

# Reconstruction of Seasonal Variation in Nutrient Budget of a Surface Mixed Layer Using $\delta^{15}\text{N}$ of Sinking Particle Collected by a Time-Series Sediment Trap System

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Seasonal variations in flux of sinking particulate nitrogen (PN), its isotopic ratios ( $\delta^{15}\text{N}$ ) and radiocarbon isotopic ratios ( $\delta^{14}\text{C}$ ) of sinking particulate organic carbon (POC) were examined at two different depths using time-series sediment trap systems in the north-western North Pacific Ocean from June 1989 to May 1990. The  $\delta^{15}\text{N}$  of sinking PN increased from July to October and then dropped down in November at the shallower trap depth (1335 m), while at deeper trap depth (4785 m) it remained high until next spring. The high  $\delta^{15}\text{N}$  and low  $\delta^{14}\text{C}$  at the deeper trap during winter could be attributed to the larger contribution of resuspended sedimentary particles there. The variations in  $\delta^{15}\text{N}$  of sinking particles at 1335 m were consistent with the expected cycle of nutrient budget in a surface mixed layer controlled by the seasonal variations of physical and biological processes. Knowledge about the relationship between nitrate concentration and its  $\delta^{15}\text{N}$  in surface water was applied for reconstruction of mixed layer processes from the sinking PN  $\delta^{15}\text{N}$ . Although there were only limited in-situ data, reasonable temporal variations on the depth and nitrate concentration of a mixed layer were obtained by this reconstruction model. While sensitivity tests demonstrated the robustness of this reconstructing procedure, it was also suggested that the complicated particle dynamics in surface water column may partly distort the result of the reconstruction during late summer and fall.

Keywords:

- Nitrogen isotopic ratio,
- sinking particle,
- nutrient budget,
- surface mixed layer,
- time-series sediment trap,
- seasonal variation,
- high latitude ocean,
- carbon 14.

## 1. Introduction

On the open ocean at high latitudes, continuous chemical and biological observations on the surface mixed layer by ship is usually very difficult, especially during the winter season. Up-to-date progress in remote sensing by satellite, although very promising, provides only very limited information on the chemical and biological properties of sea surface.

On the other hand, the flux and chemical and biological nature of sinking particles collected by time-series sediment trap experiments reflect well the temporal variation of physical, chemical and biological processes in surface water. Previous reports have covered variations in the total organic carbon flux (Deuser and Ross, 1980; Honjo, 1982), the composition of planktonic microorganism species (Deuser and Ross, 1989; Takahashi, 1989), organic molecule

components (Ittekkot *et al.*, 1984a, 1984b; Handa *et al.*, 1992) and so on. Briefly stated, sinking particles have a great potential for clarification of temporal variations in physical, chemical and biological processes occurring in the surface water.

The predominant characteristic in the biogeochemistry of  $^{15}\text{N}$  in surface water of high latitude ocean, is large isotopic fractionation during the assimilation of nitrates by phytoplankton (Wada and Hattori, 1976; Wada, 1980; Wada *et al.*, 1987). Consequently, there is a good correlation between the decrease in ambient nitrate concentration and the increase in  $\delta^{15}\text{N}$  of suspended PN during spring bloom, which reflects the increase in  $^{15}\text{N}$  in the nitrate pool due to the preferential uptake of  $^{14}\text{NO}_3^-$  by phytoplankton (Saino and Hattori, 1985; Goering *et al.*, 1990; Altabet *et al.*, 1991; Nakatsuka *et al.*, 1992).

This correlation between the  $\delta^{15}\text{N}$  of suspended PN and ambient nutrient condition in the surface mixed layer is thought applicable to sinking PN collected by time-series sediment trap in the deep water, as has been shown by Altabet *et al.* (1991). In the present study, we report the seasonal variation in  $\delta^{15}\text{N}$  of sinking PN collected by time-

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series sediment trap experiment in the northwestern North Pacific Ocean, in order to understand to what degree the  $\delta^{15}\text{N}$  of sinking PN can actually reflect the seasonal variations in the nutrient condition of the surface mixed layer. And further, we make an model to reconstruct the seasonal changes in the nutrient budget and the depth of the surface mixed layer quantitatively from  $\delta^{15}\text{N}$  of sinking PN. In this study, we consider radioactive carbon isotopic ratios ( $\delta^{14}\text{C}$ ) of sinking POC too, in order to judge the origin of sinking particulate organic matter (POM) at each of the trap depths, before applying the sinking PN  $\delta^{15}\text{N}$  data for reconstruction of the mixed layer depth and nutrient concentration of surface water.

## 2. Material and Method

### 2.1 Sediment trap samples

Sediment trap samples were obtained at station NNWP-02 (44°00' N, 154°59' E; Fig. 1) on the cruise of the Hokusei-Maru, Hokkaido University, Japan. The sediment trap experiment was conducted from June 30, 1989 to May 21, 1990 using two time series sediment traps (Mark VI) set at two different depths (1335 m, 4785 m; Water depth there: 5305 m). Each collecting cup of the sediment trap was automatically exchanged with the next one every 25 days so that integrated fluxes over this period could be collected. Thirteen time series trap samples were obtained at each depth. The collecting cups of sediment trap were filled with 5% (volume %) solutions of formaldehyde in sea water which were neutralized by sodium borate before settling to prevent in situ bacterial degradation of samples. All samples were stored in a refrigerator until analysis. Unfortunately, no sinking particle sample could be collected after January, 1990 (Cup No. 8) at depth of 1335 m. Nevertheless, because

the micro-computer of the sediment trap controlling system memorized the fact that the collecting cups were exchanged exactly at pre-determined times there, the samples of Cup Nos. 1–7 can be considered to be collected exactly in the corresponding periods at 1335 m, also.

### 2.2 Chemical analysis

A portion of each sediment trap sample was acidified in 1N HCl solution for a half day to remove carbonate materials, and washed with enough distilled water to remove salts. Carbonate-free samples were vacuum dried and their aliquots were analyzed for POC and PN by CHN analyzer (Yanaco MS-1). The residues were analyzed for stable isotope ratios of carbon and nitrogen by the combustion method described in Minagawa *et al.* (1984). The compositions of  $^{13}\text{C}$  and  $^{15}\text{N}$  of the purified  $\text{CO}_2$  and  $\text{N}_2$  gases were determined by mass spectrometers (Finnigan Mat-251 and Hitachi RMU-6R). The residues of  $\text{CO}_2$  gases were further converted into graphite carbon pellets by Fe- $\text{H}_2$  reduction method (Kitagawa *et al.*, 1993) and analyzed for their  $^{14}\text{C}/^{13}\text{C}$  ratios using a tandem accelerator mass spectrometer at Nagoya University (Nakamura *et al.*, 1987). The  $^{14}\text{C}/^{12}\text{C}$  ratios were then calculated using the predetermined  $^{13}\text{C}$  compositions.

The stable and radioactive isotopic ratios of nitrogen and carbon were calculated in terms of  $\delta X$  as follows.

$$\delta X = (R_{\text{sample}}/R_{\text{std}} - 1) \times 10^3 (\text{‰}),$$

where  $X$  (and  $R$ ) are  $^{15}\text{N}$  (and  $^{15}\text{N}/^{14}\text{N}$ ) and  $^{14}\text{C}$  (and  $^{14}\text{C}/^{12}\text{C}$ ), and the standards for  $\delta^{15}\text{N}$  and  $\delta^{14}\text{C}$  are  $\text{N}_2$  in atmosphere and “tree carbon in 1950”, whose  $\delta^{13}\text{C}$  value is  $-25\text{‰}$  (Stuiver and Polach, 1977), respectively. The standard deviation of  $\delta^{15}\text{N}$  measurement was less than  $0.2\text{‰}$ . Usually, the  $\delta^{14}\text{C}$  values are further converted to  $\Delta^{14}\text{C}$  values, considering the isotope fractionation effect (Stuiver and Polach, 1977). However, in the present study, we use the  $\delta^{14}\text{C}$  values directly, in order to discuss the mixing of POC originating from different  $^{14}\text{C}/^{12}\text{C}$  end-members, rather than discussing the  $^{14}\text{C}$  ages. Nevertheless, in this case,  $\delta^{14}\text{C}$  values are almost the same as  $\Delta^{14}\text{C}$ , because the  $\delta^{13}\text{C}$  values of sinking POC measured were near  $-25\text{‰}$ .

## 3. Results and Discussion

### 3.1 Temporal and vertical variation of sinking PN in the northwestern North Pacific

The seasonal change in primary production is principally regulated by light at high latitudes during winter months and by nutrients during summer months, while at mid and low latitudes nutrients limit production throughout the year. The station NNWP-02 in this study (Fig. 1) is located in the western region of the Subarctic Pacific where intense convective mixing in winter loads the surface water

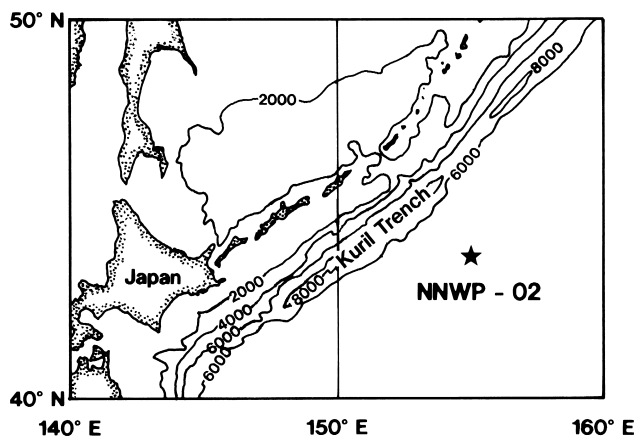


Fig. 1. The location of the sediment trap experiment (NNWP-02: 44°00' N, 154°59' E; Water depth is 5305 m; Trap depths are 1335 m and 4785 m). Contour lines in ocean are drawn at 2000 m intervals of the water depth.

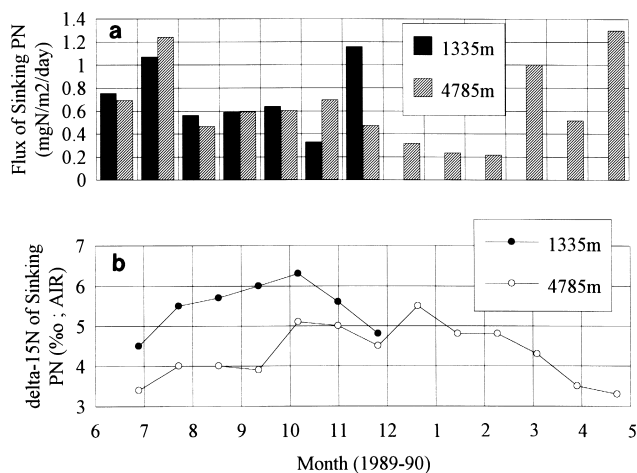


Fig. 2. Seasonal variations in the sinking fluxes of PN (a) and their  $\delta^{15}\text{N}$  values (b). Closed and shaded bars represent the fluxes at 1335 m and 4785 m, respectively (a). Closed and open circles represent the  $\delta^{15}\text{N}$  of sinking PN at 1335 m and 4785 m, respectively (b).

with nutrients with about  $20\ \mu\text{M}$  of nitrate (Wada, 1980). Therefore a large spring bloom and sometimes an autumn bloom (Parsons *et al.*, 1984) can occur by the increased light intensity in spring and by the recovery of nutrients into the surface mixed layer in fall, respectively. Because the seasonal variations in sinking particle fluxes occur in response to the seasonal cycle in primary production (Deuser and Ross, 1980; Honjo, 1982; Deuser *et al.*, 1990), such a predominant seasonal cycle of primary production in this region is expected to be observed not only in the surface biomass but also in a sinking particle flux in deep water.

Actually, large variations in sinking PN flux were observed at both depths (1335, 4785 m) of station NNWP-02 (Fig. 2a). There are three types of maxima in PN fluxes (Fig. 2a): Two large maximum (summer in 1989 and spring in 1990), and one small (large at 1335 m) maximum (late fall in 1989). These characteristic temporal variations are consistent with an annual variation of primary production in the corresponding upper water column. The former (two maximum in PN flux) must be a response to the spring bloom in this region and the latter may possibly be attributed to the bloom in fall.

In addition to the temporal variation pattern, the amount of sinking PN flux itself is similar with both traps. As shown in the  $\delta^{15}\text{N}$  values (Fig. 2b), the composition of sinking PN is thought to be different between the two depths so that this similarity between the PN flux of both depths is incidental. It seemed that a large portion of sinking PN must be decomposed and additional PN may instead be entrained within the deep water column between 1335 m and 4785 m. Especially in the continental shelf and slope region, the

sinking flux tends to increase with depth (Biscaye *et al.*, 1988). The origin of this increased flux is thought to be the downslope resuspension of particles from sediment (Walsh *et al.*, 1991). However, the station NNWP-02 in this study is located on the abyssal plain of the North Pacific and beyond the Kuril trench offshore. Where the additional particles come from is of interest for further study.

### 3.2 Temporal and vertical variation in $\delta^{15}\text{N}$ of sinking PN

The  $\delta^{15}\text{N}$  values of sinking PN at 4785 m are consistently 1‰ or more lower than those at 1335 m (Fig. 2b). This downward decrease in  $\delta^{15}\text{N}$  of sinking particle has already been reported by previous studies (Altabet *et al.*, 1991) and the reason is still not clear now. The  $\delta^{15}\text{N}$  values of total protein within cultured microorganism cells are reported to be about 3‰ higher than those of whole cells (Macko *et al.*, 1987). Therefore, this difference between the two depths may be attributed to the preferential degradation of amino acids among total nitrogenous compounds during the settling process of sinking particles. This finding of decrease in  $\delta^{15}\text{N}$  of sinking particles is potentially very important for particle dynamics in the deep water column, and will be discussed in detail in another paper (Nakatsuka *et al.*, 1997).

Although there are differences between  $\delta^{15}\text{N}$  values of sinking PN at the two depths, the temporal variations in  $\delta^{15}\text{N}$  seem nearly parallel between the two trap depths, at least until October, suggesting the constancy of the isotope fractionation during the settling process of sinking particles. The increases in those from summer to fall at both depths (Fig. 2b) can be interpreted by the change in surface nutrient budget. As shown in a previous study (Altabet *et al.*, 1991), the  $\delta^{15}\text{N}$  of suspended PN in surface water, which is isolated from subsurface water by seasonal thermocline, is primarily correlated with the proportion of biological utilization of initial nitrate, which has been injected by convective mixing in winter into surface water. During the spring and summer, the  $\delta^{15}\text{N}$  of PN increases together with the decrease of nitrate according to preferential uptake of  $^{14}\text{NO}_3^-$  by phytoplankton and consequent increase in proportion of  $^{15}\text{NO}_3^-$  in the remaining pool of nitrate. This trend must continue until the depletion of nitrate. The increase of  $\delta^{15}\text{N}$  of sinking PN at each depth from summer to fall (Fig. 2b) must indicate that such a process controlled by nitrate utilization occurred in the corresponding surface water.

On the other hand, the  $\delta^{15}\text{N}$  of sinking PN decreased in late fall at 1335 m (Fig. 2b). The same phenomenon was explained by Altabet and Deuser (1985) as due to the convective mixing of upper water column which introduces lower  $\delta^{15}\text{N}$ -nitrate into euphotic zone. Also in this study, the decrease of  $\delta^{15}\text{N}$  at 1335 m from October to December can be explained by the addition of new nitrate, which has a low  $\delta^{15}\text{N}$  value, from the layer below the thermocline.

However, the decrease of  $\delta^{15}\text{N}$  at 4785 m did not start until January and did not return to the level of last July until

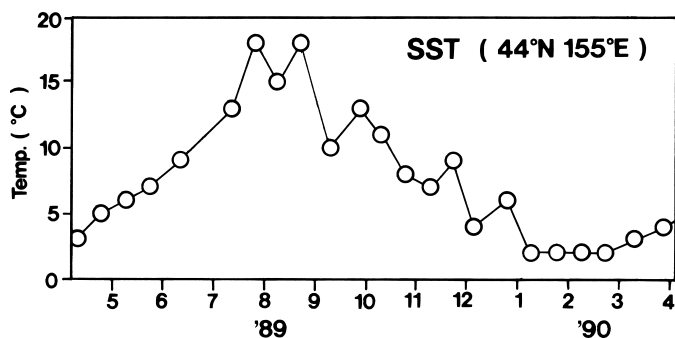


Fig. 3. Seasonal variation of the sea surface temperature at 44°N 155°E (the almost same location as the trap site). Data are derived from "Fast Report of Ocean" (Maritime Safety Agency, Japan).

the following April. At the time when the  $\delta^{15}\text{N}$  of sinking PN at 4785 m arrived at maximum, the sea surface temperature was nearly to become the lowest in the year (Fig. 3) and there must have been a high concentration of newly introduced nitrate which has a low  $\delta^{15}\text{N}$  value in surface water. In conditions of a high concentration of nitrate, low temperature and low light, the resulting large isotope fractionation must produce PN of very low  $\delta^{15}\text{N}$  value (Wada and Hattori, 1978).

The following two alternatives can explain this phenomenon. Firstly, the sinking particles in winter at 4785 m may not come from surface water at that time, but mainly consist of particles originating from late summer surface water, where PN of high  $\delta^{15}\text{N}$  was produced, because of the possible low sinking rate of particles and the very long distance from surface to 4785 m depth. Secondly, some part of sinking particle at 4785 m may comprise the resuspended particles from sediment as found in the continental shelf area (Biscaye *et al.*, 1988; Anderson *et al.*, 1994). The average value of  $\delta^{15}\text{N}$  of sedimentary PN is around 6 to 8‰ in the northern North Pacific area (Nakatsuka *et al.*, 1995; T. Nakatsuka, unpublished data) which is significantly higher than those of sinking particles in the deep water column as shown in this study, probably due to early diagenetic alteration of  $\delta^{15}\text{N}$  on the sediment surface (Altabet and Francois, 1994). This effect of resuspended sedimentary particles may also explain the relatively high  $\delta^{15}\text{N}$  value of sinking PN at 4785 m in the winter.

### 3.3 Origin of sinking particles inferred from $\delta^{14}\text{C}$

Both explanations for the winter high  $\delta^{15}\text{N}$  values at 4785 m suggest that the properties of sinking particles at greater depth cannot be applied to reconstruct the temporal variations of surface mixed layer. In order to use the  $\delta^{15}\text{N}$  data at 1335 m, however, it is also necessary to judge whether the latter mechanism affects the sinking particles at the 1335 m or not. For this purpose, the  $\delta^{14}\text{C}$  of sinking POC were

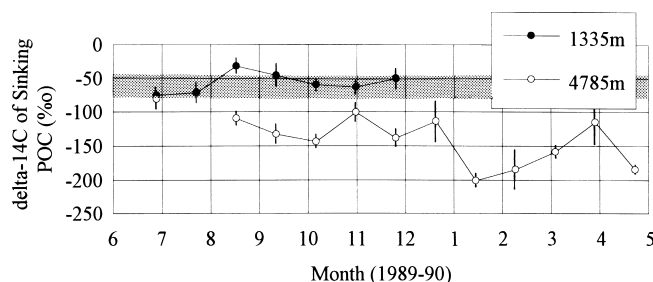


Fig. 4. Seasonal variations in the  $\delta^{14}\text{C}$  values of sinking POC. Closed and open circles represent the  $\delta^{14}\text{C}$  of sinking POC at 1335 m and 4785 m, respectively, with their measurement error bars ( $\pm 1\sigma$ ). The shaded region indicates the expected range of plankton  $\delta^{14}\text{C}$  at the corresponding surface water, which is determined on the basis of the  $\delta^{14}\text{C}$  measurement of POC in surface mixed layer in late summer at a similar area to this study ( $-44 \pm 13\text{‰}$ ; Nakatsuka, unpublished data, see text) and the reported seasonal variation of  $\Delta^{14}\text{C}$  of  $\Sigma\text{CO}_2$  in high latitudinal surface water between summer and winter (about 35‰, Broecker and Peng, 1980). The  $\delta^{14}\text{C}$  of Cup No. 2 at 4785 m was not determined.

measured at each trap. Because the surface sediments have much lower  $\delta^{14}\text{C}$  values than the plankton of surface water, the  $\delta^{14}\text{C}$  of sinking POC clearly indicates the contribution of resuspended particles into the traps (Anderson *et al.*, 1994).

The seasonal variations in  $\delta^{14}\text{C}$  of sinking POC are shown in Fig. 4. The  $\delta^{14}\text{C}$  values at 4785 m are consistently lower than those at 1335 m, indicating that the contribution of resuspended sedimentary particles is always higher at a deeper trap. Moreover, at 4785 m, the  $\delta^{14}\text{C}$  of sinking POC showed very low values during winter. This suggests that the main reason of high  $\delta^{15}\text{N}$  values in this season is the higher contribution of the resuspended sedimentary particles, whose  $\delta^{15}\text{N}$  values are higher than those of sinking particles (Altabet and Francois, 1994).

The  $\delta^{14}\text{C}$  values at 1335 m are also very low compared to those of suspended POC in surface water observed in temperate oceans, usually more than 100‰ due to the effect of bomb-produced  $^{14}\text{C}$  (Druffel *et al.*, 1992). However, in the Subarctic Pacific Ocean, very low  $\Delta^{14}\text{C}$  values of  $\Sigma\text{CO}_2$  below 0‰ have sometimes been reported in surface mixed layer (Ostlund *et al.*, 1987), and actually at the similar area to the present study, the low  $\delta^{14}\text{C}$  value of  $-44 \pm 13\text{‰}$  (T. Nakatsuka, unpublished data) was observed for the suspended POC collected at 50 m depth in late summer season (46°42.9' N, 162°22.4' E; Aug. 11, 1985). Considering the seasonal variation in  $\Delta^{14}\text{C}$  of  $\Sigma\text{CO}_2$  in surface mixed layer (about 35‰ decrease from fall to spring; Broecker and Peng, 1980), we can conclude that the  $\delta^{14}\text{C}$  values of sinking POC at 1335 m must be covered in the expected range of seasonal variation in  $\delta^{14}\text{C}$  of suspended POC in the surface mixed layer (the shaded area in Fig. 4). This indicates that there is

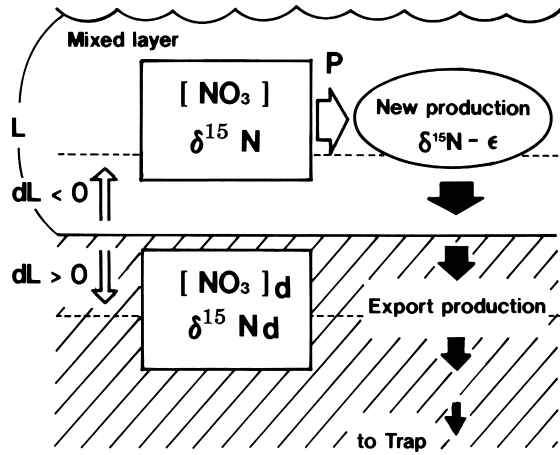


Fig. 5. The schema of a simple model on the nitrate and its isotopic ratio in the mixed layer, with special emphasis on the temporal variations caused by biological new production and change of the mixed layer depth.

very little contribution from the resuspended sedimentary particles into sinking particles at the 1335 m trap.

### 3.4 Model of temporal variations in nitrate concentration and its isotope ratio in surface mixed layer

If the temporal change of  $\delta^{15}\text{N}$  in sinking PN at 1335 m is regulated by the nitrate budget in the surface mixed layer, what processes actually control the nitrate budget and its  $\delta^{15}\text{N}$  values in the surface mixed layer? Initially, we propose the very simple mixed layer model described in Fig. 5, which is almost the same as the model proposed by Peng *et al.* (1987). The assumptions are as follows. 1) The nutrient concentration is homogeneous in the mixed layer. The mixed layer represents the surface water layer above the pycnocline, which is well-mixed by regional wind stress. 2) Supply of nitrate into the mixed layer occurs only when the depth of the mixed layer increases. The diffusion process and the nitrification from regenerated ammonium to nitrate in the mixed layer are both neglected. Based on these assumptions, the following equations can be introduced as equations of conservation of nitrate and its isotope ratio in the mixed layer.

$$d(N \cdot L) / dt = -P + (dL / dt) \cdot N^*, \quad (1)$$

$$\begin{aligned} d(\delta^{15}\text{N} \cdot N \cdot L) / dt \\ = -P \cdot (\delta^{15}\text{N} - \epsilon) + (dL / dt) \cdot \delta^{15}\text{N}^* \cdot N^* \end{aligned} \quad (2)$$

where  $N$  and  $\delta^{15}\text{N}$  are the concentration of nitrate (mg at.N/m<sup>3</sup>) in the mixed layer and its isotope ratio, respectively.  $P$  (mg at.N/m<sup>2</sup>/day) and  $\epsilon$  (‰) are the primary production of

nitrogen which originates from nitrate (new production) in the mixed layer and the magnitude of isotope fractionation during uptake of nitrate by phytoplankton.  $L(m)$  is the depth of the mixed layer. When  $dL/dt < 0$ ,  $N^*$  and  $\delta^{15}\text{N}^*$  are equal to  $N$  and  $\delta^{15}\text{N}$ . But, when  $dL/dt > 0$ ,  $N^*$  and  $\delta^{15}\text{N}^*$  are  $N_d$  and  $\delta^{15}\text{N}_d$  (the concentration and isotope ratio of nitrate in the layer below the mixed layer), respectively.

### 3.5 Reconstruction of surface mixed layer processes from $\delta^{15}\text{N}$ of sinking PN

In general, the surface mixed layer is sustained by wind stress-induced turbulent mixing (Klein and Coste, 1984). However, in summer when solar radiation flux is high, the turbulent kinetic energy could not reach to a deeper layer due to the large buoyancy of surface water and the mixed layer is restricted to shallower depth. On the other hand, in winter the cooling of sea surface makes the surface water density high and induces free convective mixing to deepen the mixed layer (Gill and Turner, 1976). In the northern North Pacific area, however, there is a strong subsurface halocline (about 100 to 200 m depth) and it prevents the winter convective mixing from deepening below it (Uda, 1963).

These seasonal variabilities of surface mixed layer depth are the most important processes, which govern the productivity of surface water via nutrient supply and light adjustment for photosynthesis (Yentsch, 1990; Mann and Laizer, 1991). Therefore, the surface mixed layer depth should be the main target for the reconstruction in present study. For this purpose, Eqs. (1) and (2) will be re-arranged. In order to apply the equations (1) and (2) to the discrete time series data sets on PN flux and  $\delta^{15}\text{N}$  values obtained from the sediment trap experiment, the differential equations (1) and (2) are first converted into the following difference equations (3) and (4), respectively.

$$\begin{aligned} N(n+1) \cdot L(n+1) \\ = N(n) \cdot L(n) - (P(n+1) + P(n)) / 2 \cdot \Delta t \\ + (L(n+1) - L(n)) \cdot N^*, \end{aligned} \quad (3)$$

$$\begin{aligned} \delta^{15}\text{N}(n+1) \cdot N(n+1) \cdot L(n+1) \\ = \delta^{15}\text{N}(n) \cdot N(n) \cdot L(n) - (P(n+1) + P(n)) / 2 \cdot \Delta t \\ \cdot \left\{ (\delta^{15}\text{N}(n+1) + \delta^{15}\text{N}(n)) / 2 - \epsilon \right\} \\ + (L(n+1) - L(n)) \cdot \delta^{15}\text{N}^* \cdot N^*, \end{aligned} \quad (4)$$

where  $n$  is the number of the time step and  $\Delta t$  is the period of one time step interval.  $(P(n+1) + P(n))/2$  and  $(\delta^{15}\text{N}(n+1) + \delta^{15}\text{N}(n))/2$  represent the averaged  $P$  and  $\delta^{15}\text{N}$  values during the time step interval, respectively. When  $L(n+1) <$

$L(n)$ ,  $N^*$  and  $\delta^{15}N^*$  are equal to  $N(n)$  and  $\delta^{15}N(n)$ . But when  $L(n+1) > L(n)$ ,  $N^*$  and  $\delta^{15}N^*$  are  $Nd$  and  $\delta^{15}Nd$  (the concentration and isotope ratio of nitrate in the layer below the mixed layer), respectively.

By substituting Eq. (3) into the left-side term of Eq. (4), we can obtain the following relation on the  $L(n+1)$ , after some re-arrangement of equation.

$$\begin{aligned} L(n+1) &= L(n) + \left[ (P(n+1) + P(n)) / 2 \cdot \Delta t \right. \\ &\quad \cdot \left\{ \varepsilon + (\delta^{15}N(n+1) - \delta^{15}N(n)) / 2 \right\} \\ &\quad \left. - (\delta^{15}N(n+1) - \delta^{15}N(n)) \cdot N(n) \cdot L(n) \right] \\ &\quad / \left\{ (\delta^{15}N(n+1) - \delta^{15}N^*) \cdot N^* \right\}. \end{aligned} \quad (5)$$

Then, substituting  $L(n+1)$  in Eq. (5) into Eq. (3), the relation on  $N(n+1)$  can be introduced also.

$$\begin{aligned} N(n+1) &= \left\{ N(n) \cdot L(n) - (P(n+1) + P(n)) / 2 \cdot \Delta t \right. \\ &\quad \left. + (L(n+1) - L(n)) \cdot N^* \right\} / L(n+1). \end{aligned} \quad (6)$$

Using Eqs. (5) and (6), we can calculate the temporal changes in  $L$  and  $N$  explicitly from the data sets of temporal variations in  $P(n)$  and  $\delta^{15}N(n)$ , which could be possibly estimated from the data sets obtained in the sediment trap experiment. When we apply the equations (5) and (6), we first judge whether  $L(n+1)$  is  $>L(n)$  or not, in order to determine the values of  $N^*$  and  $\delta^{15}N^*$ . This task can be carried out according to following criteria. Firstly, when  $\delta^{15}N(n+1)$  is less than (or equal to)  $\delta^{15}N(n)$ ,  $L(n+1)$  is obviously  $>L(n)$ . This is because there is no other mechanism than the entrainment of lower  $\delta^{15}N$  nitrate from the deeper layer which can make the  $\delta^{15}N$  lower (or keep the  $\delta^{15}N$  constant under the condition where  $P$  is not 0) in the surface mixed layer in this model. Secondly, if  $\delta^{15}N(n+1)$  is higher than  $\delta^{15}N(n)$ , it is judged based on the following equation derived from Eq. (5).

$$\begin{aligned} L(n+1) - L(n) &= \left[ (P(n+1) + P(n)) / 2 \cdot \Delta t \right. \\ &\quad \cdot \left\{ \varepsilon + (\delta^{15}N(n+1) - \delta^{15}N(n)) / 2 \right\} \\ &\quad \left. - (\delta^{15}N(n+1) - \delta^{15}N(n)) \cdot N(n) \cdot L(n) \right] \\ &\quad / \left\{ (\delta^{15}N(n+1) - \delta^{15}N^*) \cdot N^* \right\}. \end{aligned} \quad (7)$$

In this equation, the denominator of the right-side term,  $(\delta^{15}N(n+1) - \delta^{15}N^*) \cdot N^*$ , is always  $>0$ , irrespective of the result, whether  $L(n+1)$  is  $>L(n)$  or not. This is because if  $L(n+1) < (=) L(n)$ ,  $\delta^{15}N^*$  is equal to  $\delta^{15}N(n)$  which is lower than  $\delta^{15}N(n+1)$  from the assumption of this case, and if  $L(n+1) > L(n)$ ,  $\delta^{15}N^*$  is equal to  $\delta^{15}Nd$  which is the  $\delta^{15}N$  of the deeper water nitrate and always lower than the  $\delta^{15}N$  of the nitrate in surface mixed layer. Therefore whether  $L(n+1)$  is more than  $L(n)$ , or not, is judged only from the numerator of the right-side term of Eq. (7), which can be calculated without  $N^*$  or  $\delta^{15}N^*$ .

In order to calculate the temporal changes in the surface mixed layer depth ( $L$ ) and its nitrate concentration ( $N$ ) using Eqs. (5) and (6), we need the following two kinds of data set in the surface mixed layer. One is the temporal variation in new production rate of particulate nitrogen,  $P(n)$ , and the other is that in  $\delta^{15}N$  of nitrate in the surface mixed layer,  $\delta^{15}N(n)$ . Because the temporal variations in the sinking PN flux and its  $\delta^{15}N$  value at 1335 m seem to correspond well to those of  $P$  and  $\delta^{15}N$  in the surface mixed layer as discussed above, it seems reasonable to estimate this information of surface water from sinking particle data. In the present study, the following two equations are introduced to estimate the  $P(n)$  and  $\delta^{15}N(n)$  from sinking particles at 1335 m.

$$P(n) = k \cdot PN_{1335m}(n), \quad (8)$$

$$\delta^{15}N(n) = \delta^{15}N_{1335m}(n) + c \quad (9)$$

where  $PN_{1335m}(n)$  and  $\delta^{15}N_{1335m}(n)$  are “ $n$ ”th data of the sinking PN flux at 1335 m and its  $\delta^{15}N$  value at that time, respectively. “ $k$ ” in Eq. (8) is a proportional coefficient, and “ $c$ ” in Eq. (9) is an unknown constant which consists of sum of the two kinds of isotope fractionations during the nitrate uptake by phytoplankton and the degradation of sinking particles from surface water to 1335 m. Equations (8) and (9) assume the constancies of the degradation rate and isotope fractionation of sinking PN during the settling process from the surface mixed layer to 1335 m, and also require that the standing stocks of suspended PN and ammonium in the surface mixed layer be much smaller than nitrate, and newly produced PN from nitrate is immediately removed from surface water in the form of sinking PN (Altabet and Francois, 1994).

Using Eqs. (5), (6), (8) and (9), the temporal changes in the surface mixed layer depth ( $L$ ) and nitrate concentration ( $N$ ) can be calculated explicitly from the time-series data of  $PN_{1335m}(n)$  and  $\delta^{15}N_{1335m}(n)$  shown in Fig. 2. Before applying the data from sinking particle at 1335 m for the equations, however, we must determine some parameter values and initial and boundary conditions about the mixed layer as follows.

[1] The magnitude of isotope fractionation during uptake of nitrate by phytoplankton ( $\epsilon$ ). Although laboratory studies revealed the large variability of  $\epsilon$  depending on the growth conditions of phytoplankton (Wada and Hattori, 1978; Montoya and McCarthy, 1995), field studies have usually obtained similar values of  $\epsilon$ , 5 to 9‰ (Saino and Hattori, 1985; Goering *et al.*, 1990; Altabet *et al.*, 1991; Nakatsuka *et al.*, 1992), suggesting that in natural conditions the  $\epsilon$  values do not change so much. In the present study we use 5.6‰, as a fixed value for  $\epsilon$ , which has been determined by Wada (1980) at the northwestern north Pacific area near NNWP-02 site.

[2] The degradation rate of sinking PN flux from surface mixed layer to 1335 m ( $k$ ). We use 40 as the  $k$  value, which has been inferred from reported profiles of the vertical flux of sinking POM (Martin *et al.*, 1987, etc.).

[3] The initial nitrate concentration (N) in surface mixed layer at the beginning of trap experiment of NNWP-02. Unfortunately, we have no data about the nitrate concentration at the time of the deployment of the sediment trap. Therefore, in the present study, we try to estimate that concentration based on the following procedure. Kamykowski and Zentara (1986) compiled a world-wide data set of the temperature and nutrients, and concluded that the nutrient concentration in the surface mixed layer can be well predicted from temperature and/or sigma-t. Similarly, we can find a good correlation between SST and surface water nitrate below 15°C, in the northwestern North Pacific Ocean, around the NNWP-02 site, as shown in Fig. 6. Therefore, it seems reasonable to estimate the initial N at NNWP-02, using following equation derived from Fig. 6.

$$[\text{NO}_3^-]_{\text{surface}} (\mu\text{M}) = 29.1 - 1.93 \cdot [\text{SST}] (^\circ\text{C}). \quad (10)$$

Considering the time lag between the surface mixed layer and 1335 m with a sinking rate of about 50 m/day (Banse, 1990; Deuser *et al.*, 1990), an SST of 25 days prior to the central day of the first sampling in the trap experiment was used for the estimate, and the value of 13.4  $\mu\text{M}$  was obtained as the initial N value.

[4] The nitrate concentration (Nd) and its  $\delta^{15}\text{N}$  ( $\delta^{15}\text{Nd}$ ) of deep water below the mixed layer. Because we have no subsurface data at the NNWP-02 site, Nd and  $\delta^{15}\text{Nd}$  are assumed to be 20  $\mu\text{M}$ , based on a reported vertical profile of nitrate near the NNWP-02 site (Wada, 1980), and 4.0‰, respectively. This  $\delta^{15}\text{Nd}$  includes an additional unknown constant besides a real  $\delta^{15}\text{N}$  of nitrate as shown in Eq. (9). The influences of these somewhat arbitrary determinations of deep water conditions will be checked later (Figs. 8 and 9).

[5] The initial mixed layer depth (L). Because we have no information on L above the NNWP-02 site just before the beginning of this experiment, the initial L value

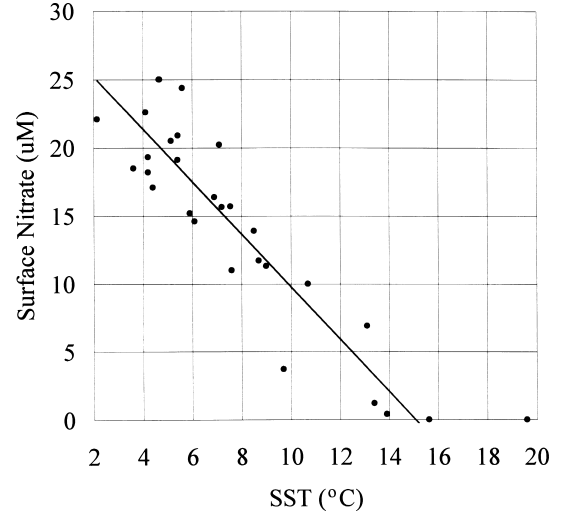


Fig. 6. Relationship between sea surface temperature and surface water nitrate concentration in northwestern North Pacific Ocean, north of 40°N and west of 180°E. Closed circles are data compiled from “Preliminary Reports of The Hakuho-Marui Cruises (KH-71-3, 74-2, 78-3, 83-3, 86-3, 85-2 and 88-3)” (Ocean Research Institute, University of Tokyo, Japan). Solid line is a regression line of data below 16°C ( $[\text{NO}_3^-] (\mu\text{M}) = 29.1 - 1.93 \cdot [\text{SST}] (^\circ\text{C})$ ;  $r = 0.91$ , Standard error =  $\pm 3.1$ ).

is set as 40 m on the basis of the inter-annual averaged value of the surface mixed layer depth in June in this area (Bathen, 1972).

### 3.6 Results of reconstruction

The reconstructed temporal variations of the surface mixed layer depth and nitrate concentration are presented in Fig. 7, together with SST and PN flux data. The time interval ( $\Delta t$ ) for the integration of Eqs. (5) and (6) was 25 days which is determined from the sampling interval of the sediment trap experiment.

Several characteristics can be found in Fig. 7. The mixed layer depth is shallowest in early summer, gradually deepens during fall and then becomes rapidly deeper at the end of fall. Considering the time lag from the surface to 1335 m, the minimum of the mixed layer depth occurs in July, or before July if the initial L is actually shallower than 20 m (Fig. 8-f). This is not the time of the highest sea surface temperature at NNWP-02 (Fig. 3) but the time of the highest solar radiation flux. This pattern in temporal variation corresponds very well to the theoretically predicted one using a kinetic and potential energy balance model conducted by seasonal solar heating and constant wind stress in mid and high latitudinal oceans (Gill and Turner, 1976). And actually in Subarctic Pacific Ocean, the regular seasonal variation in the surface mixed layer depth shows a very similar pattern to that in Fig. 7 (Takahashi, 1989).

The relatively rapid decrease in nitrate concentration in early summer is due to a large primary production associated with spring bloom, but it does not keep on decreasing much more after mid summer because of the gradual deepening of the mixed layer and relatively low new production rate during late summer and early fall (Fig. 7). At the end of fall

the concentration of nitrate increases and causes the autumn bloom as suggested in the sinking PN flux (Fig. 7). Those results are also consistent with the observed changes in the nitrate concentration of the surface water column at Station P in the northeastern North Pacific Ocean (Miller *et al.*, 1991).

The reconstructed temporal variations in the surface

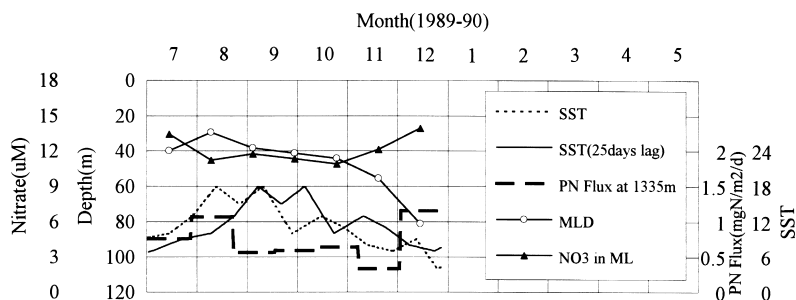


Fig. 7. A result of reconstruction of temporal variations in the surface mixed layer depth (MLD) and nitrate concentration in the mixed layer (ML) from the data of sinking PN flux and its  $\delta^{15}\text{N}$  value at 1335 m shown in Fig. 2. SST curve in this figure is the same as that in Fig. 3. See in text for the parameter values and initial conditions for this reconstruction.

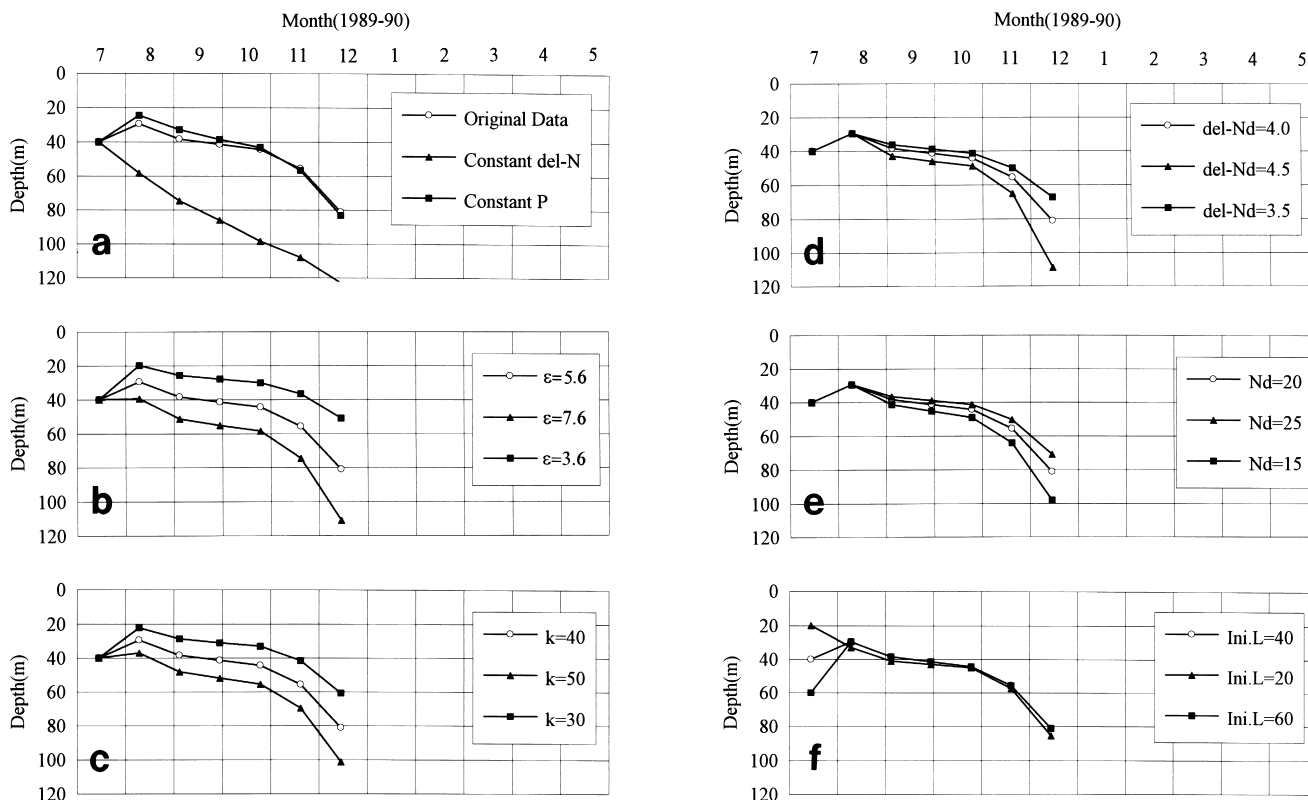


Fig. 8. Results of sensitivity tests for the reconstruction of temporal variations in the surface mixed layer depth. Each figure is corresponding to the sensitivity test for the data of sinking PN flux and  $\delta^{15}\text{N}$  themselves (see text) (a), the magnitude of isotope fractionation during nitrate uptake by phytoplankton (b), the degradation rate of sinking PN during settling process from surface mixed layer to 1335 m (c), the deep water nitrate  $\delta^{15}\text{N}$  (This contains additional unknown constant besides the  $\delta^{15}\text{N}$  of deep water nitrate itself. See Eq. (9) in text.) (d), the deep water nitrate concentration (e) and the initial depth of the surface mixed layer (f), respectively.



mixed layer depth and nitrate concentration (Fig. 7) thus seem to simulate well the general features of surface water processes in the Subarctic Pacific region. However, as we have pre-determined the parameter values and deep water conditions for the calculation of Fig. 7 somewhat arbitrarily, it is necessary to test the sensitivities of the results to those parameters and boundary conditions.

The reconstructed surface mixed layer depth and nitrate concentration with different parameter values, boundary and initial conditions are shown in Figs. 8 and 9. In most cases, the almost same pattern in temporal variations as in Fig. 7 were obtained under different parameters and deep water conditions. In the case of nitrate concentration, the temporal variations do not change at all among the different  $k$  values, the degradation rate of sinking PN (Fig. 9c). These facts suggest that, at least on the qualitative viewpoints, the results of reconstruction are not affected much by parameters and boundary conditions, while the quantitative aspects were largely affected, for example the mixed layer depth in late fall.

Figures 8a and 9a show the sensitivity tests for the “data” of sinking particles themselves, using the extreme assumption of “constant  $\delta^{15}\text{N}$  (always 5.0‰ at 1335 m)” and “constant sinking PN (always 0.75 mgN/m<sup>2</sup>/day at 1335 m)”.

Although the changes in  $\delta^{15}\text{N}$  data distort the original results completely, the results did not have a large sensitivity to sinking PN flux itself. This suggests that the reconstructed temporal patterns are completely dependent on the quality of  $\delta^{15}\text{N}$  data. In general, the  $\delta^{15}\text{N}$  values of sinking PN can be measured correctly, irrespective of the efficiency of the sediment trap at collecting the sinking particle fluxes, which are sometimes affected by uncertainties like the shape of sediment traps and/or deep water currents, etc. Consequently, Figs. 8a and 9a demonstrate the robustness of this reconstructing procedure.

### 3.7 Comparison between the reconstructed surface nitrate concentration and the SST record

In order to clarify the validity of the model presented in this study, it is necessary to compare the reconstructed results with in-situ continuous observations of surface mixed layer. However, it is very difficult to obtain such a continuous data set in the open ocean like the NNWP-02 site, except for SST data as shown in Fig. 3. Therefore, in the present study, the SST-derived nitrate concentrations, as discussed for the initial condition of calculation, are used for validation of the reconstructed results. Figure 10 shows the relationship between the reconstructed variation in surface nitrate from

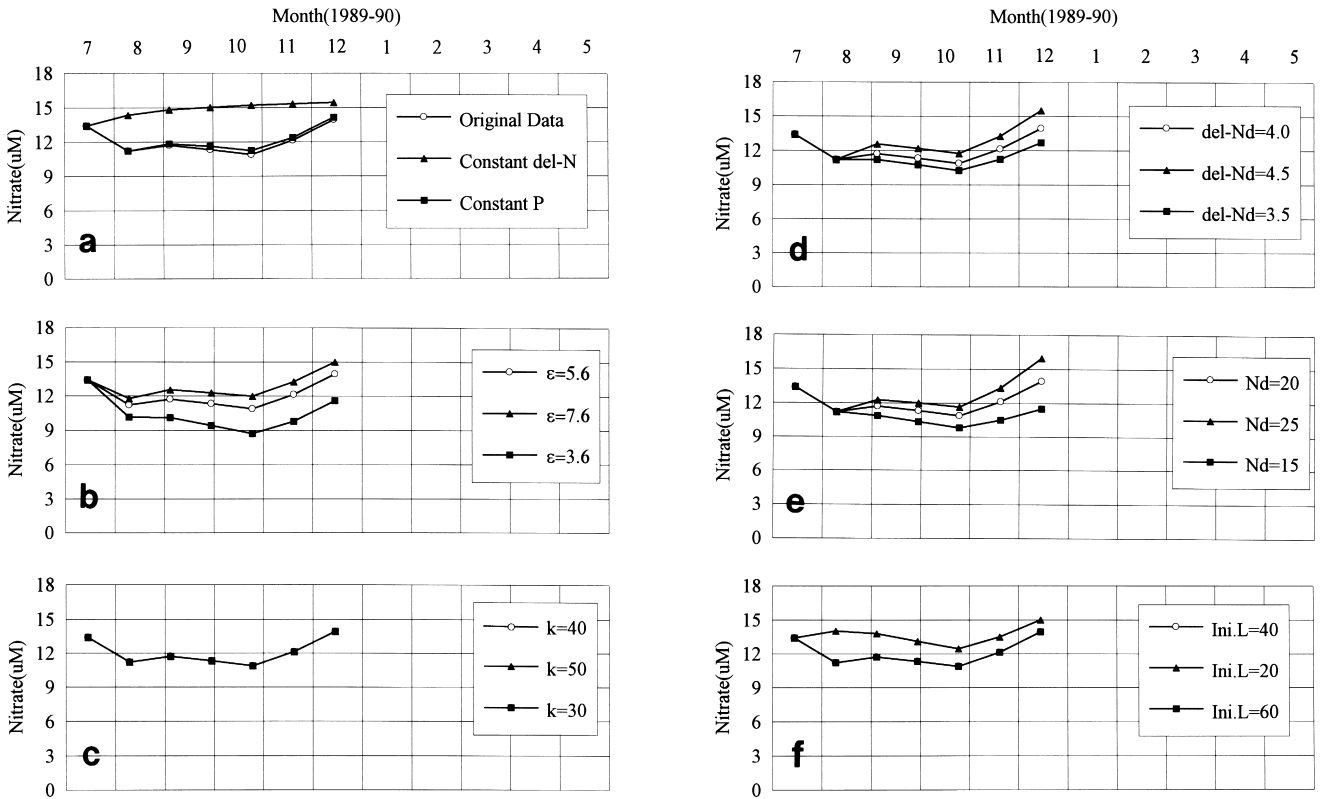


Fig. 9. Results of sensitivity tests for the reconstruction of temporal variations in the nitrate concentration of surface mixed layer. Each figure is corresponding to the sensitivity test as same as shown in Fig. 8.

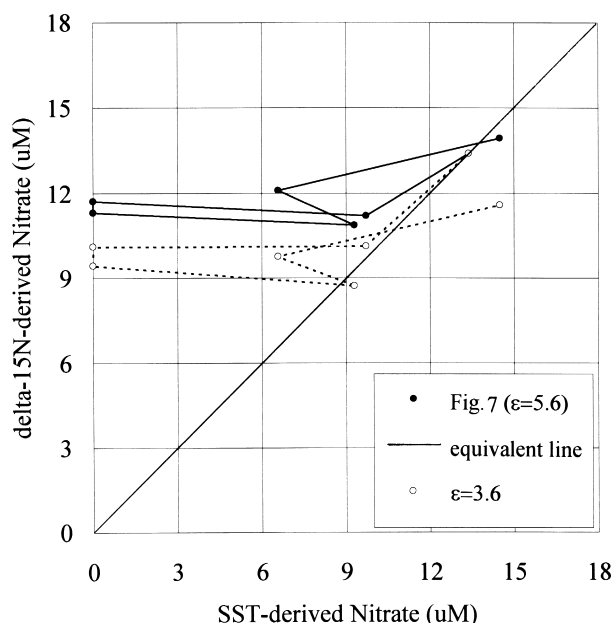


Fig. 10. Relation between the SST-derived surface nitrate concentration and the nitrate concentration reconstructed from  $\delta^{15}\text{N}$  values of sinking PN, during the experiment period. The SST-derived nitrate was calculated using Eq. (10) and SST data ( $44^\circ\text{N}$ ,  $155^\circ\text{E}$ ; Fig. 3) of “25 days before”, considering time lag in sinking process. Closed and open circles correspond to the results from Figs. 7 and 9-b ( $\epsilon = 3.6\text{‰}$ ), respectively.

$\delta^{15}\text{N}$  of sinking PN and the SST-derived nitrate estimated using Eq. (10). The SST data of “25 days before” were used for calculation as discussed earlier.

The  $\delta^{15}\text{N}$ -derived nitrate concentration in the case of Fig. 7 is always higher than the SST-derived one. When the 3.6‰ is used for the parameter value,  $\epsilon$  (magnitude of isotope fractionation during uptake of nitrate by phytoplankton), instead of 5.6‰, the correlation is somewhat improved (Fig. 10). While this suggests the possibility of low  $\epsilon$  value in this area, as found in the Arctic region of the North Atlantic (Voss *et al.*, 1996), the deviation from an equivalent line during September and October (see also Figs. 7 and 9) still cannot be explained. Because the SST-derived nitrate sometimes reaches  $0\text{ }\mu\text{M}$  in contrast to the high concentration in  $\delta^{15}\text{N}$ -derived nitrate which is always above  $8\text{ }\mu\text{M}$ , they cannot be coincided by changes in the hypothesized length of time lag (25 days).

There are several possibilities that explain this discrepancy. The first possibility is the difference between SST and the mixed layer temperature. Specifically, the mixed layer temperature must be equal to SST from the viewpoint of the definition of “surface mixed layer”. However, SST is measured at the top of the water body, and is much affected by diurnal variation of solar radiation flux (Mann and

Lazier, 1991). Therefore, the SST may be sometimes higher than the mean mixed layer temperature during summer, and thus the SST-derived nitrate may also be under-estimated compared to the true nitrate concentration because Eq. (10) assumes a negative linear correlation between SST and nitrate concentration in surface water.

Secondly, the uptake of nitrate by phytoplankton may occur below the mixed layer in late summer. It is due to the exhausted nutrient condition in the surface mixed layer as suggested by SST-derived nitrate. The model, presented for high latitudinal ocean as NNWP-02 in this study, implicitly assumes that nitrate can be assimilated by phytoplankton only in the surface mixed layer. Therefore if the nitrate uptake occurs mainly at the layer below the surface mixed layer, where the nitrate concentration is probably much higher than that in surface mixed layer during late summer, the  $\delta^{15}\text{N}$ -derived nitrate is over-estimated for nitrate in the surface mixed layer in that season.

Thirdly, the assumption of parallel changes between the  $\delta^{15}\text{N}$  of surface nitrate and sinking PN may not be always justified. Because the temporal pattern of  $\delta^{15}\text{N}$  of sinking PN are set to be equal to that of surface water nitrate in this model, Eq. (9), it rules completely the reconstructed results of nitrate variation as shown in Fig. 9a. Therefore, if this hypothesis of parallel changes in  $\delta^{15}\text{N}$  is not acceptable in some cases, other mechanisms must be proposed as alternatives for such cases. One possibility is the longer residence time of assimilated nitrate in the surface mixed layer before sinking out into deep water (Saino *et al.*, 1983). In the present model, the residence time is assumed to be nearly zero, similar to that in the model presented by Altabet and Francois (1994). However, if this is a very long period such as more than one or two months, the temporal increase in  $\delta^{15}\text{N}$  of suspended PN in the surface mixed layer, thus also in  $\delta^{15}\text{N}$  of sinking PN, must be delayed compared to the increase in  $\delta^{15}\text{N}$  of nitrate in the surface mixed layer during summer, because the “old” and “low- $\delta^{15}\text{N}$ ” nitrogen, which has been produced in the spring and early summer seasons using low- $\delta^{15}\text{N}$  nitrate, must remain in the suspended PN pool in the surface mixed layer even in late summer. This tendency has been seen in Voss *et al.* (1996). The apparent depression of the increasing rate of  $\delta^{15}\text{N}$  in sinking PN may cause the under-estimate of biological nitrate uptake rate in surface water, and thus the over-estimate of the  $\delta^{15}\text{N}$ -derived nitrate concentration.

#### 4. Conclusion

The flux and  $\delta^{15}\text{N}$  value of sinking PN showed predominant temporal variations at the NNWP-02 site. Although the  $\delta^{14}\text{C}$  of sinking POC indicated the larger contribution of resuspended sedimentary particles during the winter part of the deeper trap, it was also demonstrated that the  $\delta^{15}\text{N}$  of sinking PN at the shallower trap is primarily controlled by the seasonal variations in physical supply and biological

utilization processes of nitrate in the surface mixed layer. This can be explained well by a model of the surface mixed layer, based on certain assumptions about the mass and isotope balances of nitrate and the close connections between newly produced PN and sinking PN. The temporal variations in the depth of surface mixed layer and the nitrate concentration in it could be appropriately reconstructed by a calculation using this model.

Some problems remain, however, especially when data on temporal variations in sinking particles are used for quantitative estimation of the surface mixed layer process. We must further investigate the particle dynamics which transform the newly produced PN into sinking PN in an upper water column.

For reconstruction of the surface mixed layer processes, much other information in the sinking particle can also be utilized in addition to  $\delta^{15}\text{N}$  of PN; for example, the composition of micro-organism species, their isotope ratios, and  $\Delta^{14}\text{C}$  values of POC as measured in the present study. By synthesizing those data into a certain model of a surface mixed layer as shown in the present study, the physical, chemical and biological processes and their coupling in the upper water column will be illustrated in a comprehensive way. That will be a new subject of the study of the ocean, "Trap Oceanography".

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