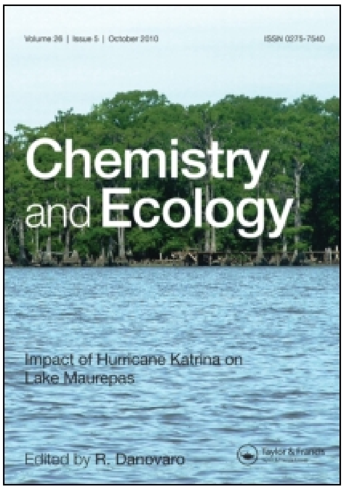

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Speciation of nitrogen in the coral reef sedimentary environment of Lakshadweep archipelago, Indian ocean

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SPECIATION OF NITROGEN IN THE CORAL REEF SEDIMENTARY ENVIRONMENT OF LAKSHADWEEP ARCHIPELAGO, INDIAN OCEAN

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This paper focuses on the spatial distributional profiles of different species of the important micronutrient element, nitrogen (nitrite, nitrate and total nitrogen) in various coral-reef sedimentary environment of Lakshadweep Archipelago. The relative abundance of the three forms of nitrogen was in the order, total-N $\gg$ nitrate-N $\gg$ nitrite-N. Relatively very low levels of nitrite in the different microenvironments of the islands are an indication of a higher rate of nitrification, so as to produce thermodynamically most stable form of nitrogen, namely nitrate under the condition of well-oxygenated shallow coastal/lagoon waters. A lagoon-ward enrichment pattern of total nitrogen in the lagoon transects of Agathy, Minicoy, Kadamath and Kiltan Islands also reflected the fact that the rate and space available for nitrogen fixation in shallow zones of the lagoon are high. Further, the nitrogenous waste materials produced from the reefs and surrounding environments have limited exchange with the sea, and all these factors, together with the organic nitrogen retention capacity of sediment types, contribute to total nitrogen.

Keywords: Coral reef; Speciation; Biogeochemical; Micronutrients; Archipelago

1 INTRODUCTION

Ever since Charles Darwin, the enigma of the existence of a rich life on coral reefs surrounded by surface oligotrophic oceanic waters depleted of nutrients has been the subject of heated discussions and investigations (Wiebe, 1988; Atkinson *et al.*, 1995; Miyajima *et al.*, 2001; Suzumura *et al.*, 2002). The basic turnover of nutrients within the coral reef ecosystem proceeds via their transformations in the food webs. The processes of nutrients turnover going on in the water column start with the consumption of the inorganic salts by the phytoplankton and bacterioplankton. Thus, the dispersed dissolved nutrients are transferred into the concentrated particulated matter in the biomass of microorganisms, and in part into the excreta by microplankton. Its larger portion is mineralized via respiratory decomposition, by planktonic and benthic filterers and by bacteria of bottom sediments (Atkinson, 1987; Suzumura *et al.*, 2002). Especially, the contribution from sediment seems to play a greater role in recycling and regeneration of organic matter and nutrients.

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Quantitative and qualitative studies of nitrogen are important for understanding the basic processes governing the distribution and biogeochemical cycling of this important micronutrient. Thus, changes in both absolute and relative concentrations of nitrogen species would help in understanding the potential availability of life-supporting elements in the coral reef sedimentary environment and are also good indicators of the intensity of the eutrophication process and of disturbances in the cycling of these elements.

Nitrogen enters rivers, estuaries and coastal marine ecosystems from natural, and in many cases anthropogenic, sources throughout their watershed and airsheds (Seitzinger *et al.*, 2002). Since nitrogen is the primary nutrient limiting resources for plant, algal and microbial production in many marine and some freshwater environments, increases in nitrogen inputs can markedly alter those ecosystems (Caraco and Cole, 2001). The biogeochemical cycling of nitrogen is of a complex nature because of varieties for chemical forms (species) in which the nitrogen is available for biological utilisation.

A baseline survey, which is needed to determine the future, if the natural and anthropogenic processes are changing the sedimentary environment of Lakshadweep Archipelago, has been attempted to formulate by assessing the nitrogen characteristics of the study area. The study focuses on six inhabited islands: Kavaratti, Kadamath, Agathy, Androth, Kiltan and Minicoy.

2 MATERIALS AND METHODS

2.1 Study Area

Lakshadweep is an archipelago consisting of 12 atolls (Fig. 1), three reefs and five submerged banks. It is located at 8–12° 13′ N 71–74° E and 220–440 km away from the coastal city of Kochi in Kerala, India. Lakshadweep has an area of 32 km^2 and has 10 inhabited islands, Kavaratti, Agathy, Amini, Androth, Kiltan, Kalpeni, Kadamath, Chetlat, Bitra and Minicoy. Among these, Androth has the largest land area and Bitra the smallest. Sediment samples were collected from six islands, Agathy, Androth, Kadamath, Kavaratti, Kiltan and Minicoy.

2.2 Sampling and Storage of Sediment Samples

Sampling sites were chosen to represent the environmental setting of the different microenvironments of the island ecosystem; these are listed in Table I, and the out-sketches of the islands are collated in Figure 2. Of the six islands studied, two sets of shallow sediment cores were retrieved from Kavaratti and Androth Islands, along with a core sample from Kiltan Island. Sediment cores were sub-sampled from various depths, as specified in Table I. Utmost care was taken to collect uncontaminated samples from all the subsurface litho-units of the sediment column. In addition to this, five lagoon transect collections, each representing a landward part, berm, low-tide level and lagoon, were made from the lagoon side of Kavaratti, Kiltan, Kadamath, Agathy and Minicoy Islands. Three seaside transect collections each representing a land, low-tide level, high-tide level and lagoon were also conducted from three islands, Agathy, Kiltan and Kadamath, where marked beach development was observed. To assess the nitrogen-loading capacity of sediments/soils inside the island, two soil samples separated by a distance of approximately 500 m were taken after removing the top 2–3 cm layer. Minicoy Island is famous for its mangrove vegetation among the Lakshadweep Archipelago. Hence, two sediment samples were collected from the mangrove ecosystem located in the southern lagoon side of Minicoy Island.

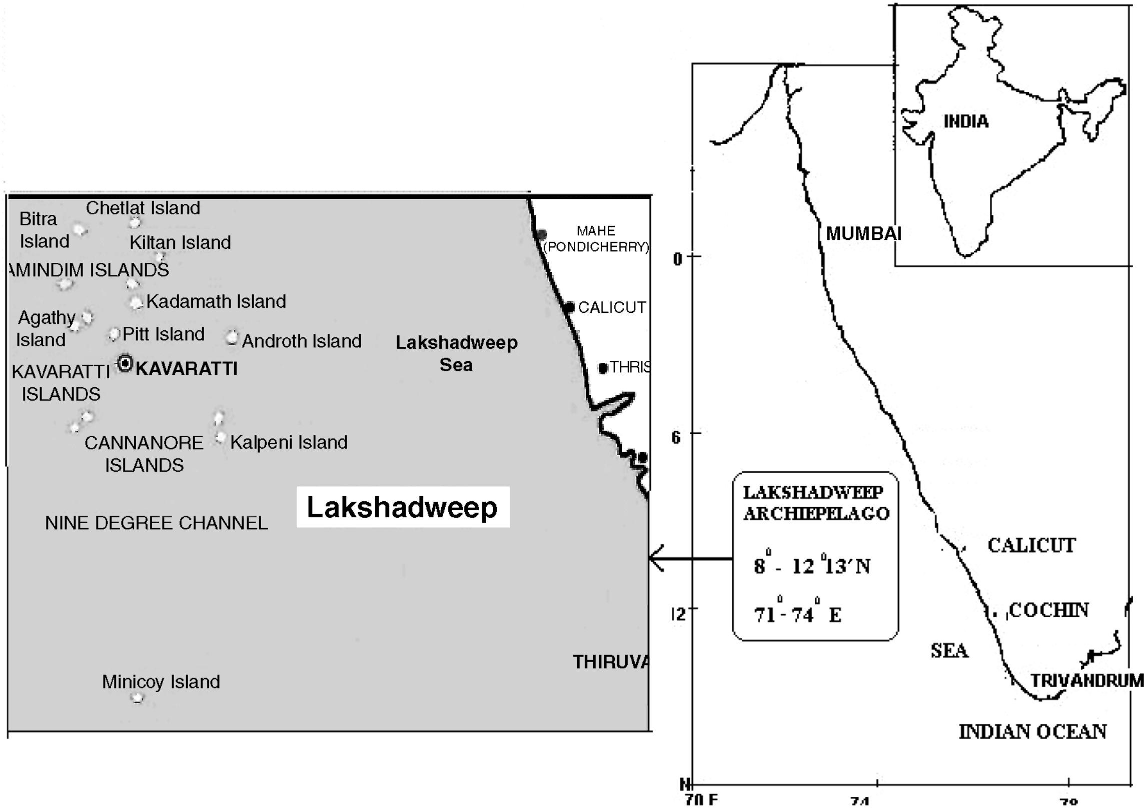

FIGURE 1 Location of Lakshadweep Archipelago.

TABLE I Location of the sampling sites in the six islands of Lakshadweep Archipelago (station numbers used in figures are given in parentheses).

Kavaratti	*Kiltan*	*Kadamath*	*Minicoy*	*Androth*	*Agathy*
Lagoon transect: • Land (1) • Berm (2) • Lowtide level (3) • Lagoon (4)	Lagoon transect: • Land (1) • Berm (2) • Lowtide level (3) • Lagoon (4)	Lagoon transect: • Land (1) • Berm (2) • Lowtide (3) • Lagoon (4)	Lagoon transect: • Land (1) • Berm (2) • Lowtide (3) • Lagoon (4)	Core-1 (seaside): • Surface (1) • 50 cm (2) • 100 cm (3)	Lagoon transect: • Land (1) • Berm (2) • Lowtide (3) • Lagoon (4)
Core-1 (south): • Surface (5) • 30 cm (6) • 60 cm (7) • 90 cm (8) • 120 cm (9)	Seaside transect: • Land (5) • Lowtide level (6) • Hightide level (7) • Lagoon (8)	Seaside transect: • Land (5) • Lowtide level (6) • Hightide level (7) • Lagoon (8)	Mangrove samples: • Sample-1 (5) • Sample-2 (6)	Core-2 (landslide): • Surface (4) • 15 cm (5) • 30 cm (6) • 50 cm (7) • 90 cm (8) • 150 cm (9)	Seaside transect: • Land (5) • Lowtide level (6) • Hightide level (7) • Lagoon (8)
Core-3 (north): • Surface (10) • 30 cm (11) • 60 cm (12) • 90 cm (13) • 120 cm (14)	Core profile: • Surface (9) • 50 cm (10) • 130 cm (11)				
Soil samples: • Soil—1 (15) • Soil—2 (16)	Soil samples: • Soil—1 (12) • Soil—2 (13)	Soil samples: • Soil—1 (9) • Soil—2 (10)	Soil samples: • Soil—1 (7) • Soil—2 (8)	Soil samples: • Soil—1 (10) • Soil—2 (11)	Soil samples: • Soil—1 (9) • Soil—2 (10)

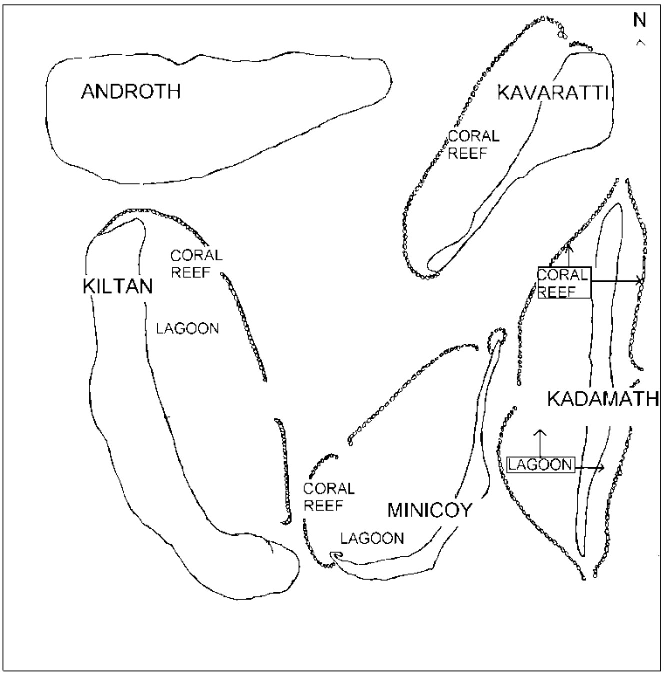

FIGURE 2 Outline sketches of the Lakshadweep islands studied.

Sediment samples were collected using a specially fabricated stainless steel (with a plastic lining) coring device, and subsamples were carefully skimmed (excluding shells and leafy materials) and put into ice-box storage in closed polythene containers for transport to the laboratory and stored at $-5\,^{\circ}C$ until analysis. The sampling reproducibility was checked by duplicate subsampling measurements.

2.3 Analytical Techniques

Keeney and Nelson's (1982) method was used to sediment samples for nitrite and nitrate extraction. This method involves the determination of sedimental forms of nitrite and nitrate by shaking a known quantity of sediment with 2 N KCl for 1 hr. Nitrite was then estimated using the standard procedure suggested by Grasshoff *et al.* (1999), in which the nitrite formed an azo dye, with sulphanilamide and *N*-(1-naphthyl) ethylene diamine dihydrochloride, which was then quantified spectrophotometrically at 540 nm. Nitrate was estimated by reduction to nitrite using a copper-coated cadmium column and quantifying the nitrite as outlined above (Grasshoff *et al.*, 1999). Total N was estimated using a procedure of persulphate oxidation (Libby and Wheeler, 1994; Bronk *et al.*, 2000), and the resulting nitrate-N was subjected to reduction using a cadmium column, which was then analysed spectrophotometerically by the method referred to above (Grasshoff *et al.*, 1999).

Sedimentary organic carbon was determined using the procedure of Gaudette *et al.* (1974), with some modifications as per the APHA (1995) procedure for those sediment samples which have chloride interference. Textural characteristics were determined by dry sieving using a Ro-Tap sieve shaker and classified as Wentworth size classes (Folk, 1974).

The apparatus used was thoroughly cleaned using HCl, washed in deionized water and finally rinsed with Milli-Q water. The reagents and standards used were of Merck/Suprapur

BDH grade. All spectroscopic readings were taken on a Hitachi UV-visible spectrophotometer (model 150-20). Cell-to-cell and blank corrections were applied to all sets of readings. The analytical reproducibility was checked by performing duplicate analyses of each of the duplicated subsamples. Thus, final analytical data are presented on a weighted-average bases of quadruplet values.

3 RESULTS

In this study of speciation of nitrogen, three forms were estimated, namely nitrite, nitrate and total nitrogen. The sediments collected from different microenvironments of the Lakshadweep Islands marked the presence of very low levels of the reduced form of nitrogen, namely nitrite-nitrogen (Fig. 3). In the lagoon transect of the Kavaratti Island, nitrite concentration showed an increasing trend from lagoon (0.01 μmol g^{-1}) to landward (0.025 μmol g^{-1}). The other two forms of nitrogen, nitrate-nitrogen and total nitrogen (Figs. 4 and 5), also showed more or less a similar pattern of variability in the lagoon transect, *i.e.* a landward flourishing, but their absolute values were several times higher than nitrite. Nevertheless, on Kavaratti Island, the contribution of nitrate fraction to total nitrogen is less than one-tenth of the total

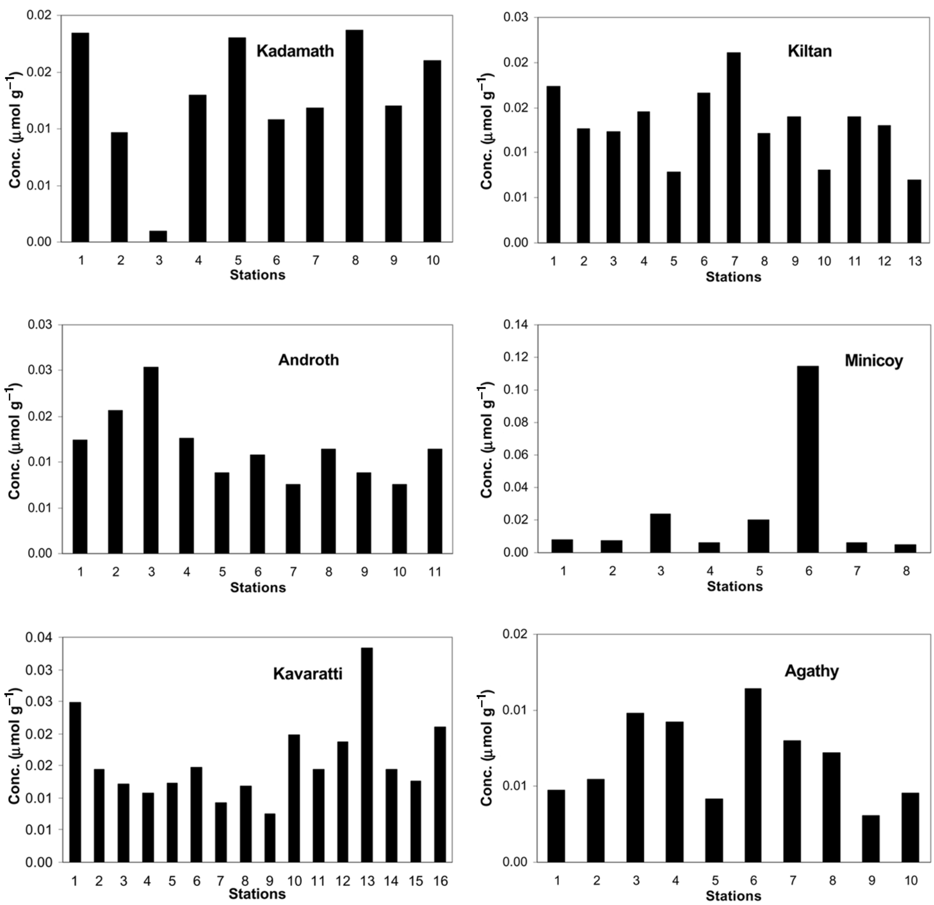

FIGURE 3 Spatial variability of nitrite in the sediments of the six islands of Lakshadweep Archipelago.

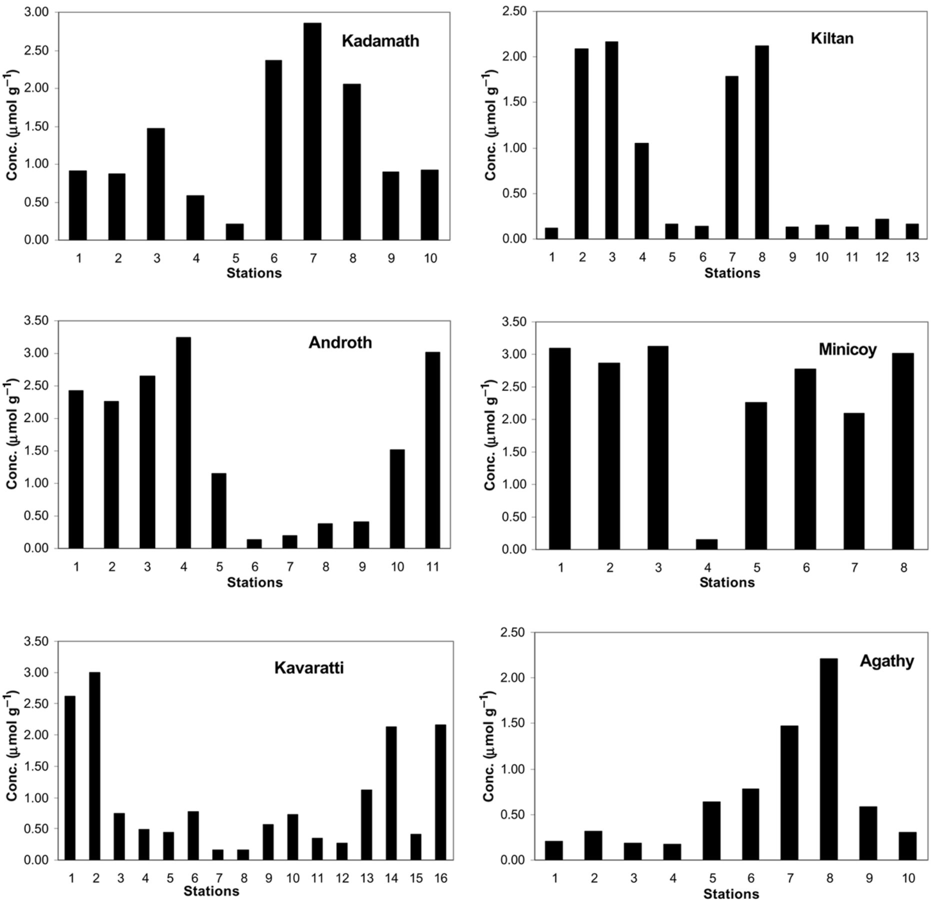

FIGURE 4 Spatial variability of nitrate in the sediments of the six islands of Lakshadweep Archipelago.

nitrogen. A depthwise decrease in concentration of nitrite in the core sample taken from the southern part of this island was noted, while a zigzag pattern was depicted for the distribution of both nitrate and total nitrogen in the southern core sample. On tracing the depthwise variability of nitrate and total nitrogen in core samples collected from the northern part of the island (stations 10–14), an increasing trend in concentration was seen (2.124 and 24.596 μmol g^{-1} at 120 cm depth). The nitrite levels of the surface soil samples (stations 15 and 16) collected were in accordance with the surface values of the respective northern and southern core samples, whereas the nitrate and total nitrogen values seemed to fit within the ranges recorded above, without any particular resemblance to core/lagoon transect samples.

On Agathy Island, as observed in the case of Kavaratti, wide fluctuations were not reflected in the nitrite-nitrogen concentrations of different sedimentary environments. A uniform pattern of nitrite distribution was observed at various stations of the lagoon transect. This trend was also shown by the seaside transect as well as the soil samples of this island. In the lagoon transect of Agathy, maximum nitrate concentrations were reported at the berm (0.32 μmol g^{-1}), and these decreased towards the lagoon (0.18 μmol g^{-1}). In this transect, total nitrogen was observed to be at a maximum in the low-tide samples (101.81 μmol g^{-1}). In the seaside transect, both nitrate-nitrogen and total nitrogen increased from land to lagoon. The soil samples of this island also showed comparatively higher values of total nitrogen (10.23–34.86 μmol g^{-1}).

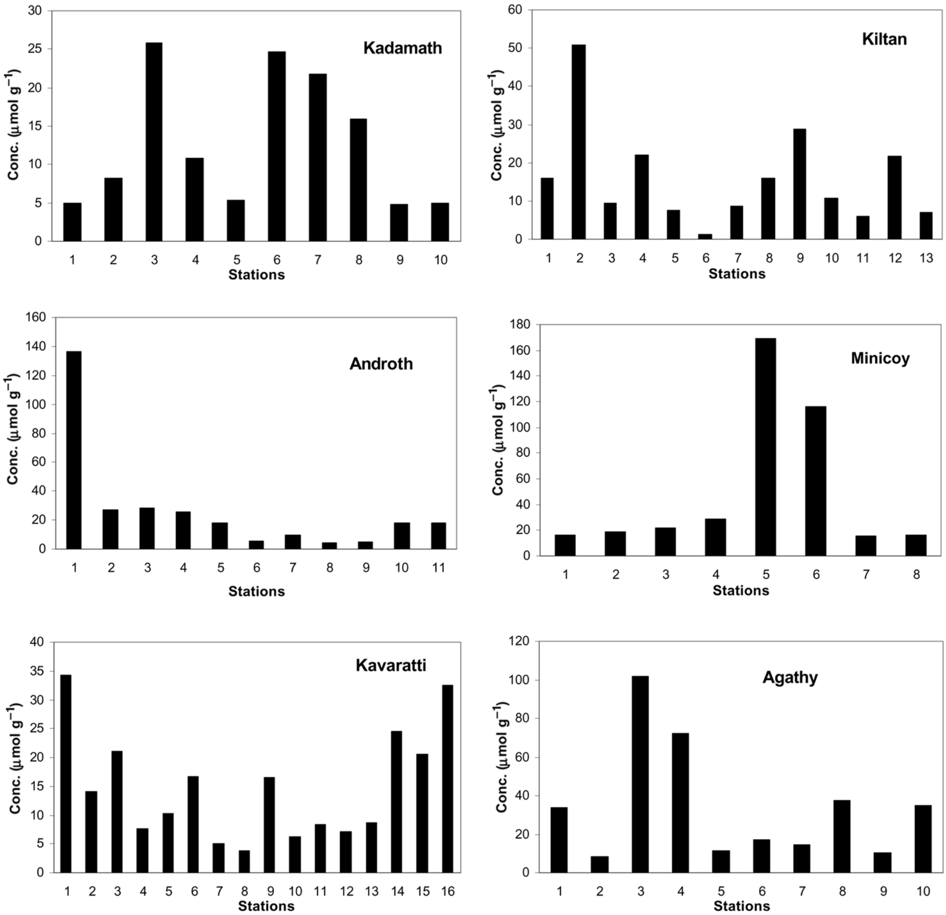

FIGURE 5 Spatial variability of total nitrogen in the sediments of the six islands of Lakshadweep Archipelago.

In the distribution pattern of nitrogen forms on Minicoy Island, the lagoon transect samples showed a maximum nitrite concentration in the low-tide region, an observation similar to that of Agathy Island. The mangrove environment of this island showed a slightly higher concentration of nitrite than the other environments. Nitrate-nitrogen in the lagoon transect of Minicoy showed a maximum concentration in the low-tide samples (3.12 μmol g^{-1}) and a minimum in the lagoon (0.15 μmol g^{-1}). In this transect, total nitrogen increased from land (16.33 μmol g^{-1}) to lagoon (28.63 μmol g^{-1}). The mangrove samples had a nitrate concentration in the range of 2.25–2.78 μmol g^{-1}, whereas the soil samples also showed a narrow range of nitrate concentration, *i.e.* 2.09–3.01 μmol g^{-1}. Comparing the variations of total nitrogen in the different sedimentary environments of Minicoy Island, maximum concentrations were found in the mangrove environment (116.42–168.87 μmol g^{-1}), and these were the maximum reported values for this island.

The core samples collected from Kiltan Island did not show wide fluctuations in the pattern of nitrite distribution with depth. Regarding the variations in the seaside transect, concentration gradually increased from land (0.01 μmol g^{-1}) to high-tide samples (0.02 μmol g^{-1}). In the lagoon transect the concentration range observed for different stations in this transect was very narrow.

In the lagoon transect of Kiltan, maximum nitrate-nitrogen concentration was reported for low-tide samples (2.16 μmol g^{-1}) and minimum in the land (0.12 μmol g^{-1}). In the distribution pattern of total nitrogen, maximum was at berm (50.81 μmol g^{-1}) and minimum at low tide (9.50 μmol g^{-1}). In the core samples of this island, nitrate showed no wide variation with the depth of the core. For total nitrogen, a depthwise decreasing trend in concentration was observed (28.77–5.99 μmol g^{-1}). In the distributional characteristics of nitrate and total nitrogen in the seaside transect of Kiltan, a lagoonward increasing trend was observed. The maximum values reported in this transect were 2.12 μmol g^{-1} for nitrate and 15.86 μmol g^{-1} for total nitrogen.

Kadamath Island, in the lagoon transect, maximum nitrite concentration was observed in the land (0.02 μmol g^{-1}) and minimum for low-tide stations (0.001 μmol g^{-1}). In the seaside transect, the distributional pattern of nitrite was zigzag, *i.e.* the concentration decreased from land (0.02 μmol g^{-1}) to berm (0.01 μmol g^{-1}), which again increased towards lagoon (0.02 μmol g^{-1}).

In Kadamath, nitrate as well as total nitrogen displayed their maximum at low-tide stations (1.47 and 25.82 μmol g^{-1}, respectively) of the lagoon transect. The distributional characteristics of nitrate and total nitrogen were random in the seaside transect of Kadamath. Both in the lagoon and in the seaside transect, minimum values of total nitrogen were reported in the land samples.

The nitrite concentration in the core sample taken from the seaside of Androth was found to be within a very narrow range (0.02–0.03 μmol g^{-1}) but increased with core depth, whereas in the landside core, no definite distribution pattern was observed, *i.e.* it showed a maximum at the surface (0.02 μmol g^{-1}), decreasing towards 1.5 m depth (0.01 μmol g^{-1}) except for a slight increase at 90 cm depth (0.02 μmol g^{-1}). In the seaside core, the maximum nitrate concentration was at 1 m depth (2.66 μmol g^{-1}), whereas that for the landside core was at the surface (3.25 μmol g^{-1}). Both the core samples (landside and seaside) showed a decreasing trend in the distribution of total nitrogen with core depth (1 m for the seaside core and 1.5 m for the landside core). The maximum values of total nitrogen found for these cores transects were 136.68 μmol g^{-1} (seaside) and 25.38 μmol g^{-1} (landside).

4 DISCUSSION

The various nitrogen forms, namely nitrite, nitrate and total nitrogen, quantified in the lagoon transects of some islands like Kavaratti generally showed an increasing trend from lagoon to land. This landward enrichment suggests either that inshore sources of nitrogen are not transported offshore in measurable quantities or that they are metabolized before they can accumulate in offshore sediments. The relatively low levels of intermediate nitrite can be explained by the fact that ammonia oxidation to nitrite and subsequent oxidation to nitrate may be closely coupled in the different microenvironments of this island. A comparatively higher concentration of nitrate is partially an indication of the higher rate of nitrification in sediments using the available ammonia and oxygen released from the root zone of plants or diffusing from the sediment surface. Further, the shallowness of the coastal water column comprising the lagoon and seaside transect stations favours increases in the dissolved oxygen level, which also enhances nitrification processes. The major source of nitrogen in sediments was attributed to the nitrogen fixation by benthic algal communities, which strongly influence the nitrogen budget of coral reefs (Wiebe, 1988).

A decreasing trend in the concentration of nitrogen forms with depth was observed in the core samples of Kavaratti Island (southern part). On Kiltan Island also, total nitrogen showed a decreasing trend from the surface to 1.3 m depth. A similar pattern of nitrogen distribution was displayed in the landside and seaside cores collected from Androth Island. The contribution from high benthic biodiversity, besides the death and decay of seagrass and seaweeds, results in a higher rate of release of organic materials and nutrients. These compounds are retained by sediments according to the nature of the sediment texture, which contributes to the pool of organic nitrogenous compounds. The surface stratum of the above-mentioned islands was characterized by a higher percentage of organic carbon content (Fig. 6), which may act as a good source of organic nitrogenous compounds, thereby enhancing the nitrification mechanism in the surface stratum of the soil. On Kavaratti and Androth, the organic carbon content showed a remarkable decrease with depth (17.20–0.56% in Androth and 22.27–0.595% in Kavaratti). Thus, the enrichment of organic nitrogenous compounds together with the presence of nitrifying agents in the well-oxygenated surface stratum of the soil may account for the accumulation of nitrogen forms in these surface layers. On Kiltan Island, the organic carbon content was high at the surface as well as at 1.3 m depth; still, the concentration of nitrogen forms decreased with depth, which may be due to the absence of nitrifying agents in the deoxygenated zone (higher depths) of the soil.

Total nitrogen showed an increasing trend from land to lagoon, in the lagoon transects of Agathy, Minicoy, Kadamath and Kiltan Islands. The lagoons are shallower than

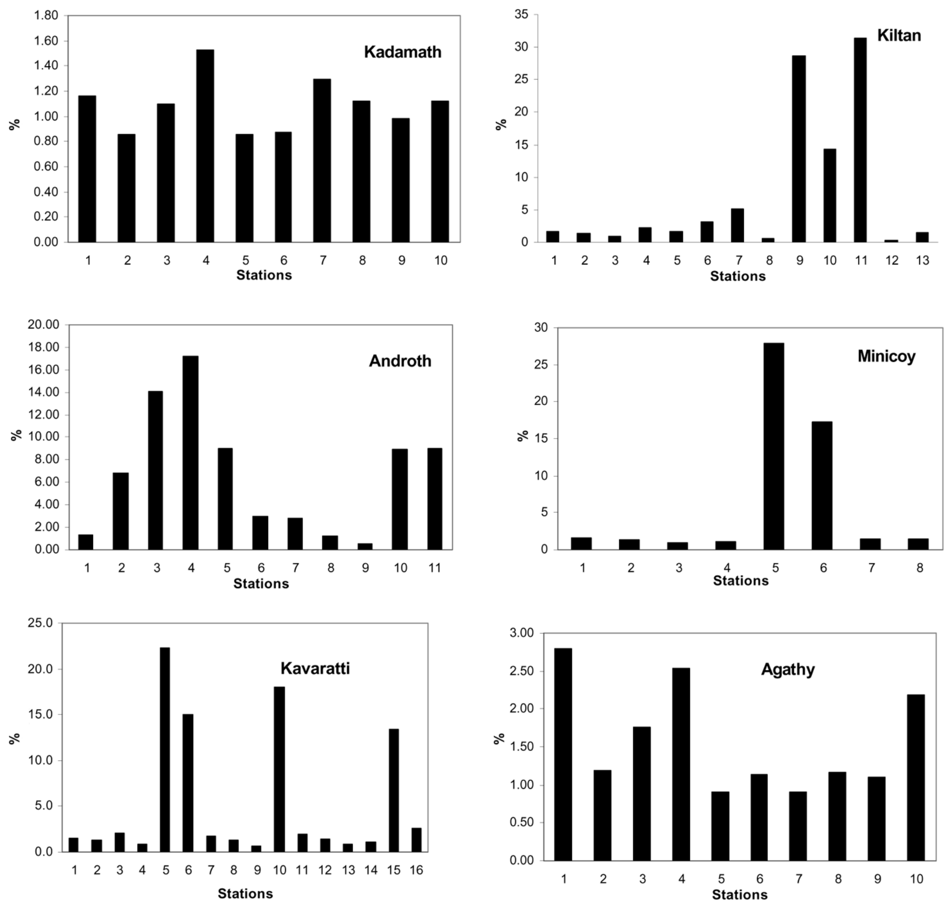

FIGURE 6 Spatial variability of Org. C in the sediments of the six islands of Lakshadweep Archipelago.

the surrounding area. The rate of nitrogen fixation and the space and surface available for nitrogen fixation are comparatively higher in these zones. The amount of nitrogenous waste materials produced from the reefs and surrounding area introduced into the lagoon will have limited exchange with the sea. The settlement and aggregation of organic matter through migratory fishes may contribute a certain amount of total nitrogen (Vinithkumar *et al.*, 1999). Sea grasses play an important role in contributing amino acids to the water and sediments, which adds to a higher concentration of total nitrogen in the sediments (Jagtap and Untawale, 1984), particularly in the shallow environments of the lagoon and low-tide samples. On Kiltan Island, the land samples were of medium-sandy nature, whereas on moving towards lagoon, the particle size changed to coarse sand. On Minicoy there was also a variation in sedimentary characters; here, land samples had a medium sandy nature, which changed to fine sand for the rest of the lagoon transect. Sediments with a higher percentage of sand were found to be low in organic carbon and nitrogen (Jagtap and Untawale, 1984). The fine sandy nature of the sediments seems to impart a greater retention capacity for organic nitrogenous compounds than coarse and medium sandy sediments, thus leading to a higher percentage of total nitrogen in the fine sandy lagoon sediments of the above-mentioned islands.

The low values of total nitrogen reported at some stations may be due to the utilization of nitrogenous compounds and lower rate of nitrogen fixation. Coral-reef flats are known to export dissolved organic nitrogen (Wiebe, 1988). The nitrogen released may be lost permanently from the system and/or exchanged among the different compartments of the system, and this could be the reason for the changes observed in the inter-environmental variations of nitrogenous compounds. In addition to these sedimentological factors (organic matter, carbon content, textural characteristics of the sediments, etc.), benthic producers also provide information regarding the nitrification process, as the sediment contributes the major nitrogen source for benthic producers in the shallow water environment of the reefs (Szmant, 1997; Costa Jr *et al.*, 2000).

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