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^{15}N enrichment in the surface Particulate Organic Nitrogen of the north-eastern Arabian Sea from the middle to the waning phase of the winter monsoon: possible causes

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Abstract

A temporal increase of ∼5‰ in the average nitrogen isotopic composition ($\delta^{15}N_{PON}$) of surface particulate organic nitrogen was observed in the open north-eastern Arabian Sea during January to late February-early March 2003, despite the presence of *T. erithraeum* (up to ∼11%), a diazotroph that fixes atmospheric N_2, in the latter period. Hydrographic conditions and residence time of nitrate in the water column suggested that this increase could be a combined effect of denitrification in the subsurface layer and inefficient utilization of nitrate entrained in the water column during January.

1 Introduction

Abundance of ^{15}N in marine organic and inorganic pools is known to vary significantly over a range of spatial and temporal scales (e.g., Saino and Hattori, 1980; Altabet, 1996; Rau et al., 1998). The nitrogen isotopic composition, $\delta^{15}N$, of naturally occurring land derived particulate matter has been reported to have lower values (1.5–2.5‰; Miyake and Wada, 1967), while those of marine organisms have a strong dependence on the $\delta^{15}N$ of the source and are reported to vary significantly (e.g., Saino and Hattori, 1980; Altabet, 1996; Wada and Hattori, 1991; Rau et al., 1998). The $\delta^{15}N$ of phytoplankton averages around 7‰ while zooplankton and fishes, which are at higher trophic levels in the food web, have values around 10‰ and 15‰ respectively (Wada and Hattori, 1976). Isotopic variations in different chemical species of nitrogen are caused by mass dependent fractionations associated with various biogeochemical transformations such as N_2 fixation, denitrification, nitrification and nitrate assimilation. Denitrification leads to ^{15}N enrichment of the remaining (dissolved) nitrate while nitrification causes enrichment of ^{15}N in the (dissolved) ammonium pool (Mariotti et al., 1984). Isotopic fractionation up to 20‰ has been found due to denitrification (Miyake and Wada, 1971; Cline and Kaplan, 1975; Liu and Kaplan, 1989), nitrification (Miyake and Wada, 1971) and nitrate assimilation (Wada et al., 1971). Fixation of atmospheric nitrogen is known to lower the δ^{15}N values of particulate organic nitrogen (PON), atmospheric nitrogen being the isotopic reference, is zero ‰; the areas with lower δ^{15}N may thus be attributable to N_2 fixation. However, lower δ^{15}N could also be caused by the preferential removal of ^{15}N enriched matter by sinking material, leading to a depletion of ^{15}N in the remaining suspended matter (Altabet, 1988). ^{15}N measurements on marine organic matter can provide information on the mechanisms and rates of these transformations and largely reflect the isotopically selective processing of nitrogen by biota (Rau et al., 1998).

Isotopic fractionation during the formation of PON governs its isotopic signature and therefore δ^{15}N of PON ($\delta^{15}N_{PON}$) records the nitrogen availability in the euphotic zone (Wada and Hattori, 1991). The isotopic signature inherited during the biological fixation of nitrogen by particulate matter varies with the substrate concentration as well as with algal species, physiology and growth rate (e.g. Wada and Hattori, 1978; Wada, 1980; Montoya and McCarthy, 1995). Significant correlations found in the world ocean between $\delta^{15}N_{PON}$ and nitrate concentrations and variations in $\delta^{15}N_{PON}$ have been explained by Rayleigh fractionation kinetics for a closed system (Altabet, 1996) implying that $\delta^{15}N_{PON}$ can be an index of nutrient availability and utilization. Consequently, δ^{15}N in sediments has been used to reconstruct paleonutrient conditions and biological productivity (Calvert et al., 1992; Francois et al., 1992; Altabet and Francois, 1994; Farrell et al., 1995).

Direct measurements of surface $\delta^{15}N_{PON}$ in the Indian Ocean are so far quite limited (e.g., Kumar et al., 2004, 2005; Montoya and Voss, 2006). Here we present new data on the surface $\delta^{15}N_{PON}$ in the north-eastern Arabian Sea from the middle and the waning phases of the northeast monsoon (NEM). We also investigate the effectiveness of nutrient utilization in the photic zone and whether the observed increase is a result of surficial phenomena or it is related to denitrification in the subsurface layer, prevalent here during the sampling period.

2 Material and methods

Surface water was collected using a clean plastic bucket during two cruises in the northeastern Arabian Sea during the winter monsoon, 2003. The first cruise was onboard ORV *Sagar Kanya* (SK-186) from 4–17 January and the second cruise was onboard FORV *Sagar Sampada* (SS-212) during late February–early March (28 February–5 March). The sampling stations were not exactly the same during both cruises; however, they were mostly in the same region i.e., off Gujarat, India (Fig. 1a and b). Thirteen stations during January and five stations during late February-early March were sampled. The latter was a typical bloom period (e.g., *Noctiluca miliaris*) and significantly higher productivity was observed compared to January (Kumar et al., 2007[1]). Biological measurements from these two cruises have been recently reported (Parab et al., 2006). Samples were filtered on precombusted (4 h @ 400°C) 47 mm Whatman GF/F filters and subsequently dried and stored for further mass spectrometric analysis. The analysis was performed using a CarloErba elemental analyser interfaced via Conflo III to a Finnigan Delta Plus mass spectrometer. The variability in particulate organic nitrogen between duplicate samples was less than 10% whereas it was less than 0.3‰ for δ^{15}N. Procedures have been elaborated earlier by Kumar et al. (2004, 2005). Nitrate concentrations were measured using an Autoanalyzer (SKALAR) based on standard spectrophotometric technique.

3 Meteorological and hydrographic conditions

The present study was conducted during the middle and waning phases of the northeast monsoon. During this period the Arabian Sea witnesses a surface water temperature difference of almost 5–6°C from the south to the north (north being cooler).

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During January 2003 the sea surface temperature (SST) showed a general northward decrease, with a maximum SST of 28.6°C at one of the southernmost stations (Stn. 2) and a minimum SST of 23.9°C at one of the northernmost stations i.e., Stn. 11 (Fig. 2a). The SST pattern during late February–early March was almost the same as in January i.e., decreasing SST northwards (Fig. 2b).

The depth profile of temperature during January suggested a general northward increase in temperature-based mixed layer depth (henceforth MLD) with ~40 m at southern station 2 to ~107 m at one of northern (Stn. 10) stations (Fig. 2c). The MLD pattern during late February-early March was quite different from that of January. The MLD for the first two stations was not clearly defined and the depth profile showed a continuous decrease in temperature with slight undulations (Fig. 2d). This might be an indication of the varying degree of mixing with depth between cooler water and relatively warmer surface water. The cooler water might have been supplied from deeper sources or due to horizontal advection from a nearby region. Interestingly, temperature profiles of stations 4–6 were marked by a relatively sharper decrease in temperature with depth, indicating a shallow mixed layer (33 to 55 m) for these stations. Although SSTs at these locations were not drastically different from January, the MLDs were markedly shallower (it was near 80 m in the same region during January). Relatively shallow mixed layer at these locations persisted despite higher wind speed (8 ms^{-1}) indicating the limited role played by wind in the deepening of MLD during this period. Therefore, the shoaling of MLD from January to late February-early March indicates the temporal decrease in effectiveness of winter cooling in the region.

Winds during January were north/north-easterly with an average speed of around 4 ms^{-1}. During late February-early March, the wind speed at the first station was around 2.74 ms^{-1}, typically in the range observed during January. However, it was more than 8 ms^{-1} at other stations.

4 Results

4.1 January, 2003

Of the 13 sampling stations, 9 were located in the open ocean (Stn. 2 to Stn. 10) and 4 in the coastal region (Stn. 11 to Stn. 14). Overall, the PON content varied widely ranging from 0.16 μM N at station 8, which was an open ocean station to 2.10 μM N at station 13, a coastal station. Overall, PON averaged around 1.04 (with a std. dev. of ±0.47) μM N. The PON at open ocean locations varied from 0.16 to 1.27 μM N averaging around 0.90 (std. dev.±0.41) μM N (Table 1), whereas at coastal stations variation was between 0.89 to 2.1 μM N with relatively higher average of 1.35 (std. dev.±0.52) μM N. Among the open ocean stations, there was a clear demarcation in PON concentration, where the first four stations (towards south) had PON more than 1 μM N with an average of 1.23 μM N. The rest of the open ocean locations (towards north) had PON less than 1 μM N, averaging around 0.64 μM N.

The $\delta^{15}N_{PON}$ also showed a wide variation in the region during this month with minimum of 1.7‰ at station 7, which was an open ocean location, to 7.5‰ at station 13, a coastal location. Interestingly, PON at station 13 was also a maximum during the whole study period. No systematic spatial pattern was observed in $\delta^{15}N_{PON}$. Overall, the $\delta^{15}N_{PON}$ averaged around 4.7 (±1.7) ‰. The average $\delta^{15}N_{PON}$ for open ocean locations was around 4.4 (±1.5) ‰ and no clear demarcation was observed as in the case of PON content. The $\delta^{15}N_{PON}$ of coastal locations averaged around 5.3 (±2.2) ‰. Barring three locations during the study period (Stations 3, 7 and 12) the $\delta^{15}N_{PON}$ was $\geq$ 4‰ during January.

The community composition, based on averages of cell counts was dominated by diatoms ~84% of total population (Parab et al., 2006).

4.2 Late February–early March, 2003

During this period, five open ocean stations were sampled. The PON content varied from 1.0 to 2.25 μM N with an average value of 1.6 (std. dev. ±0.5) μM N. The $\delta^{15}N_{PON}$ varied from 7 to 11‰ with an average of 9.4 (±1.7) ‰. The PON content and $\delta^{15}N_{PON}$ are listed in Table 1.

Community structure was ~50% diatoms, with the rest domintaed by *Noctiluca miliaris* and some *T. erythraeum* (Parab et al., 2006).

A plot of $\delta^{15}N_{PON}$ and PON content for whole data set revealed a positive relationship between the two with $r^2 = 0.41$(Fig. 3a). No significant ($r^2 = 0.14$) relationship was observed between the surface nitrate and $\delta^{15}N_{PON}$ (Fig. 3b). A strong negative correlation is expected between the two, as in regions where the surface nitrate concentration is very high. The observed poor relationship may be because of low nitrate concentrations and/or the limited number of measurements. Nevertheless, an overall increase in $\delta^{15}N_{PON}$ of the region during late February-early March is clearly borne out by the data. A comparison of $\delta^{15}N_{PON}$ of open ocean locations of January to late February-early March revealed an average increase of ~5‰, which is highly significant (at 0.00012 level). A significant increase of ~0.7 μM N in PON content was also concurrently observed.

Montoya and Voss (2006) have also reported enriched $\delta^{15}N_{PON}$ (average ~8.34‰) in upper 100 m of water column during May–June 1995. However, they encountered surface $\delta^{15}N_{PON}$ as low as ~ 4‰ at 10° N latitude which they attributed to a *Trichodesmium* bloom. In the present study, although up to ~11% of N_2-fixer (*T. erythraeum*) was present at one station (Stn. 6) along with a predominantly diatom population, $\delta^{15}N_{PON}$ values did not decrease significantly.

5 Discussion

Our observations in winter, combined with the enriched $^{15}N_{PON}$ observed by Montoya and Voss (2006) suggest that the enrichment that happens during late winter could persist till the intermonsoon. Montoya and Voss (2006) have argued that isotopic signature of denitrification in the form of enriched $^{15}N_{NO3}$ propagates upward to the surface through vertical mixing and subsequent uptake by phytoplankton results in ^{15}N enriched PON. However, we did not see the signature of denitrification i.e., enriched $^{15}N_{PON}$, immediately after the entrainment of enriched nitrate in the surface layer; there was a delay in the response of about one month, consistent with the residence time of nitrate in the water column. Therefore, the observed increase in $\delta^{15}N_{PON}$ during the present study appears to be the combined effect of subsurface (denitrification) and surficial processes.

The observed enrichment from January to late February–early March may be due to: (a) the overall increase in the nitrogen isotopic composition of the source nutrient taken up by the phytoplankton, (b) change in the fractionation during uptake by the phytoplankton, if the $\delta^{15}N$ of the source nutrient was the same during both periods, and (c) a combination of both (a) and (b). Very high new production during late February-early March (Kumar et al., 2007[1]) suggested nitrate to be the principal source of nutrient, which could be deeper nitrate (> euphotic depth) or nitrate already present in the water column which got entrained during January due to the deepening of the mixed layer caused by winter cooling (Madhupratap et al., 1996). Shallow mixed layers at the last three stations during late February–early March preclude the possibility of nitrate supply from deeper layers. However, the undulating and continuously sloping mixed layer at the first two stations suggest such a possibility: nitrate supply might be from deeper layers or due to advection from a nearby region where the effect of winter cooling had not completely vanished. If the nutrient source during both the months was deeper nitrate, the dramatic increase in $\delta^{15}N$ of nitrate was possible only when there was an intensification of denitrification in the intermediate waters leading to more enriched nitrate. However, if the nitrate already entrained in the surface layer is assumed to be the only source, the continuous use of available nitrate might probably leave the remaining pool enriched enough to explain the observed variation. The following subsections explore both the above possibilities.

5.1 Possible role of denitrification

Although most part of the open Arabian Sea is well oxygenated, an acute depletion of oxygen leading to denitrification occurs at intermediate depths (100–500 m) particularly in the eastern and the central Arabian Sea. The total area affected by denitrification has been estimated to be around 1.37×10^6 km^2 (Naqvi, 1991). The variation in the denitrification intensity based on seasonal scale has also been observed (Naqvi et al., 1990).

Denitrification leads to the enrichment of ^{15}N in the remaining nitrate, which eventually acts as a source for phytoplankton. But why was there so significant difference in δ^{15}N of PON from January to late February–early March? Was that the signature of relative intensification of denitrification at intermediate depths, which eventually lead to more enriched nitrate from January to late February–early March? Banse (1984) observed the relative suppression of denitrification during winter due to the supply of oxygen to the upper part of the oxygen minimum zone as a result of enhanced diffusion due to deepening of the mixed layer. The deepening of mixed layer up to 107 m was observed at the northern stations during January. This might have helped in reducing the denitrification leading to lower δ^{15}N values of nitrate during January, as reflected by δ^{15}N of PON. This situation changed during late February–early March as the effect of winter cooling decreased. The shallow mixed layer at a few stations suggested that the environment during late February-early March was not so supportive of vertical mixing and hence the decrease in the aeration of deeper layers. This could have lead to increase in the intensity of denitrification and consequent increase in δ^{15}N of nitrate.

Brandes et al. (1998) estimated the δ^{15}N of nitrate in the Central Arabian Sea with highest value of 15‰ at 350 m during September and January. However, they observed a decrease in δ^{15}N of nitrate (6‰) at 80 m due to the dilution by lighter isotopes added due to nitrogen fixation in the surface layer. Assuming that there was a similar level of dilution of nitrate isotopic composition at around 80 m and the same fractionation factor for both months during the uptake of nitrate by phytoplankton, an increase in δ^{15}N of nitrate due to the intensification of denitrification from January to late February–early March appears likely.

5.2 Varying degree of dilution of deeper nitrate

Assuming that there was no intensification of denitrification from January to late February–early March and the δ^{15}N of dissolved nitrate produced in the denitrification layer remained same (say a steady state value of ~15‰), with no change in the ^{15}N fractionation by phytoplankton during the uptake of nitrate, the observed difference of 5‰ in the δ^{15}N of surface PON could be explained by the varying degrees of dilution of enriched nitrate due to nitrogen fixation in the surface layer. The dilution during January should be more than late February-early March. To estimate the level of dilution during both the months the highest value of δ^{15}N of PON of respective months would be considered as the δ^{15}N of ambient nitrate available for uptake, assuming these values to be a reflection of the δ^{15}N of nitrate consumed without fractionation. These values during January and late February–early March were 7.5 and 11‰ respectively. The January value (7.5‰) is closer to the known δ^{15}N of nitrate in World Ocean (~ 5‰; Miyake and Wada, 1967). The dilution level required to obtain the 7 and 11‰ can be obtained by simple isotopic mass balance:

15x + (1-x)*5 = 7.5 (January) and 15x +(1-x)*5 = 11 (March)

Where x is the proportion of deeper nitrate and (1–x) is fraction of required dilution by lighter nitrate. 5‰ and 15‰ are the assumed δ^{15}N compositions of natural and deeper nitrate respectively. From this equation it is quite clear that ~75% dilution would be required during January whereas only ~30% during late February–early March.

5.3 Change in the ^{15}N fractionation by phytoplankton

The third scenario might be varying ^{15}N discrimination with the same denitrification intensity and level of dilution during both the months. We assume that the δ^{15}N of nitrate in the denitrification layer was 15‰ which underwent similar dilution in the surface layer such that the isotopic composition changed to 11‰, the maximum observed $\delta^{15}N_{PON}$. This 11‰ nitrate would be acting as source during both the months. The observed difference of 5‰ now can be explained by different degree of fractionation during uptake in January and late February-early March to reach the observed isotopic composition. This depends upon the pool of available dissolved nitrate to the phytoplankton. The nitrate concentrations in surface water of the open Arabian Sea during these two months suggested the abundant nitrate (average ~1.8 μM) during January whereas its relative dearth (average ~0.3 μM) during late February-early March except at one station where it was 3.5 μM. This high nitrate during January and lower during late February–early March indicated the flexibility for phytoplankton to fractionate during January, and not during March; consequently, reflecting lower and higher δ^{15}N values during January and late February-early March respectively

5.4 Utilization of nitrate present in the surface layer

One important possibility of the nutrient during late February–early March was the inefficient utilization of the nitrate introduced in the water column during January. It is interesting to note that the euphotic zone integrated nitrate concentration in the water column at most of the stations during January was more than 150 mmol m^{-2} but the average new production was only ~2.3 mmol N m^{-2}d^{-1}. However, the integrated euphotic zone column nitrate during March was only around 80 mmol m^{-2} but the average new production was as high as 13 mmol Nm^{-2}d^{-1}. Also, the residence time of nitrate entrained during January was found to be more than 50 days (Kumar et al., 2007[1]). These observations indicate that the nutrients entrained in the water column due to convective mixing during January did not get completely consumed and remained in the water column. Assuming the surface layer of the ocean as a closed system and nitrate with initial isotopic composition (δ_o) of 7‰ (close to maximum observed $\delta^{15}\mathrm{N}_{\mathrm{PON}}$) as the source for phytoplankton during January, the isotopic composition of remaining nitrate (δ) would keep on changing with the consumption of the substrate pool (i e., fraction of substrate remaining, f) according to well known relationship:

$\delta=\delta_o+\varepsilon$ * ln (f) where, ε = (α-1)*1000; α is the fractionation factor.

Assuming that the isotopic composition of nitrate during late February-early March was 11‰ and taking ε = –5‰ (typical value assumed for fractionation; Waser et al., 1998) the f from the above equation comes to be 0.4. It reveals that at the start of the uptake during late February-early March 60% of the nitrate introduced in the water column during January had already been consumed. If we take ε = –9‰ as in some cases (e.g., Rau et al., 1998) only 40% of the nitrate was consumed and rest 60% was available for further consumption (Fig. 4).

6 Conclusions

During the north-east monsoon δ^{15}N of surface PON in the northeastern Arabian Sea increases from January to March by at least ~5‰. A combination of two factors, viz., intensification of denitrification and consequent ^{15}N enrichment of the dissolved nitrate pool, and inefficient utilization of the entrained nitrate by plankton, despite the patchy occurrence of N_2-fixers, appears responsible for this increase.

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Table 1. Particulate organic nitrogen (PON) and nitrogen isotopic composition (δ^{15}N) observed during present study.

Month	Stations	PON (μM N)	δ^{15}N (‰)
January (SK-186)	2	1.61	6.7
	3	1.06	2.7
	4	0.99	4.6
	5	1.27	5.8
	6	0.75	3.8
	7	0.79	1.7
	8	0.16	5.1
	9	0.65	4.7
	10	0.86	4.6
	11	1.2	5.7
	12	0.89	2.3
	13	2.1	7.5
	14	1.21	5.6
Late February–early March (SS-212)	2	1.04	10.4
	3	2.25	10.6
	4	1.61	10.9
	5	2	8.1
	6	1.13	7

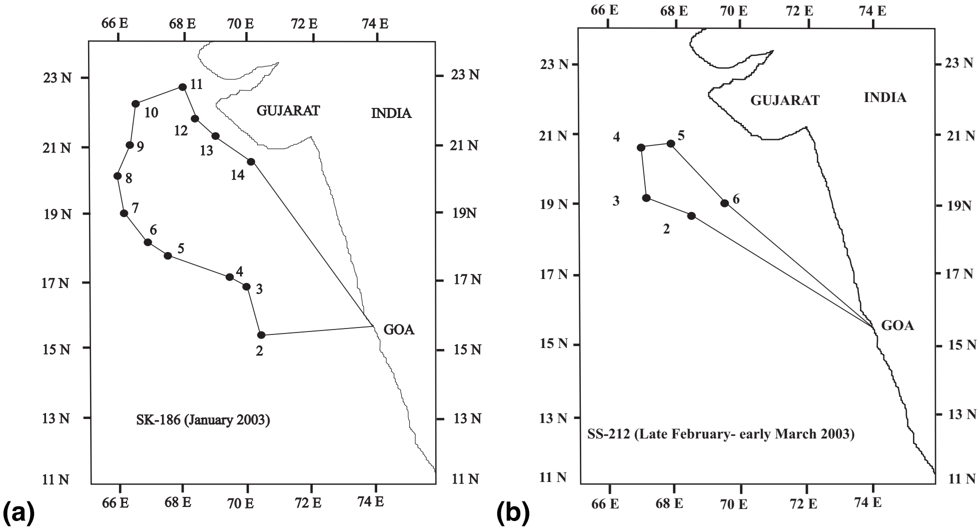

Fig. 1. Stations sampled during **(a)** January and **(b)** late February–early March.

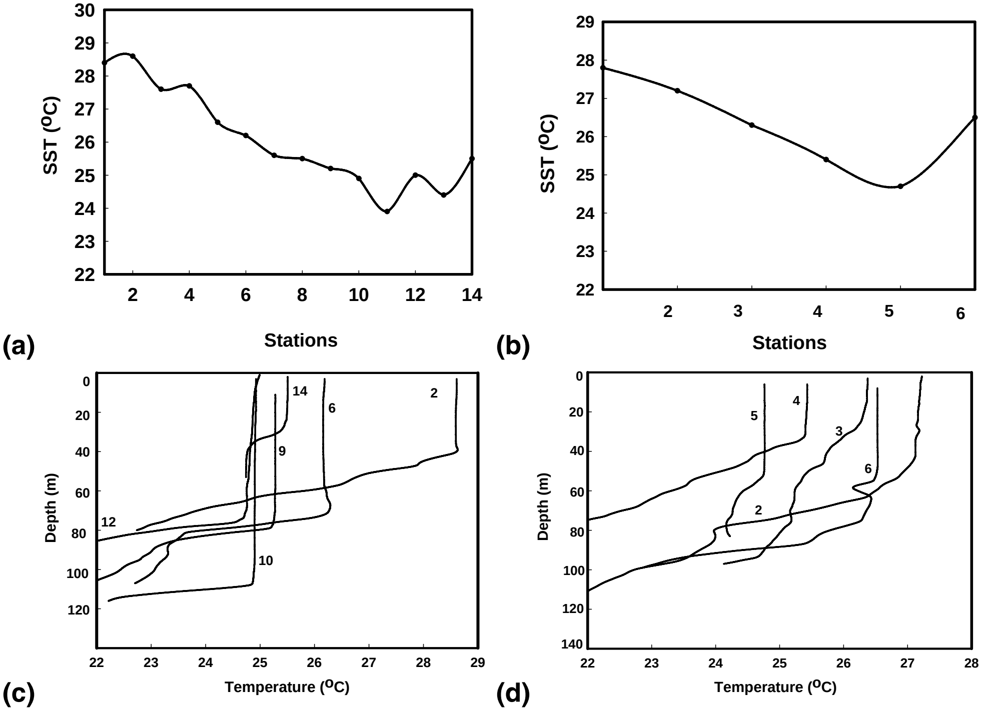

Fig. 2. Hydrodynamic conditions during sampling **(a)** sea surface temperature-January **(b)** sea surface temperature-late February–early March **(c)** temperature based mixed layer depth–January **(d)** temperature based mixed layer depth – late February–early March.

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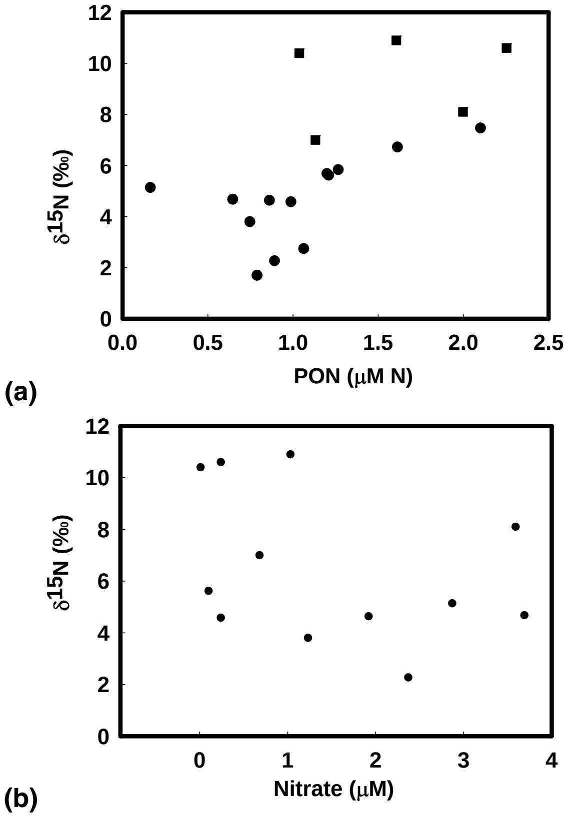

Fig. 3. Relationship between **(a)** PON and δ^{15}N **(b)** δ^{15}N and nitrate during present study.

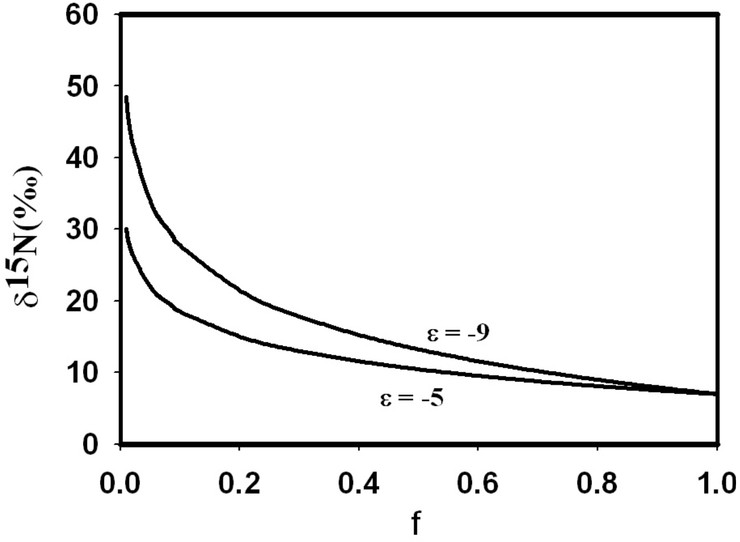

Fig. 4. Relationship between δ^{15}N and fraction of remaining substrate (f) assuming different fractionation factors.