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Nutrients and Phytoplankton Blooms in the Southern Coastal Waters of Korea: I. The Elemental Composition of C, N, and P in Particulate Matter in the Coastal Bay Systems

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An investigation was conducted to determine limiting nutrients in the bay systems of the southern coastal area of Korea. The elemental composition of C, N, and P in suspended particulate matter was monitored nearly monthly in Chinhae and Koje Bays and seasonally in Deukryang Bay for 2 years. Atomic C:N ratio in particulate matter ranges from 4.3 to 9.6, typical of marine phytoplankton. C:P and N:P ratios vary from the Redfield ratio to 229 (C:P) and 37 (N:P). A constant C:N ratio of 6.87 from regression of particulate C and N concentrations demonstrates that the particulate matter in the systems originates from primary production. C:P and N:P ratios from regression of C on P and N on P are well associated with changes in salinity. The low N:P ratio of 13.1 implies N limitation in the environments of the systems. This seems to result from the low N:P ratio of nutrients released across sediment-water interface. Phytoplankton response, expressed here as the increase of chlorophyll *a*, to N addition also verifies N limitation for phytoplankton communities. In heavy rainfall season (from June to September), the addition of excessive N via streams into the stratified coastal water proliferates phytoplankton greatly. During the phytoplankton blooms, C:P and N:P ratios are much higher than the Redfield ratio, implying P limitation. This results from the high N:P ratio in nutrients supplied from stream waters. Strong stratification during the blooms also interrupts the supply of nutrients, particularly P, from bottom waters. Dependent upon precipitation, this tendency shows great inter-annual variation.

INTRODUCTION

In coastal eutrophic systems, when no balance between the production and the loss of organic matter is sustained, the accumulation of intensive production material may reach to the point where it exceeds the carrying capacity of the water. These extreme conditions lead to the development of anoxic and eventually abiotic environments (Ryther *et al.*, 1972). Since the 1970's, eutrophication has been accelerated in the southern coastal bay systems of Korea because of the continuous supply of nutrients by industrial and domestic waste waters. Extreme algal bloom (red tide), has occurred regularly in Chinhae Bay and its neighboring waters from April to October every year since 1961 (Park and Kim, 1967; Park *et al.*, 1989; NFRDI, 1997). Anoxic water mass at the bottom layer has also formed concurrently (Lee *et al.*, 1993). Eutrophication in the sediment in Koje Bay and Deukryang Bay have also been revealed to be in progress and in the early stage, respectively (Cho and Kim, 1977; Cho *et al.*, 1982).

To control the eutrophication, it is essential to assess "Which nutrient is limited?" and "Which nutrient has to be reduced?" Although chemical composition, physiological measurements, and nutrient enrichment bioassays have been used to assess the nutrient status of phytoplankton, particulate C:N:P composition ratio may be one of the simplest ways to obtain the information on nutrient limitation for phytoplankton growth in natural waters (Hecky and Kilham, 1988).

There is a remarkable constancy of 106:16:1 (by atoms) in the elemental composition of three major constitutents C, N, and P in marine particulate matter. This ratio has been referred to as the Redfield ratio, which is a clue in treating stoichiometrically the biological activity in aquatic systems. Such chemical composition of marine plankton is strongly influenced by the chemistry of the surrounding waters. Numerous laboratory and field studies have shown that nutrient deficiency alters the chemical composition of algae in a relatively predictable way, that is, the cellular composition ratio coupled with the changes of composition of the medium (Redfield, 1958; Perry, 1976; Myklestad, 1977; Rhee, 1978; Goldman *et al.*, 1979; Healey and Hendzel, 1980; Hecky *et al.*, 1993; Mastuda, 1993).

The current dogma that phytoplankton tends to be N limited in marine waters and P limited in fresh waters is difficult to generalize for the coastal waters that are affected by the supply of nutrient from the outer systems such as the open sea and rivers. While N is limited in the New England coastal phytoplankton populations (Yentsch *et al.*, 1977), P is limited in the Norwegian fjords and coastal waters (Sakshaug and Olsen, 1986) and in the Chinese coastal waters (Harrison *et al.*, 1990). D'Elia *et al.* (1986) reported the great seasonal variability in N or P limitation in a tributary of the Chesapeake Bay estuary.

Despite numerous studies on the distribution and cycle of nutrients, the characteristics of particulate matter and phytoplankton populations in Korean coastal waters, particulate C, N, and P compositions as well as nutrient limitation of phytoplankton have been little investigated. Therefore, we examine here the relationship between the input of nutrient and the seasonal variation of particulate C:N:P ratios in the southern coastal bay systems of Korea. We discuss the inter-annual variations of the ratios in comparison with the data set on Chinhae Bay in 1993 reported by Lee *et al.* (1994a). The primary objective of this study is to assess temporal

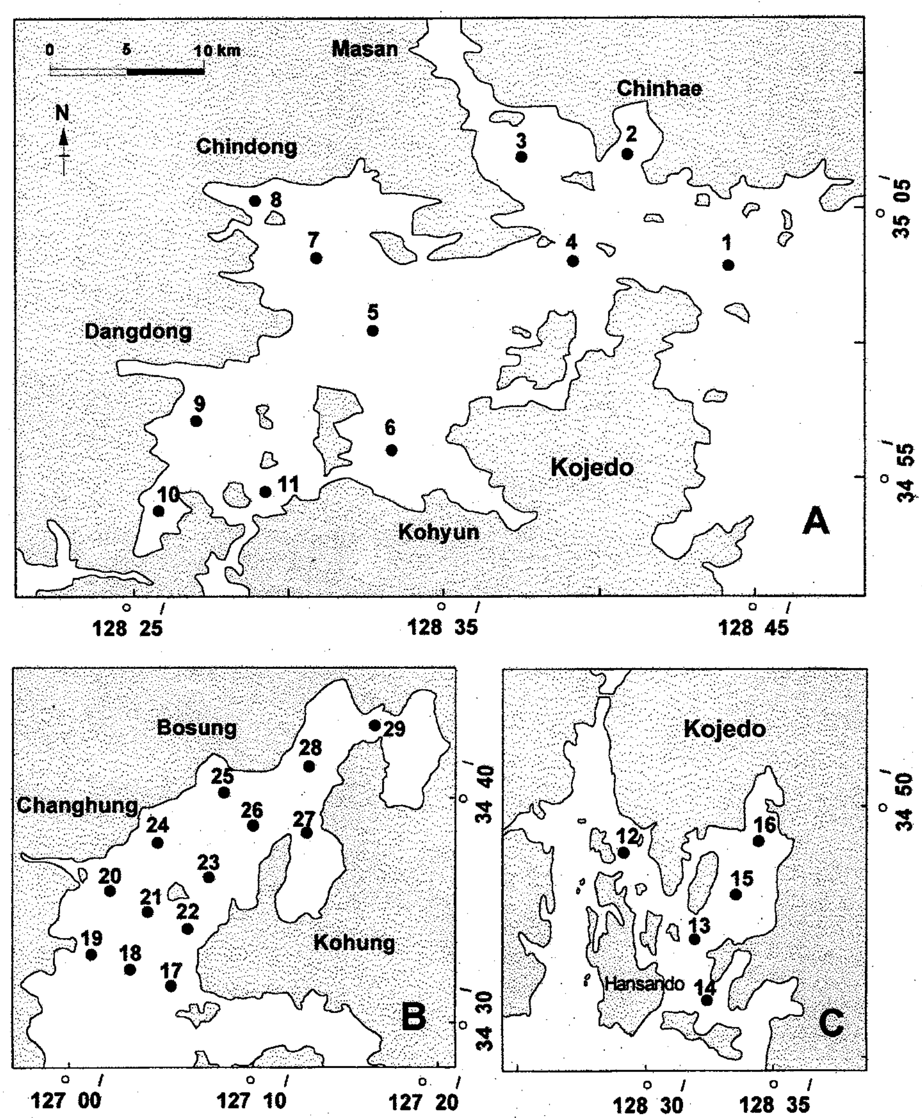

Fig. 1. Map showing sampling stations in the southern coastal bay systems of Korea.
(A: Jinhae Bay, B: Deukryang Bay, C: Koje Bay).

variations in N or P limitation from monitoring the ratios and the chlorophyll *a*. The influence of precipitation on the inter-annual fluctuation of summer phytoplankton blooms is also discussed.

MATERIALS AND METHODS

Site description

Twenty-nine sampling stations were located from Chinhae and Koje Bays to Deukryang Bay in the southern coast of Korea (Fig. 1). Chinhae Bay is subdivided into two zones (Lee *et al.*, 1981). One comprises the western part of the bay and is a less polluted area; the other includes Masan Bay (Station 3) and Haengam Bay (Station 2) with apparently severe pollution problems with red tide. The water column of Chinhae Bay is stratified in summer and well mixed in winter (Hong *et al.*, 1991; Lee *et al.*, 1993).

In Koje Bay, chlorophyll *a* contents are lower than those of the Chinhae Bay. Even during the summer stratification period, no oxygen depletion is found in the bottom layer because of the rapid current velocity (Yoo *et al.*, 1980).

Deukryang Bay is characterized as permanently well-mixed coastal waters (Hong *et al.*, 1988). Pollutant load from land is relatively lower and nitrate is mainly supplied by the inflow of offshore coastal water (Yang *et al.*, 1995).

Sampling and analytical procedure

Samplings were carried out nearly monthly in Chinhae and Koje Bays and seasonally in Deukryang Bay during a two-year (1994-1995) period. Surface water samples were collected using a 3-l van Dorn water sampler. Temperature and salinity were measured *in situ* using a CTD meter (Seabird Electronics, Inc.). For nutrient analysis, water samples were filtered through Whatman GF/F glass fiber filters and deep frozen for later analysis. Dissolved inorganic nutrients were measured spectrophotometrically following Parsons *et al.* (1984).

For particulate matter analysis, two samples were prefiltered by using a 350 micron-mesh net to exclude macrozooplankton. Particulate organic carbon (POC) and nitrogen (PON) were determined after filteration by Whatman GF/F glass fiber filters precombusted onto 450°C. The filters were exposed in concentrated HCl to eliminate calcium carbonate and then determined after dry combustion in a Perkin Elmer 2400 elemental analyzer. The instrument was calibrated with acetanilide standard. Particulate phosphorus (PP) was determined colorimetrically after filteration. Details are given in De Lange (1992). Chlorophyll *a* was determined on aceton extracts using the fluorometric method of Holm-Hansen *et al.* (1965) as modified by Parsons *et al.* (1984) with a 10 AU Fluorometer (Turner Designs). All statistical analyses were done on SPSS program (Version 7.5). All available data on Chinhae Bay in 1993 (Lee *et al.*, 1994a) was also employed.

RESULTS

Hydrography

Fig. 2 presents the monthly variations of mean surface temperature and salinity during the survey period. Mean surface temperatures varied from *ca.* 6°C in February 1993 to higer than 28°C in August 1994. The time series of mean temperature in which the minimum appears in winter and the maximum in summer is typical in the temperate zone. Generally, mean surface salinity is lower in summer than in the

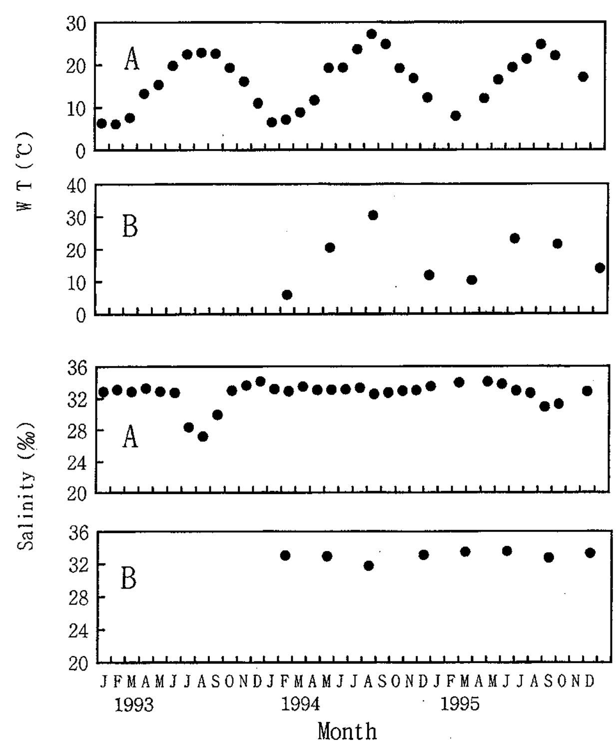

Fig. 2. Time series of mean surface temperature and salinity during the surveying period. (**A**) Chinhae and Koje Bays, (**B**) Deukryang Bay.

other seasons. Surface salinity in summer also shows the great inter-annual variability. In Chinhae and Koje bays in August, mean surface salinities were 32.50‰ and 30.89‰ in 1994 and 1995, respectively, while it was 27.15‰ in 1993 in both areas. In Deukryang Bay, mean surface salinity of August was 32.98‰ (SD= ± 0.61) and the seasonal variation was not apparent.

Nutrients and chlorophyll a

In the surface waters of Masan (Station 3) and Haengam (Station 2) Bays, concentrations of dissolved nutrient varied irregularly and were remarkably higher than at the other stations (Fig. 3). Mean DIN ($NH_4+NO_2+NO_3$) concentrations ranged from 0.64 μM (November 1995) to 167.72 μM (September 1995). Phosphate concentrations were in the range of 0.17 μM (June 1993) to 8.26 μM (September 1995). DIN:PO_4 ratios varied from 0.6 to 67.0.

In the western part of Chinhae Bay, nutrient concentrations pulsed in the low saline summer period and then declined abruptly (Fig. 3). Mean DIN and phosphate concentrations were in the range of 2.21 to 39.72 μM and 0.07 to 2.08 μM, respectively. The concentrations peaked in July 1993 and September 1995. Phosphate concentrations were, however, less than 1 μM except for 2.08 μM in September 1995. DIN:PO_4 ratios varied from 5.1 to 82.3.

In Koje Bay, the mean DIN concentrations ranged from 3.20 to 16.07 μM and no apparent seasonal variation patterns appeared over the survey period (Fig. 3). Mean phosphate concentrations were in the range of 0.08 to 0.63 μM. The concentrations peaked in late summer and fell and declined in spring. DIN:PO_4 ratios varied from 4.9 to 50.3.

In Deukryang Bay, the mean DIN and phosphate concentrations were in the range of 0.66 to 4.71 μM and 0.17 to 0.65 μM, respectively, and were in lower level than those in the other bays (Fig. 3). DIN concentrations were generally higher in summer but seasonal variations were moderate. DIN:PO_4 ratios varied from 3.8 to 28.1 and were lower than in the other bays.

Annual variations of chlorophyll *a* concentrations in the surface waters are presented in Fig. 4. The

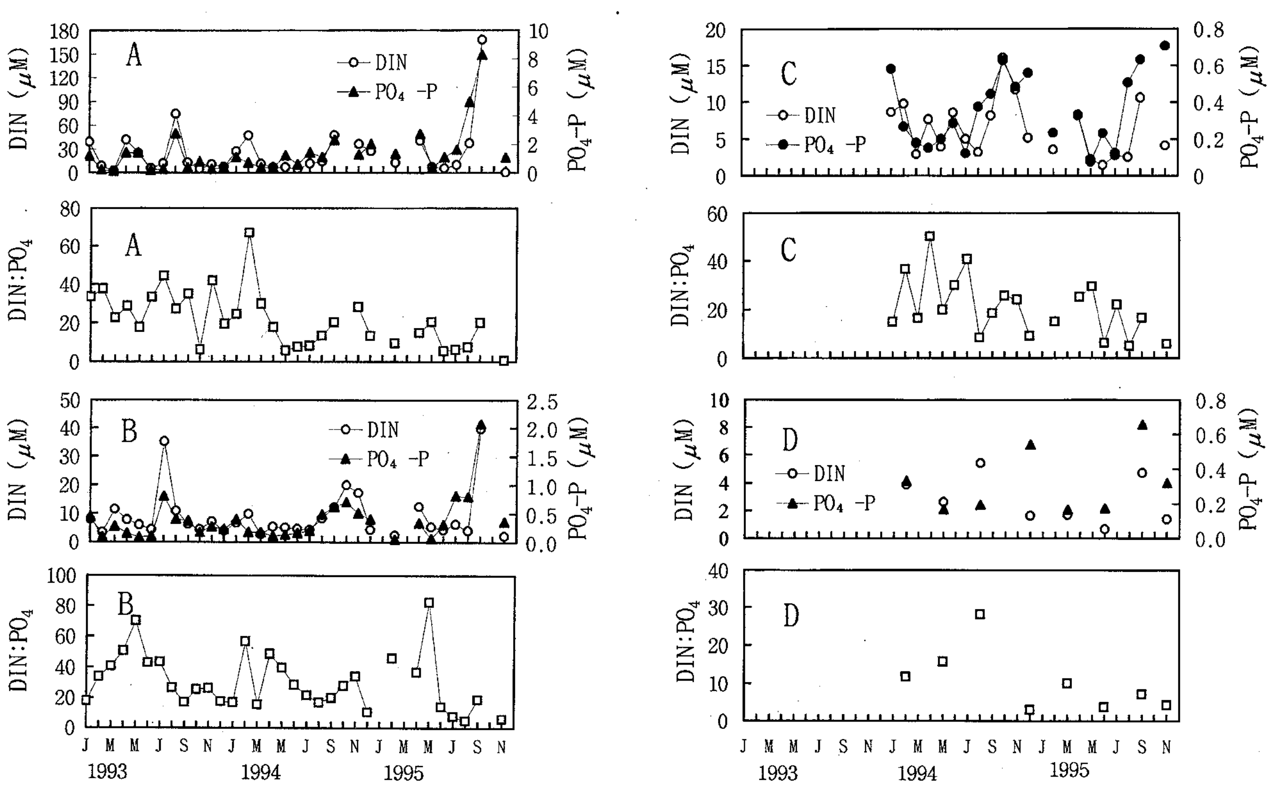

Fig. 3. Annual variations of mean DIN ($NH_4+NO_2+NO_3$) and phosphate concentrations and N/P ratios in the surface waters. (**A**) Stations 2 and 3, (**B**) the western part of Chinhae Bay, (**C**) Keoje Bay, (**D**) Deukryang Bay.

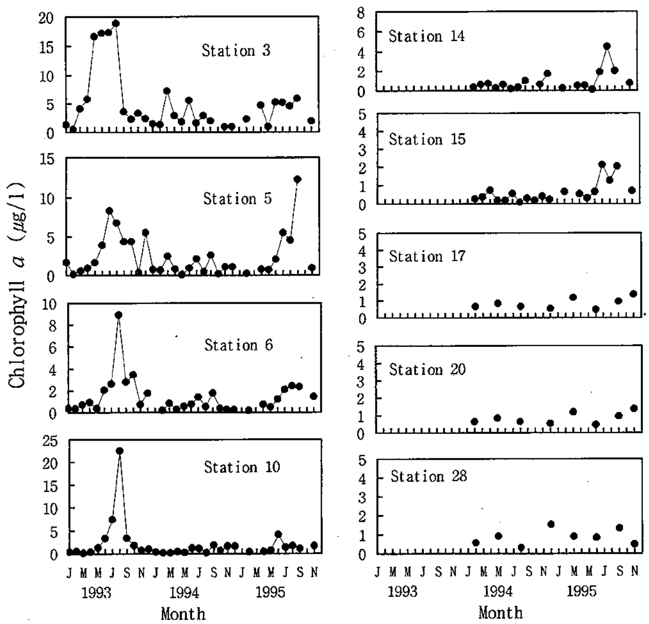

Fig. 4. Annual variations of chlorophyll *a* concentrations in the surface waters.

Table 1. Mean concentration (μM) of particulate organic C and N and particulate P for n samples. The elemental ratios are based on the mean elemental concentrations

Systems	n	C	N	P	C:N	C:P	N:P
Chinhae Bay	316	61.6	8.4	0.50	7.3	123.1	16.8
Keoje Bay	94	35.5	5.6	0.30	6.4	118.2	18.5
Deukryang Bay	101	30.9	5.1	0.31	6.1	98.7	16.2
Mean	511	50.8	7.2	0.43	7.0	119.0	17.0

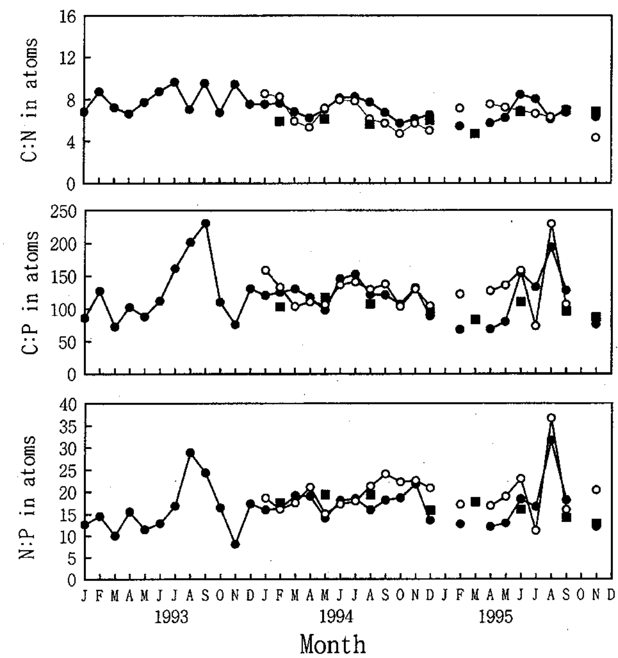

Fig. 5. Annual variations of atomic C:N, C:P, and N:P ratios during the survey period. Values are calculated from the mean monthly concentrations (μM/l) of C, N, and P in particulate matter. ○ =Chinhae Bay, ● =Keoje Bay, ■ =Deukryang Bay.

high concentrations of more than 5 μg/l appeared irregularly at the mouth of Masan Bay (Station 3). Time series patterns of chlorophyll *a* concentrations in the surface waters of Chinhae and Koje bays were well correlated to the salinity variations. The concentrations peaked during the low saline periods in summers 1993 and 1995 but not in 1994. Chlorophyll *a* concentrations ranged from 0.09 to 22.59 μg/l in the western part of Chinhae Bay and 0.12 to 4.49 μg/l in Koje Bay. The chlorophyll *a* concentrations in Deukryang Bay were in the range of 0.24 to 1.52 μg/l and the seasonal variations were less apparent than in the other bay systems.

C:N:P composition ratios in particulate matter

Mean concentrations of particulate C, N, and P were in the range of 30.9 to 61.6 μM, 5.1 to 8.4 μM and 0.30 to 8.4 μM, respectively (Table 1). The concentrations were much higher ($P<0.001$) in Chinhae Bay than in Koje and Deukryang Bays where the concentratios were similar ($P>0.05$). The mean elemental C:N:P composition in particulate matter was 119:17:1 close to the Redfield ratio, in all bay systems (Table 1). There was little difference in the atomic ratios of C:N and N:P among the bays. The atomic C:P ratio was the highest (123.1) in Chinhae Bay and the lowest (98.7) in Deukryang Bay.

Figure 5 shows the annual variations of atomic C:N, C:P, and N:P ratios in the bays. C:N ratio fell within a narrow range of 4.3 to 9.6, typical of marine phytoplankton. C:P and N:P ratios varied from near the Redfield ratio to 229 (C:P) and 37 (N:P). Extremely high C:P and N:P ratios were observed during the low saline period with the peak chlorophyll *a* in 1993 and 1995 but not in 1994. None of the particulate concentrations presented here were corrected for detrital contribution. Instead, we investigated the particulate contents only where the surface waters were characterized by a high algal production. That is because the linear regression between particulate elements (C, N, or P) and chlorophyll for correcting detrital contributions to these elements is not efficient in estimating the nonalgal particulate C, N, and P. We also examine here C:N, C:P, and N:P ratios from linear regressions (Fig. 6). All of the correlation coefficients are highly significant at the 0.01 level. The particulate

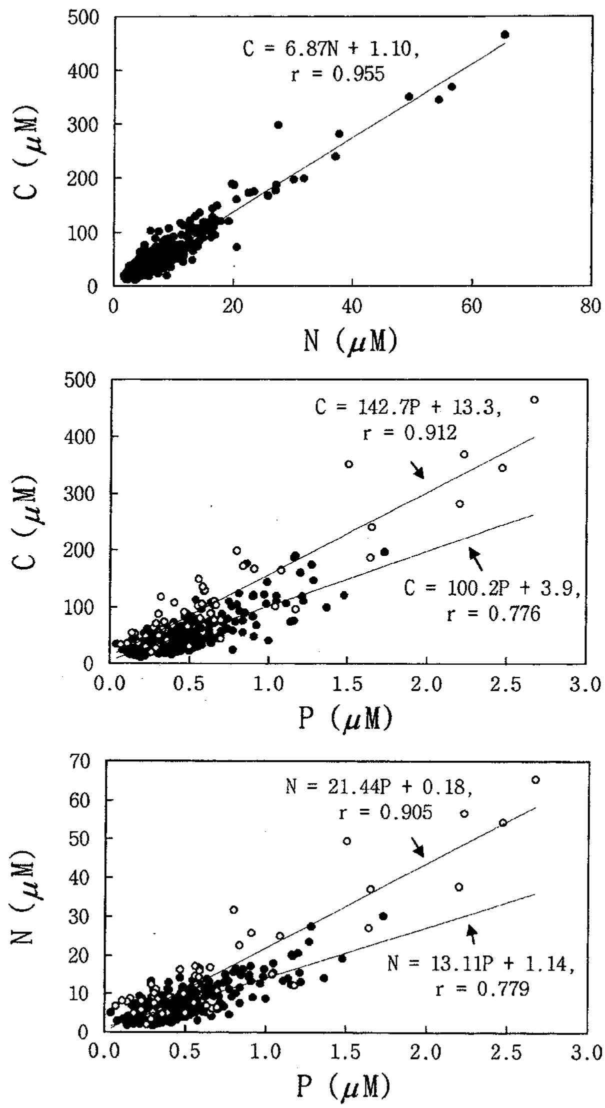

Fig. 6. (A) Particulate C and N concentrations for 511 samples. (B) Particulate C and P concentrations. (C) Particulate N and P concentrations. All the correlation is significant at the 0.01 level (2-tailed). ○ represents 54 samples in July, August, and September 1993 and August and September 1995; ● 457 samples for the other months.

C and N concentrations are closely correlated ($r=0.955$, $p<0.001$, $n=511$). The C:N ratio of 6.87, a regression coefficient from all particulate C and N concentrations, is well consistent with the Redfield ratio which is characteristic of oceanic particles. The relationships between particulate C and P concentrations and between particulate N and P concentrations are shown separately according to the salinity variation. The regression lines ($r=0.912$ (C:P) 0.905 (N:P), $p<0.001$, $n=54$) during the low saline (mean<32‰, in convenience) period have much steeper slopes of 143 (C:P) and 21.4 (N:P), compared to these of the other survey periods. These high ratios correspond to chlorophyll maxima. The general ratios of 100 (C:P) and 13.1 (N:P), which are regression results ($r=0.776$ (C:P), 0.779 (N:P), $p<0.001$, $n=457$) of the concentrations except for the low saline period, are more or less lower than the Redfield ratio.

DISCUSSION

Lee *et al.* (1994a) observed marked seasonal variations in particulate C, N, and P concentrations in the surface waters of Chinhae Bay whose values were much higher than those in the other coastal waters of Korea. No apparent seasonal variations in the concentrations were, however, observed in Koje and Deukryang bays (Lee *et al.*, 1994b; 1995). Although the mean C, N, and P concentrations in Koje and Deukryang Bays were much lower than those in Chinhae Bay (Table 1), the concentrations were still much higher than the maximum values (24 μM, 4 μM and 0.2 μM for C, N and P, respectively) in the Atlantic, Indian, and Antarctic oceans, and the Mediterranean Sea surveyed by Copin-Montegut and Copin-Montegut (1983).

In general, the C:N atomic ratio of marine phytoplankton varies from 3 to 9 (Parsons *et al.*, 1961), while the ratio of land-derived particulate matter is over 12 (Pocklington and Leonard, 1979) because of the high ratios in terrestrial soils and C_4 plants. The atomic ratio of POC and PON ranged from 4.3 to 9.6 in the bay systems of study area (Fig. 5). This implies that particles in these systems were derived from *in situ* phytoplankton production. From C:N ratio and stable carbon isptope ratio ($\delta^{13}C$) of the particles in the eastern part of Chinhae Bay, Kim *et al.* (1993, 1994) also reported that increased POC and PON levels in the surface waters in the spring and summer were caused by high primary productivity. C:P and N:P ratios were closely linked with the variation of salinity and showed marked inter-annual variations. In Chinhae and Koje bays, extremely high ratios were observed during the low saline summer period in 1993 and 1995, but not in the summer of 1994. In Deukryang Bay, no summer increase of the ratios was found.

Hecky and Kilham (1988) reviewed that phytoplankton can become limited by the availability of nutrients when sunlight intensity and water temperature are adequate and the loss rates of the nutrients are not excessive. In general, the ratio of C:N:P in algal cells reflects the N:P ratio of ambient water. Therefore, the C, N, and P contents of particulate matter reflect the supply ratios between each nutrient as well as nutrient deficiencies in natural phytoplankton assemblage (Dodds and Priscu, 1990). The compositional responses of diverse alga groups to nutrient limitation are similar (Healey and Hendzel, 1980; Hecky and Kilham, 1988). From the fundamental view that algae rather than the environment provide the most direct and relevant answers regarding algal behavior, Sakshaug and Olsen (1986) also demonstrated that when the chemical composition of a natural population is known, information on the limiting nutrient for phytoplankton growth can be extracted.

Overall variation patterns of C:N, C:P, and N:P ratios of particulate matter are revealed from the regressions of C on N, C on P, and N on P (Fig. 6). The C:N ratio was independent of salinity variation and the ratio of 6.87 is closely consistent with the Redfield ratio, while C:P and N:P ratios in Chinhae and Koje bays were different according to the salinity variation. In the 8-time investigations for 2 years, the low saline waters (mean$<$32‰) were not found in Deukryang Bay (Fig. 2). General C:P and N:P ratios in the data set except for the low saline period in Chinhae and Koje bays were 100 and 13.1, respectively. No consistency is, however, found in the DIN:PO_4 ratio of nutrients in ambient waters (Fig. 3). There being no apparent relationship between particulate nutrients and ambient nutrients, it is fruitful to examine the process of nutrient supply (Sakshaud and Olsen, 1986).

Nutrients released across sediment-water interface could affect the primary productivity. In the western part of Chinhae Bay, nutrients released in the *in situ* chamber were maximum with 1268 $\mu M/m^2$/day for DIN and 228 $\mu M/m^2$/day for PO_4 in August and were minimum with 131 $\mu M/m^2$/day for DIN and 19 $\mu M/m^2$/day for PO_4 in February (NFRDI, 1992). The DIN:PO_4 ratio in the released nutrients was less than 7, which implies that enriched PO_4 over DIN was supplied from the bottom sediment to the water column. This low released ratio was also found in Koje Bay (NFRDI, unpublished data). Yang and Hong (1982) and Lee (1993) also suggested that dissolution of phosphate from the sediments could be the possible source of high phosphates. Consequently, the low N:P ratio of particulate matter is regarded to reflect the low supply ratio of DIN:PO_4 and N-limitation for phytoplankton growth.

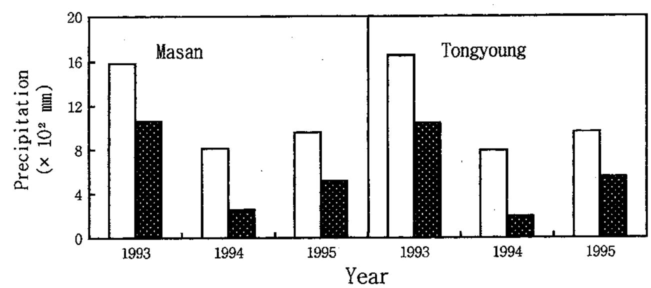

Fig. 7. Annual precipitation in Masan and Tongyoung areas. Open bar, total precipitation over a year; shaded bar, total precipitation over a summer period (from June to September).

Nutrient limitation has been tested using nutrient enrichment bioassay experiments of various types to determine if algal growth is stimulated by addition of one or more nutrient. Of these experiments, Heckey and Kilham (1988) concluded that the test results for large mesocosms (rivers, lakes, bays and oceans) must have been very clear results. We here consider Chinhae Bay a good example for large mesocosm. Phytoplankton-growth response to the addition of one nutrient may elucidate the limitation by the nutrient.

Figure 7 shows the variation in precipitation in Masan and Tongyoung areas near Chinhae and Koje Bays. High variability of the annual precipitation is evident. The annual precipitation was about 1600 mm in 1993, about 800 mm in 1994, and 950 mm in 1995. The annual precipitation in 1993 was twice of that in 1994. Of the annual precipitation in 1993 and 1995, over 60% was concentrated in the stratified period (from June to September) but in 1994 only 30% was recorded in the stratified period. Consequently, low salinity of the surface waters in 1993 and 1995 can be explained from the high precipitation. Nutrient-loading ratios from streams investigated by MOE (1991) and NFRDI (1995) in Chinhae Bay are presented in Table 2. Being dependent on the precipitation (NFRDI, 1995), DIN:PO_4 ratio loaded from stream waters varied from 61

Table 2. Nutrient load from streams into Chinhae Basin

		Nutrient loading rate (kg/day)		N/P molar ratio	Reference
		[1]DIN	[2]DIP		
1991	June	28448	560	112.5	MOE
	August	28532	242	261.1	(1991)
1995	June	5023	180	61.8	NFRDI
	July	14489	526	61.0	(1995)

[1]DIN=$(NH_4+NO_2+NO_3)$-N
[2]DIP=PO_4-P

to 261, which indicates that DIN was overenriched in the stream waters. A high DIN:PO_4 ratio of about 80 was also reported from freshwater introduced to Masan Bay (KORDI, 1987). As a result, during the low saline period in 1993 and 1995, excessive N in comparison to P entered the Masan Bay due to the increase of volume of stream waters by the intense precipitation. Additions of N enhanced the phytoplankton biomass, expressed here as chlorophyll *a* concentration, greatly during the summer stratified periods in 1993 and 1995 (Fig. 4). In the summer of 1994, no increase in chlorophyll *a* concentration was found. Irregularly high chlorophyll *a* concentrations at the mouth of Masan Bay resulted from allochthonous input of N throughout the year (Hong *et al.*, 1991). Such growth response of natural algal community to N enrichment at long-time scale confirms us that the low N:P ratio indicate N limitation. In Deukryang Bay, there was no apparent increase in chlorophyll *a* concentration in the survey period. Hong *et al.* (1988) reported that the addition of nutrients via freshwater runoff during the late summer enhanced phytoplankton growth and the depletion of nitrate terminated phytoplankton bloom in Deukryang Bay.

By contrast, during phytoplankton blooms in the low saline period in Chinhae and Koje Bays, C:P and N:P ratios were 143 and 21, respectively, which were much higher than the Redfield ratio (Fig. 6). High ratios of C:P$>$129 and N:P$>$22 indicate P limitation for phytoplankton growth (Healey and Hendzel, 1980). When chlorophyll *a* in the surface waters peaked due to N addition from the stream waters in the low saline period, the water column was strongly stratified. PO_4 flux across sediment-water interface in August was 6 times greater than that in February (NFRDI, 1992). Even though PO_4 concentrations in the bottom waters increased as oxygen deficiency became more and more severe from June to August, strong stratification of water column interrupted the supply of PO_4 to the surface waters (Yang and Hong, 1982; Lee, 1993; Lee *et al.*, 1993). This fact supports P limitation during the low saline summer period. In general, the nutrient supply limits plankton growth at the end of a bloom. It is therefore suggested that direct phosphate supply from the bottom waters by a mixing which breaks discontinuous layers may cause temporarily intensive phytoplankton blooms (red tide).

In conclusion, a generally low N:P ratio implies N limitation in the southern coastal bay systems of Korea. In the low saline (mean$<$32‰) summer period, the addition of excessive N via streams enhances phytoplankton biomass greatly. C:P and N:P ratios during phytoplankton blooms are much higher than the Redfield ratio, implying P limitation. Dependent upon precipitation, this tendency shows a great inter-annual variation.

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