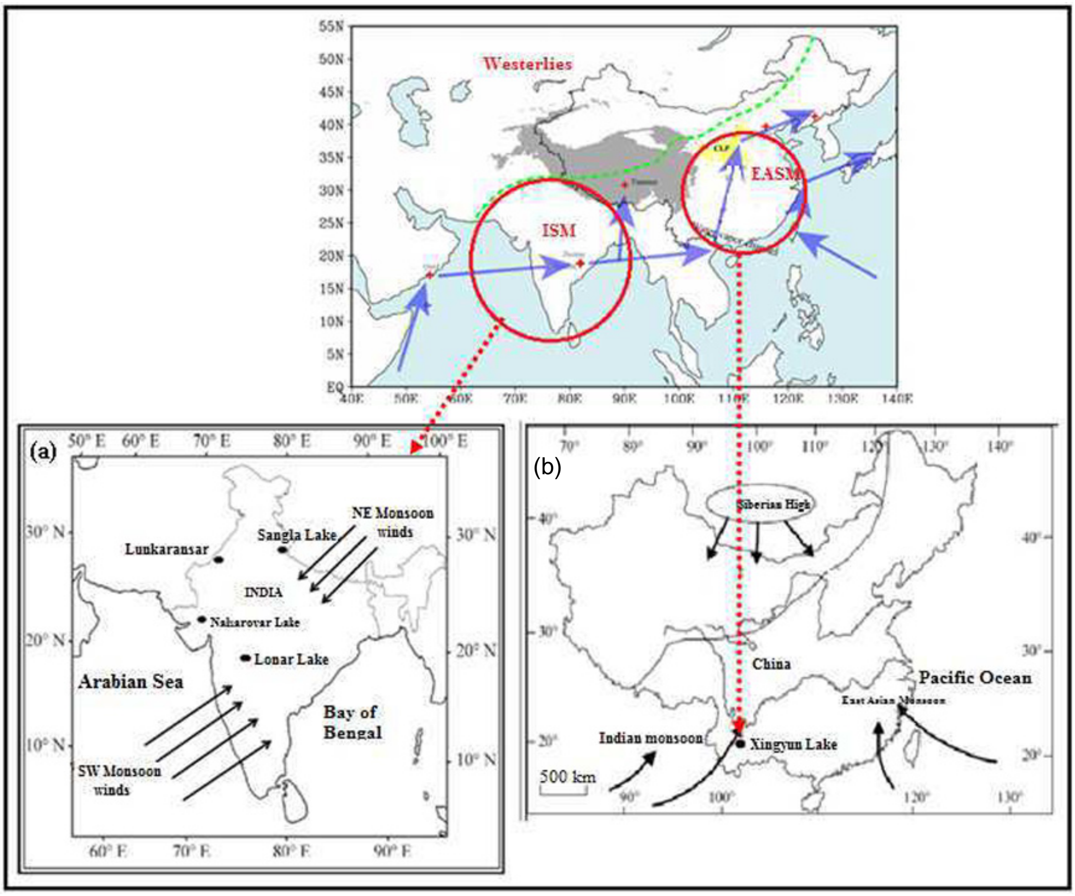

Fig.1. Location map of lake sites from (**a**) Indian subcontinent and (**b**) Xingyun lake, China.

past 1600 years. High atomic C:N ratios in the sediment suggest that a large amount of OM in the sediments is derived from vascular plants and soil in the catchment (Chu et al., 2009). Integrated textural, magnetic, and geochemical signatures from lacustrine turbidite deposits of lake Tahoe enabled discrimination of turbidites initiated by seismic induced debris flow generated by severe storms and associated hyperpycnal currents over the last 7 kyr BP (Osleger et al., 2008). The multi-proxy data from the lake provide strong evidence for extreme hydrologic events affecting the basin watershed over the past 7 kyr BP years. The data suggest that the shift towards significantly lower C:N values within the turbidites in the sediment core suggests increased algal productivity in response to the input of nutrients associated with a major hydrologic event. The lacustrine sedimentation in westernmost section of Sierra Nevada, which is controlled by geomorphic processes occurring over different temporal and spatial scales revealed a gradual decrease in C:N ratio since ~4.2 kyr BP, suggesting a sparser vegetation extent and a lower biological productivity in the lake catchments (Das et al., 2007; Oliva and Schulte, 2010).

The records obtained from the lacustrine sediment sequences have frequently provided longer and similar regional paleoclimatic shifts (Pu et al., 2013). The Xingyun lake (SW China) (Fig.1b) sediments reveal multi-proxy indicators of past climate change and human activities since the Holocene period in the lake catchment and four major climatic stages have been identified during the Holocene (Zhang et al., 2014) (Fig.3e). The C:N ratio, $\delta^{13}C$ value from 11.06 to 9.98 kyr BP indicate that OM in the lacustrine sediments was mainly derived from C_4 land plants that grew in warm and humid climate, which likely resulted from an enhanced Asian southwest monsoon. The high TOC and TN content, stable C:N and $\delta^{13}C$ values from 9.98-5.93 kyr BP suggests that the OM was mainly derived from land and aquatic plants, and that vegetation in the lake catchment was flourishing. The warmest stage identified lies in the 8.98- 6.10 kyr BP interval, coinciding with the global Holocene Optimum. The high TOC, TN content, and C:N ratio from 5.93-1.35 kyr BP indicates significant climatic fluctuation and a decrease in plant productivity in the lake catchment. The last stage from 1.35 kyr BP to present showed consistently low content of $CaCO_3$ and TOC reflecting the low biomass of the Xingyun catchment during this stage. In addition, the C:N ratio and $\delta^{13}C$ values were found to be consistent with high contribution of OM from aquatic plant life in the lake sediments and negligible terrestrial OM input suggesting an abrupt change in the plant community and intensified human activity from 1.35 kyr BP. This is

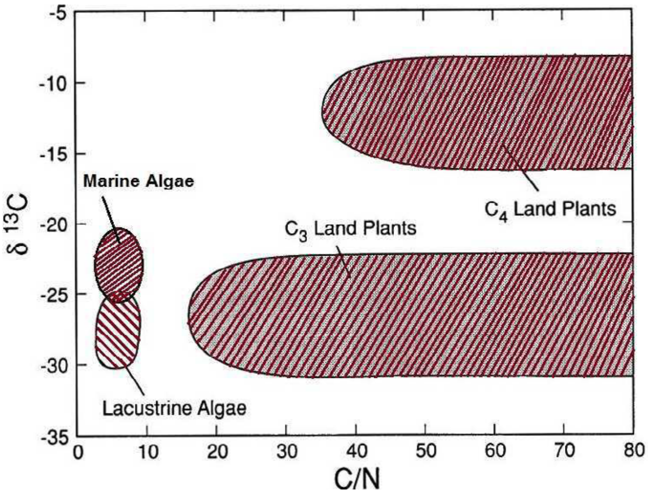

Fig.2. A plot of graph showing the representative elemental and carbon isotopic compositions of OM from marine algae, lacustrine algae, C_3 and C_4 land plants (modified after Meyers and Lallier-Vergès, 1999; Meyers, 1994).

consistent with the period of Dian culture established within the Xingyun catchment.

The nature of sediments deposited in lakes also serve as important indicators of past vegetation and environment, for example, whether they have OM fractions with isotopic signatures suggestive of C_3 or C_4 vegetation, amount of carbonate content, type of mineral(s) present (Fig.2). For example, Figure 2 represents a plot of δ^{13}C values vs C:N ratio for inferring OM contribution from different sources. In this plot the δ^{13}C values of -26‰ to -20‰ with C:N values from 3 to 10 represents OM contributions by marine algae or in the same plot the values of δ^{13}C from -23‰ to -30‰ with C:N values from 20 to 80 and beyond corresponds to the OM contribution by C_3 vegetation.

RESULTS (Compilation of available data)

A compilation of the available data from the five lake sites studied indicated that the C:N ratio fluctuated in all the lakes since the early to late Holocene period. Higher values of C:N ratio greater than 20 indicate periods of arid or dry climate, reduced precipitation and lower lake levels. On the contrary lower C:N values suggest wetter conditions. The reason for such alternation is that times of wetter climate results in enhanced algal productivity in lakes as a result of greater input of soil nutrients, and thus these periods are characterized by lower organic C:N ratios. However, lowering of lake water levels, associated with drier, rain depleted climate typically depresses autochthonous algal productivity and thus cause an apparent increase in the C:N ratio. From the data, it is also evident that the period from middle to late Holocene was an extremely arid with lakes receiving less precipitation and thus having enhanced C:N values. During this period, the OM accumulation fluctuated in all the three lakes with a wide range of C:N values (Lonar lake 10 to 30, Sangla lake 10 to 16, Nal Sarovar lake 10 to 25). On the other hand, decreased values of C:N ratio (3-10) indicated periods with relatively greater precipitation and high lake levels. Based on the OM records, the Holocene is divided into different periods comprising early, mid and late Holocene indicating large and small-scale dry and wet phases. The period of early Holocene from ~11.1 to ~10 kyr BP is characterised by dry conditions with higher C:N ratios, shallow lake levels, reduced monsoon precipitation. This is followed by strong moist conditions with higher lake levels (enhanced precipitation), low C:N ratios due to enhanced SWM activity between ~10 to ~6 kyr BP. The climate in mid Holocene was characterised by strong arid conditions from ~6.6 to ~3 kyr BP that witnessed a strong dry climate resulting in shallow lake beds, higher C:N ratio and almost complete absence of aquatic vegetation from the lake sediments (Nal Sarovar lake). The Holocene paleoclimate reconstruction from core monsoon zone (CMZ) of Indian peninsula revealed a similar trend with an arid climate from the onset of mid Holocene period. Carbon isotopes of sedimentary leaf waxes from Godavari delta revealed a gradual increase in aridity adapted vegetation from 4.0 kyr BP until 1.7 kyr BP followed by perseverance of aridity adapted plants (Ponton et al., 2012). The lakes shrunk significantly during this period. However, from the late Holocene to present, the climate remained more or less similar with minor fluctuations in C:N ratio and lake levels. The available data also revealed significant small-scale changes in the accumulation of OM content, although at different time scales. Lonar lake which lies in the monsoon trough region shows a well marked shift in C:N ratio at around 4.2 kyr BP (C:N=35-40). This period of intense aridity is also distinctly seen in other lakes as revealed by increased C:N values (Sangla lake C:N=12-16, Nal Sarovar C:N=15-20). The small-scale climatic fluctuations caused migration and decline of earlier civilisations (Dixit et al., 2014) such as the Harappan due to dramatic changes in the intensity of precipitation patterns around 4.2 kyr BP (Fig.3a, b). The shift in precipitation pattern is also linked with the changes in local climate around these lakes. Figure 3 also indicates continuous deterioration of paleoclimate and the intensity of precipitation, but with milder amelioration in precipitation. Lake sediment proxy records from Lunkaransar and Sangla exhibit distinctly these shifts from the onset of late Holocene. However, the Lonar lake sediments do not exhibit the subsequent climate shifts.

Changes in OM and C:N proxy records (Intra and inter regional comparison)

The use of multi-proxy data (C:N ratios and δ^{13}C) for paleoclimatic reconstruction (Chakraborty et al., 2006; Prasad and Enzel, 2006) revealed lake level changes and alternate phases of arid and dry periods during early to late Holocene period. The fluctuating C:N ratios from the paleo-lake sediments reveal alternate phases of

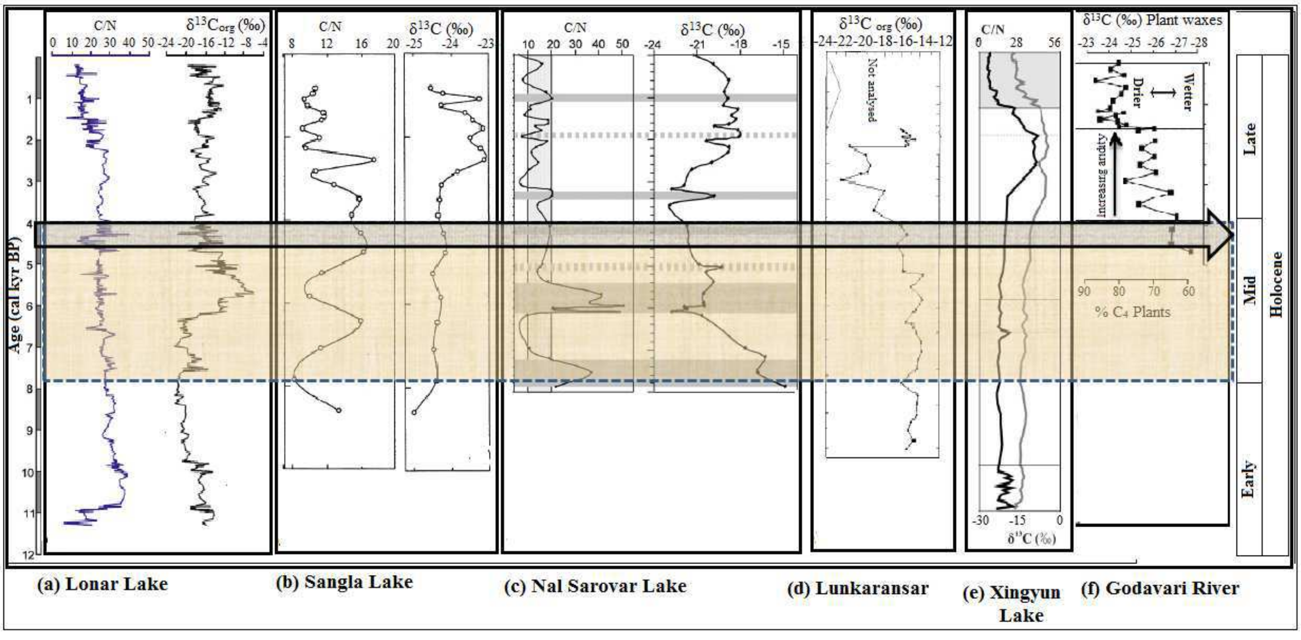

Fig.3. A comparison of the palaeoclimatic interpretation data of C:N and δ^{13}C in sediment samples from (**a**) Lonar Lake (**b**) Sangla paleolake (**c**) Nal Sarovar Lake (**d**) Lunkaransar Lake (**e**) Xingyun Lake and (**f**) Godavari Delta. The data show major and minor climatic fluctuations in India and China as archived from lake sediments. The shaded arrow indicates 4.2 kyr BP drought event associated with the decline of the Harappan civilisation.

dry and wet conditions from early to mid Holocene, although high lake level between 10.0 to 4.0 kyr BP (Fig.3) with one of the values around 3.0 kyr BP indicating the driest phase. This period of strong aridity caused the decline and migration of important civilisations, including the collapse of great Harappan civilisation of Indian subcontinent around 4.2 kyr BP (Achyuthan et al., 2007). The decline in the monsoon rains around 4.2 kyr BP lead to weakened river dynamics, and played a critical role in the collapse of the Harappan civilization, which relied on monsoon rains to fuel their agricultural surpluses (Possehl, 1997; Staubwasser et al., 2003; Dixit et al., 2014). Another possible explanation is that a reduction of the average annual rainfall over the Indus river watershed (4.2 kyr BP drought event) restricted Harappan farming in the Indus valley and left large city populations unsustainable (Weiss, 2000; Singh et al. 1990). In addition multiproxy records from sediments of the lake Nal Sarovar (Fig.3c) revealed successive phases of arid and dry periods from 7.8 kyr BP to present. The records from C:N values and $\delta^{13}C$ along with other proxy records showed periods of high lake level (~7.2-6.1 kyr BP) and low lake level (~6.1-5.4 kyr BP) in lake Nal Sarovar (Prasad et al., 1997). A comparison of these with climate records from the other regions displays a strong similarity, signifying that these fluctuations were of global extent (Fig.3e). However, the changes in proxy records from such lakes are not observed commonly in all the lake-based records as shown in the Fig. 3. This is due to the timing and magnitude of limnological response to the climate change and local, direct and indirect influences such as the response to micro-climate, local atmospheric temperature and wind pattern, stratification, nutrient availability and type of lake system (open or closed).

Lake systems in India have also been studied in detail for Holocene environmental and paleomonsoon records. These lakes provide a continuous record of the declining monsoonal shifts in this region (Menzel et al. 2014). Lonar lake revealed climate shifts from wet conditions during early Holocene to dry conditions from late Holocene. A period of strong dry phase in this region from 4.6-3.9 kyr BP correlates well with the decline of Indus civilisation around 3.9 kyr BP. In addition, Holocene records from Lunkaransar Lake (Fig.3d) also reveal local and regional lake level fluctuations due to changes in southwest monsoon. In early Holocene, the lake level was shallow and then rose abruptly around 6.3 kyr BP reflecting the effect of regional precipitation (Enzel et al., 1999). However, none of these lakes hold signatures of extreme short events such as the Roman Warm Period (RWP), the Medieval Warm Period (MWP) and the Little Ice age (LIA).

In summary, it is evident that the intermittent fluctuations in the OM content, $\delta^{13}C$ values and C:N ratio provide a record of paleoclimate and past environmental changes. Our compilation of the data from the lakes chosen show significant spatial variations in organic carbon proxies for the lakes that reflect changes in the environment, which in turn affected the processes transporting OM within the lake. The earlier available paleoclimate studies carried out using OM as a proxy indicate that the accumulation of OM in sediments is affected by many variables, including local and regional climate, lake bathymetry, catchment stability, anthropogenic sources etc. However, the deposition of OM and the apparent changes in C:N ratio due to changes in above variables need to be thoroughly investigated. Similarly, the amount of OM contributed from different sources in lake systems need to be quantified while interpreting the data for better understanding of the influence of natural and human activities on the relative amount that is deposited in lake sediments.

CONCLUSIONS

The available proxy records suggest that the climate has undergone dramatic changes during the early to late Holocene period and lake bathymetry fluctuated during the Holocene period owing to the shifts in precipitation pattern. A comparison of these records with climate records from the other regions displays a strong similarity, signifying that these fluctuations were of global extent. However, the episodes of changes in the C/N and $\delta^{13}C$ proxy data do not show a common trend in all the lakes. The reason for such contrast is that the components of lake sediments are mediated by timing and magnitude of limnological response to the climate change and other influences such as the response to micro-climate, local atmospheric temperature, geomorphic and hydrologic setting, stratification and nutrient availability.

Combined proxy records available from the C:N ratio and ä^{13}C values also reveal fluctuating lake levels due to corresponding changes in climate and local environment from early Holocene period to Present. These fluctuations include:

1 Changes in the source of OM from autochthonous to allochthonous nature.
2 Changes in the lake biodiversity and bathymetry when the sediments received greater amount of terrestrial OM.
3 Small-scale disturbances (anthropogenic and natural) in the local catchment supplying OM to the lake basin.

More detailed and simultaneous analysis of the results from these proxies will help in better comprehension of the palaeolimnological shifts and processes.

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