

琵琶湖の環境化学的研究 (第1報)

全リン及び全リンの形態別組成を指標とする琵琶湖の水圏環境評価

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要 旨

琵琶湖水中の全リンを5形態に分けて観測し、この結果に基づいて湖水の栄養状態の変動と推移を研究した。観測したリンの形態P(I)~P(V)の中で、P(III)とP(V)は注目すべきものであって、それらの比P(V)/P(III)は湖水の栄養状態によく対応した。ここで、P(III)はトリリン酸、フィチン酸、フミン酸などの口過可能な化合物に含有されるリンの形態、また、P(V)は生物体やその他の懸濁体に含有されるリンである。一方、全リンそのものの観測値を用いると湖水環境の長期的な変化を捉えることができる。水深70mの観測地点Ie-1に10cm×10cm×70mの単位水柱を想定し、この中の全リン量すなわち総リン量(GWP)を計算する。このGWP値の時間に対する変化を調べると、湖全体で進行する長期的なリンの循環の様子を捉えることができる。この手法を1963~81年の観測結果に適用し、琵琶湖湖水相における19年間のリンの増加率を得た。この増加率は、湖周辺からこの期間に流入したはずのリン量から予測される値とくらべると、極めて低いことが解った。この不一致は、湖に流入負荷されたリンの殆どの部分が活発な基礎生産によって湖岸部で不溶化して沈積し、残りの僅かな部分が湖心部のIe-1にプランクトンの流れとなってもたらされたことによる、と解してよい。

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Environmental Chemistry of Lake Biwa.

Part 1, Characterization of Lake Biwa Based on the Total-phosphorus and Its Fractional Composition

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Summary A method to measure the fractional composition of the so-called total-phosphorus in lake waters has been established and applied for monitoring the eutrophic nature of Lake Biwa. Among the five fractions of total-phosphorus measured, P(III) and P(V) were so noticeable that the P(V)/P(III) ratio corresponded to the environmental condition of the lake. Here, P(III) denotes phosphorus in filtrable compounds such as triphosphates, phytates, and humic acid, and P(V) denotes phosphorus in unfiltrable substances like living organisms or in other suspended substances. In parallel with the fractionation of the total-phosphorus, gross weight of phosphorus (GWP), the amount of phosphorus contained in a unit column of water having a dimension of 10 cm × 10 cm × 70 m at Ie-1 sampling site, was calculated. The variation of GWP against time in the period 1980–82 revealed a circulation profile of phosphorus which occurred in the northern basin of the lake throughout the years. A statistical treatment of the GWP values observed in the period 1963–81, gives the average rate of increase of phosphorus during the 19 year term. The rate of the increase, however, was much lower than that estimated from the phosphorus which might have been brought in by human activities. This discrepancy suggests that most of the phosphorus was fixed rapidly at the coast where phytoplankton and algae propagate highly while remaining phosphorus migrated as a stream or as a cloud of plankton into sampling site Ie-1 being located in the pelagic part of the lake.

INTRODUCTION

Self-multiplication, heredity, and energy metabolism are the essence of life. Biochemistry and molecular biology currently describe life as a unique assemblage of chemical reaction systems. In these systems, interestingly, phosphorus participates in the most fundamental reactions, forming compounds which constitute cell membranes, genes, and energy-rich materials. Nevertheless, it is difficult to propose a hypothesis for the chemical role of phosphorus in chemical evolution, or the development of life.

The occurrences of phosphorus and sulfur in the earth's crust, *viz.*, the Clarke numbers, are 0.08 and 0.06, respectively, and relatively close to each other. In contrast, their occurrences in seawater

are extremely different, the content of phosphorus (0.06 ppm) being much lower than that of sulfur (900 ppm). The large difference in seawater is due mainly to the high activity of living organisms toward phosphate rather than sulfate ion. It has been observed commonly in every aquatic environment that where phytoplankton propagate the concentration of phosphorus in the orthophosphate form is lower than 0.1 ppb. Such biological preference for phosphorus rather than sulfur and the chemical interpretation for that preference is of great interest in many fields of science.

In limnology, the eutrophic conditions of lakes are classified according to their phosphorus contents; lakes containing phosphorus lower and higher than 20 ppb are termed as oligotrophic and eutrophic lakes, respectively. According to this

classification, Lake Biwa was apparently an oligotrophic lake in 1935¹⁾. In recent years, however, the phosphorus content has varied between 10 and 40 ppb, and the lake is now considered in various respects to be eutrophic. Eutrophication of Lake Biwa was undoubtedly caused by human activities around the lake; and similar phenomena have been recognized in various lakes and rivers and in some parts of the oceans of the world. As a subject of study by environmental chemists, however, Lake Biwa provides an unique opportunity because many chemical components, including phosphorus, have been monitored continuously through the periods before and after eutrophication of the lake.

In the present paper, we shall analyze the pattern of variation of total-phosphorus content in Lake Biwa as a function of time, and we shall propose criteria useful for evaluation of concurrent as well as future environmental conditions of the lake.

GEOLOGICAL AND LIMNOLOGICAL CHARACTERISTICS OF LAKE BIWA

Lake Biwa is located at 35°15'N latitude and 136°05'E longitude. The lake, which developed about five million years ago²⁾, is one of the oldest lakes in the world. Many kinds of biota, including more than 1,000 species, live in the lake; indigenous species are abundant. The lake is geometrically divided into two parts, the northern and southern basins³⁾. The northern basin is the main basin, and has a surface area of 616 km² (91.4%) and a capacity of 27.3 km³ (99.3%), with average and maximum depths of 44 m and 104 m. The southern basin has a surface area of 58 km² (8.6%) and a capacity of 0.2 km³ (0.7%), with average and maximum depths of 3.5 m and 8 m. The watershed area of the lake is 3,848 km² and covers essentially the whole area of Shiga Prefecture. The total precipitation on the watershed, 7.31 km³ yr⁻¹, flows mainly into the northern basin and down to the southern basin, and then flows out through the River Seta and canals at the respective rates of 5.1 and 0.8 km³ yr⁻¹ (for geography, see also Fig. A1 in Appendix). The residence times of water in the northern and southern basins are estimated to be 5.5 yr and 0.04 yr, respectively.

Lake Biwa is a subtropical lake. Thermal stratification typically occurs in the northern basin from

May to December, and convection of the lake water occurs regularly from January to April. In the south basin, however, there is little stratification and the vertical mixing of water occurs throughout the year.

The human population in the watershed area of the lake began to increase significantly from 1960 and has been increasing very rapidly in recent years. The annual industrial production has also been increasing along with the population. A local law set in July of 1980 now regulates the use of phosphate-containing detergents in Shiga Prefecture in order to retard the eutrophication of Lake Biwa. Today, the northern basin and southern basins seem to be mesotrophic and eutrophic, respectively, in character.

SETTING OF OBSERVATIONS

In a 1935 study of Lake Biwa water, concentration of sodium, calcium, magnesium, sulfate, chloride, carbon dioxide, silicate, and nitrogen, pH, conductivity, COD, and residue on evaporation were determined by Yoshimura¹⁾.

In August, 1962, and January, 1963, as part of a project for an integrated assessment of animate resources in Lake Biwa (Biwako Seibutsu-shigen Tyosa: BST), the water research group headed by Fujinaga carried out a survey of the chemical components of the lake. It was the first and the most elaborate survey ever performed on the lake. The BST-group collected water samples from more than 100 locations in 4 km × 4 km square areas sectioned and designated by lines of longitude and latitude (Fig. A1 in Appendix). The results of the BST-observations are compiled in a report⁴⁾.

Following the BST-observations, Fujinaga and his coworkers, Hydrosphere Research Group (HRG), made a new series of observations from May, 1963, until June, 1981. Taking samples several times a year at four representative sampling sites, *i.e.*, at Ie-1 in the northern basin and at Nb-2, Nb-5, and Na-3 in the southern basin, they measured the levels of silicate, phosphate, nitrate, nitrite, ammonia, Kjeldahl-nitrogen, chloride, calcium, magnesium, sodium, and potassium as well as pH, water temperature, COD, and dissolved oxygen. The accumulated data of the HRG-observations are compiled in book form⁵⁾, which is the source of the data pertaining to phosphorus levels

cited herein (Table A1) and used for the discussion below.

One of the authors (T. H.) observed phosphorus levels at locations Ie-1, Nb-2, Nb-5, and Na-3 during the period 1980–1982. Samples were taken also at locations Kc-3(KIDO), Lc-4(WANI), Lc-3(OHASHI), and Mb-3(OGOTO) in order to examine the horizontal distribution of phosphorus from the northern to the southern basins. The results are compiled in Table A2 entitled "PHOS-observation".

METHODS

Classification of Phosphorus Compounds Distributed in Lake Water

Various forms of phosphorous compounds are known to be present in lakes. Most of these compounds, however, are not yet precisely specified, but are usually classified into several groups according to their chemical and physical properties. The conventional classification is shown in Fig. 1. The so-called total-phosphorus is divided first into two large groups: particulate-phosphorus and soluble-phosphorus. The former group consists broadly of inorganic phosphorus and organic phosphorus while the latter consists of dissolved organic phosphate (DOP), dissolved inorganic phosphate (DIP), and orthophosphate. The quantification of these fractions, however, is tedious and time con-

suming, and have been performed only for some special or limited samples.

For convenience, in the present work the total-phosphorus was divided into five fractions as P(I)-P(V). In this classification, P(I) denotes orthophosphate-phosphorus; P(II), orthophosphate-phosphorus released from suspended substances by the action of 0.5 M HCl; P(III), filtrable inorganic and/or organic phosphorus forms (other than orthophosphate-phosphorus) like triphosphates, phytates, and humic acid; P(IV), unfiltrable phosphorus made filtrable by the action of 0.5 M HCl; P(V), unfiltrable phosphorus incorporated in living organisms or bound firmly to suspended substances. These new classifications are enclosed by dashed lines (---) and superimposed on Fig. 1 indicate their correspondence to the classifications used conventionally.

Pre-treatment of Samples for Differentiation and Differentiative Determination of P(N), P(F), P(A), P(FA), and P(HFA)

The principle of sample treatment⁶⁾ is outlined in Fig. 2. Five 35 ml-aliquots from each water sample are treated separately in preparation for determination of phosphorus; Procedure-N (non-treatment): addition of 3.5 ml of 6 M HCl; Procedure-F: filtration with a membrane filter having a pore size of 0.40 μm , followed by the addition of 3.5 ml of 6 M HCl; Procedure-A: incineration with 2 ml

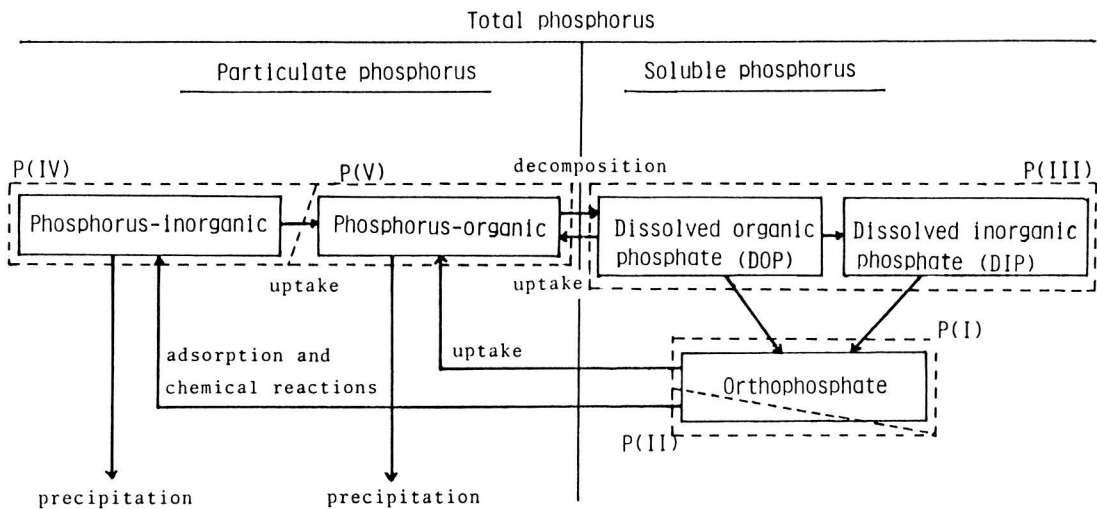


Fig. 1. Classification of phosphorus compounds distributed in natural waters. The so-called total-phosphorus is classified into five fractions, P(I)-P(V), enclosed with dashed lines and superimposed on the classifications used conventionally.

of HClO_4 ; Procedure-FA: filtration followed by the incineration as above; Procedure-HFA: acidification with 3.5 ml of 6 M HCl, followed by Procedure FA (filtration and incineration). Residues from the incinerated samples are dissolved by warming with 3.5 ml of 6 M HCl and allowed to stand at room temperature for 2 h.

All the samples thus treated were transferred to 50 ml-centrifuge tubes and prepared for indirect spectrophotometric measurement of phosphorus by the following procedure: To each pre-treated sample is added 3.5 ml of 0.12 M Na_2MoO_4 solution. The mixture is shaken for 5 min with 7 ml of an 11:9 (v/v) mixture of 4-methyl-2-pentanone and cyclohexane. After centrifugation, a 5 ml-portion of the organic extract is transferred to a 10 ml-centrifuge tube and shaken for 1 min with 2.5 ml of 1 M NaOH. To the mixture is added, successively, 1.5 ml of 2.5 M HCl, 1 ml of 0.2% pyrocatechol sulfonphthalein-aqueous solution, and 0.5 ml of 0.5% trioctylmethylammonium chloride-ethanol solution, followed by shaking again for 1 min. After centrifuging for 3 min, the absorbance

of the organic extract at 545 nm is measured and the phosphorus content is determined using an absorbance-phosphorus calibration curve. Phosphorus ranging from 0.3 to 120 ppb in the original lake water samples can be determined by this procedure⁷⁾. The relative standard deviations in six determinations of phosphorus at 0.3, 1.8, and 6.2 ppb concentrations are 10, 5, and 3%, respectively.

The phosphorus concentrations thus determined after pre-treatment procedures N, F, A, FA, and HFA are noted temporarily as P(N), P(F), P(A), P(FA), and P(HFA), respectively.

RESULTS AND DISCUSSION

The Fractional Composition of Total-phosphorus and the Environmental Condition of Lake Biwa

The Fractions Noticed.—The analytical results of P(N), P(F), P(A), P(FA), and P(HFA) can be converted to the previously mentioned new classification of fractions [P(I)-P(V)] of total-phosphorus according to the following relations⁶⁾: $\text{P(F)} = \text{P(I)}$; $\text{P(N)} = \text{P(I)} + \text{P(II)}$; $\text{P(FA)} = \text{P(I)} + \text{P(III)}$;

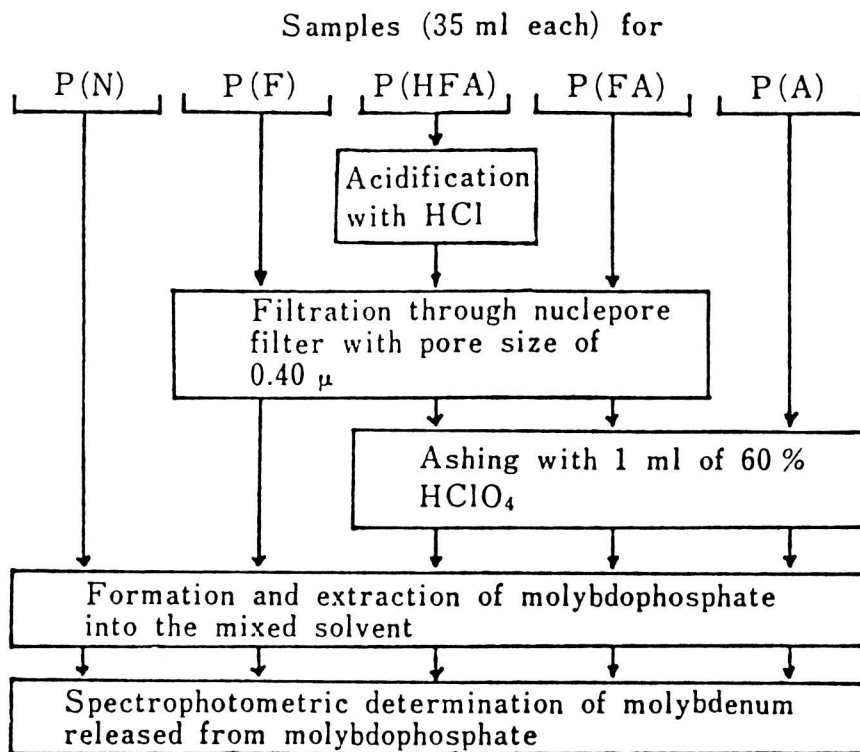


Fig. 2. General procedures for the differentiative determination of P(N), P(F), P(HFA), P(FA), and P(A).

$P(\text{HFA}) = P(\text{I}) + P(\text{II}) + P(\text{III}) + P(\text{IV})$; and $P(\text{A}) = P(\text{I}) + P(\text{II}) + P(\text{III}) + P(\text{IV}) + P(\text{V})$. As an example of the fractionation, the analytical results ob-

served for samples taken on May 14, 1981, in Lake Biwa (# PHOS-13A in Table A2), are plotted in Fig. 3. In the figure, for instance, the difference be-

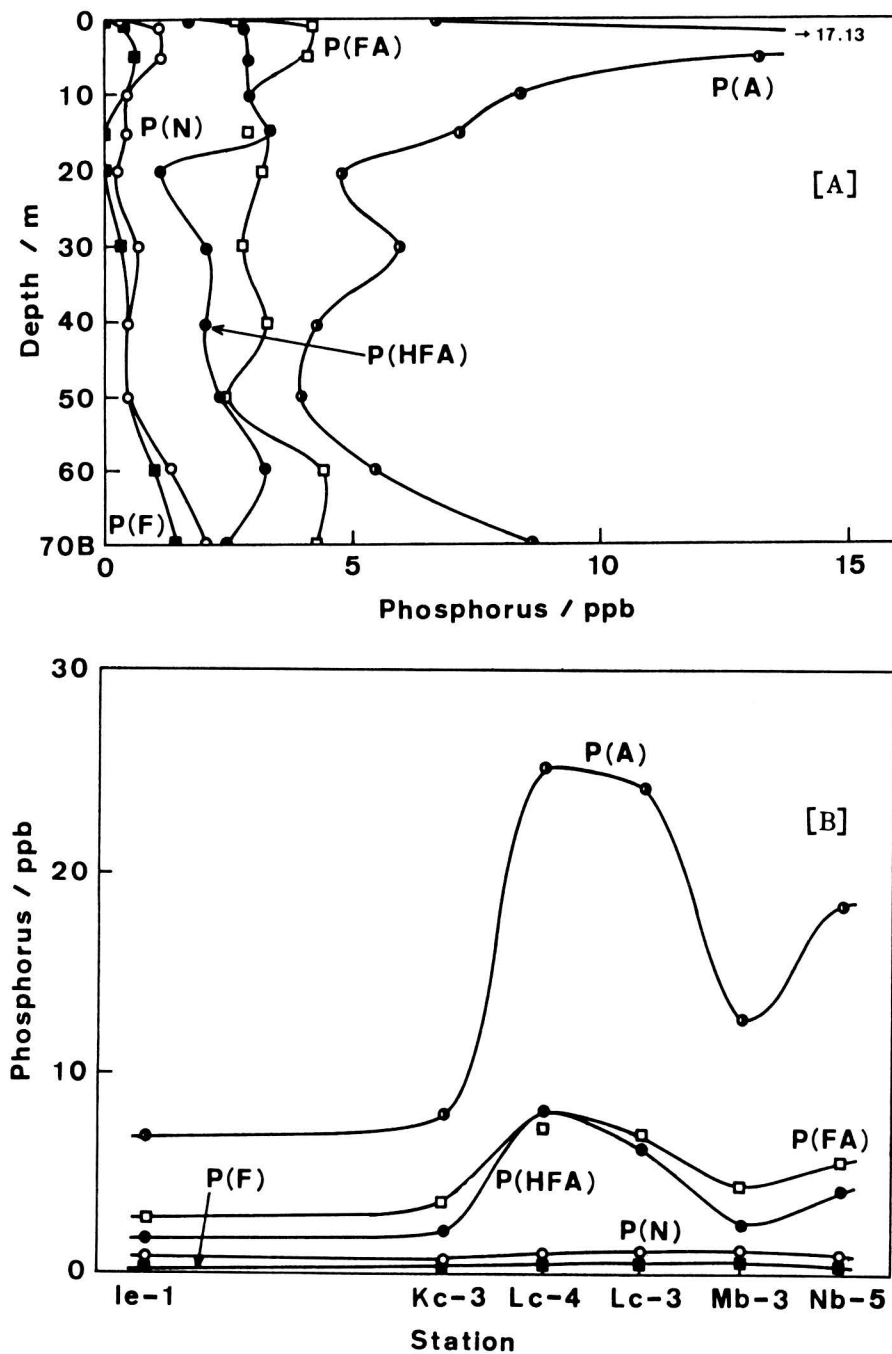


Fig. 3. Distribution profiles of phosphorus on May 14, 1981, in Lake Biwa: [A] vertical distribution profile observed at Ie-1 and [B] horizontal distribution profile from the northern basin (Ie-1) to the southern basin (Nb-5). Locations of the sampling sites are shown in Fig. A1 in Appendix.

tween curves of P(HFA) and P(A) gives the fraction of P(V) and, similarly, the difference between P(FA) and P(F) gives the P(III).

Curve P(A) in Fig. 3[A] suggests that certain phosphorus compounds accumulate markedly at depths of 1, 30, and 70 m (bottom) at sampling site Ie-1. At 1 m depth, unfiltrable P(V) is the main fraction, whereas at 70 m soluble P(I) and P(III) fractions, as well as P(V), are apparent. It appears that phosphorus is made unfiltrable by the primary production at the surface layer and, simultaneously, phosphorus products also precipitate on the bottom and degrade to soluble fractions. The presence of P(I), orthophosphate-phosphorus, at the bottom layer indicates that bottom degradation

takes place to an appreciable extent. As discussed in the later part of this paper, however, amounts of phosphorus insolubilized at the surface exceed that solubilized at the bottom and, consequently, the amounts of phosphorus held in the aqueous phase are decreased, overall, in warm seasons. The relatively small maximum of P(A) appearing at the middle layer (at 30 m depth) is due to the unfiltrable P(V) fraction produced at the surface layer and trapped temporarily on the cooler and more dense water lying below the thermocline. Soluble fractions are also noticeable at the thermocline, indicating that degradation of P(V) proceeds extensively at that depth as well as at the bottom.

The seasonal variations of P(I), P(III), and P(V)

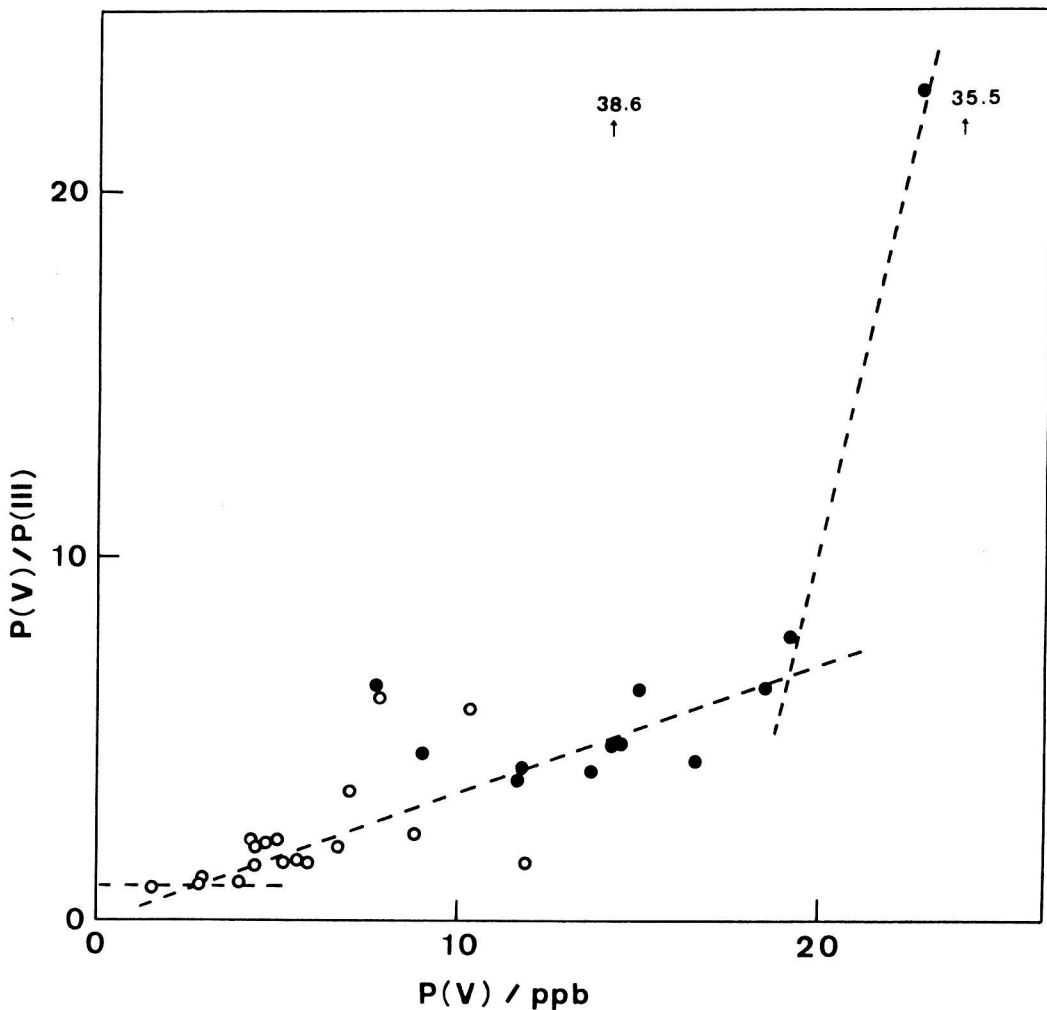


Fig. 4. Correlation between the P(V)/P(III) ratio and the P(V) fraction, observed at Ie-1, 5 m depth (○), and at Nb-5, 0 m depth (●), during the period 1980–1982.

fractions observed in the years 1980–81 at location Ie-1 are featured in Figs. A2-A5. Although the real forms of P(III) and P(V) are not yet known, the relative abundances of P(III) and P(V), in particular, seem to be related to the eutrophic condition of the lake.

The P(V)/P(III) Ratio and the Primary Production.—Growth and decay of various species of plankton are observed in the surface layers at locations Ie-1 and Nb-5 throughout the year. In these layers, the fractions of P(III) and P(V) are related to each other. That is, when P(V) accumulates due to primary production, P(III) decreases markedly. The P(V)/P(III) ratio increases as a function of P(V). P(V) itself is a practical measure of the degree of primary production in the surface layers. Figure 4 shows a plot of P(V)/P(III) against P(V). As suggested earlier⁸⁾, the P(V)/P(III) ratio depends on P(V) in three different ways; when P(V)

< 3 ppb, the ratio remains almost constant, whereas in the 5–20 ppb range, it increases linearly with P(V). The slope of the linear relation rises sharply at P(V) > 20 ppb, where extraordinary propagation of plankton occurs. The P(V)/P(III) ratio is thus an indicator of the concurrent situation of the lake: the oligotrophic and eutrophic natures of the lake are associated, respectively, with conditions P(V)/P(III) ~ 1, P(V) < 3 ppb and with conditions P(V)/P(III) > 7, P(V) > 20 ppb.

Environmental Characteristics of Lake Biwa Viewed from the Variation of Total-phosphorus

Annual Variation of Total-phosphorus.—The total-phosphorus observed periodically by HRG⁵⁾ at location Ie-1 from May, 1963 (# HRG-1), to May, 1973 (# HRG-20), is plotted against time in Fig. 5. Since Lake Biwa had an apparently oligotrophic nature in 1963 and exhibited a truly eu-

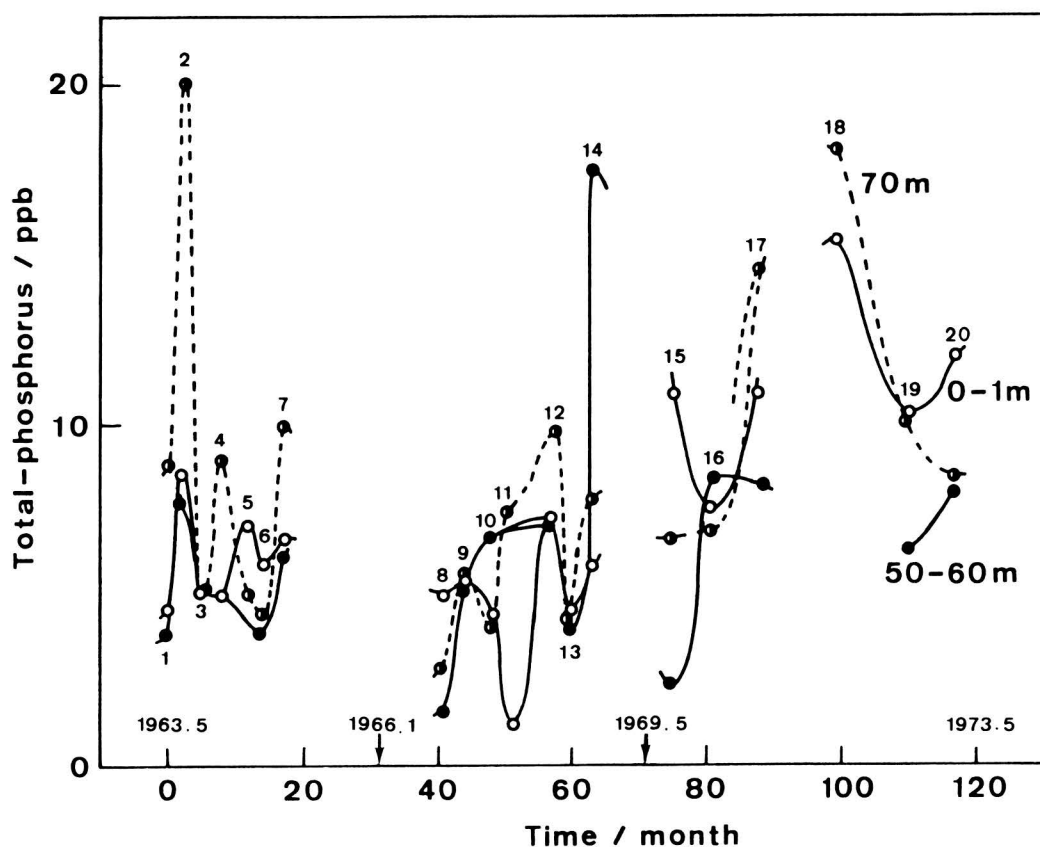


Fig. 5. Annual variation of total-phosphorus at Ie-1 during the period from May, 1963, to May, 1973. The arrows at the bottom of the figure indicate two events, the first is the propagation of unusual phytoplankton, and the second is the occurrence of malodor in the tap water of Kyoto City.

trophic nature in 1973, the variation shown in Fig. 5 should represent the eutrophication processes in terms of total-phosphorus concentration.

In 1965, it was found that unusual species of *Staurastrum dorsidentiferum* propagated occasionally in Lake Biwa⁹⁾. In May of 1969, *i.e.*, at $t=72$ months in Fig. 5, a musty odor was first noticed in the tap water supplied to Kyoto City from Lake Biwa. Comparing the phosphorus concentrations before and after the appearance of the odor, Koyama *et al.* noticed¹⁰⁾ that the amplitude of the variation of the phosphorus became noticeable after February, 1968, *i.e.*, at $t=57$ months. Until the present, no explanation for the wide variation of the phosphorus concentration has been deduced. In order to describe these variations with time, it seems profitable now to use the term "gross weight of phosphorus (GWP)" contained in a unit column of water.

Evaluation of GWP and Its Variation in a

Year.—In Fig. 3[A], the area enclosed by curve P(A) and the abscissa of the graph is calculated by integrating the phosphorus concentration along the depth. In principle, this area corresponds to the gross weight of phosphorus (4.59 mg for the present calculation) which is contained in a water column having a dimension of $10\text{ cm} \times 10\text{ cm} \times 70\text{ m}$ at sampling location Ie-1. It also indicates the amount of phosphorus remaining in the aqueous phase after the balance between the loading and the removal of phosphorus is established. In Fig. 6, the GWP value evaluated similarly for each observation performed at Ie-1 in the period from November, 1980 (# PHOS-7), to March, 1982 (# PHOS-23) is plotted against time, along with the extent of vertical circulation of the lake water. Throughout these 16 observations, the GWP varied about a mean value of 4.9 mg with a maximum amplitude of 1.5 mg, depending to some extent on the mixing of lake water. Based on the geometry

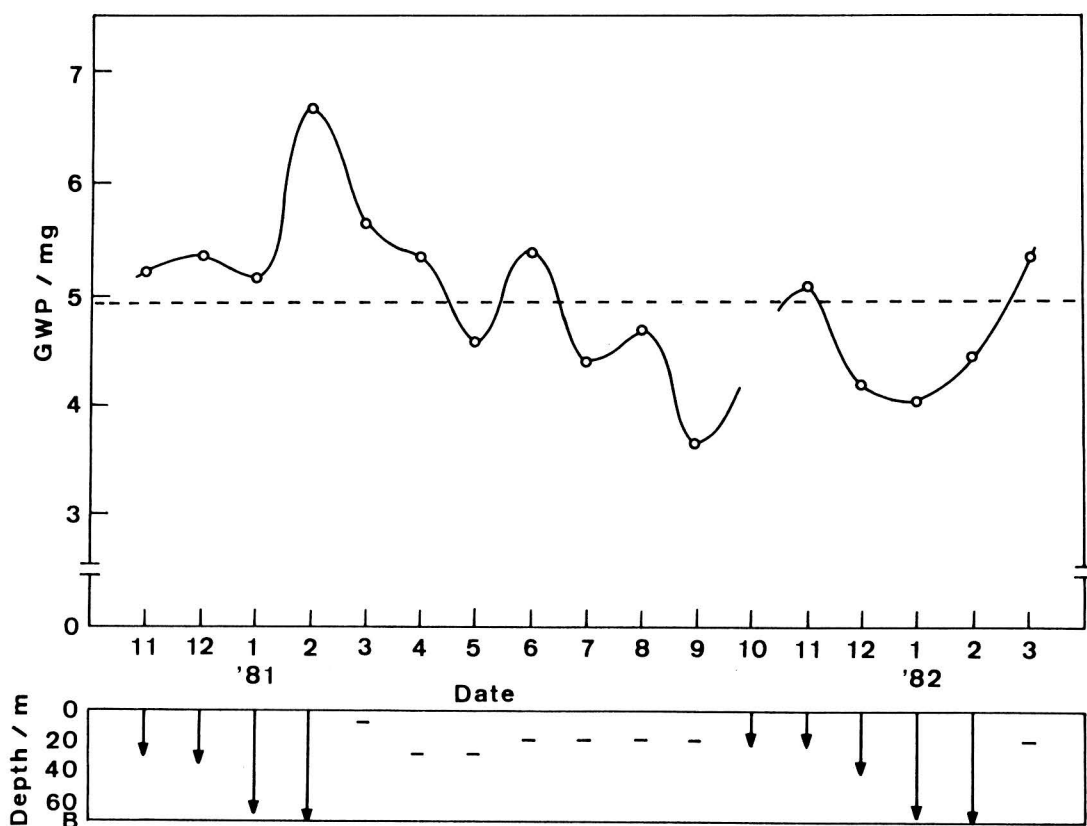


Fig. 6. Seasonal variation of GWP (upper) and the depths where circulation and stratification occur (lower), at Ie-1. Here, GWP denotes the gross weight of phosphorus contained in a unit column of water having a dimension of $10\text{ cm} \times 10\text{ cm} \times 70\text{ m}$.

of the lake, it can be said that the increment of GWP by 1 mg in the unit column of water (0.7 m^3) corresponds to an increase of 39 tons of phosphorus in the whole lake (27.3 km^3). The GWP showed a maximum in February, 1981, at the time when convection in the lake water reached the bottom; GWP decreased gradually in March through September when primary production takes place actively, and increased rapidly in early spring.

In order to examine the uneven distribution of phosphorus with depth, GWP was calculated in the shorter columns of water assumed to be at the surface (0–20 m), middle (20–40 m), deep (40–60 m), and bottom (60–70 m) layers. The corresponding GWP values are plotted against time in Fig. 7. The figure shows that in the warm season phosphorus accumulates mostly in the surface layer (○—● distance in Fig. 7), suggesting that large amounts

of phosphorus fixed in the surface layer settle into the sediments without being suspended in the hypolimnion. The phosphorous compounds in the sediments are degraded gradually during the hot season to soluble compounds. These are returned to the surface layers by violent convections which occur in the cold season. The phosphorous eluted from the sediment is mostly in the orthophosphate form. The released orthophosphate-phosphorus is estimated to be ca. 20% of the GWP; it is dispersed by water circulation during a relatively short period in early spring (see Fig. A5 for detail).

Variation of GWP over a Long Term (1963–1981).—Based on the phosphorus contents measured by HRG⁴, GWP values were calculated at location Ie-1 and plotted against time in Fig. 8. Although extraordinarily high (# HRG-18, 30, 40,

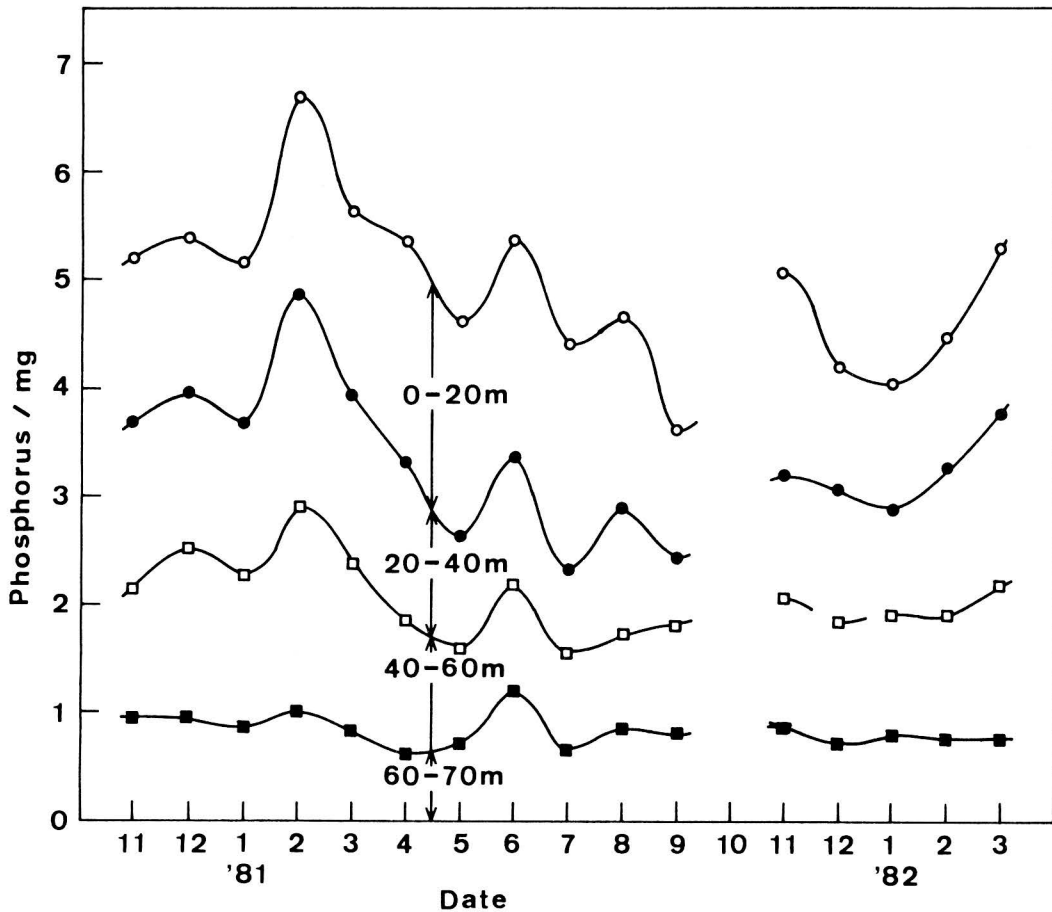


Fig. 7. Distribution of the GWP in the respective, short columns of water at the surface (○—●: 0–20 m), middle (●—□: 20–40 m), deep (□—■: 40–60 m), and bottom (■—■: 60–70 m) layers at Ie-1. In the figure, the GWP value for each short column is added to give the total GWP in the entire 70 m-column.

and 58) and low (# HRG-21 and 60) values are included, all the data points in the figure can be approximated by least squares analysis to a straight line as

$$\text{GWP/mg} = 0.015 \times T + 4.1, \text{ with } r = 0.30$$

Here, T and r denote the time in a term of months elapsed since May, 1963, and the correlation coefficient, respectively. In the same figure, the GWP values observed in 1980–1982 (# PHOS-7 through PHOS-23) are plotted for comparison. For these more recent years, the data points are clustered in a relatively small area below the HRG regression line. The regression line of Fig. 8 can be used to indicate a general trend for a eutrophic lake over a rather long period of time.

The slope of the regression line, 0.015 mg

month⁻¹, suggests that the amount of phosphorus in the unit column has increased at an average rate of 0.18 mg yr⁻¹; for the whole lake the rate of increase is 7.0 ton yr⁻¹. It should be noted that the increase of phosphorus thus estimated is much smaller than that calculated from the net amount of phosphorus presumably emptied into the lake from the watershed area. In 1981, in fact, the total amount of the phosphorus loading was estimated to be 230 ton yr⁻¹ by Kunimatsu¹¹⁾ and 144 ton yr⁻¹ by Yoshioka *et al.*¹²⁾ Assuming a uniform mixing of the loaded phosphorus with the lake water, these estimates lead to rates of GWP increase of 5.9 mg yr⁻¹ and 2.7 mg yr⁻¹, respectively, in the unit column at location 1e-1. The discrepancy (5.9 or 2.7 versus 0.18 mg yr⁻¹) suggests that most of the exogenous phosphorus brought to

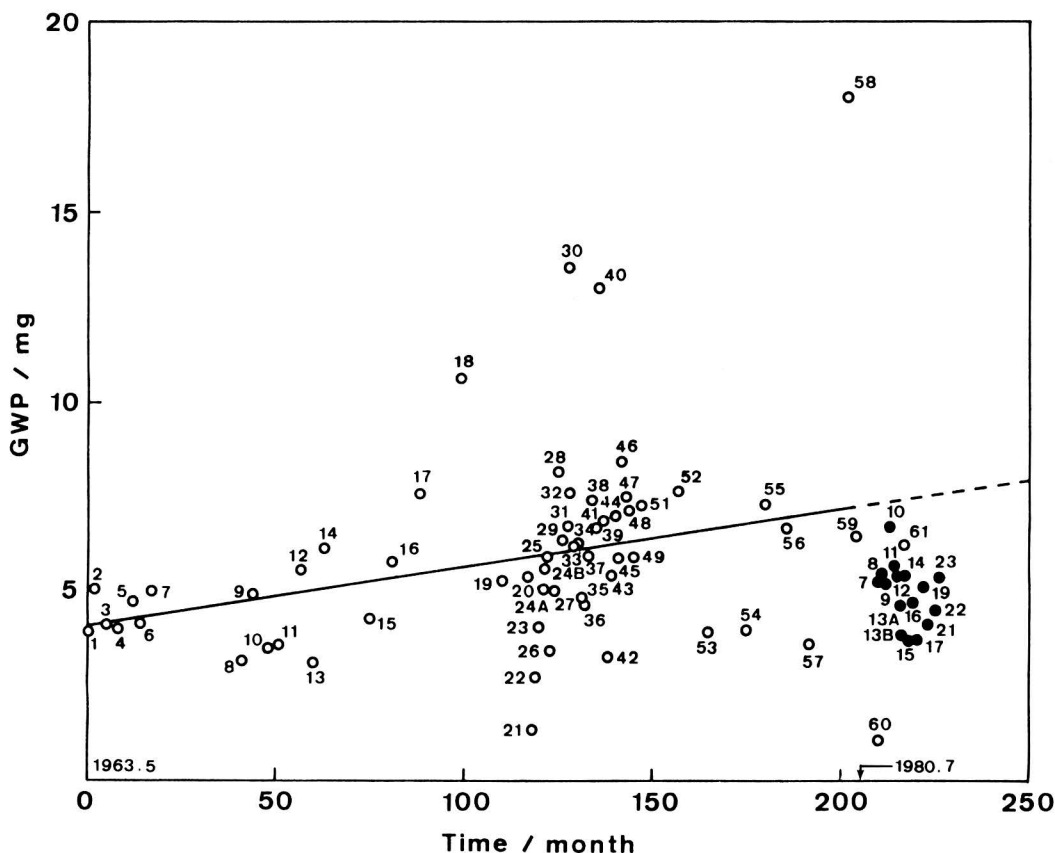


Fig. 8. Change of GWP values from 1963 through 1982. The GWP values, listed in Table A1 in the Appendix, are plotted against the time elapsed since May, 1963. The GWPs of HRG-observations (○) are approximated to a straight line by least squares analysis. The regression line is extrapolated toward the more recent years and compared with the GWPs of PHOS-observations (●). The arrow at the bottom of the figure indicates the date, July, 1980, when the use of phosphorus was first regulated by the local law set in Siga Prefecture.

the lake from the watershed area was fixed rapidly by primary production occurring actively along the coastline, while the remaining phosphorus migrated, as a stream or as a cloud of plankton, to sampling site Ie-1 located in the pelagic part of the lake. Phytoplankton propagating in shallow, coastal zone served to retain phosphorus there and retard the eutrophication of the deep, pelagic part of the lake. This conclusion is supported by photos taken by the LANDSAT satellite, which visualizes a heavy distribution of chlorophyll-a along the coast of the lake.

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References

- 1) S. Yoshimura, "*Koshogaku (Limnology), 1937*", Reprinted, Seisan-gijutsu Center, Tokyo, 1976.
- 2) S. Horie, "*Lake Biwa*", Dr W. Junk Publ., 1984.
- 3) Lake Biwa Research Institute and National Institute for Research Advancement, "*Data Book of World Lakes*", the LECS secretariat, Otsu, Japan (1984).
- 4) BST, "*Biwako Seibutsu-shigen Tyosa-dan, Chukan Hokokusho (An Interim Report on the Assessment of Animate Resources in Lake Biwa)*", Kinki Regional Construction Bureau, Ministry of Construction, Japan (1965).
- 5) T. Fujinaga and T. Hori, "*Biwako no Kankyo Kagaku (Environmental Chemistry of Lake Biwa)*", The Japan Society for Promotion of Sciences, Tokyo (1982).
- 6) T. Hori, Y. Kanada, and T. Fujinaga, *Bunseki Kagaku*, **31**, 592-597 (1982).
- 7) T. Fujinaga, T. Hori, and Y. Kanada, *Bunseki Kagaku*, **28**, 222-226 (1979).
- 8) T. Hori, *Summary Reports of Research Project on Environmental Science, B-202-R-50-6*, pp. 175-178, Japanese Ministry of Education, Culture and Sciences (1983).
- 9) K. Negoro, "*Biwako no Dotai (Environmental Aspects of Lake Biwa)*", pp. 181-199, T. Fujinaga, Ed., Jiji Press, Tokyo (1974).
- 10) M. Koyama, T. Hori, O. Itasaka, M. Kawashima, and T. Hori, *Proc. International Congress of Scientist for Human Environment (HESC)*, pp. 391-397, Kyoto (1975).
- 11) T. Kunimatsu, *Personal communication*.
- 12) R. Yoshioka, *Personal communication*.

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Appendix

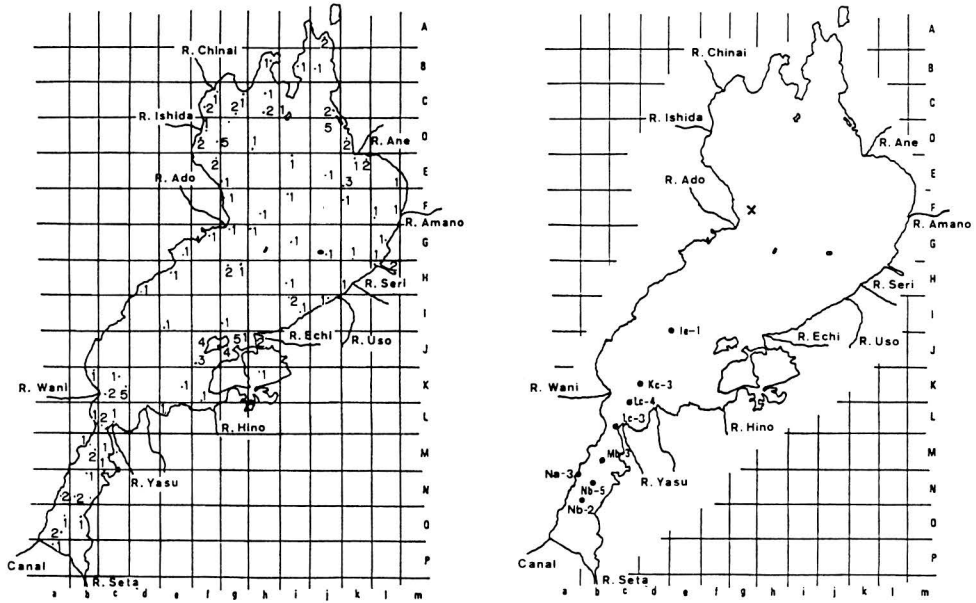


Fig. A1, Location of sampling sites chosen at BST-observation (Left) and at HRG-observation (Right), in Lake Biwa. The deepest point (104 m) is marked by a cross.

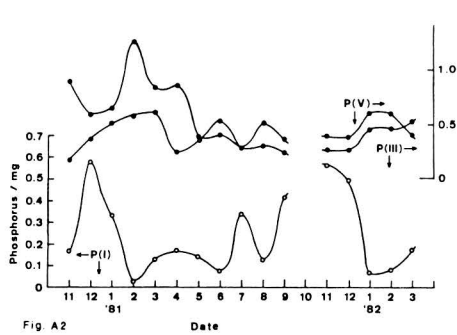


Fig. A2

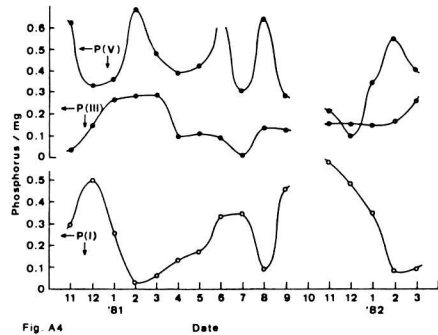


Fig. A4

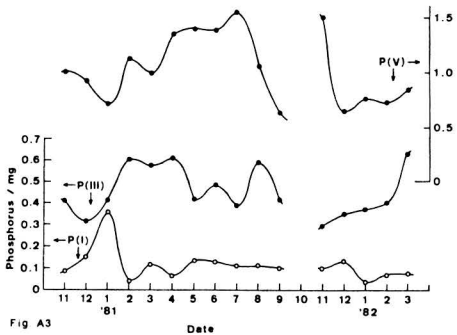


Fig. A3

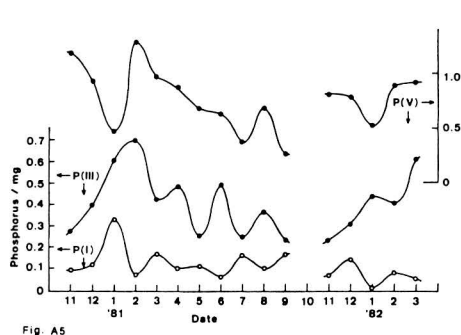


Fig. A5

Figs. A2–A5, Seasonal variation of P(I), P(III), and P(V) fractions contained in water-columns which were assumed at surface (0–20 m; Fig. A2), middle (20–40 m; Fig. A3), deep (40–60 m; Fig. A4), and bottom layers (60–70 m; Fig. A5) of Ie-1. See text for the definitions of the water-columns and the phosphorus fractions of P(I), P(III), and p(V).

Table A1. Date of Sampling, and Variations of Total-phosphorus Observed at Ie-1 and Nb-5

Observation #	Date	Time ⁽¹⁾ /months	GWP ⁽²⁾ /mg at Ie-1	Concn. of P/ppb at Nb-5
BST-1	Aug. 4, 1962	*	2.91	
2	Jan. 10, 1963	*	2.62	
HRG-1	May 12	0	3.89	12.3
2	July 23	2	5.02	19.0
3	Oct. 18	5	4.10	13.0
4	Jan. 12, 1964	8	3.98	17.0
5	May 30	12	4.71	14.5
6	July 21	14	4.12	6.8
7	Oct. 26	17	4.97	12.1
8	Oct. 19, 1966	41	3.16	7.8
9	Jan. 18, 1967	44	4.89	9.6
10	May 22	48	3.47	10.8
11	Aug. 2	51	3.55	
12	Feb. 7, 1968	57	5.52	21.3
13	May 21	60	3.09	11.4
14	Aug. 5	63	6.10	15.3
15	Aug. 4, 1969	75	4.25	18.6
16	Feb. 26, 1970	81	5.73	18.0
17	Sep. 9	88	7.55	11.0
18	Aug. 16, 1971	99	10.61	21.2
19	July 31, 1972	110	5.23	11.3
20	Feb. 27, 1973	117	5.35	19.8
21	Mar. 13	118	1.35	
22	Apr. 17	119	2.72	12.0
23	May 14	120	4.01	13.2
24A	June 14	121	5.03	8.6
24B	June 30	121	5.52	
25	July 16	122	5.85	17.1
26	Aug. 9	123	3.40	14.4
27	Sep. 11	124	4.89	31.3
28	Oct. 15	125	8.13	30.0
29	Nov. 14	126	6.31	32.5
30	Dec. 6, 1973	127	13.58	
31	Dec. 12	127	6.69	21.1
32	Jan. 16, 1974	128	7.58	26.6
33	Feb. 14	129	6.15	17.2
34	Mar. 12	130	6.23	15.2
35	Apr. 15	131	4.79	19.4
36	May 15	132	4.62	15.3
37	June 14	133	5.88	9.1
38	July 14	134	7.38	16.1
39	Aug. 14	135	6.63	25.7
40	Sep. 12	136	13.04	58.3
41	Oct. 14	137	6.83	15.9
42	Nov. 14	138	3.25	10.9
43	Dec. 13	139	5.37	12.6
44	Jan. 16, 1975	140	6.93	22.1
45	Feb. 14	141	5.83	18.1
46	Mar. 12	142	8.40	15.9
47	Apr. 14	143	7.47	23.5
48	May 14	144	7.09	34.1
49	June 12	145	5.84	24.3
50	July 14	146		
51	Aug. 14	147	7.24	12.7
52	June 14, 1976	157	7.60	43.5
53	Feb. 14, 1977	165	3.91	22.3
54	Dec. 12	175	3.96	18.9
55	May 31, 1978	180	7.26	8.0
56	Nov. 2	186	6.63	36.4
57	May 10, 1979	192	3.60	37.9
58	Mar. 13, 1980	202	17.89	26.3
59	May 12	204	6.42	37.0
60	Nov. 12, 1980	210	1.09	3.9
61	June 15, 1981	217	6.21	20.0
PHOS-1	May 12, 1980	204		
2	June 11	205		
3	July 10	206		
4	Aug. 13	207		
5	Sep. 10	208		
6	Oct. 15	209		
7	Nov. 12	210	5.22	24.10
8	Dec. 11	211	5.43	18.24
9	Jan. 19, 1981	212	5.16	20.66
10	Feb. 9	213	6.67	15.86
11	Mar. 11	214	5.62	10.51
12	Apr. 15	215	5.34	18.47
13A	May 14	216	4.59	27.42
13B	May 14	216	3.85	15.04
14	June 15	217	5.38	22.62
15	July 15	218	3.67	10.93
16	Aug. 11	219	4.67	20.12
17	Sep. 8	220	3.63	18.27
18	Oct. 15	221		
19	Nov. 11	222	5.08	25.77
20	Dec. 9	223	4.10	28.89
21	Jan. 11, 1982	224	4.05	11.80
22	Feb. 15	225	4.48	16.72
23	Mar. 17	226	5.32	16.69

⁽¹⁾ Time elapsed since May of 1963

⁽²⁾ Gross Weight of Phosphorus contained in a unit column of water with a dimension of 10 cm x 10 cm x 70 m (see text for detail)

Table A2. Distributions of Phosphorus in Lake Biwa

Sampling Date: May 12, 1980 (PHOS-1)				Sampling Date: June 11, 1980 (PHOS-2)				Sampling Date: Sept. 10, 1980 (PHOS-5)				Sampling Date: Oct. 15, 1980 (PHOS-6)			
Sample	DO (%)	pH	Temp (°C)	Sample	DO (%)	pH	Temp (°C)	Sample	DO (%)	pH	Temp (°C)	Sample	DO (%)	pH	Temp (°C)
1:1e-1.0m	105.3	7.4	15.9	1:1e-1.0m	101.7	7.5	20.75	1:1e-1.0m	90	7.5	24.6	1:1e-1.0m	101	8.9	20.7
2:1e-1.1m	106.5	7.4	15.1	2:1e-1.1m	102.5	7.4	20.6	2:1e-1.1m	90	7.5	24.51	2:1e-1.1m	99	8.9	20.7
3:1e-1.5m	102.4	7.4	13.0	3:1e-1.5m	101.6	7.4	19.55	3:1e-1.5m	89	7.5	24.51	3:1e-1.5m	100	8.8	20.50
4:1e-1.10m	99.9	7.3	11.6	4:1e-1.10m	97.5	7.3	16.0	4:1e-1.10m	87	7.5	24.5	4:1e-1.10m	99	8.8	20.50
5:1e-1.15m	97.5	7.2	11.2	5:1e-1.15m	95.1	7.3	15.05	5:1e-1.15m	61	7.3	16.1	5:1e-1.15m	96	8.7	20.20
6:1e-1.20m	94.4	7.1	10.5	6:1e-1.20m	93.3	7.3	13.1	6:1e-1.20m	67	7.0	12.9	6:1e-1.20m	92	8.4	19.60
7:1e-1.30m	91.8	7.1	9.4	7:1e-1.30m	89.3	7.2	10.6	7:1e-1.30m	78	7.0	9.8	7:1e-1.30m	88	7.6	18.20
8:1e-1.40m	88.5	7.0	8.9	8:1e-1.40m	88.5	7.0	9.1	8:1e-1.40m	76	7.0	8.9	8:1e-1.40m	73	7.0	10.28
9:1e-1.50m	87.2	7.0	8.0	9:1e-1.50m	84.9	7.0	8.35	9:1e-1.50m	71	7.0	8.35	9:1e-1.50m	72	6.9	9.20
10:1e-1.60m	85.1	6.9	7.5	10:1e-1.60m	85.9	7.0	8.05	10:1e-1.60m	70	6.9	8.0	10:1e-1.60m	70	6.9	8.55
11:1e-1.70m	79.9	6.9	7.4	11:1e-1.70m	79.5	6.9	7.8	11:1e-1.70m	48	6.7	7.6	11:1e-1.70m	57	6.6	8.20
12:Nb-2.0m	117.7	8.9	17.49	12:Nb-2.0m	114.8	8.2	22.8	12:Nb-2.0m	88.9	7.3	24.3	12:Nb-2.0m	97	8.5	21.5
13:Nb-5.0m	114.9	8.7	17.2	13:Nb-5.0m	119.7	8.5	23.4	13:Nb-5.0m	86.9	7.4	24.7	13:Nb-5.0m	97	8.4	21.4
14:Nb-3.0m	111.6	7.6	19.0	14:Nb-3.0m	114.2	8.4	24.0	14:Nb-3.0m	91.0	7.3	24.95	14:Nb-3.0m	98	7.7	21.5
15:KIDO.0m				15:KIDO.0m				15:KIDO.0m				15:KIDO.0m			
16:WANI.0m				16:WANI.0m				16:WANI.0m				16:WANI.0m			
17:OHASHI.0m				17:OHASHI.0m				17:OHASHI.0m				17:OHASHI.0m			
18:OGOTO.0m				18:OGOTO.0m				18:OGOTO.0m				18:OGOTO.0m			

Sampling Date: July 10, 1980 (PHOS-3)				Sampling Date: Aug. 13, 1980 (PHOS-4)				Sampling Date: Nov. 12, 1980 (PHOS-7)								
Sample	DO (%)	pH	Temp (°C)	Sample	DO (%)	pH	Temp (°C)	Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)
1:1e-1.0m	120.8	9.3	23.7	1:1e-1.0m	103.3	8.9	26.5	1:1e-1.0m	0.18	0.55	2.40	2.39	7.84	94.5	7.6	15.65
2:1e-1.1m	121.0	9.3	23.6	2:1e-1.1m	104.4	8.9	25.9	2:1e-1.1m	0.54	0.54	2.45	2.34	6.16	95.6	7.5	15.5
3:1e-1.5m	118.6	9.3	23.0	3:1e-1.5m	102.1	8.9	25.3	3:1e-1.5m	0.24	0.43	3.25	3.40	7.63	94.9	7.6	15.2
4:1e-1.10m	102.0	8.7	21.1	4:1e-1.10m	90.1	8.6	23.7	4:1e-1.10m	0.41	0.43	2.65	3.02	5.90	90.8	7.5	15.10
5:1e-1.15m	85.0	7.1	16.85	5:1e-1.15m	62.8	8.0	15.6	5:1e-1.15m	Trace	0.34	2.18	1.97	9.10	89.8	7.4	15.05
6:1e-1.20m	85.8	7.0	12.1	6:1e-1.20m	73.1	7.0	12.9	6:1e-1.20m	Trace	0.54	1.47	2.91	9.14	89.8	7.5	15.05
7:1e-1.30m	85.1	7.0	9.9	7:1e-1.30m	79.8	6.8	10.0	7:1e-1.30m	0.18	0.49	2.08	2.75	7.94	73.6	7.2	13.60
8:1e-1.40m	83.6	7.1	9.05	8:1e-1.40m	76.2	6.8	8.8	8:1e-1.40m	Trace	0.29	1.55	2.50	6.06	68.2	7.0	9.20
9:1e-1.50m	69.9	7.0	8.6	9:1e-1.50m	75.1	6.8	8.4	9:1e-1.50m	0.99	1.05	1.88	2.39	5.85	76.0	7.0	8.95
10:1e-1.60m	78.4	7.0	8.1	10:1e-1.60m	73.0	6.8	8.1	10:1e-1.60m	0.45	0.89	1.65	3.28	6.17	58.8	6.9	8.40
11:1e-1.70m	67.8	6.9	7.8	11:1e-1.70m	55.3	5.2	7.7	11:1e-1.70m	3.25	5.05	4.99	8.00	12.91	49.5	6.7	7.95
12:Nb-2.0m	120.2	8.7	25.6	12:Nb-2.0m	111.3	8.4	27.0	12:Nb-2.0m	0.55	2.12	3.96	4.27	15.28	114.8	8.1	13.5
13:Nb-5.0m	107.9	8.8	25.5	13:Nb-5.0m	107.6	8.8	27.2	13:Nb-5.0m	1.33	2.69	5.63	7.62	24.10	95.8	7.7	13.9
14:Nb-3.0m	115.0	8.9	25.3	14:Nb-3.0m	108.4	8.6	28.2	14:Nb-3.0m	2.25	2.96	5.74	8.04	30.59	91.3	7.3	14.6
15:KIDO.0m				15:KIDO.0m				15:KIDO.0m	0.55	0.46	2.04	4.17	6.37			
16:WANI.0m				16:WANI.0m				16:WANI.0m	Trace	0.28	3.02	3.15	7.48			
17:OHASHI.0m				17:OHASHI.0m				17:OHASHI.0m	0.18	0.24	3.31	5.33	9.20			
18:OGOTO.0m				18:OGOTO.0m				18:OGOTO.0m	0.24	0.43	2.80	3.70	8.04			

Remarks: observation # HRG-60

Table A2 (continued).

Sampling Date: Dec.11. 1980 (PHOS-8)									Sampling Date: Feb.9. 1981 (PHOS-10)								
Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)	Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)
1:1e-1.0m	0.21	0.20	1.84	1.89	5.87	88.9	7.2	12.85	1:1e-1.0m	0.67	0.54	1.79	2.30	8.42	90.5	7.2	6.8
2:1e-1.1m	0.48	0.56	1.89	1.74	6.45	90.0	7.2	12.70	2:1e-1.1m	Trace	0.13	2.64	2.45	8.48	91.2	7.1	6.8
3:1e-1.5m	Trace	0.42	2.67	2.71	7.40	89.1	7.2	12.45	3:1e-1.5m	0.26	Trace	3.26	2.11	8.89	90.8	7.2	6.8
4:1e-1.10m	0.20	1.39	2.33	2.34	7.23	87.7	7.1	12.40	4:1e-1.10m	Trace	Trace	3.60	2.56	8.84	91.0	7.2	6.8
5:1e-1.15m	Trace	0.65	2.31	2.12	6.52	87.3	7.1	12.40	5:1e-1.15m	91.0	7.2	6.75
6:1e-1.20m	Trace	0.55	2.23	2.24	7.33	87.5	7.1	12.40	6:1e-1.20m	Trace	0.75	3.09	2.98	9.27	90.2	7.2	6.72
7:1e-1.30m	0.18	0.35	2.23	1.89	7.33	86.5	7.1	12.35	7:1e-1.30m	0.34	Trace	4.67	2.96	10.98	91.6	7.2	6.72
8:1e-1.40m	1.31	1.10	3.59	2.96	7.11	64.8	6.8	9.83	8:1e-1.40m	Trace	0.28	2.80	3.39	9.87	90.6	7.2	6.69
9:1e-1.50m	2.86	3.30	5.00	4.85	8.33	59.0	6.7	8.75	9:1e-1.50m	Trace	Trace	2.91	2.62	9.06	92.6	7.2	6.69
10:1e-1.60m	3.67	3.92	5.48	5.63	8.30	57.6	6.7	8.55	10:1e-1.60m	Trace	0.18	3.26	2.64	8.89	80.8	7.1	6.62
11:1e-1.70m	5.83	6.02	7.44	7.68	11.17	52.7	6.6	8.17	11:1e-1.70m	Trace	0.49	3.07	4.86	11.03	90.4	7.0	6.52
12:Nb-2.0m	0.51	2.64	4.05	4.75	18.97	96.8	7.4	9.2	12:Nb-2.0m	96.9	7.0	5.0
13:Nb-5.0m	0.40	0.73	3.74	3.93	18.24	97.5	7.2	9.5	13:Nb-5.0m	0.24	1.11	4.19	6.45	15.86	96.2	7.1	6.05
14:Nb-3.0m	0.30	2.01	3.85	5.58	22.85	96.9	7.2	9.5	14:Nb-3.0m	Trace	0.37	3.39	4.05	14.04	96.2	7.1	6.05
15:KIDO.0m	Trace	Trace	1.72	1.70	6.67				15:KIDO.0m	Trace	0.27	3.82	3.12	9.96			
16:WANI.0m	Trace	0.31	2.11	1.89	7.97				16:WANI.0m	Trace	0.22	3.90	3.50	9.49			
17:OHASHI.0m	Trace	0.30	2.67	2.07	7.81				17:OHASHI.0m	Trace	Trace	4.48	3.28	9.91			
18:OGOTO.0m	Trace	0.70	2.93	3.10	13.56				18:OGOTO.0m	0.17	3.37	7.39	8.84	24.05			

Sampling Date: Jan.19. 1981 (PHOS-9)									Sampling Date: Mar.12. 1981 (PHOS-11)								
Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)	Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)
1:1e-1.0m	0.93	1.53	3.67	3.33	6.39	83.7	6.6	7.75	1:1e-1.0m	0.32	0.43	2.31	2.14	5.23	95.6	7.0	6.95
2:1e-1.1m	1.26	1.64	3.34	3.18	7.02	85.1	6.6	7.74	2:1e-1.1m	0.34	0.80	3.24	2.55	5.03	96.6	7.0	6.95
3:1e-1.5m	1.76	1.54	3.76	3.01	8.26	85.6	6.6	7.72	3:1e-1.5m	0.83	0.34	3.66	2.70	8.87	93.8	7.0	6.42
4:1e-1.10m	1.97	1.64	3.82	3.33	7.06	85.5	6.6	7.72	4:1e-1.10m	0.54	0.43	3.66	2.91	8.97	94.5	7.0	6.35
5:1e-1.15m	1.59	(2.40)	4.03	3.96	7.48	86.6	6.6	7.72	5:1e-1.15m	0.64	0.77	3.28	3.18	9.26	95.0	7.0	6.35
6:1e-1.20m	1.44	1.44	4.08	(5.14)	6.91	85.3	6.6	7.72	6:1e-1.20m	(1.12)	0.66	3.18	2.24	8.39	93.7	6.9	6.35
7:1e-1.30m	1.66	1.64	4.82	3.25	6.76	83.9	6.6	7.72	7:1e-1.30m	0.85	0.79	3.47	3.23	7.83	93.6	6.9	6.30
8:1e-1.40m	(2.08)	1.74	4.87	3.20	7.55	83.5	6.6	7.72	8:1e-1.40m	0.85	1.09	3.66	2.22	7.23	93.3	6.9	6.30
9:1e-1.50m	1.84	1.72	3.87	2.80	6.89	83.7	6.6	7.72	9:1e-1.50m	0.64	0.45	3.81	2.65	7.81	94.6	6.8	6.30
10:1e-1.60m	1.79	1.54	4.34	3.17	8.71	83.5	6.6	7.70	10:1e-1.60m	0.48	0.53	3.39	3.23	8.39	93.8	6.8	6.30
11:1e-1.70m	1.64	1.60	3.97	3.33	6.86	86.1	6.5	7.65	11:1e-1.70m	0.43	0.66	3.41	2.65	7.96	93.7	6.8	6.30
12:Nb-2.0m	(24.80)	7.04	13.52	34.36	67.56	93.9	7.0	2.7	12:Nb-2.0m	4.96	11.50	10.89	21.12	61.56	123.5	8.8	8.90
13:Nb-5.0m	(1.69)	0.22	4.08	5.05	20.66	95.5	7.0	2.5	13:Nb-5.0m	Trace	0.77	3.90	3.37	10.51	97.9	7.0	6.60
14:Nb-3.0m	(2.35)	0.43	5.36	5.88	18.97	91.9	7.0	3.6	14:Nb-3.0m	1.73	2.26	6.46	4.48	20.82	94.2	6.9	6.57
15:KIDO.0m	1.30	1.26	3.55	2.40	6.57				15:KIDO.0m	0.32	0.69	3.76	2.80	6.52			
16:WANI.0m	1.14	1.27	3.75	3.44	7.53				16:WANI.0m	0.23	0.55	4.24	1.83	8.00			
17:OHASHI.0m	0.93	0.88	3.24	3.35	7.06				17:OHASHI.0m	0.21	0.77	4.48	3.08	11.47			
18:OGOTO.0m	1.93	0.32	4.61	5.75	20.66				18:OGOTO.0m	Trace	0.61	4.19	3.28	8.97			

Remarks: These samples were taken on Jan. 16, 1981 and analyzed immediately.

Following compositions were determined again (15 days after the sampling) and compared with those determined immediately. P(FA) 7: 3.38 8: 4.44 9: 4.65 10: 4.23 15: 4.35

P(HFA) 7: 2.73 8: 3.15 9: 2.78 10: 2.84 15: 2.99

Remarks: P(HFA) < P(FA) at 0 - 70 m of 1e-1.

The such relation occurred also in the southern basin.

Table A2 (continued).

Sampling Date: Apr. 15, 1981 (PHOS-12)									Sampling Date: May 14, 1981 (PHOS-13B)								
Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)	Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)
1:1e-1.0m	0.35	0.26	3.63	3.56	12.17	101.8	7.4	9.95	1:1e-1.0m	1.93	1.28	2.96	3.18	6.72	112.4	7.6	16.50
2:1e-1.1m	0.75	0.39	4.63	3.96	12.11	101.5	7.3	9.80	2:1e-1.1m	9.00	7.71	10.16	10.49	14.05	111.4	7.8	14.82
3:1e-1.5m	0.30	0.40	4.33	2.85	13.16	99.9	7.1	9.25	3:1e-1.5m	7.53	7.71	9.23	9.67	10.35	105.0	7.4	13.18
4:1e-1.10m	0.20	0.60	2.92	1.84	8.54	100.0	7.1	9.10	4:1e-1.10m	0.98	0.71	3.28	2.98	8.00	102.0	7.2	12.85
5:1e-1.15m	0.26	Trace	2.57	3.30	8.34	99.5	7.1	8.95	5:1e-1.15m	0.69	0.38	2.03	2.94	6.14	100.8	7.2	12.55
6:1e-1.20m	0.33	0.20	2.74	3.05	9.07	99.0	7.1	8.80	6:1e-1.20m	0.21	0.25	1.61	1.21	3.85	98.8	7.2	12.05
7:1e-1.30m	0.37	0.68	2.87	2.35	7.40	96.1	7.0	8.40	7:1e-1.30m	Trace	0.16	1.54	1.92	4.02	95.8	7.0	10.55
8:1e-1.40m	(1.36)	0.42	3.12	3.66	5.16	96.6	7.0	7.62	8:1e-1.40m	0.38	0.16	1.28	0.77	3.42	89.8	6.8	8.40
9:1e-1.50m	0.68	0.76	1.61	2.95	6.73	93.6	7.0	7.38	9:1e-1.50m	0.27	Trace	1.24	1.05	3.12	86.7	6.8	7.30
10:1e-1.60m	0.55	1.31	1.71	2.45	6.29	90.5	7.0	7.05	10:1e-1.60m	Trace	Trace	1.31	1.19	3.58	80.2	6.8	6.85
11:1e-1.70m	0.98	1.41	2.92	3.27	6.13	87.6	7.0	6.75	11:1e-1.70m	Trace	0.22	1.64	2.41	6.31	81.0	9.8	6.72
12:Nb-2.0m	0.86	4.71	5.87	14.13	55.24	108.7	7.6	12.6	12:Nb-2.0m	19.04	19.04	18.72	24.78	37.02	130.7	9.2	22.5
13:Nb-5.0m	0.68	1.19	3.53	5.32	18.47	106.3	7.4	11.0	13:Nb-5.0m	2.01	1.69	4.57	6.67	15.04	122.4	9.2	20.4
14:Nb-3.0m	111.7	7.4	12.8	14:Nb-3.0m	10.57	10.42	13.77	14.72	17.06	120.6	9.0	21.2
15:KIDO.0m	Trace	0.15	3.34	2.65	7.33				15:KIDO.0m	1.25	0.77	3.21	3.12	(30.20)			
16:WANI.0m	0.47	0.45	4.23	4.16	13.78				16:WANI.0m	11.01	10.55	14.03	13.76	23.38			
17:OHASHI.0m	0.57	0.91	3.85	3.86	17.96				17:OHASHI.0m	11.62	11.84	13.16	13.43	22.84			
18:OGOTO.0m	0.65	1.48	3.55	5.91	19.63				18:OGOTO.0m	4.47	4.34	6.33	6.61	12.31			

Remarks: P(HFA) < P(FA) occurred in 0 - 15 m depth at 1e-1.

Remarks: In order to examine the degree of degradation of phosphorus compounds during storage, phosphorus analysis was performed 25 days after the sampling and the results were compared with that performed immediately (see #PHOS-13A).

Sampling Date: May 14, 1981 (PHOS-13A)									Sampling Date: June 15, 1981 (PHOS-14)								
Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)	Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO(%)	pH	Temp(°C)
1:1e-1.0m	Trace	0.27	1.64	2.67	6.71	112.4	7.6	16.50	1:1e-1.0m	Trace	0.32	3.13	2.57	7.20	106.5	7.4	20.8
2:1e-1.1m	0.42	1.06	2.81	4.22	17.13	111.4	7.8	14.82	2:1e-1.1m	0.38	0.32	3.40	3.91	7.63	102.8	7.4	20.2
3:1e-1.5m	0.57	1.15	2.91	4.11	13.17	105.0	7.4	13.18	3:1e-1.5m	0.18	0.37	2.35	4.21	9.30	100.0	7.3	18.15
4:1e-1.10m	0.45	0.45	2.91	2.96	8.39	102.0	7.2	12.85	4:1e-1.10m	0.65	0.64	3.34	3.10	12.16	92.8	7.4	14.90
5:1e-1.15m	Trace	0.45	3.31	2.93	7.17	100.8	7.2	12.55	5:1e-1.15m	Trace	1.21	2.97	3.26	10.97	91.4	7.4	13.70
6:1e-1.20m	Trace	0.27	1.13	3.18	4.78	98.8	7.2	12.05	6:1e-1.20m	0.38	0.34	3.56	2.72	7.63	87.8	7.2	11.20
7:1e-1.30m	0.35	0.70	2.04	2.78	5.95	95.8	7.0	10.55	7:1e-1.30m	0.50	0.19	2.37	2.34	5.61	86.7	7.1	9.0
8:1e-1.40m	0.50	0.50	1.99	3.28	4.30	89.8	6.8	8.40	8:1e-1.40m	1.11	0.31	2.58	2.50	4.58	85.5	7.0	7.8
9:1e-1.50m	0.40	0.48	2.35	2.42	4.00	86.7	6.8	7.30	9:1e-1.50m	1.02	0.41	3.04	2.23	5.25	83.6	7.1	6.8
10:1e-1.60m	1.05	1.36	3.21	4.40	5.44	80.2	6.8	6.85	10:1e-1.60m	1.19	0.41	3.45	2.77	4.98	83.0	7.0	6.7
11:1e-1.70m	1.41	2.07	2.45	4.25	8.64	81.0	6.8	6.72	11:1e-1.70m	1.89	6.26	5.02	9.14	19.28	76.6	6.9	7.0
12:Nb-2.0m	0.37	1.78	4.28	9.65	39.62	130.7	9.2	22.5	12:Nb-2.0m	0.37	1.08	3.62	3.20	10.92	95.7	7.2	23.3
13:Nb-5.0m	0.45	0.90	4.28	5.42	18.53	122.4	9.2	20.4	13:Nb-5.0m	0.49	0.92	3.43	6.22	22.62	117.7	7.5	23.2
14:Nb-3.0m	0.46	1.29	5.17	6.33	27.42	120.6	9.0	21.2	14:Nb-3.0m	0.36	2.32	4.42	9.73	28.83	105.4	7.3	24.5
15:KIDO.0m	0.24	0.63	2.15	3.49	7.90				15:KIDO.0m	Trace	0.30	3.56	2.88	6.66			
16:WANI.0m	0.45	1.01	8.45	7.20	25.39				16:WANI.0m	Trace	0.22	4.32	3.66	7.77			
17:OHASHI.0m	0.42	1.11	6.31	6.82	24.37				17:OHASHI.0m	0.49	0.49	3.67	3.32	10.32			
18:OGOTO.0m	0.35	1.21	2.53	4.32	12.86				18:OGOTO.0m	0.44	0.22	4.26	3.31	9.68			

Remarks: P(HFA) < P(FA) occurred at 5 - 60 m depth of 1e-1 and in the southern basin excepting Nb-5.

Table A2 (continued).

Sampling Date: July 15, 1981 (PHOS-15)									Sampling Date: Sept. 8, 1981 (PHOS-17)								
Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)	Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)
1:le-1.0m	Trace	0.58	3.24	2.71	6.88	119.2	9.2	27.2	1:le-1.0m	0.24	0.34	1.49	1.85	5.79	92.6	8.1	24.7
2:le-1.1m	0.19	0.63	2.64	3.69	9.18	122.6	9.2	26.4	2:le-1.1m	0.14	0.44	2.70	2.40	7.01	91.7	8.1	24.7
3:le-1.5m	0.19	0.66	3.01	4.79	8.15	129.5	9.2	25.55	3:le-1.5m	0.26	0.34	3.10	4.41	6.01	89.2	8.0	24.7
4:le-1.10m	0.24	0.51	2.78	2.65	6.24	94.6	7.4	17.8	4:le-1.10m	0.21	0.44	2.30	3.20	6.26	88.6	8.0	24.6
5:le-1.15m	0.19	0.34	1.97	2.42	6.07	83.2	7.2	13.3	5:le-1.15m	0.35	0.50	2.30	3.42	5.41	69.3	7.2	18.6
6:le-1.20m	0.37	0.81	1.85	1.95	3.72	81.6	7.1	11.3	6:le-1.20m	0.83	1.15	2.80	2.74	3.65	69.2	7.0	11.9
7:le-1.30m	0.55	0.75	1.97	2.24	3.82	83.1	7.1	8.5	7:le-1.30m	0.61	0.44	1.50	2.60	3.10	74.9	7.0	8.7
8:le-1.40m	1.04	1.00	2.43	1.84	4.45	82.1	7.1	7.4	8:le-1.40m	0.57	1.26	2.15	2.60	2.98	77.1	7.0	7.6
9:le-1.50m	1.64	1.54	3.61	1.86	3.99	80.0	7.0	6.8	9:le-1.50m	1.83	1.83	3.10	3.72	5.21	75.3	6.8	7.0
10:le-1.60m	2.00	2.74	2.66	1.72	5.32	75.3	7.0	6.66	10:le-1.60m	3.13	3.39	4.54	2.40	6.61	62.7	6.8	6.7
11:le-1.70m	3.98	4.16	4.39	4.21	7.81	66.4	6.9	6.5	11:le-1.70m	4.96	5.73	6.63	4.71	10.11	55.2	6.6	6.55
12:Nb-2.0m	1.15	4.79	6.13	18.14	56.67	128.3	9.0	27.9	12:Nb-2.0m	1.00	3.69	7.71	20.88	94.97	119.0	9.8	24.6
13:Nb-5.0m	0.19	2.02	3.24	4.59	10.93	115.4	9.0	27.2	13:Nb-5.0m	0.47	1.09	4.53	4.71	18.27	100.8	8.9	24.85
14:Nb-3.0m	0.73	0.72	2.43	3.34	11.04	119.1	9.0	26.9	14:Nb-3.0m	1.18	2.33	7.51	9.21	23.33	101.4	8.9	25.4
15:KIDO.0m		0.77	3.70	2.99	7.46				15:KIDO.0m	0.47	1.26	2.00	2.50	6.06			
16:WANI.0m	Trace	0.37	3.01	4.04	10.52				16:WANI.0m	0.24	1.15	2.38	3.00	7.07			
17:OHASHI.0m	0.19	0.65	4.45	4.90	14.05				17:OHASHI.0m	0.47	0.70	2.55	2.13	7.45			
18:OGOTO.0m	0.61	1.13	4.88	8.43	32.97				18:OGOTO.0m	0.77	1.15	5.36	10.86	45.41			

Remarks: P(HFA) < P(FA) occurred at 40 - 60 m of le-1. Remarks: P(HFA) < P(FA) occurred at 60 - 70 m of le-1.

Sampling Date: Aug. 11, 1981 (PHOS-16)									Sampling Date: Nov. 11, 1981 (PHOS-19)								
Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)	Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)
1:le-1.0m	0.20	0.56	4.07	1.96	10.36	103.5	9.0	26.9	1:le-1.0m	0.52	0.34	1.67	2.38	5.25	92.2	7.2	15.0
2:le-1.1m	Trace	0.91	5.06	2.98	9.70	102.8	9.0	26.9	2:le-1.1m	0.52	0.84	2.81	3.13	7.57	91.2	7.2	14.9
3:le-1.5m	Trace	0.65	4.34	4.17	10.24	102.3	9.0	25.9	3:le-1.5m	0.16	0.52	1.78	2.81	9.72	90.2	7.0	14.8
4:le-1.10m	Trace	0.48	3.19	3.27	9.20	87.8	7.5	20.0	4:le-1.10m	(0.58)	0.26	2.21	3.02	10.31	91.7	7.1	14.7
5:le-1.15m	0.16	0.48	2.74	3.27	7.44	80.1	7.1	14.7	5:le-1.15m	0.37	0.63	1.59	2.70	10.74	91.1	7.0	14.7
6:le-1.20m	0.20	0.46	2.50	3.54	7.08	76.5	7.1	11.5	6:le-1.20m	0.21	0.40	1.88	3.19	7.46	88.8	7.0	14.6
7:le-1.30m	Trace	0.46	2.20	2.54	5.48	77.0	7.0	8.55	7:le-1.30m	Trace	0.21	1.11	2.57	5.20	68.5	6.8	9.75
8:le-1.40m	Trace	0.51	2.17	1.49	4.58	78.3	7.0	7.6	8:le-1.40m	0.60	0.58	1.95	2.54	4.18	72.4	6.7	8.20
9:le-1.50m	0.39	0.74	1.90	2.12	3.81	78.8	7.0	7.1	9:le-1.50m	(3.14)	2.81	4.02	4.42	5.90	67.9	6.7	7.10
10:le-1.60m	0.23	0.48	2.29	1.63	6.31	73.7	6.9	6.8	10:le-1.60m	4.66	5.03	6.38	6.25	8.16	56.5	6.7	6.72
11:le-1.70m	0.39	1.26	2.02	2.08	10.71	61.5	6.8	6.6	11:le-1.70m	(6.18)	5.61	7.17	6.56	9.50	49.2	6.7	6.70
12:Nb-2.0m	10.68	14.28	21.72	31.66	58.21	120.9	8.8	27.9	12:Nb-2.0m	1.80	(7.23)	6.89	13.79	44.60	98.0	7.0	11.2
13:Nb-5.0m	1.04	0.74	8.93	7.74	20.12	105.1	8.8	27.25	13:Nb-5.0m	0.52	1.99	2.97	6.52	25.77	94.6	7.0	11.9
14:Nb-3.0m	0.78	1.65	7.26	8.69	28.15	112.8	8.8	27.95	14:Nb-3.0m	----	----	----	----	----	96.2	7.0	12.15
15:KIDO.0m	0.78	0.87	5.74	3.99	10.83				15:KIDO.0m	0.55	0.13	1.92	2.90	5.90			
16:WANI.0m	0.52	0.48	5.02	4.76	10.95				16:WANI.0m	0.11	0.21	2.35	2.97	6.28			
17:OHASHI.0m	0.59	0.56	6.93	5.12	12.80				17:OHASHI.0m	0.21	0.89	2.35	3.24	8.64			
18:OGOTO.0m	0.65	1.16	5.86	6.01	17.02				18:OGOTO.0m	0.44	0.84	3.93	6.34	17.25			

Remarks: P(HFA) < P(FA) occurred at 0 - 5 m of le-1 and in the southern basin.
P(V) fraction accumulated at 70 m of le-1. Remarks: P(AMFA) > P(FA)

Table A2 (continued).

Sampling Date: Dec. 9, 1981 (PHOS-20)

Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)
1:1e-1.0m	0.14	0.24	2.26	1.99	4.34	91.6	7.2	11.04
2:1e-1.1m	Trace	0.59	2.31	2.85	4.77	91.1	7.2	11.02
3:1e-1.5m	0.24	0.46	2.23	2.31	3.67	91.7	7.3	10.96
4:1e-1.10m	0.14	0.72	2.42	3.23	6.87	91.3	7.3	10.95
5:1e-1.15m	0.16	0.72	2.66	2.48	6.28	90.7	7.3	10.94
6:1e-1.20m	Trace	0.73	2.15	2.94	6.38	90.1	7.3	10.92
7:1e-1.30m	0.16	0.51	2.05	2.31	6.28	90.4	7.2	10.88
8:1e-1.40m	0.68	0.96	2.55	2.64	5.41	73.4	7.0	9.82
9:1e-1.50m	2.11	2.39	3.61	3.28	5.52	63.7	6.8	7.63
10:1e-1.60m	3.50	4.07	5.30	5.68	6.25	55.8	6.8	7.00
11:1e-1.70m	4.89	5.49	7.27	6.41	8.16	48.8	6.7	6.80
12:Nb-2.0m	2.89	(28.49)	5.81	21.54	66.03	95.0	7.2	7.46
13:Nb-5.0m	0.63	4.07	4.79	16.20	28.89	94.6	7.1	7.68
14:Nb-3.0m	1.00	3.80	5.49	9.42	24.69	93.7	7.2	8.35
15:KIDO.0m	0.25	0.51	3.07	1.99	5.31			
16:WANI.0m	0.36	0.38	3.18	2.31	6.06			
17:OHASHI.0m	0.14	0.72	3.12	3.41	8.21			
18:OGOTO.0m	0.97	(5.61)	3.77	11.52	32.93			

Remarks: P(HF) 2: 0.29 5: 0.14 10: 4.58 13: 5.49 P(HFA) 2: 2.42 5: 2.11 10: 5.71 13: 8.02
P(AmF) 2: Trace 5: Trace 10: 3.16 13: 0.66

Sampling Date: Jan. 11, 1982 (PHOS-21)

Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)
1:1e-1.0m	Trace	0.33	1.53	2.85	4.01	70.2	7.0	8.80
2:1e-1.1m	Trace	0.35	1.92	3.27	4.85	69.2	7.0	8.70
3:1e-1.5m	Trace	Trace	2.19	3.18	6.53	69.6	7.0	8.55
4:1e-1.10m	0.20	0.30	2.42	4.32	6.64	70.3	7.0	8.55
5:1e-1.15m	0.13	0.30	1.88	3.79	5.14	68.7	6.9	8.50
6:1e-1.20m	Trace	Trace	1.80	3.71	5.80	69.2	6.9	8.50
7:1e-1.30m	Trace	Trace	2.19	---	3.99	68.5	6.9	8.50
8:1e-1.40m	(0.98)	0.24	2.74	2.85	5.50	69.0	6.9	8.50
9:1e-1.50m	0.27	0.23	2.42	2.95	5.22	67.8	6.9	8.50
10:1e-1.60m	0.50	0.60	2.64	2.45	6.01	56.5	6.9	8.20
11:1e-1.70m	4.90	6.20	6.96	6.90	10.43	34.7	6.7	6.98
12:Nb-2.0m	1.55	(11.75)	5.46	11.12	47.30	76.0	7.2	6.50
13:Nb-5.0m	0.71	0.78	2.81	5.47	11.80	72.3	7.0	7.18
14:Nb-3.0m	0.54	3.16	3.32	7.91	21.11	72.1	7.0	7.90
15:KIDO.0m	Trace	0.44	2.55	2.53	4.19			
16:WANI.0m	0.20	Trace	2.76	2.79	5.48			
17:OHASHI.0m	0.22	0.27	3.53	3.48	6.64			
18:OGOTO.0m	0.27	1.03	3.71	4.11	8.54			

Remarks: P(AmF) 2: 0.76 5: 0.44 10: 1.31 13: 1.41 P(HFA) 2: 2.21 5: 2.32 10: 2.85 13: 3.14
P(HF) 2: 0.54 5: 1.41 10: 1.34 13: 2.01

Sampling Date: Feb. 15, 1982 (PHOS-22)

Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(AmFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)
1:1e-1.0m	0.350	0.304	1.77	2.18	2.39	4.98	89.9	7.0	6.60
2:1e-1.1m	0.355	Trace	2.49	3.63	2.86	5.19	88.8	7.0	6.80
3:1e-1.5m	0.355	0.203	2.75	2.89	4.31	5.61	88.7	7.0	6.75
4:1e-1.10m	0.324	0.304	2.49	2.23	3.48	6.65	88.5	7.0	6.70
5:1e-1.15m	0.507	0.639	1.87	3.63	3.53	5.09	87.0	7.0	6.70
6:1e-1.20m	0.791	0.558	2.44	2.70	2.75	8.41	88.7	7.0	6.70
7:1e-1.30m	0.831	0.355	2.28	2.60	3.06	6.75	87.7	7.0	6.65
8:1e-1.40m	0.608	0.304	2.63	3.22	2.82	5.40	88.1	7.0	6.65
9:1e-1.50m	0.355	0.405	2.91	2.39	3.79	5.09	87.7	7.0	6.60
10:1e-1.60m	0.912	0.508	2.21	3.22	2.82	6.96	85.6	7.0	6.50
11:1e-1.70m	0.811	1.085	2.62	3.01	4.57	8.72	85.1	7.0	6.48
12:Nb-2.0m	2.210	(15.15)	5.42	21.06	11.63	47.00	94.6	7.1	5.50
13:Nb-5.0m	0.405	2.017	2.39	4.05	6.39	16.72	95.6	7.0	5.71
14:Nb-3.0m	0.385	1.541	2.75	4.57	6.54	15.16	93.8	7.0	5.78
15:KIDO.0m	0.203	0.223	1.77	1.79	2.28	5.82			
16:WANI.0m	0.405	0.507	1.87	2.00	2.75	5.50			
17:OHASHI.0m	Trace	0.304	1.61	4.15	3.17	6.36			
18:OGOTO.0m	0.608	0.608	2.49	3.55	5.12	9.35			

Remarks: P(AmF) 2: 0.152 5: 0.203 10: 0.507 13: 0.791 P(N) < P(F) occurred at 20 - 60 m of 1e-1.
P(HF) 2: 0.324 5: 0.912 10: 1.064 13: 3.142

Sampling Date: Mar. 17, 1982 (PHOS-23)

Sample	P(F)/ppb	P(N)/ppb	P(FA)/ppb	P(HFA)/ppb	P(A)/ppb	DO (%)	pH	Temp (°C)
1:1e-1.0m		0.95	3.54		8.10	95.8	7.0	7.81
2:1e-1.1m		0.52	2.35		5.20	95.3	7.0	7.52
3:1e-1.5m		0.48	4.11		8.14	94.4	7.0	7.16
4:1e-1.10m		Trace	3.54		8.61	94.0	7.0	7.10
5:1e-1.15m		0.62	3.60		7.53	93.5	7.0	7.10
6:1e-1.20m		0.26	3.44		7.94	93.3	7.0	7.09
7:1e-1.30m		0.23	3.13		7.89	93.2	7.0	6.91
8:1e-1.40m		0.31	3.47		7.68	93.2	7.0	6.90
9:1e-1.50m		1.14	3.44		7.06	91.2	7.0	6.74
10:1e-1.60m		0.88	3.51		7.48	89.6	7.0	6.61
11:1e-1.70m		0.96	3.33		7.32	87.7	7.0	6.60
12:Nb-2.0m		3.73	6.44		23.30	99.3	7.3	11.50
13:Nb-5.0m		1.99	4.89		16.69	99.8	7.2	10.40
14:Nb-3.0m		2.38	6.44		21.97	100.4	7.3	11.08
15:KIDO.0m		0.62	1.80		6.08			
16:WANI.0m		0.43	2.51		9.13			
17:OHASHI.0m		0.67	3.96		11.51			
18:OGOTO.0m		1.04	3.08		10.06			