原著論文

Environmental chemistry of rivers and lakes, Part VIII[†]. General trends in the variations of phosphorus, nitrogen and the N/P ratio over 540 months (May 1963-May 2008) in Lake Biwa, Japan

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Abstract

Based on a series of observations of chemical components occurring in Lake Biwa over a period of 540 months from May 1963 (at which the time was set as t = 0) to May 2008 (t = 540) and by applying regression analysis, the average rates of increase of total-phosphorus, TP, and total-nitrogen, TN, in the Northern Basin were evaluated as TP_{avg} (μM/month) = $3.0 \times 10^{-4} \cdot t + (0.2 \pm 0.1)$ and TN_{avg} (μM/month) = $6.2 \times 10^{-3} \cdot t + (20 \pm 7)$. By referring to these regression lines over the 540 months (t = 0 to 540), the trophic state in the recent, second stage (t = 347 to 540) was compared with that in the early, first stage (t = 0 to 226) in the Northern Basin, represented by the sampling station Ie-1. A similar comparison was also made between the Northern Basin (Ie-1) and Southern Basin (Nb-5). The variations in the TN/TP ratios against time were also discussed in relation to propagation of plankton. As a result, the lower TN/TP ratios observed in more recent observations were well coincident with the more frequent and denser propagation of N_2-fixing cyanobacteria. A first sign for the restoration of water quality in the Southern Basin has thus been observed.

Keywords: Lake Biwa, Long term variation, Total-phosphorus, Total-nitrogen, TN/TP ratio

Introduction

A period of 540 months (45 years) is relatively short compared with the time needed for geological and meteorological changes in the natural environment. Over 540 months, starting from May 1963 (at which the time was set as t = 0) to May 2008 (t = 540), however, human activity increased rapidly due to industrialization and upset the material balance in the ecosystem. The perturbation has entered the air, soil and water environments. Especially, for the water environment, eutrophication has proceeded in various water reservoirs even over such a short period as 540 months.

Over the 540 months, several groups of limnologists have been observing the variations in nutritious elements in Lake Biwa. In 1962, Japan's Ministry of Construction organized an investigative committee named Biwa-ko Seibutsu-shigen Tyosa-dan, BST.

As part of the BST general survey, the chemistry division headed by Fujinaga twice observed the distribution of chemical components over the whole lake. The results were published in (BST 1965), whose data we refer to here as "BST."

Following the BST survey, Fujinaga and his coworker, the Hydrosphere Research Group (HRG),

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[†]For Part VII, see Nagai et al. (2001)

started a new series of observations from May 1963 to June 1981 (t = 0 to 217). They took water samples several times a year at four representative sampling stations, including Ie-1 in the Northern Basin and Nb-5 in the Southern Basin. The data termed "HRG" have been published in a book (Fujinaga and Hori 1982). After HRG, one of the present authors (T.H.) conducted a new series of observations termed "PHOS" during the period 1980 to 1982 (t = 204 to 226). The results were made public (Hori et al. 1992), and a new parameter of Gross Weight of Phosphorus, GWP, was first introduced for the statistical treatment of the widely varied phosphorus concentrations at Ie-1. Focusing on the distribution of silicon and aluminum, the next series of observation termed "SIAL" was carried out monthly from April 1992 to March 1995 (t = 347 to 382), during which period usual [1992–93] and unusually wet [1993–94], as well as unusually drought [1994–95] conditions were included (Hori et al. 1996). The present authors started to observe phosphorus and nitrogen levels from May 1996 to May 2008 (t = 396 to 540) and the results are referred to as "NUTR". From the HRG to the recent NUTR, the whole data set thus covers 540 months.

In parallel to these researchers, Yoshida (1997) monitored the variations in the TP, TN and N/P ratio from 1983 to 94 in the Southern Basin and concluded that *Microcystis aeruginosa* dominantly occurred in water with low DIN, DIP, and DIN/DIP ratios, while the species also occurred in waters with meteorological conditions of high air temperature and long sunshine time. Harashima et al. (2006) also reported that loading of TP and TN into Lake Biwa increased from 1960 to 75 and then decreased, reflecting the economic growth and regulation of P and N loading. Okamura (2008) reported that Lake Biwa has been investigated by many institutes to date, but the Fisheries Experimental Station program is the longest continuous conducting since 1915, monitoring water temperature, transparency, water color, DO, pH, COD, nutrients and plankton biomass; the study also reported that during 1970s and 1980s several eutrophication-related phenomena were observed, such as excessive growth of *Egeria densa,* and an outbreak of freshwater red tied caused by blooming of *Uroglena americana.* Nowadays, eutrophication in Lake Biwa has some ways improved, because transparency has increased and the number of red tides has decreased.

It is scientifically significant that the 540-month period includes the two important stages for Lake Biwa : The first stage covers the years of 1963–1982 (t = 0 to 226 months) when human activities were initially so subtle that the lake retained its original state. Subsequently, eutophication of the lake proceeded rapidly due to industrialization, and later local laws were legislated for the restoration of the lake. The second stage covers 1992–2008 (t = 347 to 540) when self-cleaning effects may have appeared.

The effects of the inflows to Lake Biwa are very complicated because different water flows such as riverwater, groundwater, agriculture or industrial drain water, or surface water due to heavy rainfall or flood containing various chemical materials, non-point sources, and most of them were not measured well for analysis (Aota et al. 2003; Rimmer et al. 2005). By using time (t), we are trying to correlate our nutrients (total-nitrogen and total-phosphorus) data with the impact of anthropogenic influences on Lake Biwa, such as implementation of Shiga prefecture ordinance for prevention of eutrophication in Lake Biwa in 1979 (t = 192), promotion of septic tanks in 1981 (t = 212); recently, improvement in TN/TP ratio due to self-cleaning effects.

In the present study, the data of BST, HRG, SIAL, NUTR (2009)-observations were analyzed by least-square regression analysis against time and approximated to straight lines to deduce long term trends in variations of chemical components such as phosphorus and nitrogen which occurred in Lake Biwa. The variation in the N/P ratio with time is also discussed in relation to the propagation of plankton. A general view of the past, present and future trends in the eutrophic state of Lake Biwa is presented.

Observation settings

The geological and limnological characteristics of Lake Biwa are summarized in Table 1. Water samples were collected from the surface at Ie-1 (35°12′58″N 135°59′55″E) and Nb-5 (35°04′02″N 135°54′27″E). The location of Ie-1 and Nb-5 are shown in Fig. 1. At Ie-1, samples were also taken at depths of 1, 5, 10, 15, 20, 30, 40, 50, 60, 70 and 73 m (bottom) using a Van Dorn sampler.

Table 1. Geological and limnological characteristics of Lake Biwa

Item	Data
Coordinates	35°15′N, 136°05′E
Primary inflow	More than 100 small rivers
Primary outflow	Seta river
Catchment Area	3,174 km^2
Surface area	
Northern Basin	616 km^2
Southern Basin	58 km^2
Capacity	
Northern Basin	2.73×10^{10} m^3
Southern Basin	0.02×10^{10} m^3
Round	235.2 km
Length	63.5 km
Width	22.8 km (max.), 1.35 km (min.)
Water depth	
Average	41 m (whole lake)
	43 m (N. Basin), 8 m (S. Basin)
Maximum	104 m (N. Basin)
Ie-1	70 m
Nb-5	3.5 m
Residence time of water	5.5 yrs (N. Basin)
	0.04 yrs (S. Basin)
Stratification period	May-December

Fig. 1. Map of Lake Biwa and the location of sampling stations Ie-1 and Nb-5

Methods for chemical analysis

Total-phosphorus, TP; a fixed amount of unfiltered sample (200 ml) was taken into a beaker and dried on a hotplate to dryness, followed by both incineration with 2 ml of 60% $HClO_4$ and quantitization of phosphorus contents according either to the heteropoly blue spectrophotometric method at 700 nm (APHA, AWWA, WPCF 1965) for HRG-observation and or to the indirect spectrophotometry at 545 nm (Hori et al. 1982) for PHOS-, SIAL- and NUTR-observation. The P (A) thus evaluated was the same as the so-called total-phosphorus, TP.

KjN (A); a fixed amount (100 ml) of unfiltered sample was subjected to Kjeldahl digestion (APHA, AWWA, WPCF 1971a) and the resulting ammonia-N was analyzed spectrophotometrically at 660 nm according to the indophenol method (Koyama et al. 1976).

NO_3-N; a fixed amount (100 ml) of unfiltered sample was dried in a beaker and the nitrate in the residue was quantitized spectrophotometrically at 410 nm (APHA, AWWA, WPCF 1971b) by using phenoldisulphonic acid.

Total-nitrogen, TN; as the sum of KjN (A) and NO_3-N, TN was evaluated. As usual, TN was nearly the same as KjN (A), since the NO_3-N contents were relatively low compared with KjN(A).

Results and Discussion

Evaluation of the averaged rate of increase in total-phosphorus, TP

TP was observed periodically over the 540 months at the surface of Ie-1. As a representative result, the TP at Ie-1 (0 m) is plotted against time in Fig. 2. The variation in TP could be approximated to a straight line by least-square regression analysis

$$\mathrm{TP}\ (\mu\mathrm{M}) = 2.4 \times 10^{-4} \cdot t + (0.2 \pm 0.1) \text{ with } r^2 = 0.06 \qquad (1)$$

Here, t denotes the time in months elapsed since May 1963. Although the slope of eqn (1) suggests an averaged increase of TP at a rate of $2.4 \times 10^{-4}\,\mu$M per month, the certainty of the regression expressed as $r^2 = 0.06$ is rather low.

Apart from such a regression analysis, during that observation period it was pointed out by Negoro (1967), that in 1967 (t = 44) unusual species of micro-cyanobacteria increased the populations to 1.3×10^6 (*aphanothece clathrata*) and $2.2 - 14.8 \times 10^6\ \mathrm{m}^{-3}$ (*Lyngbya limnetica*). In May 1969 (t = 72), a musty odor was first noticed in the tap water which was supplied from Lake Biwa to Kyoto city. Actually, in 1979 (t = 192), a Shiga prefecture ordinance on prevention of eutrophication in Lake Biwa was enacted as a countermeasure to freshwater red tides. Comparing the phosphorus concentrations before and after the appearance of the musty odor, Koyama (1974) pointed that the amplitude of the TP variation became noticeable. Even when such limnological and social events are taken into consideration, it is still difficult to deduce any general trends from eqn (1) or Fig. 2.

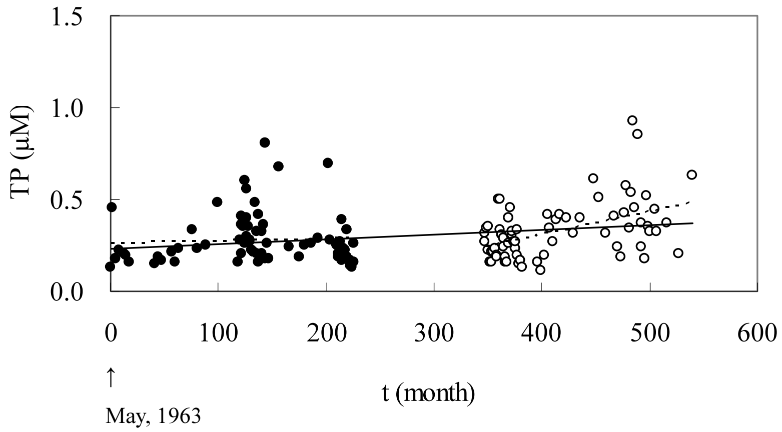

Fig. 2. Variation of TP with time at Ie-1 (0 m). The regression line calculated over the whole stage [eqn (1) in the text] is shown by a solid line, and those calculated separately over the first (whose data points are shown by closed circles, ●) and the second stages (by open circles, ○) are also shown by dotted lines, respectively

Hori et al. (1992) introduced a new parameter of Gross Weight of Phosphorus (GWP) for the statistical treatment of such a widely varying TP. The parameter was calculated by integrating the vertical profile of TP concentrations along the depth from the surface (0 m) to the bottom (70 m) at Ie-1. In principle, the result of the integration corresponds to the GWP, which is contained in a water column with dimensions of 0.1 m × 0.1 m × 70 m (with a volume of 700 dm^3) being assumed at Ie-1. The GWP values thus evaluated are plotted against time over the whole stage in Fig. 3 (A), where the regression line is given as

$$\mathrm{GWP}\ (\text{mg-P in } 700\ \mathrm{dm}^3) = 6.4 \times 10^{-3} \cdot t + (5 \pm 3) \text{ with } r^2 = 0.13 \qquad (2)$$

From the slope of the line, it is known that the increasing rate of TP in the Northern Basin can be estimated as 6.4×10^{-3} mg-P per month per a 70 m-column of 700 dm^3. Here, r^2 improved from 0.06 to 0.13. Only for convenience, in the present paper the parameter GWP was converted further to TP*avg,* which denotes the averaged concentration of TP in terms of μM through the full depth of

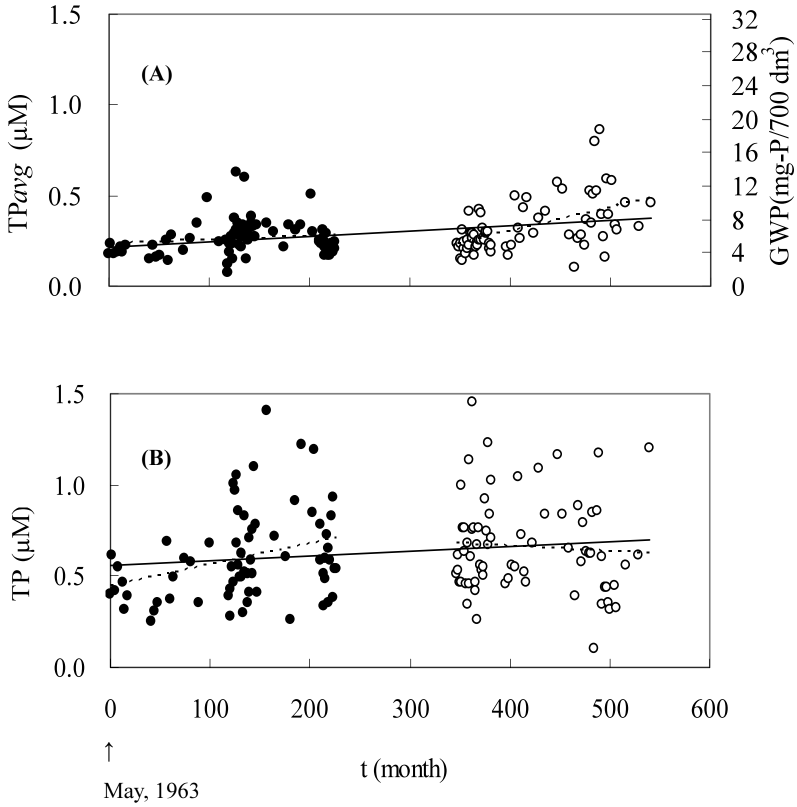

Fig. 3. (A) Variation with time of TP*avg* at Ie-1 (0–70 m) and (B) that of TP at Nb-5. As a reference, the GWP axis is also indicated on the right side of Fig. 3 (A). The conversion from GWP to TP*avg* is given by eqn (3) in the text. The regression lines calculated over the whole stage are shown by solid lines, and those calculated separately over the first and second stages by dotted lines, respectively. Open and closed circles are as in Fig. 2

0–70 m at Ie-1. Obviously, the mutual relation between GWP and TP*avg* should be given as

$$\mathrm{TP}avg\ (\mu\mathrm{M}) = \mathrm{GWP}\ (\mathrm{mg}) \times 10^{3} / 10^{-6} \times 700 \times 31.0 \qquad (3)$$

As can be seen from the calculation process of TP*avg,* the highly, non-uniform distributions which occur in every stratification period (see Table 1) at Ie-1 are made uniform mathematically. Thus, the seasonal effect on the vertical distribution of TP was taken into account. Although extraordinarily high values (t = 99, 127, 136, 202, 485 and 489) and low values (t = 118, 119 and 465), which may arise from the non-uniform, horizontal distribution of phosphorus are included, all the data points in Fig. 3 (A) can be approximated to a straight line as

$$\mathrm{TP}avg\ (\mu\mathrm{M}) = 3.0 \times 10^{-4} \cdot \mathrm{t} + (0.2 \pm 0.1) \text{ with } \mathrm{r}^2 = 0.13 \qquad (4)$$

Here, t again denotes the time in months elapsed since May 1963. On average, it can be said that TP at Ie-1 in the Northern Basin has been increasing at a rate of 3.0×10^{-4} μM per month; in other words, the amount of phosphorus contained in the water phase of whole lake (2.75×10^{13} dm^3), (see also Table 1) has increased by 3.0 ton yr^{-1}.

In more detail, the increasing rate of TP*avg* can be separately calculated for the early, first stage and the recent, second stage. The respective rates are 2.2×10^{-4} μM per month with an r^2 value of 0.02 and 1.3×10^{-3} μM per month with an r^2 value of 0.28, and the latter rate is 6 times larger than the former irrespective of the strong actions taken against eutrophication. As will be discussed later in comparison with the corresponding rates at Nb-5 in the Southern Basin, the recent increase at Ie-1 can better be attributed to supply from a coastal region where phosphorus compounds accumulated and also from the Southern Basin (Hori et al. 1992). Thus, the monthly rate seems to give a general view of the past, present and future trends in eutrophic states of Lake Biwa.

Increasing rates of TP at Nb-5

The variation in TP at the surface of Nb-5 is plotted against time in Fig. 3 (B), and the data points are approximated to a straight line expressed as

$$\mathrm{TP}\ (\mu\mathrm{M}) = 2.4 \times 10^{-4} \cdot \mathrm{t} + (0.6 \pm 0.2) \text{ with } \mathrm{r}^2 = 0.02 \qquad (5)$$

Since the water depth at Nb-5 is around 3.5 m and stratification hardly occurs throughout the year, the concentration profile of TP along this depth is mostly uniform and, in consequence, the concentration at the surface may well represent the average concentration of TP to the full depth at Nb-5.

Also, from the comparison of eqns (4) with (5) or from the comparison of Figs. 3 (A) with 3 (B), it is known that the regression lines over the whole stage at Ie-1 and Nb-5 have virtually the same slope, but different intercept values. The latter fact means that the TP at Nb-5 (0.57 μM) is 2.5 times higher than that at Ie-1 (0.23 μM). Such a high concentration of TP in the Southern Basin can be attributed to higher activity of human beings in the surrounding urbanized area, secondly to the geological nature of the Southern Basin having a shallow and gradual shaped-bottom (Oonishi and Imasato 1974; Kumagai et al. 1997), and thirdly to the high population level of seston (Tezuka 1985).

In contrast to such differences between the TP levels, however, the rates of increase at the two stations during the 540 month period resemble each other ($2.4 - 2.4 \times 10^{-4}$ μM per month). In order to examine the rate of increase in more detail, the data points over the whole stage were divided into two groups, i.e., those in the first and the second stages, the regression lines were calculated separately and the results were superimposed on the corresponding figures. It is seen from the regression lines that at Nb-5, the slope for the first stage (1.1×10^{-3} μM per month) is higher than that of Ie-1 (1.3×10^{-4} μM per month) for the same duration, indicating that the Southern Basin was strongly influenced by the input of phosphorus in the first stage. In the second stage, however, the lower slope (3.0×10^{-4} μM per month) at Nb-5 than that at Ie-1 (1.3×10^{-3} μM per month) was evaluated. This suggests that the restoration of water quality regarding TP may proceed much faster in the Southern Basin than in the Northern Basin.

One of the reasons for restoration of water quality, regarding total phosphorus concentration in the Southern Basin, proceeded much faster in the Northern Basin was due to the improvement of the water quality of tributary streams entering the Southern Basin. The quality was much better at the beginning of the 1990s, compared with the 1980s (Hamabata and Kobayashi 2002). In fact, the mean concentration of total phosphorus contained in the river water inflowing to Southern Basin during 1978–1999 was decreased from 455 to 93 mg/l (Shiga Prefecture Government Environmental Policy Division 2001). Whereas, total phosphorus concentration in the river water flowing to the Northern Basin during 1996–2007 was increased or stable (White Paper Shiga prefecture 2008).

Evaluation of averaged rate of total-nitrogen, TNavg

According to the same manner as applied to TP, firstly the Gross Weight of Nitrogen (GWN) was calculated at Ie-1 (0–70 m) and then converted to TN*avg*. Although extraordinarily high ($t = 516$) and low ($t = 14, 60, 468$ and 483) values are included, all the data points are plotted against time in Fig. 4 (A) and approximated to a straight line expressed as eqn (6).

$$\mathrm{TN}avg\ (\mu\mathrm{M}) = 6.2 \times 10^{-3} \cdot t + (20 \pm 7) \text{ with } r^2 = 0.03 \qquad (6)$$

As is known from the slope of the line, the averaged rate of TN at Ie-1 over the whole stage is 6.2×10^{-3} μM per month. In the first and the second stages, the respective increasing rates of TN*avg* were 1.5×10^{-2} μM per month with an r^2 value of 0.03 and 8.8×10^{-2} μM per month with an r^2 value of 0.16.

The variation in TN at Nb-5 is plotted against time in Fig. 4 (B), and all the data points in the figure can be approximated to a straight line as

$$\mathrm{TN}\ (\mu\mathrm{M}) = -1.2 \times 10^{-2} \cdot t + (29 \pm 11) \text{ with } r^2 = 0.04 \qquad (7)$$

Equation (7) shows that in the Southern Basin represented by sampling station Nb-5, TN decreased at a rate of minus 1.2×10^{-2} μM per month, with increasing rates in the first and second stages of plus 3.9×10^{-2} μM per month with an r^2 value of 0.05 and plus 5.2×10^{-2} μM per month with an r^2 value of 0.04, respectively. The decline in TN in the whole stage at Nb-5 was principally the result of a marked decline in KjN (A), but not of NO_3-N.

By comparing eqn (6) with (7) or Fig. 4 (A) with 4 (B), it is found that the slope for the first stage at Nb-5 is 2.6 times higher than that of Ie-1; in other words, the Southern Basin was strongly

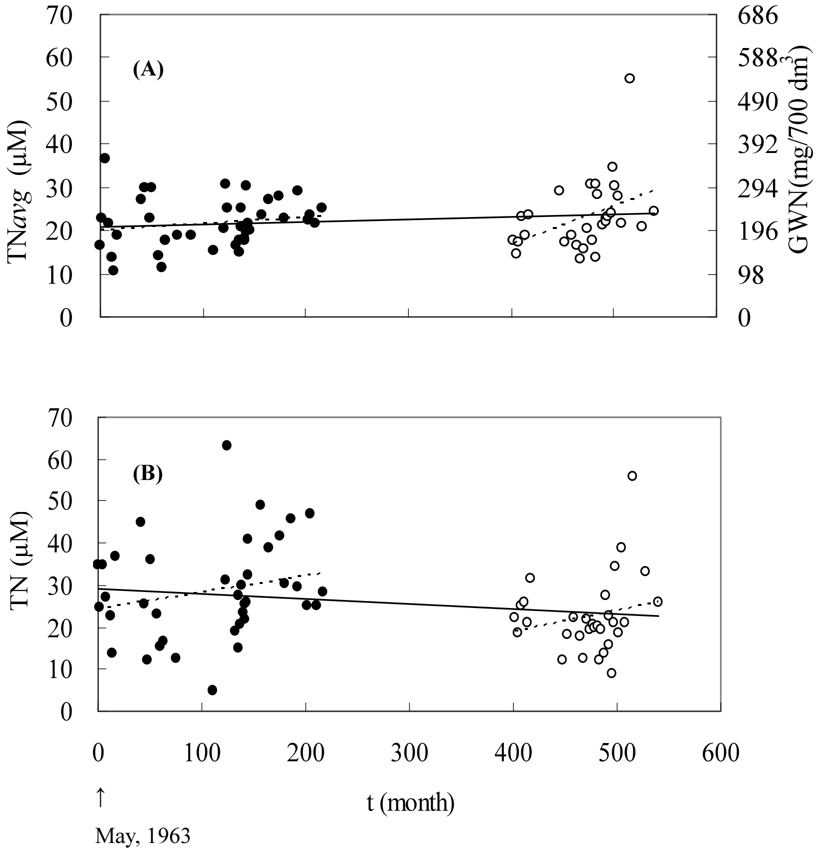

Fig. 4. (A) Variation with time of TNavg at Ie-1 and (B) that of TN at Nb-5 (0 m). As a reference, the GWN axis is also indicated on the right side of Fig. 4 (A). Open and closed circles are as in Fig. 2

influenced by the input of nitrogen compounds over the period. In the second stage, however, the lower slope at Nb-5 (5.2×10^{-2} μM per month) than at Ie-1 (8.8×10^{-2} μM per month) suggests that water quality regarding TN is improving faster than in the Northern Basin. In fact, Hamabata and Kobayashi (2002) reported that the water quality was much more improved in the Southern Basin after 1994 ($t = 368$) due to the development of macrophyte. Faafeng and Mjelde (1998) postulated the following mechanism by which macrophyte may maintain a clear-water state; competition with phytoplankton for nutrients, competition with epiphytic algae for light, and the release of allelopathic substances. In view of these relationships, the recovery of macrophyte presumably played an important role in the recent improvement.

Variations in TN/TP ratio at Ie-1 and Nb-5

Phytoplankton can store phosphorus and nitrogen intracellularly (e.g. polyphosphate bodies, cyanophycin granules in cyanobacteria) which can be utilized for growth after external inorganic dissolved N and P are exhausted. Thus, TN and TP are better estimators of the nutrient pool utilized for algal growth than their respective dissolved fractions (Sommer 1999). Tilman (1982) suggested that cyanobacteria should typically be dominant in a lake with a low TN/TP ratio.

At Ie-1 (0 m), the mean TN/TP ratio over the whole stage was 76 (min. 14; max. 177), whereas in the first and the second stages the mean values were 87 (min. 31; max. 177) and 62 (min. 14; max. 176), respectively (see also Fig. 5A). At Nb-5 over the whole stage, the mean was 50 (min. 11; max. 190), and those in the first and second stages were 53 (min. 21; max.179) and 46 (min. 11; max. 190), respectively (Fig. 5B). The low TN/TP ratios in the Southern Basin have the capacity to support large cyanobacterial blooms (Tezuka and Nakano 1993). Even in the Northern Basin, which is considered mesotrophic, there have been summer blooms of cyanobacteria since 1994 (t = 368). From 1997 (t = 405) onwards, bloom-forming cyanobacteria have been observed more frequently in the offshore water of the Northern Basin (Kumagai et al.1999; Ishikawa et al. 2002). The mechanism proposed by Smith (1983) to link cyanobacteria dominance to such a low TN/TP ratio is that all species of cyanobacteria are better able to compete for nitrogen than other phytoplanktons when N is scarce. Therefore, when excessive P loading creates a surplus supply of phosphorus, nitrogen becomes relatively scarce and cyanobacteria are predicted to become dominant.

Smith (1983) compiled extensive data sets for several lakes in which cyanobacteria dominance

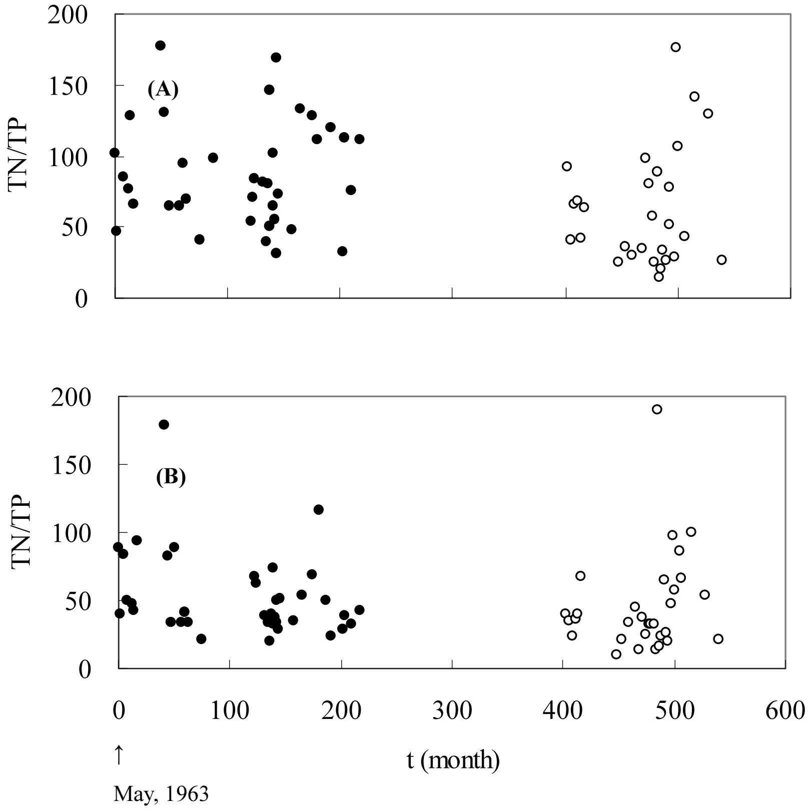

Fig. 5. (A) Variation of the TN/TP ratio with time at Ie-1 (0 m) and (B) that of Nb-5. Open and closed circles are as in Fig. 2

and TN/TP ratios varied, and showed that cyanobacteria dominance became low when the TN/TP ratios were larger than 65 (by atoms). However, when the ratio was smaller than 65, the potential for cyanobacteria dominance increased dramatically, and in the special case where lakes showed smaller ratios than 25 the dominance of cyanobacteria became mostly 100 %.

At the Northern and the Southern Basins of Lake Biwa, TN/TP ratios have been reported by several other researchers; Ichise et al. (2001) reported that during the period of 1979 (t = 192) to 1999 (t = 429), the mean of TN/TP ratios in the Northern and the Southern Basins were increased, respectively, from 67 to 104 and from 29 to 48, with the decrease of population of phytoplanktons. According to Environmental White Paper Shiga prefecture (2001), the mean of TN/TP ratios at the Northern and the Southern Basins during 1993–2000 were found to be 91 and 43, respectively. Nakanishi et al. (2001) reported that the mean in the respective Basins were 77 and 41 during 1980–1992 and 134 and 99 during 1993–2000 and pointed that the higher TN/TP ratios observed at the Northern than the Southern Basin and such high TN/TP ratios compared to the other lakes seemed to be one of the limnological characteristics of Lake Biwa.

Recently, more high TN/TP ratios were observed at the Southern Basin due to decrease of the total phosphorus concentration in inflow river waters and increase of the macrophyte growth in the basin.

Conclusion

Based on a series of observations of BST, HRG, SIAL, and NUTR conducted at Lake Biwa, the variations with time of the TP, TN and TN/TP ratios have been approximated into straight lines by using least-square regression analysis. As a result, it was found that TP was still increasing at a similar rate in both the Northern and Southern Basin, whereas TN in the Northern Basin increased slightly and TN decreased in the Southern Basin. Particularly, in the recent, second stage, the decreases in TP and TN and the slight increase in TN/TP at Nb-5 were noted as first signs of the restoration of water quality in the Southern Basin. Based on 540 months (45 years) of limnological records for Lake Biwa it is suggested that the water quality of the Southern Basin is improving much faster than in the Northern Basin.

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