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Nitrogen isotopic fractionation of particulate organic matter production and remineralization in the Prydz Bay and its adjacent areas

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Abstract

During the 29th Chinese National Antarctic Research Expedition, spatial variations in nitrogen isotopic composition of particulate nitrogen ($\delta^{15}N_{PN}$) and their controlling factors were examined in detail with regard to nitrate drawdown by phytoplankton and particulate nitrogen (PN) remineralization in the Prydz Bay and its adjacent areas. To better constrain the nitrogen transformations, the physical and chemical parameters, including temperature, salinity, nutrients, PN and $\delta^{15}N_{PN}$ in seawater column were measured from surface to bottom. In addition, the nitrogen isotopic fractionation factor of nitrate assimilation by phytoplankton in the mixed layer, and the nitrogen isotopic fractionation factor of PN remineralization below the mixed layer were estimated using Rayleigh model and Steady State model, respectively. Our results showed that suspended particles had its lowest $\delta^{15}N_{PN}$ in the surface layer, which was due to the preferential assimilation of ^{14}N in nitrate by phytoplankton. The $\delta^{15}N_{PN}$ in the mixed layer of the Prydz Bay and its adjacent areas decreased from the inner shelf to the outer basin, ascribing to the effect of isotope fractionation during phytoplankton assimilation. In mixed layer, the spatial distribution of $\delta^{15}N_{PN}$ associated with particulate organic matter (POM) production can be well interpreted according to Rayleigh model and Steady State model. The nitrogen isotope fractionation factor during phytoplankton assimilating nitrate was estimated as 10.0‰ by Steady State model, which was more reasonable than that calculated by Rayleigh model. These results validate the previous reports of fractionation factor during nitrate assimilation by phytoplankton. Increasing $\delta^{15}N_{PN}$ with depth below the euphotic zone correlated with the decreasing PN contents, and it was attributed to preferential remineralization of ^{14}N in PN by bacteria. In subsurface and deep layer, the $\delta^{15}N_{PN}$ distributions also conformed to Rayleigh model and Steady State model during PN remineralization, with a fractionation factor of about 3.6‰ and 3.2‰, respectively. It is the first time to estimate the fractionation factor during POM production and remineralization in the Prydz Bay and its adjacent areas. Such fractionation may provide a useful tool for the follow-up study of the nitrogen dynamics in the Southern Ocean.

Key words: PN, $\delta^{15}N_{PN}$, isotopic fractionation, POM production, remineralization, Prydz Bay

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1 Introduction

The nitrogen (N) isotopic composition of particulate N ($\delta^{15}N_{PN}$) has been used to trace nitrogen sources and transformations in marine systems (Altabet and McCarthy, 1985, 1986; Altabet, 1988; DiFiore et al., 2009; Liu et al., 2005; Lourey et al., 2003; Karl et al., 1997; Rau et al., 1991). For example, the utilization of nitrate, ammonium, and dissolved organic N, each process with potentially distinct isotopic fractionations, may significantly influence the $\delta^{15}N_{PN}$ in the water column (Carpenter et al., 1997; Minagawa and Wada, 1986; Montoya et al., 1991). The $\delta^{15}N_{PN}$ in sediments has been proposed as a proxy of surface water nitrate utilization in past or paleo oceanic environments, such as glacial/interglacial periods (Altabet and Francois, 1994a, b; Farrell et al., 1995; Francois et al., 1992; Francois et al., 1997; Ganeshram et al., 1995; Nakatsuka et al., 1995). $\delta^{15}N_{PN}$ is also a useful tracer of particulate organic matter (POM) formation and transportation in the deep sea (Altabet, 1988).

In typical open oceans, nitrate uptake by phytoplankton and organic matter remineralization are two key processes that influence spatial variations of $\delta^{15}N_{PN}$. In surface layer, the POM is usually ^{15}N-depleted because of the preferential utilise of ^{14}N in nitrate during biological assimilation (Altabet, 2006; Sigman and Casciotti, 2001). This isotopic fractionation varies with substrate contents as well as algal species, physiology and growth rate (Montoya and McCarthy, 1995; Pennock et al., 1996; Wada and Hattori, 1978; Wada, 1980; Waser et al., 1998a, b). Whereas below

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the euphotic zone, the preferential remineralization of ^{14}N in POM leads to a progressive increase in residual $\delta^{15}N_{PN}$ and shape the vertical profiles of $\delta^{15}N_{PN}$ in deep water (Altabet, 1988, 1996).

The isotopic fractionation factor (ε) is an important parameter in quantitatively understanding N transformations among different biological processes. For N fixation in surface ocean, the POM produced by diazotrophs is relatively ^{15}N-depleted with $\delta^{15}N_{PN}$ as low as –2‰ to 0.5‰, because diazotrophs assimilate dissolved dinitrogen gas (N_2) with the value of $\delta^{15}N$ around 0.6‰ (Carpenter et al., 1997; Minagawa and Wada, 1986). As for nitrate assimilation by phytoplankton, the ε value is typically within a range of 1‰–10‰, with most estimates close to 5‰–8‰ (Montoya and McCarthy, 1995; Needoba et al., 2003, 2004; Pennock et al., 1996; Wada and Hattori, 1978; Waser et al., 1998a, b). Culture studies suggest an ε value up to 20‰ for ammonium assimilation while less than 1‰ for assimilation of nitrite and urea, both of which are nearly exhausted in surface mixed layer of open ocean (Pennock et al., 1996; Wada and Hattori, 1978; Waser et al., 1998a, b). In general, complete remineralization includes several stages participated by different microbial metabolisms. Organic N is usually degraded prior to ammonium by aerobic respiration and then oxidized to nitrate via the intermedia nitrite by nitrification. Isotopic fractionation occurs at all steps involved in remineralization with a net isotope effect of ≤5‰ estimated by Sigman and Casciotti (2001).

$\delta^{15}N_{PN}$ data in the Southern Ocean are really scarce especially for the deep water. Rau et al. (1991) reported that the $\delta^{15}N_{PN}$ in the surface Weddel Sea ranges from 5.4‰ to 6.1‰. The $\delta^{15}N_{PN}$ was about 2.3‰ in the mixed layer and about 6‰ in the waters below 300 m in the Southern Ocean (DiFiore et al., 2009). Lourey et al. (2003) showed that $\delta^{15}N_{PN}$ was relatively constant at 1‰ in the surface water of Subantarctic Zone while ranging between –4‰ and 0‰ in the Polar Frontal Zone in summer. Due to the advantages of N isotopes in providing constraints on biological utilization of nutrients, both the $\delta^{15}N$ signals of nitrate and particulate organic N in the water column have been widely used to diagnose supply and uptake of N in the Southern Ocean during the past decades (Altabet and Francois, 1994b, 2001; DiFiore et al., 2006, 2009; Francois et al., 1992; Francois et al., 1997; Sigman et al., 1999a, b, 2000). The negative linear relationship between nitrate concentration and $\delta^{15}N_{PN}$ indicates a strong link between nitrate utilization and N isotopes in the Southern Ocean. (Altabet et al., 1991; Altabet, 1996; Altabet and McCarthy, 1985, 1986). Fractionation factor during nitrate assimilation by phytoplankton was estimated in the Southern Ocean by using the Rayleigh model (Altabet and Francois, 1994a, 2001; DiFiore et al., 2006, 2009; Sigman et al., 2000). For instance, according to profiles of nitrate and $\delta^{15}N_{PN}$ in the upper 100 m of Antarctic Polar Frontal Zone in summer, Altabet and Francois (2001) estimated the fractionation factor to be 6‰–8‰ by the Rayleigh model and 7‰–10‰ by the Steady State model.

In the present study, variations of $\delta^{15}N_{PN}$ and their controlling factors in the Prydz Bay and its adjacent areas were examined during the Chinese National Antarctic Research Expedition. Based on the Rayleigh model and the Steady State model, the isotopic fractionation factors with regard to both POM production and remineralization were estimated. Our results suggest that $\delta^{15}N_{PN}$ is a good tracer in evaluating POM transportation from the surface to the deep sea.

2 Materials and methods

2.1 *Study area*

The Prydz Bay and its adjacent areas are key sea areas for scientific exploration in the Southern Ocean, which is a triangular-shaped embayment with an area of 80 000 km^2 in the Indian sector (Stagg, 1985; Zhang et al., 2014). It is located at the end of graben occupied by the world's largest Lambert Glacier, and forms the vast Amery Ice Shelf. The southwestern bay is connected to the Amery Ice Shelf, and bounded by the West Ice Shelf (around 80°E) to the east and the Cape Darnley (around 70°E) to the west.

The water masses in the Prydz Bay and its adjacent areas in summer are composed of Antarctic surface water (AASW), shelf water (SW) or ice shelf water (ISW), circumpolar deep water (CDW) and Antarctic bottom water (AABW) (Gao et al., 2003; Pu and Dong, 2003; Yu et al., 1998; Wong et al., 1998). The characteristics of these water masses were listed in Table 1. The AASW is divided into Antarcitc summer surface water (AASSW) with high temperature and low salinity, and the winter water (WW) with low temperature and high salinity. The depth of AASSW is usually less than 50 m with the temperature and the thickness increase with the increasing latitude in the bay, although with seasonal and annual variations (Chen et al., 1995). A temperature minimum below AASSW exists at depths of 50–100 m, which is called the winter water resulting from winter convection. The SW in the Prydz Bay is divided into high salinity shelf water (HSSW) and low salinity shelf water (LSSW). The SW expands northward at a depth of 50 m, and its intensity decreases gradually (Shi and Zhao, 2002; Pu et al., 2000). A water mass with the temperature lower than –1.9°C occurs near the bottom of the continental shelf, which is called ice shelf water (ISW) or supercooling water. The CDW is the largest water mass in the Southern Ocean, characterized by high temperature, high salinity and low dissolved oxygen (Le et al., 1996), rising from a depth of more than 1 000 m to the subsurface and even to the surface. The AABW is the densest water mass in global oceans with low temperature and high salinity, which is closely related to global deep circulations and heat transfers. The Weddell Sea and the Ross Sea are recognized the regions for AABW formation. The AABW is formed by mixing of the high-density shelf water and the CDW (Foster et al., 1987; Jacobs et al., 1970; Orsi et al., 1999; Orsi and Wiederwohl, 2009).

2.2 *Sample collection*

Seawater and suspended POM samples were collected throughout the entire water column onboard the icebreaker R/V

Table 1. The characteristics of water masses in the Prydz Bay and its adjacent areas in summer

Water masses		Temperature/°C	Salinity
Antarctic surface water (AASW)	Antarctic summer surface water (AASSW)	–0.5 to 2	32.5–34.5
	winter water (WW)	<–1.5	34.2–34.5
Shelf water (SW)	High salinity shelf water (HSSW)	<–1.5	>34.5
	Low salinity shelf water (LSSW)	<–1.5	<34.25
Ice shelf water (ISW)		<–1.9	34.3–34.5
Circumpolar deep water (CDW)		0.5–2	34.5–34.75
Antarctic bottom water (AABW)		<0	34.6–34.72

Xuelong from January to March 2013 during the 29th Chinese National Antarctic Research Expedition. A total of 24 stations were sampled in the Prydz Bay and its adjacent areas, with 9 stations located over the shelf, 8 stations over the slope and 7 stations in the basin (Fig. 1). Seawater samples were collected using a CTD rosette system. A total of 10 L of seawater was filtered through a precombusted Whatman GF/F membrane (400°C, 4 h). The filter was dried at 60°C for 15 min onboard and placed in a petri dish and stored frozen until analysis.

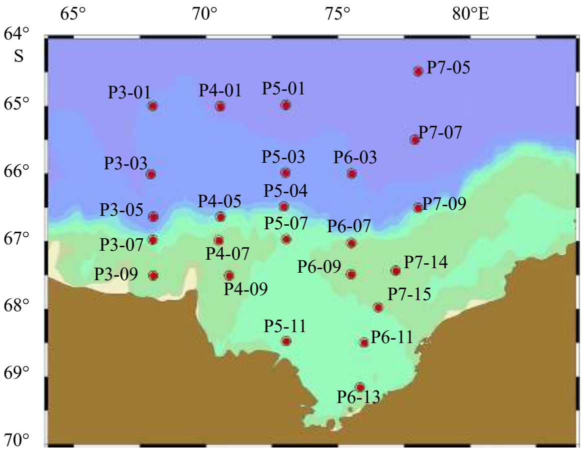

Fig. 1. Sampling locations in the Prydz Bay and its adjacent areas during summer 2013.

2.3 *PN and $\delta^{15}N_{PN}$ measurement*

When back to the land laboratory, the filter sample was dried at 60°C for 24 h prior, and wrapped into a tin capsule. The stable N isotopic composition ($\delta^{15}N_{PN}$, in terms of per mil) and PN content were measured using a Delta^{plus} XP mass spectrometer interfaced with a Carlo Erba NC2500 elemental analyzer (Thermo Finnigan). PN content was calculated according to a working curve with $C_6H_6N_2O$ as a standard. The $\delta^{15}N_{PN}$ value was defined as follow:

$$\delta^{15}N_{PN} = \left(\frac{R_{sample}}{R_{standard}} - 1\right) \times 1\ 000‰, \qquad (1)$$

where R_{sample} means the ratio of $^{15}N/^{14}N$ of samples measured by mass spectrometer; $R_{standard}$ means the ratio of $^{15}N/^{14}N$ of standard N_2.

In the isotopic measurement, atmospheric N_2 was adopted as a standard. The detection limit of PN content is 0.1 μmol/L. The analytical precision for PN and $\delta^{15}N_{PN}$ were 0.2% and 0.2‰, respectively.

2.4 *Temperature, salinity, nutrient measurement*

Temperature, salinity and nutrient data were provided by the Second Institute of Oceanography, Ministry of Natural Resourses. Temperature and salinity were recorded using a Seabird 911 plus CTD system. The analytical precision of conductivity and temperature was ±0.000 2 S/m and ±0.001°C. Seawater samples for dissolved nutrients (including nitrate, nitrite and ammonium) analysis were filtrated through a 0.45 μm cellulose acetate membrane. Nitrate, nitrite and ammonium were measured with cadmium reduction, pink azo dye and sodium hypobromite oxidation, respectively. The detection limit for nitrate, nitrite and ammonia were 0.05 μmol/L, 0.02 μmol/L and 0.020 μmol/L, respectively (GB 17378.4—2007).

3 Results

3.1 *In the basin*

Profiles of temperature, salinity, nitrate, ammonium, PN and $\delta^{15}N_{PN}$ in the basin were shown in Fig. 2. Temperature varied from –1.74°C to 1.58°C with an average of –0.08°C. Salinity varied from 33.68 to 34.71 with an average of 34.36. Generally, the temperature dropped to the lowest at 75 m or 100 m, and then increased to the highest at 500 m. Below 500 m, a distinct dicothermal layer occurs with temperature decreasing with the depth (Fig. 2a). At all stations, the lowest salinity occurs in the surface water and increases to a maximum at about 500 m, and then remained relatively constant below (Fig. 2b). The concentrations of nitrate varied from 27.54 μmol/L to 35.37 μmol/L with an average of 31.77 μmol/L (Fig. 2c). The concentrations of ammonium varied from 0.09 μmol/L to 2.79 μmol/L with an average of 0.63 μmol/L (Fig. 2d). The contents of PN varied in a range of 0.05–0.78 μmol/L with an average of 0.24 μmol/L (Fig. 2e). The PN content sharply decreased with the increasing depth in the euphotic zone, just being opposite to that of nitrate (Fig. 2c), which is attributed to phytoplankton photosynthesis in the euphotic zone. $\delta^{15}N_{PN}$ varied from –5.2‰ to 9.5‰ with an average of 1.6‰. The profiles of $\delta^{15}N_{PN}$ showed a minimum at surface and increased to ~5‰ at 100 m or 200 m, and then slightly increased with the depth (Fig. 2f).

3.2 *In the slope*

Profiles of temperature, salinity, nitrate, ammonium, PN and $\delta^{15}N_{PN}$ in the slope were shown in Fig. 3. Temperature varied from –1.85°C to 0.85°C with an average of –0.75°C. A dicothermal layer marked by a temperature minimum occurrs at 75 m. Salinity varied from 33.67 to 34.68 with an average of 34.30. The vertical patterns of temperature and salinity were similar to those in the basin. At most stations, the profiles of PN content showed a maximum at the surface, and decreased with depth (Fig. 3e). A slightly increase of PN content was observed in the bottom layers, which was different from those in the basin. The $\delta^{15}N_{PN}$ increased just below the surface and reached a maximum at roughly depth where the PN minimum occurred, while the $\delta^{15}N_{PN}$ decreased instead with the further increasing depth (Fig. 3f).

3.3 *In the shelf*

Profiles of temperature, salinity, nitrate, ammonium, PN and $\delta^{15}N_{PN}$ in the shelf were shown in Fig. 4. Temperature varied from –2.03°C to 0.24°C with an average of –1.39°C. The averaged temperature in the mixed layer and the subsurface was (–1.09±0.47)°C and (–1.54±0.51)°C, respectively. Salinity varied from 32.89 to 34.54 with an average of 34.18, and the average in the mixed layer and the subsurface was (33.67±0.39)°C and (34.33±0.22)°C, respectively (Table 2). The hydrological characteristics in the shelf was characterized with a lower temperature and salinity than those in the slope and basin, most probably due to the melts of sea ice and winter residual waters. Nitrate concentration decreased sharply from the subsurface ((28.89±4.06) μmol/L) to the mixed layer ((23.81±3.57) μmol/L). This upward decrease was most likely driven by phytoplanktonic assimilation of nitrate in the mixed layer. The vertical distribution of PN content was mirrored to those of nitrate (Figs 4c, e). The shallower mixed layer tends to have a greater drawdown of nitrate. Water-

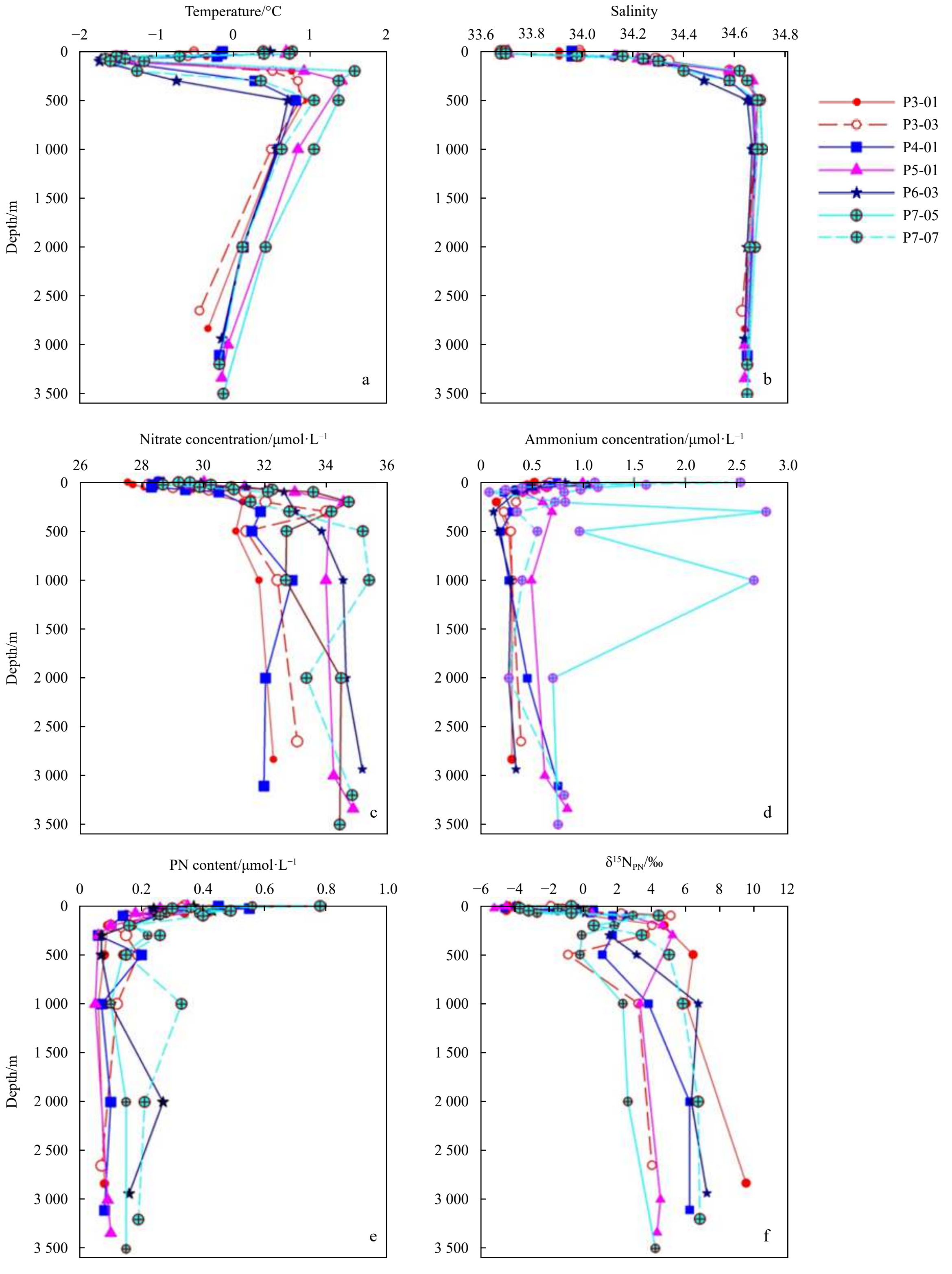

Fig. 2. Vertical profiles of temperature, salinity, nitrate, ammonium, PN, $\delta^{15}N_{PN}$ in the basin region.

depth-related increase in $\delta^{15}N_{PN}$ has been found in the whole shelf region. Temperature, nitrate and DIN concentrations in the shelf were lower than those in the slope and the basin. Ammonium and PN contents were higher in the whole water column of the shelf compared to the slope and the basin. Overall, salinity, nitrate, DIN and $\delta^{15}N_{PN}$ tended to increase from the mixed layer to the subsurface, while temperature, ammonium and PN concentrations tended to decrease from the mixed layer to the subsurface layer (Table 2).

3.4 *At Sections P3 and P7*

The spatial and vertical distributions of temperature, salinity, nitrate, ammonium, PN and $\delta^{15}N_{PN}$ at Sections P3 and P7 were shown in Fig. 5 and Fig. 6, respectively. Surface temperature and

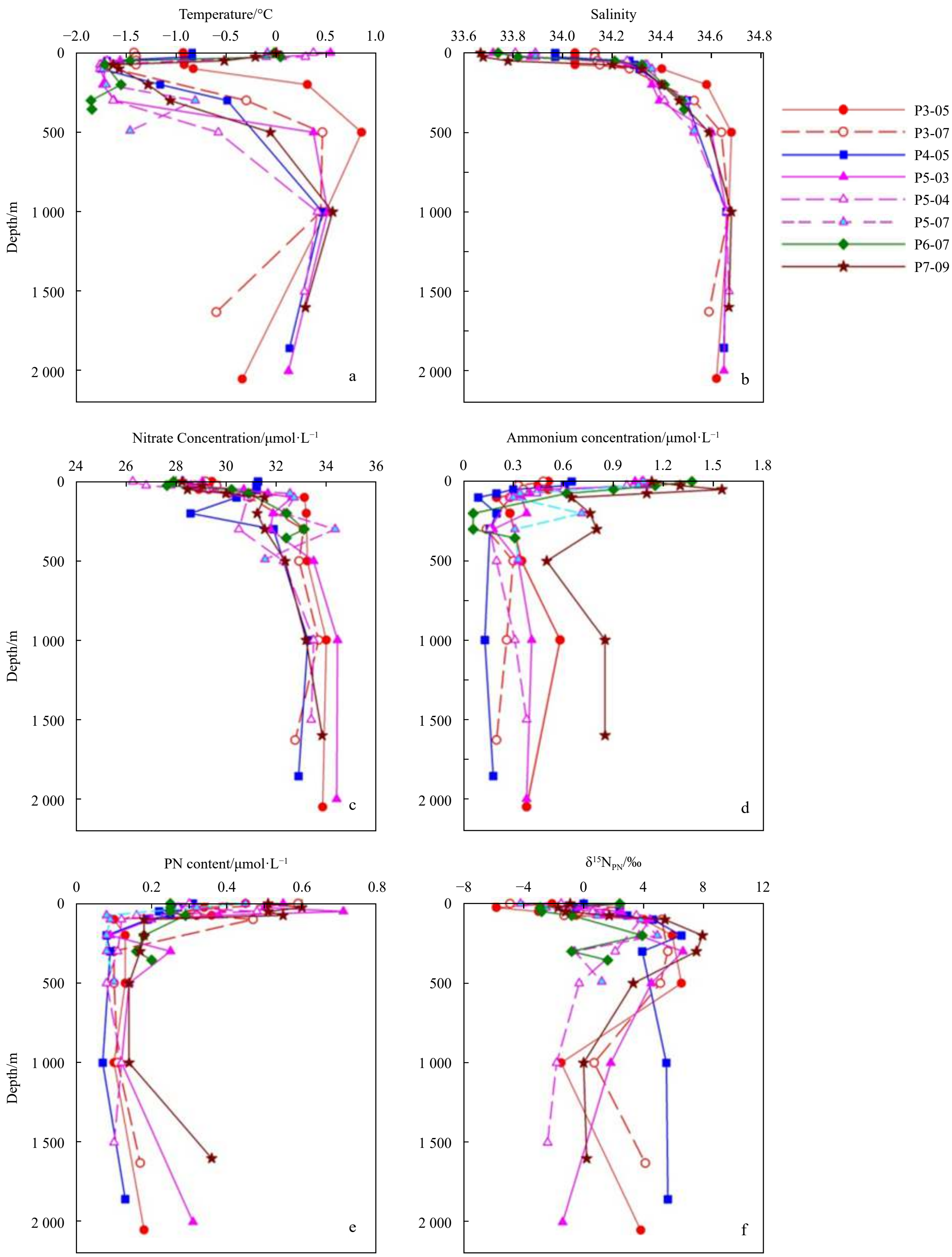

Fig. 3. Vertical profiles of temperature, salinity, nitrate, ammonium, PN, $\delta^{15}N_{PN}$ in the slope region.

salinity showed a general increase from the shelf to the basin, indicating a significant influence of sea ice melting in the Amery Ice Shelf. Distributions of temperature and salinity depicted the impact of various water masses across our study areas, showing an evident transition from the CDW with high temperature and salinity in the basin to the WW and SW with low temperature in the shelf. Furthermore, the low temperature (< –1.5°C) was generally found in the shelf, likely resulted from the residual waters in winter.

Nitrate concentrations in surface waters increased from the

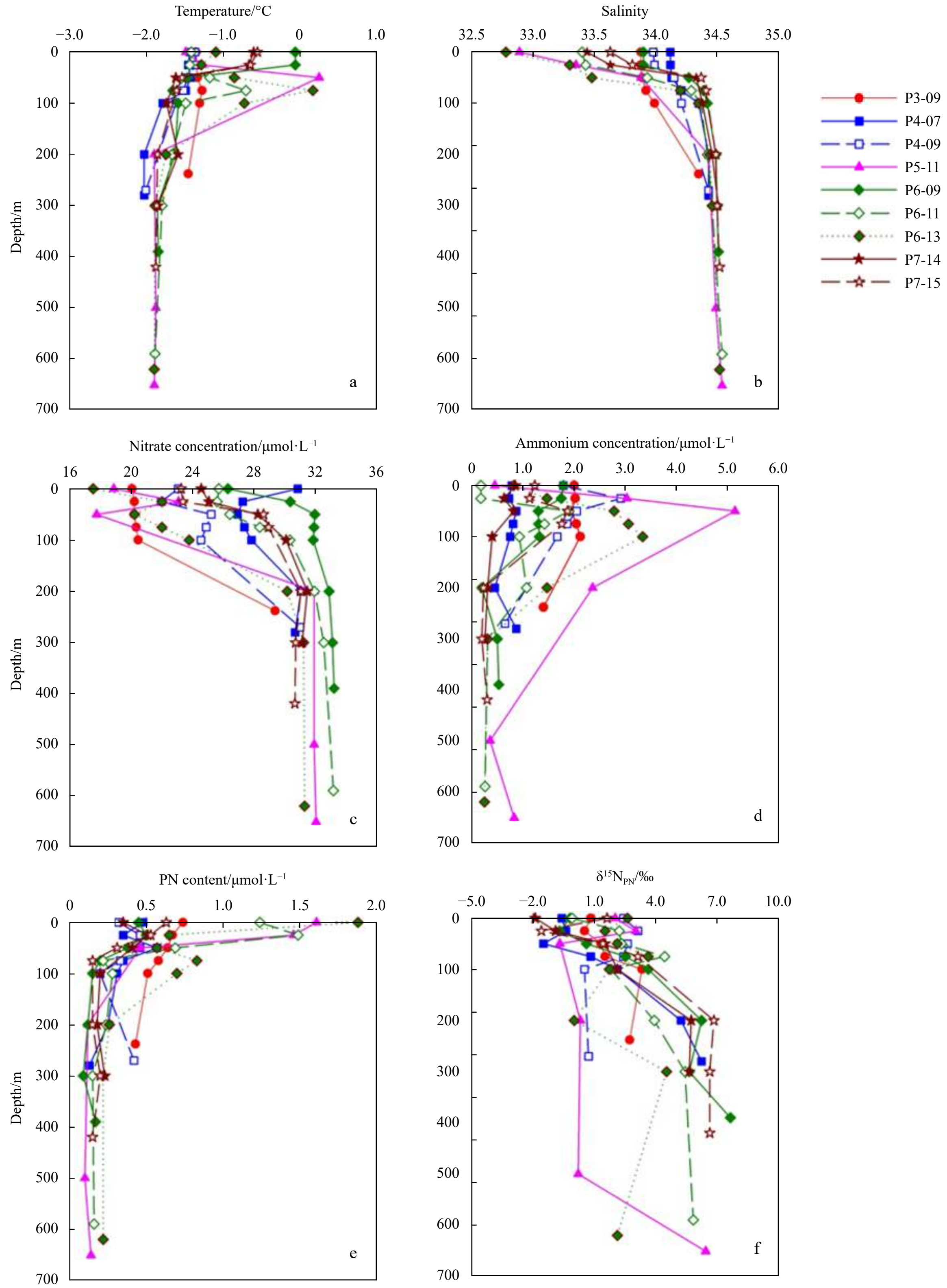

Fig. 4. Vertical profiles of temperature, salinity, nitrate, ammonium, PN, $\delta^{15}N_{PN}$ in the shelf region.

shelf to the basin. In general, low concentrations of nitrate, high concentration of ammonium and content of PN were found in the shelf, indicating the influence of enhanced biological productivity in the shelf. PN contents decreased, while nitrate concentrations increased with the increasing depth, indicating a major role of biological processes in controlling POM distribution. PN contents in the upper water column were elevated by the biogenic POM production from photosynthesis, while the decrease of PN in the deep water was due to the POM degradation during particle settling. The increase of $\delta^{15}N_{PN}$ with depth suggests that

Table 2. Temperature, salinity, nitrate, nitrite, ammonium, DIN, PN and $\delta^{15}N$ in the mixed layer, subsurface layer and deeper layer in the basin, the slope and the shelf (the variables are expressed as the mean value±standard deviation)

Region	Layer	Depth /m	Data number	Temperature /°C	Salinity	Nitrate concentration /μmol·L⁻¹	Nitrite concentration /μmol·L⁻¹	Ammonium concentration /μmol·L⁻¹	DIN concentration /μmol·L⁻¹	PN content /μmol·L⁻¹	$\delta^{15}N_{PN}$ /‰
Basin	mixed	0–50	17	0.10±0.48	33.83±0.13	28.91±0.85	0.31±0.06	0.90±0.52	30.12±1.26	0.41±0.17	−2.73±1.87
	subsurface	75–500	34	−0.40±1.23	34.43±0.21	32.14±1.49	0.14±0.08	0.54±0.48	32.81±1.67	0.22±0.12	1.65±2.52
	deep	⩾1 000	19	0.20±0.43	34.66±0.02	33.62±1.20	0.06±0.03	0.60±0.54	34.28±1.30	0.13±0.07	5.24±1.86
Slope	mixed	0–50	18	−0.43±0.65	33.91±0.16	28.78±1.28	0.30±0.05	0.84±0.35	29.93±1.12	0.38±0.12	−1.37±2.36
	subsurface	75–500	40	−1.19±0.74	34.37±0.17	31.57±1.35	0.17±0.08	0.41±0.30	32.15±1.21	0.21±0.15	2.99±2.69
	deep	⩾1 000	12	0.23±0.36	34.65±0.03	33.59±0.54	0.10±0.03	0.41±0.24	34.10±0.65	0.16±0.09	1.22±2.89
Shelf	mixed	0–50	20	−1.09±0.47	33.67±0.39	23.81±3.57	0.32±0.11	1.33±0.82	25.46±3.42	0.78±0.48	0.66±1.61
	subsurface	75–651	42	−1.54±0.51	34.33±0.22	28.89±4.06	0.28±0.11	1.23±1.04	30.41±3.25	0.30±0.18	3.27±2.27

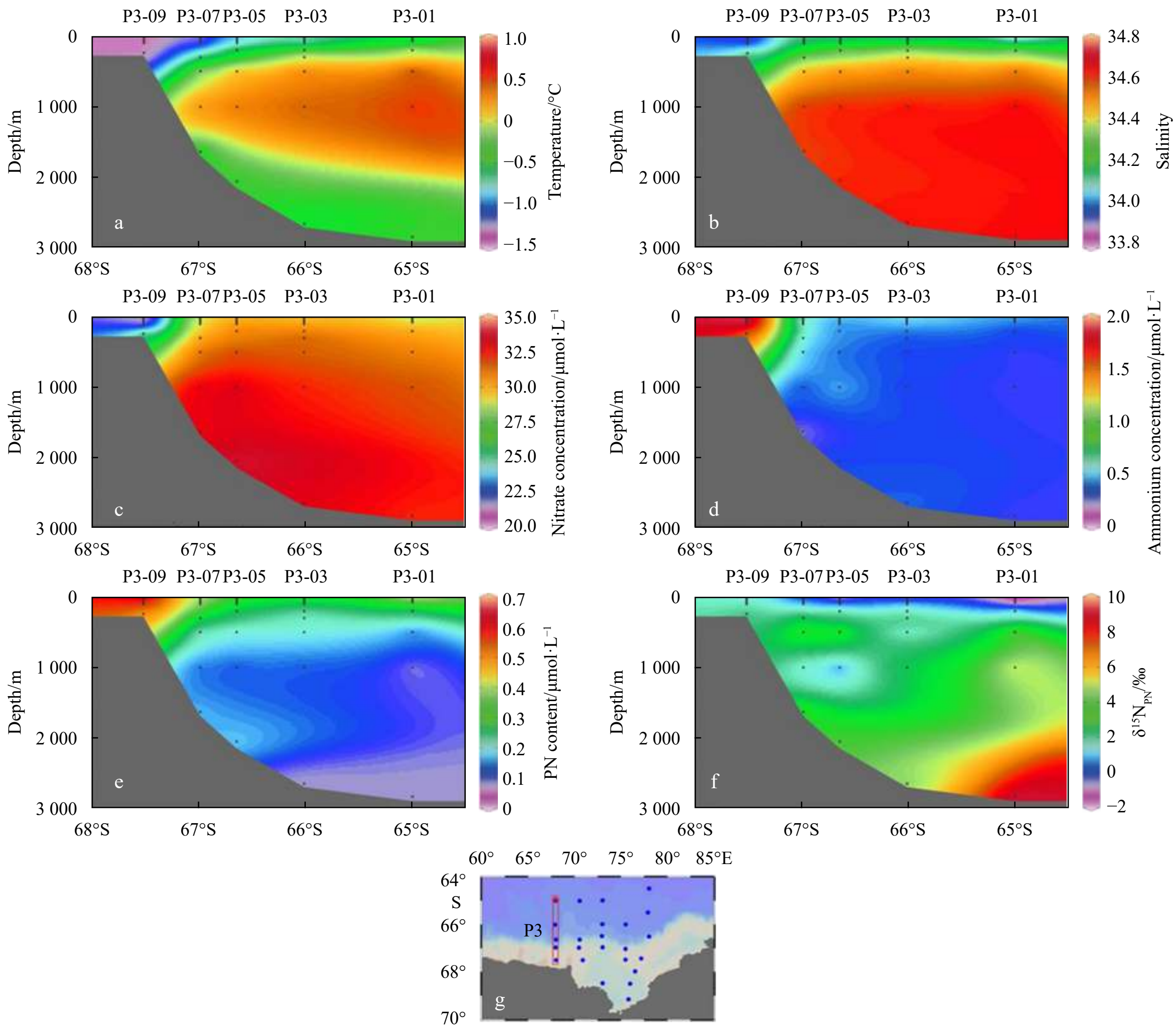

Fig. 5. Distributions of temperature (a), salinity (b), nitrate (c), ammonium (d), PN (e), $\delta^{15}N_{PN}$ (f) at Section P3 (68°E) (g) in the Prydz Bay and its adjacent areas.

bacteria preferentially degrade the ^{14}N in PN, leading to relative ^{15}N enrichment in the residual PN at depth.

4 Discussion

4.1 *$\delta^{15}N_{PN}$ in the Prydz Bay and its adjacent areas*

Surface $\delta^{15}N_{PN}$ in the Prydz Bay and its adjacent areas varied from −4.9‰ to 2.6‰ with an average of −0.9‰, similar to those reported in the Weddel Sea (−5.4‰ to 6.1‰) (Rau et al., 1991) and the Polar Frontal Zone (−4‰ to 0‰) (Lourey et al., 2003), but lower than previous studies in the Southern Ocean (−2‰ to 7.1‰) (Biggs et al., 1989; Liu et al., 2005; Wada et al., 1987) and other low latitudinal areas (−3‰ to 9‰) (Altabet et al., 1991; Mino et al., 2002). There are two possible explanations contributing to the surface low $\delta^{15}N_{PN}$ in the Prydz Bay, that is N_2 fixation and nitrate assimilation. However, N_2 fixation is expected to produce fixed N with $\delta^{15}N$ signal as low as about −1‰ to 0‰, which are not fully explain the low value we observed here. The low

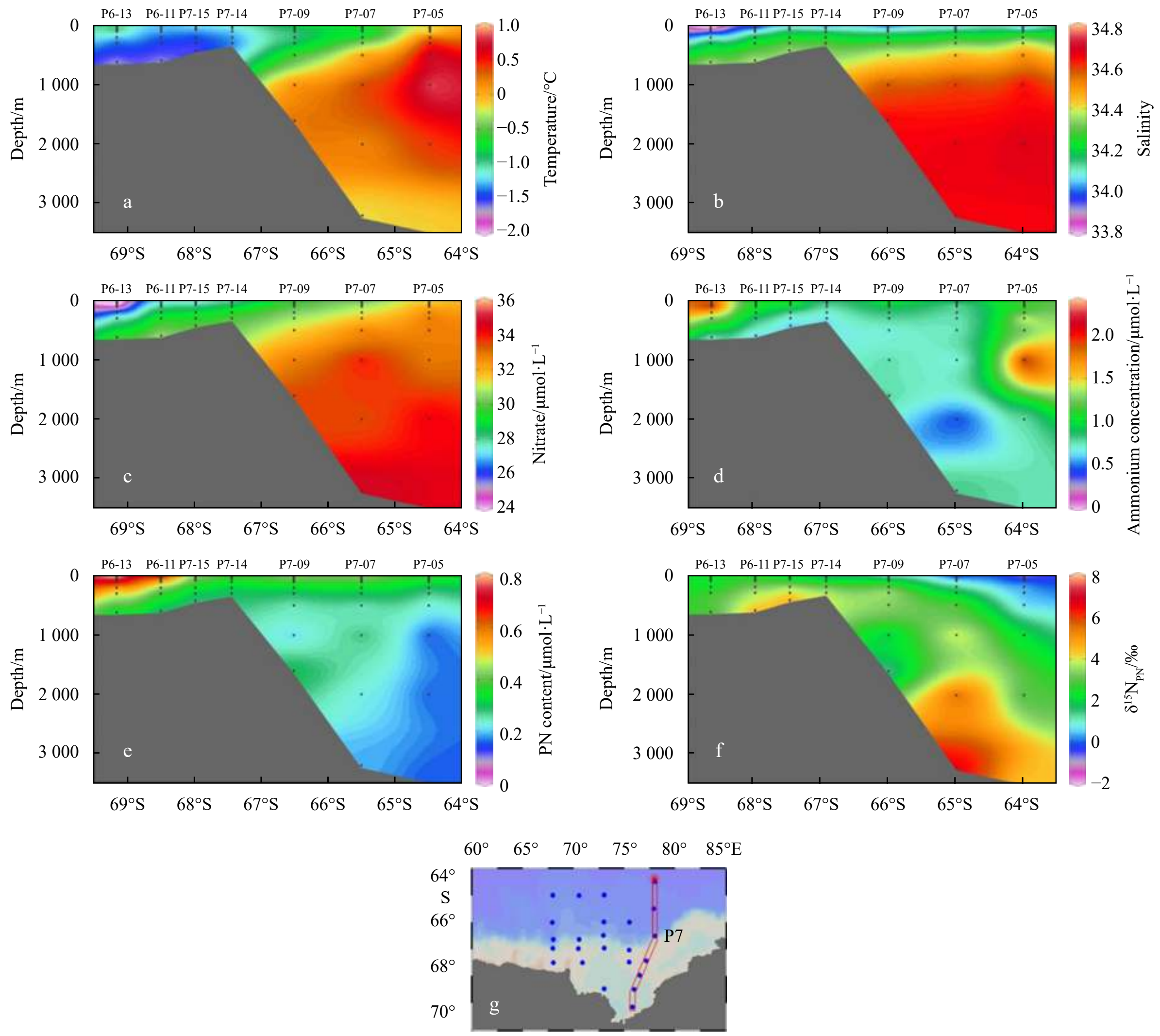

Fig. 6. Distributions of temperature (a), salinity (b), nitrate (c), ammonium (d), PN (e), $\delta^{15}N_{PN}$ (f) at Section P7 (78°E) (g) in the Prydz Bay and its adjacent areas.

$\delta^{15}N_{PN}$ in the study regions was more likely due to nitrate assimilation by phytoplankton. Phytoplankton preferential discrimination against ^{15}N relative to ^{14}N during nitrate assimilation, resulting in ^{15}N enrichment in the residual nitrate and ^{15}N depletion in organic matter.

The surface pattern of increase of $\delta^{15}N_{PN}$ from the outer toward the inner shelf is shaped by biological process. The inner bay is a favorable environment for phytoplankton growth under the influence of sea ice melting (Rigual-Hernández et al., 2015; Waldron et al., 1995; Zhang et al., 2014), and primary production in the inner shelf is significantly higher than that in the outer shelf (Liu et al., 2001, 2002, 2004; Qiu et al., 2004). The high primary production resulted in efficient utilization of nitrate, and thus higher $\delta^{15}N_{PN}$ values in the inner shelf. The average $\delta^{15}N_{PN}$ values were lowest in the mixed layer followed by the subsurface layer, and the highest in the deeper layer (Table 2). In the mixed layer, the mean $\delta^{15}N_{PN}$ values in the basin, the slope and the shelf were –2.73‰±1.87‰, –1.37‰±2.23‰ and 0.66‰±1.61‰, respectively. In the subsurface layer, the mean $\delta^{15}N_{PN}$ values in the basin, the slope and the shelf were 1.65‰±2.52‰, 2.99‰±2.69‰ and 3.27‰±2.27‰, respectively. In the deep water, the mean $\delta^{15}N_{PN}$ values in the basin, the slope were 5.24‰±1.86‰ and 1.22‰±2.89‰, respectively. The $\delta^{15}N_{PN}$ value in the subsurface was typically about 6‰, which is higher than that of suspended particles in the surface. The highest PN content in the shelf, slope and basin were observed in the mixed layers with shelf > slope ≈ basin. As for the $\delta^{15}N_{PN}$, the highest values in both the shelf and slope occurred in the subsurface layer, while in the deep layer in the basin. The $\delta^{15}N_{PN}$ in deep water was typically 5‰–10‰, which is consistent with those studies in the Southern Ocean (DiFiore et al., 2009; Liu et al., 2005; Rau et al., 1991). On average, the particulate matter in deep water was enriched in ^{15}N by about 2.5‰, 4.3‰ and 8‰ in the shelf, slope and basin, respectively, relative to those in surface layer. The $\delta^{15}N$ of deep particles is consistent with the notion that deep particles are breakdown products of settling particles exported from the surface, and bacteria preferentially remineralize low $\delta^{15}N$ particulate N. That means that the higher $\delta^{15}N_{PN}$ in deep water is a result of isotopic discrimination during degradation and consumption by bacteria (Sigman and Casciotti, 2001).

4.2 *Estimates of fractionation factor during nitrate assimilation by phytoplankton*

The $\delta^{15}N_{PN}$ in mixed layer is regulated by isotopic fractionation during nitrate assimilation, which leads to ^{15}N enrichment in the substrate and depletion in the product, because of preferentially consuming ^{14}N relative to ^{15}N when nitrate uptake by phytoplankton (Altabet, 2006; Sigman and Casciotti, 2001). The Rayleigh modle and Steady State model are frequently used to describe mathematically the change of isotopic compositions between the product and the reactant, with the consumption degree of the reactant N pool as a central parameter (Sigman and Casciotti, 2001). The Rayleigh model is suitable for an irreversible reaction occurring within a closed system, in which the substrate supply can be neglected and the isotopic fractionation factor is constant (Sigman and Casciotti, 2001). The equations are expressed as follow for the reactant:

$$\delta^{15}\mathrm{N}_{\mathrm{reactant}} = \delta^{15}\mathrm{N}_{\mathrm{initial}} - \varepsilon \ln(f), \tag{2}$$

and for the integrated product:

$$\delta^{15}\mathrm{N}_{\mathrm{integrated}} = \delta^{15}\mathrm{N}_{\mathrm{initial}} + \varepsilon \ln(f)\,\{f/(1-f)\}, \tag{3}$$

where f is the fraction of the reactant remaining, $\delta^{15}\mathrm{N}_{\mathrm{initial}}$ is the δ^{15}N of the initial reactant N pool, and ε is the kinetic isotope effect during the transformation.

The Steady State model refers to a system in which reactant N is continuously supplied and partially consumed, and consumption proceeds with a constant isotope effect ε (Sigman and Casciotti, 2001). The equations are expressed as follow:

$$\delta^{15}\mathrm{N}_{\mathrm{reactant}} = \delta^{15}\mathrm{N}_{\mathrm{initial}} + \varepsilon(1-f), \tag{4}$$

and

$$\delta^{15}\mathrm{N}_{\mathrm{production}} = \delta^{15}\mathrm{N}_{\mathrm{initial}} - \varepsilon f. \tag{5}$$

During the assimilation of nitrate by phytoplankton, nitrate is a reactant and PN is an accumulating end product. After choosing the salinity-normalized nitrate from deep water below 1 000 m to represent the initial nitrate, the $f_{\mathrm{NO}_3^-}$ is calculated according to following equation:

$$f_{\mathrm{NO}_3^-} = \frac{\bar{S}_{\geqslant 1\,000\ \mathrm{m}}}{S} \cdot \frac{[\mathrm{NO}_3^-]}{[\mathrm{NO}_3^-]_{\geqslant 1\,000\ \mathrm{m}}}, \tag{6}$$

where NO_3^- represents nitrate, $[\mathrm{NO}_3^-]$ is the concenteation of nitrate.

Due to the lack of $\delta^{15}\mathrm{NO}_3$ data, the fractionation factor ε during nitrate assimilation in the upper 50 m waters was estimated by Eqs (2) and (4) given by both models. In each case, the relationship between $\delta^{15}\mathrm{N}_{\mathrm{PN}}$ and nitrate drawdown may be degraded by complex hydrological and energetic nature in study areas. Our results showed that a linear relationship existed between $\delta^{15}\mathrm{N}_{\mathrm{PN}}$ and ln $(f_{\mathrm{NO}_3^-})\,\{f_{\mathrm{NO}_3^-}/(1-f_{\mathrm{NO}_3^-})\}$ (p=0.001), and between $\delta^{15}\mathrm{N}_{\mathrm{PN}}$ and $f_{\mathrm{NO}_3^-}$ (p=0.001) (Fig. 7). The calculated ε values ranged between 14‰ and 20‰ by assuming a closed system (Rayleigh model) and between 7‰ and 13‰ by assuming an open system (Steady State model). The relationship between nitrate utilization and δ^{15}N of nitrate has been used to estimate the fractionation factor ε during nitrate assimilation by phytoplankton in the Southern Ocean. For example, the ε values are 8‰, 6‰ and 8‰–9‰ in the Weddell Sea (Biggs et al., 1988), the Polar

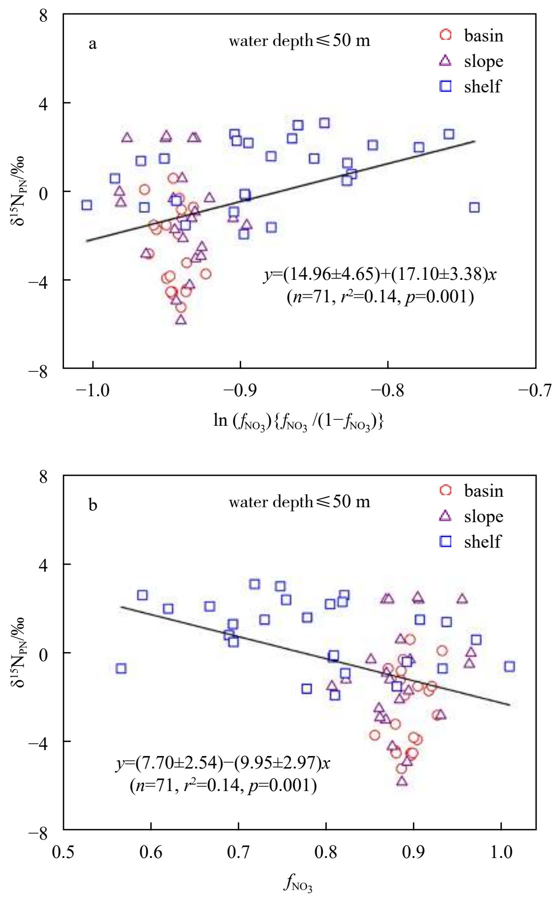

Fig. 7. Nitrogen isotope fractionation during nitrate assimilation in the upper 50 m. a. Rayleigh model and b. Steady State model.

Antarctic Zone (DiFiore et al., 2009) and the Subantarctic Zone (DiFiore et al., 2006), respectively. Field estimates of ε consistently fall between ~4‰ and 10‰ according to the relationship between nitrate-δ^{15}N and nitrate concentration described by Rayleigh model (Altabet and Francois, 1994a, 2001; DiFiore et al., 2006, 2009; Sigman et al., 2000). Our estimates of ε based on the Steady State model are consistent with above reports, but the estimates based on the Rayleigh model is significantly higher. This implies that the Steady State model is more suitable for describing the relationship between $\delta^{15}\mathrm{N}_{\mathrm{PN}}$ and nitrate drawdown in the Prydz Bay and its adjacent areas. Actually, the PN produced by phytoplankton exports continuously, mostly like an open system. Some studies suggested that the degree of efflux and isotope effect of nitrate assimilation may also vary with algal growth conditions (Granger et al., 2004, 2010; Wada and Hattori, 1978; Waser et al., 1998a). For example, the isotopic fractionation factor under light-limited condition is higher than that under iron-limited or temperature-limited condition for *Thalassiosira weissflogii* (Granger et al., 2004).

4.3 *Estimates for fractionation factor ε during POM remineralization*

The profiles of $\delta^{15}\mathrm{N}_{\mathrm{PN}}$ showed an increase with the increasing depth (Table 2; Figs 2f, 3f and 4f) due to isotopic fractionation during POM remineralization in the Prydz Bay and its adjacent areas. The recycle of organic N back to nitrate is comprised of two steps, i.e., the degradation of organic N to ammonium and then the oxidation of ammonium to nitrate, with the latter step called nitrification. This recycling pathway is quite efficient that ammonium does not accumulate in significant quantity in the mixed layer of oceanic systems (Sigman and Casciotti, 2001). Isotopic fractionation of N occurs in both steps. Field studies suggested that bacteria preferentially degrade low $\delta^{15}N_{PN}$, leaving the residual organic matter enriched in ^{15}N (Altabet, 1988; Altabet and McCarthy, 1985, 1986; Saino and Hattori, 1980, 1987; Sigman and Casciotti, 2001). However, in natural marine environments, it is difficult to quantify isotopic fractionation during degradation due to the diverse reactions involved in organic N degradation as well as the heterogeneous composition in organic matter.

In calculation of the fractionation factor during POM remineralization, PN is not only a reactant, but also an accumulating end product. Here, the average PN content ([PN]) in the upper 50 m waters at each station ($[PN]_{\leqslant 50\ m}$) is adopted as the initial $([PN])_{initial}$, and the fraction of degraded PN (f_{PN}) is calculated as below:

$$f_{PN} = \frac{[PN]_{initial}}{[PN]_{\leqslant 50\ m}}. \tag{7}$$

Eqs (2) and (4) given by the Rayleigh and the Steady State model is used to estimate fractionation factor ε. Our results showed that a linear relationship between $\delta^{15}N_{PN}$ and ln (f_{PN}) $\{f_{PN}/(1-f_{PN})\}$ (p=0.002) and between $\delta^{15}N_{PN}$ and f_{PN} (p=0.000 7) occurs (Fig. 8). The ε values ranged between 2.4‰ and 4.6‰ by

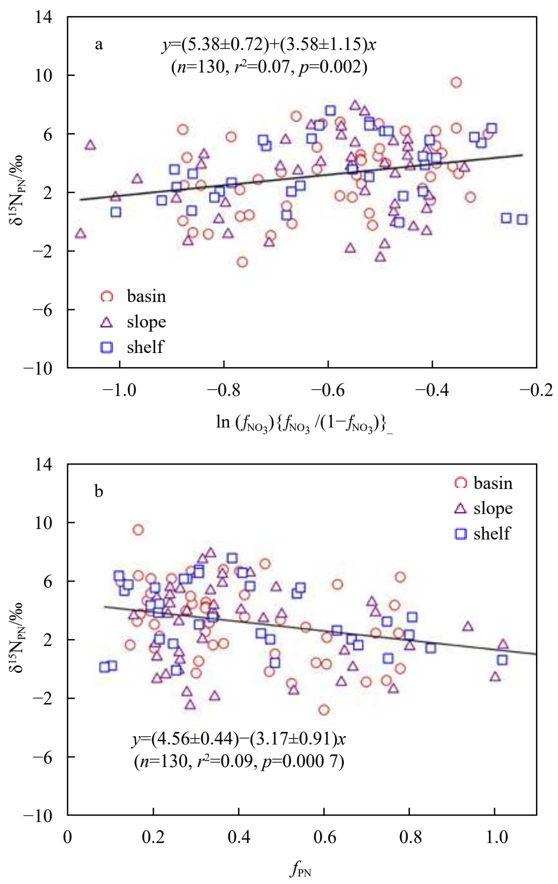

Fig. 8. Nitrogen isotope fractionation during PN remineralization below 50 m. a. Rayleigh model and b. Steady State model.

assuming a closed system (Rayleigh model) and from 2.3‰ to 4.1‰ by assuming an open system (Steady State model). Similar results are obtained by both models. A few laboratory studies have quantified the isotope effects of individual processes in organic matter degration, such as thermal peptide bond cleavage, bacterial amino acid uptake and transamination, and zooplankton ammonium release. A net isotope effect was estimated to be 5 after regarding degradation as a whole process (Sigman and Casciotti, 2001). Considering the similar ε values obtained by both models in the Prydz Bay and its adjacent areas, it is possible that both models are suitable for estimating the fractionation factor during POM remineralization when using $\delta^{15}N_{PN}$ as a parameter.

Applying the two models to estimate the isotope effect, physical and biochemical constraints on systems can ultimately result in very complicated isotopic calculation. The circulation in the Southern Ocean seems to necessitate a more complex model to interpret N isotope data. However, a simple model remains a useful starting point to describe the distribution of N isotopic compositions. Any deviation between the model and data needs to be explored in a hierarchy of more advanced model to resolve additional constraints.

5 Conclusion

Both assimilation of nitrate and remineralization of PN show an important role in shaping distribution of $\delta^{15}N_{PN}$ in the nitrate-repleted Prydz Bay and its adjacent areas. The spatial pattern of $\delta^{15}N_{PN}$ in study areas provides a clue to the sources of N supporting biological production, and the major pathways of N transformation. The isotopic fractionation factor during the assimilation of nitrate in the mixed layer is estimated to be 7‰–13‰ by the Steady State model with $\delta^{15}N_{PN}$ as a key parameter. The isotopic fractionation factors during POM remineralization in subsurface and deep water are estimated to be 3.6‰ and 3.2‰ by the Rayleigh model and the Steady State model, respectively. To our knowledge, this is the first time to quantify the isotopic fractionation of N during production and remineralization of POM in the Prydz Bay and its adjacent areas.

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