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An Indian Perspective

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6.1 INTRODUCTION

Primary productivity in the past can be reconstructed by multiple biogeochemical proxies such as nutrients, amino acids, pigments and isotopes. Besides many other proxies, biogenic silica (bSi) has emerged as a potential paleoproductivity proxy and is widely used to reconstruct palaeoceanography and paleoclimate changes.

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Biogenic silica in lake sediments has received considerably more attention in recent years as it indicates climate and environmental changes over a wide geographical area on a different time scale (Carter and Colman 1994; Colman et al. 1995).

Biogenic silica is a type of amorphous silica derived from diatoms and other siliceous microorganisms (Liu et al. 2014). Biogenic silica documents direct measure of biological products from the siliceous algae and diatoms (Conley 1998; Kaplan et al. 2002). It is produced in the euphotic zone by the diatoms, predominantly in the high-latitude areas. Although the biogeochemical cycle of silicon is slow as compared to other elements like oxygen, carbon and nitrogen, its sedimentation rate is rapid (Shan et al. 2011); therefore, silicon can be preserved in the form of biogenic silica in sediments (Birks et al. 2004; Shan et al. 2011). In lakes, nutrient concentration in water and surface temperature affects productivity. Therefore, variation in biogenic silica content can reflect changes in past climate conditions. Earlier studies from lakes of different parts of the globe such as Lake Baikal, Lake Pipa, Lake Huguangyan Maar and lakes from the Arctic region reported that a high concentration of bSi represents warm and humid climate conditions. Simultaneously, lower values indicate cold and dry climatic conditions (Shan et al. 2011). These studies suggested that changes in diatom production are related to changes in climate. Therefore, biogenic silica can be used to reconstruct past climate changes in lake sediments.

6.2 BIOGEOCHEMICAL CYCLE OF SILICA

The silicon cycle is a complex interaction of biological, chemical and geological processes occurring on a wide variety of spatial and temporal scales. The transport of Si from the land to the oceans through terrestrial ecosystems and river catchments to the estuarine and coastal zone is controlled by a complex set of terrestrial and aquatic processes. The weathering of silicates is an important sink for atmospheric CO_2. Silicon is also an essential element in terrestrial ecosystems, in soil formation processes and in regulating species composition of vegetation. The import of Si into coastal zones from the terrestrial environment is essential to sustain diatom growth. Diatoms play a vital role in the oceanic carbon sink and eutrophication of coastal zones. Because of its global environmental impact, the silicon cycle is receiving considerable attention in recent decades.

Atmospheric CO_2 plays an essential role in silicate weathering. During silicate weathering, dissolved CO_2 is consumed and dissolved silicate (DSi), i.e., ortho-silicic acid (H_4SiO_4), is released from the crystalline structure of silicate minerals. For example, in the weathering of anorthite (over kaolinite) to gibbsite, DSi is produced, and CO_2 is consumed (Stumm and Morgan 1974).

$$CaAl_2Si_2O_8 + 2CO_2 + 8H_2O \rightarrow Ca2^{+} + 2Al(OH)_3 + 2H_4SiO_4 + 2HCO_3$$

$$\text{(Anorthite)} \qquad\qquad\qquad\qquad \text{(Gibbsite)} \ \text{(Ortho-silicic acid)}$$

Further, DSi (H_4SiO_4) is transported through rivers and released eventually to the lakes and oceans. The significant silica sources include rivers, groundwater flux, seafloor weathering inputs, *hydrothermal vents*, and atmospheric deposition (*aeolian flux*)

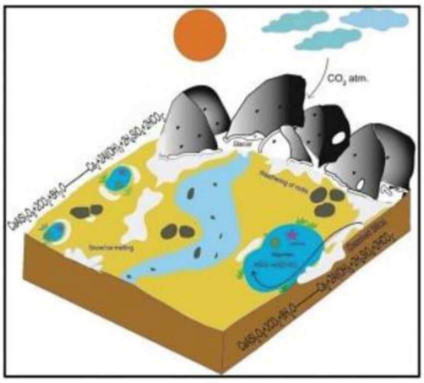

FIGURE 6.1 Cycling of silica in the lake environment.

and glacial weathering of rocks. Rivers are the largest silica source to the marine environment, contributing up to 90% of the total silica delivered to the aquatic systems (Gaillarde et al. 1999). In contrast, in the closed systems like lakes, silica is mainly derived through runoff and glacial weathering of rocks in the high-latitude regions (Figure 6.1). The DSi (H_4SiO_4) released to the lakes and oceans is consumed by primary producers like diatoms (40% of the total phytoplankton) as they extract silica from the water and use for the construction of their hard part known as diatom frustule in the form of biogenic silica or hydrated silica ($SiO_2.nH_2O$). They are responsible for extracting a large amount of silica from the water column (Wang et al. 2014), and the diatoms are critical players in the global biogeochemical cycle.

6.3 MATERIALS AND METHODOLOGY

6.3.1 Study Area and Sample Collection

The samples were collected from lakes around Ny-Ålesund, the International Arctic Research Base, located in the Spitsbergen Island of Svalbard archipelago (Figure 6.2) as part of the Indian Arctic Programme (summer phase) during August 2016.

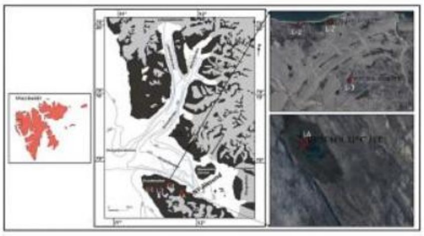

FIGURE 6.2 Map showing the study area. (Modified after Svendsen et al. 2002.)

Sediment core samples were collected from four shallow freshwater lakes. The lakes were fed by meltwater from glaciers, snowdrift and local precipitation. For the sediment core collection, a PVC hand-held corer was inserted by hammering manually into the lake sediment bed and then retrieved, and the cores were sub-sampled at 2 cm. Samples were labelled and transported to the laboratory in a frozen condition. In the laboratory, each sub-sample was dried at 60°C in the oven and utilised for further analysis.

6.3.2 Sample Analysis

Biogenic silica concentration in the sediment was measured by the wet alkaline extraction method, modified by Mortlock and Froelich (1989) and Muller and Schneider (1993), where the blue silicon-molybdenum complex intensity was measured using a visible spectrophotometer (UV-1800 Shimadzu) at 810 nm. The reagents used for the determination of biogenic silica were prepared using the following procedures.

The sulphuric acid solution was prepared by adding 250 ml of concentrated sulphuric acid slowly and continuously to 750 ml of Milli-Q water while stirring. 38 g of ammonium heptamolybdate tetrahydrate, (NH4)6Mo7O_{24}, was dissolved in 300 ml of Milli-Q water, and this solution was added to 300 ml of sulphuric acid solution to prepare ammonium molybdate solution. 10 g of oxalic acid was dissolved in 100 ml of Milli-Q water. 2.8 g of ascorbic acid powder was added to 100 ml of Milli-Q water to prepare an ascorbic acid solution.

After the preparation of all the reagents required, 0.2 g of the sediment sample was weighed accurately and transferred to a 50 ml polypropylene centrifuge tube with 25 ml of 1% Sodium Carbonate (Na_2CO_3) solution as the alkaline treatment (Na_2CO_3) has a high bSi extraction efficiency (Liu et al. 2014). The sample was then placed in a preheated water bath at 85°C for 5 hours along with the blank to extract the biogenic silica from the sediment. Then, the sample was centrifuged to remove the suspended matter, and 1 ml of supernatant was released in a 100 ml volumetric flask containing 35 ml of Milli-Q water. To this solution, four drops of 1:100 HCl was added to bring the pH 7–8, i.e., close to seawater's pH. Subsequently, 1 ml of mixed reagent was added and kept for half an hour to form the yellow silicomolybdate complex. 1 ml of oxalic acid was added to the solution immediately (to decompose silicomolybdate complex) followed by 1 ml of ascorbic acid, which reduces the acid complex to a blue-coloured complex. Then, the solution was allowed to cool at room temperature for the reaction to complete, and the entire content was made up to 100 ml using Milli-Q water. After a wait of 30 minutes, the sample solution was taken in the cuvette of 1 cm. Against a reagent blank at a wavelength of 810 nm, the blue colour intensity was measured using the spectrophotometer. Using a standard Sodium Hexafluorosilicate, the stock solution was prepared, known as the working solution, which is utilised to prepare a calibration curve.

6.3.3 Quality Control (QC) and Quality Assurance (QA)

The biogenic silica concentration was estimated from the calibration curve. Duplicate measurements were conducted on each sample, and relative error was noted to be less than 3%.

6.4 SOURCE AND DISTRIBUTION OF BIOGENIC SILICA (bSi) IN ARCTIC LAKES

Biogenic silica documents direct measure of biological products from the siliceous algae and diatoms (Conley 1998; Kaplan et al. 2002; Choudhary et al. 2018a–d), and biological productivity in lake sediment is characterised mainly by the diatom production (Birnie 1990; Roberts et al. 2001; Choudhary et al. 2018a). bSi concentration was found low in lake LA, L-1, L-2 and L-3 (Figure 6.3a–d), suggesting the presence of a low concentration of siliceous microfossils in these lakes, maybe due to the carbonate-rich sediments resulting in poor preservation of diatoms as also reported by Guilizzoni et al. (2006). However, increased concentration of biogenic silica from 6 cm to the surface and clay in core LA and L-2 (Figure 6.3a and c) indicated high productivity suggesting warming conditions in the region due to the lakes' exposure to the ice meltwater influx. In core L-1, bSi was relatively high in the middle portion of the core from 22 to 6 cm along with TOC and clay suggesting deposition of finer particles from suspension facilitating high productivity. The average C/N ratio in all the cores was found to be >15, indicating both autochthonous and allochthonous terrestrial sources for organic matter. Relatively large input of terrestrial organic matter derived from mosses growing in the catchment region of the lake is responsible for variation in bSi in the Arctic lakes. During the warmer conditions, lake water becomes warmer suitable for the growth of phytoplankton and zooplanktons resulting in overall increased productivity. Increased diatoms production can lead to an increased accumulation of biogenic silica in sediments, ultimately resulting in a decline in the water column reservoir of DSi (Conley et al. 1993). However, during the colder conditions, catchment area of the lakes would be covered with snow resulting in less solar radiation and nutrients, making it difficult for plants to

survive and ultimately low diatom production. Therefore, the change in concentration of biogenic silica in the sediment column is related to the climatic conditions, nutrient concentration and variations in solar radiation. Paleolimnological evidence based on an accumulation of biogenic silica in the sediments has been used to study

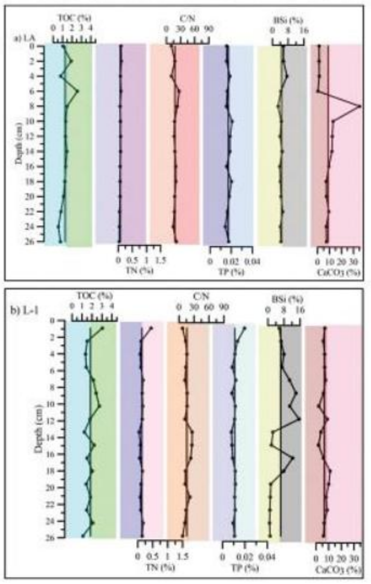

FIGURE 6.3 Depth-wise distribution of organic components of cores: (a) LA, (b) L-1, (c) L-2 and (d) L-3.

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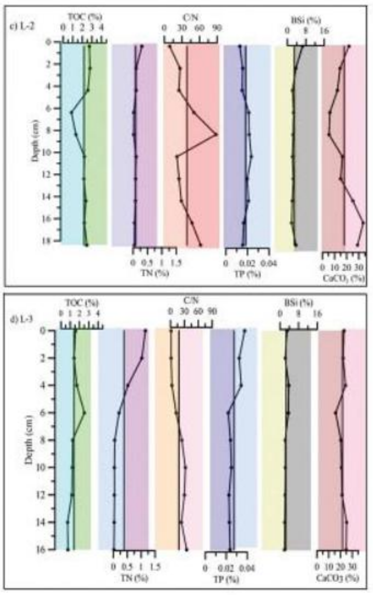

FIGURE 6.3 (*CONTINUED*) Depth-wise distribution of organic components of cores: (a) LA, (b) L-1, (c) L-2 and (d) L-3.

the change in climatic conditions and time course of depletion of DSi in the water column (Schelske et al. 1983, 1986a). Therefore, biogenic silica is an ideal proxy to understand the past environmental conditions archived in lacustrine sediments of the Arctic (Jiang et al. 2011).

6.5 FACTORS CONTROLLING THE DISTRIBUTION OF BIOGENIC SILICA IN LAKES

6.5.1 Grain Size

In a closed environment like lakes where the exchange of water is relatively less and the supply of nutrients is limited, only small-sized diatoms can survive and constitute most phytoplankton. Hydrodynamic condition and resulting grain size affect the growth of diatoms and, ultimately, bSi concentration (Wang et al. 2014). Availability of relatively coarser sediments may reduce the concentration of bSi due to the dilution effects. However, in some regions, where the sediment is relatively coarse to medium, bSi concentration may also be high due to high primary productivity and low sedimentation rate. The high abundance of diatoms reduces the dilution effect of terrigenous sediments. Therefore, the sediment's grain size should be taken into account while using bSi as a proxy for paleo-productivity.

6.5.2 Biogeochemical Proxies (TOC, TN and TP)

Biogenic Silica and TOC often show similar variations indicating that biogenic silica has originated from the organic source. C/N ratio <10 suggests the source of organic matter exclusively derived from algae. A close association between bSi and other geochemical proxies indicates that the lake's organic matter is predominantly autochthonous and derived from algae (Choudhary et al. 2020).

6.5.3 Detrital Al and Al/SiO_2 Ratio

A similar distribution of biogenic silica, sand and Al in sediment indicated that dissolved silica either due to the incorporation of aluminium in diatom frustules (Van Bennekom et al. 1988; Choudhary et al. 2018a) or low silica solubility in a freezing environment might have caused precipitation of dissolved biogenic silica with Al in sediment (Rickert et al. 2002; Choudhary et al. 2018b). Low values of Al/SiO_2 ratio in the sediment support precipitation of biogenic silica with detrital Al, while higher values of Al/SiO_2 indicate replacement of Si by Al in diatom frustules.

6.5.4 Availability of Carbonate-Rich Sediments

Generally, in the Polar regions, CO_2 dissolution is high, leading to an increase in carbonic acid concentration, which explains the low $CaCO_3$ content. However, the availability of carbonate-rich sediments in the catchment area may dilute biogenic silica concentration, resulting in low siliceous microfossils (Choudhary 2019).

6.6 CONCLUSIONS

In Arctic lakes, bSi concentration was low, suggesting the presence of low siliceous microfossils in these lakes, which may be due to the carbonate-rich sediments resulting in poor preservation of diatoms. However, a relatively high concentration of bSi in the upper section indicated high primary productivity due to the warmer conditions and glacier retreat. Therefore, bSi is a robust proxy to understand the past environmental changes archived in lacustrine sediments of the Arctic. bSi content in Arctic lacustrine sediments is influenced by the input of terrestrial organic matter and many other factors like grain size, different biogeochemical proxies and detrital Al in the water column.

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