Chemical Studies in Lake Metabolism.

By Ken SUGAWARA.

(Received June 12, 1939.)

Contents.

I. Introduction.

It will be necessary to begin with a brief explanation of the term "lake metabolism", which the writer believes he first introduced.

After A. Thienemann, the writer regards a lake as a "Lebenseinheit." The word "Lebenseinheit", which is used here in its physiological sense, was primarily understood to represent an individual living thing, animal or plant. After R. Hesse,(1) every animal or plant individual is a "Lebenseinheit", because it is "ein abgeschlossener, unabhängig für sich bestehender Organismus, bei dem alle Einzelteile derart zusammenwirken, dass der Fortbestand des Ganzen dadurch gesichert ist; mit anderen Worten, es ist selbsterhaltungsfähig."

What has just been said here concerning an individual organism is also true of what is called biocoenosis, defined as follows by the creator of the idea, Karl Möbius:(2) "Die Biocönose ist eine Gesellschaft von lebenden Wesen, eine den durchschnittlichen äusseren Lebensverhältnissen entsprechende Auswahl und Anzahl von Arten und Individuen, welche sich gegenseitig bedingen und durch Fortpflanzung in einem abgemessenen Gebiete dauernd erhalten."

---

(1) Hesse, "Der Tierkörper als selbständiger Organismus" 33, (1910).

(2) Möbius, *Sitzungsber. d. Ges. naturforsch. Freunde*, Berlin, (**1901**), 267.

When Hesse's word "Organismus" is substituted by "biocoenosis" and "Einzelteile" by "animals and plants", the "Lebenseinheit" idea is rightly applicable to the case of biocoenosis. Biocoenosis, then, is a "physiologische Lebenseinheit der zweiten Ordnung."

Now, in a lake, there may be distinguished three regions, pelagic, littoral, and abyssal, which are inhabited respectively by a characteristic biocoenosis. Thus plankton and necton inhabit the pelagic region, rooted vegetation and other plants, and the animals depending on them, the littoral region, and the larvae of *Chironomus* and *Corethra,* shellfishes, and Tubificidae worms the abyssal or third region.

Each of these biocoenoses depends on its milieu, that is, the biotope. On the other hand, all these biocoenoses and biotopes of a lake interact and give rise to a cyclic process on a larger scale between the dissolved and the floating or deposited substances, whereby the dynamical equilibrium of these biocoenoses and the permanency of the lake, as a whole, are maintained.

Fig. 1 gives a general scheme of the cyclic process and the correlations between biocoenoses and biotopes, which can be explained briefly by the following three points (1) supply of material, (2) changes in the substance in the lake water, and (3) the circulation of matter in the lake.

(1) The materials necessary for the maintenance of the lake water come from three different sources, namely, influent waters, the atmosphere, and the decomposition products that have been formed in the lake water.

(2) The nutrient materials thus supplied are then assimilated by phytoplankton and transformed into their body substances. Part of the phytoplankton that are suspended in the water are eaten by zooplankton, while the remainder, after death, together with dead zooplankton, sink to the bottom and form the lake deposits, which, filling the lake bottom, gradually make it shallower.

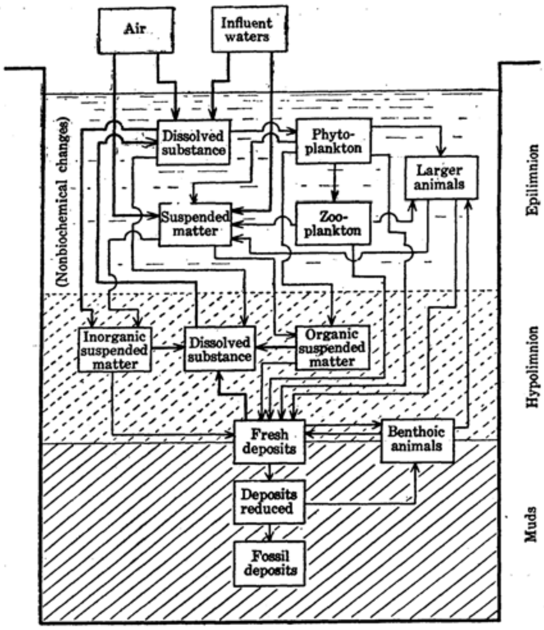


Fig. 1. A general scheme of the cyclic process and the correlation between biocoenoses and biotopes in a lake.

However, not all sediments are destined to remain buried. Not a small part of the deposited substance is given a chance of being returned to the overlying free water.

As a matter of fact, the deposited dead plankton are food for the benthoic animals which, in their turn, are relished by fish. Another part of the deposit undergoes bacterial action and decomposes into simpler products.

(3) The decomposition products which are fairly soluble, tend to dissolve into the bottom water, where they accumulate in abundance.

These accumulated products are then carried back into the upper water layers by the water current which is caused either by wind or by the lowering of temperature in the night, especially in that season between autumn and winter, thus completing the cycle, to be repeated again and again.

This is the general view (the details being unknown) of the circulation of matter in a lake, by means of which biocoenotic and biotopic equilibrium, that is, the permanency of the lake, is actualized.

Thus G. Werestchagin(3) was led to the idea of "limnologische Einheit", as he says: "Der See ist die Einheit der einzig und reel in der Natur vorhandenen Summe von Lebensraum (Biotop) plus Lebensgemeinschaft (Biocönose), eine 'limnologische Einheit'."

Thus the idea of Werestchagin was accepted by Thienemann(4) with certain modifications, as follows:

"Das ist richtig, nur sollte man das Wort "Summe" überhaupt lieber vermeiden, denn es handelt sich ja gerade darum, dass der See nicht die Summe, sondern die Einheit von Biotop und Biocönose darstellt. Ja wir glauben, dass hier wohl vielleicht sogar der Drieschsche Begriffe der Ganzheit eine Rolle spielt, noch wissen wir es allerdings nicht."

Thus with lakes, we have come to the idea of "Lebenseinheit der dritten Ordnung."

When, therefore, we regard a lake as a "Lebenseinheit" and investigate from such a point of view the changes that occur in the various substances forming a lake, the system of knowledge thus brought together is called "lake metabolism."

This is the reason why the term "lake metabolism" is introduced into the present study, in which the writer primarily aims to interpret the significance of individual lake factors, their variability, and their relations to one another for the existence of a lake as a "Lebenseinheit," and thus to infer the extent to which the idea "Lebenseinheit" holds for the case of a lake.

Because of lack of available data regarding these lake factors, it was necessary for the writer to begin with the observation of these factors, so that the greater part of the work so far done had to do with the accomplishment of this purpose.

This and other circumstances retarded the writer in the solution of a number of problems, notwithstanding which the writer is glad to be able to report that it was possible to reach a number of results, whereby certain questions were clearly solved, some new questions raised, and new relations discovered, the solutions and interpretations of which latter, it is hoped, will be forthcoming with the aid of further studies.

---

(3) Werestchagin, *Verhandl. d. Internat. Vereinigung f. theoret. u. angew. Limnologie*, Stuttgart, **1** (1923), 225.

(4) Thienemann, *Naturwissenschaften*, **13** (1925), 595.

II. General Descriptions of the Lakes Studied.

Takasuka-numa.

Takasuka-numa, where the greater part of the writer's investigations were carried out, and which belongs to that peculiar body of water called "Oppori", meaning a swamp formed by flood from a broken river bank, lies about 2.5 km. northe-east of Satte-mati Saitama Prefecture, having an area of about 0.021 km.^{2} and a maximum depth of about 6 m. The water is humus brown, and so turbid that Secchi's disc was often invisible at 0.8 m. The lake is protected from north-easterly winds by a bank 7 m. high, running close to the northern shore, and by the village with its old trees, which occupies a part of the eastern beach, while the remaining greater part of the beach stretches directly into extensive corn- and rice-fields.

It is said that the bed of Sima-kawa, a branch of the River Gongendo which formerly was the main stream of the River Tone, was filled with volcanic ashes that were ejected during the severest recorded eruption of Mt. Asama, in 1783, and carried down by the river water, until the river shallowed and caused repeated floods up to the beginning of the Meizi era when the work of river improvement was completed.

The lake is said to owe its origin to the flood of 1786. The earth and sand then loosened seem to have been carried for a considerable distance, their remnants being found to-day as the low sandy hills called "Botti", distributed here and there in the south-western field, each covering from 3000 m.^{2} to 4000 m.^{2}

Table 1.

Altitude	(m.)	10
Area	(m^2.)	21500
Maximum depth	(m.)	6.0
Mean depth	(m.)	3.0
Volume	(m^3.)	65000

The morphometrical data of the lake basin are given in Table 1.

The basin is steeper in the east and north than elsewhere, and the littoral is narrow along these sides, while in

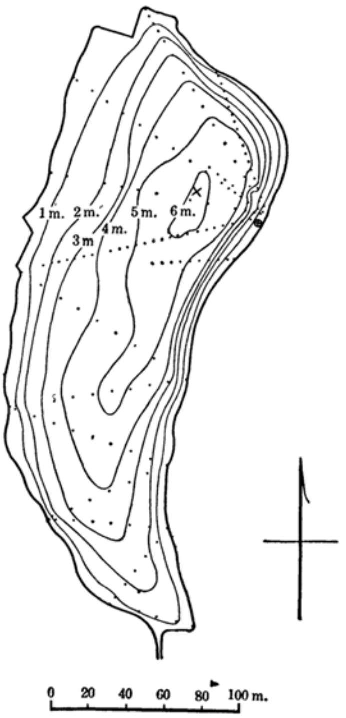


Fig. 2. Bathymetrical map of Takasuka-numa constructed with the data from the latest surveying and sounding in 1939.

- ● ··· Points sounded.
- × ··· The point where the water and the mud were sampled.
- ⊗ ··· The writer's hut for observation.

the south and west it covers a wider sandy area, with rooted vegetation. The muddy abyssal region is evidently separated from the littoral, with its center near the head, where it occupies about a quarter of the total area of the lake basin.

The Nisina Lake Group in Nagano Prefecture. Kizaki-ko(5), Nakatuna-ko, and Aoki-ko.

These three lakes are links in a chain in a valley that runs along the northern foot of the North Japan Alps. Their important morphometrical data are given in Table 2.

Table 2.

		Aoki-ko	Kizaki-ko	Nakatuna-ko
Area	(km^2.)	1.863	1.413	0.141
Depth	(m.)	62.0	29.5	12.0
Altitude	(m.)	822	764	815

Aoki-ko, the deepest of them, has a transparency of 10 m., the water of the hypolimnion containing abundant oxygen, even late in the period of summer stagnation.

Kizaki-ko, which is the lowest in level and partly surrounded by cultivated land and villages, shows an advanced state of eutrophication. Its transparency does not exceed 8 m., while the hypolimnion water exhibits during the summer stagnation period a marked decrease in its oxygen content.

In Nakatuna-ko, which receives the water from Aoki-ko and sends it into Kizaki-ko, and is the smallest and shallowest of all, the oxygen content in deep water often becomes zero in summer, the transparency being usually less than 5 m.

The Five Lakes at the Foot of Mt. Huzi. Motosu-ko, Syozi-ko, Nisino-umi, Kawaguti-ko, and Yamana-ko.

These five lakes, which nestle at the foot of Mt. Huzi, owe their origin to damming up at different periods of the valleys between Mt. Huzi, the Misaka or Dosi mountain ranges, and their branches by volcanic ejecta from Mt. Huzi.

Their important morphometrical data are given in Table 3.

Table 3.

		Motosu-ko	Syozi-ko	Nisino-umi	Kawaguti-ko	Yamanaka-ko
Area	(km^2.)	4.87	0.87	2.30	6.13	6.46
Depth	(m.)	119.0	16.0	73.0	15.2	13.2
Altitude	(m.)	908	908	908	830	982

---

(5) "ko" is a termination given to all lakes.

Motosu-ko, the most westerly of the five lakes, is surrounded by a branch of the Misaka mountains, except at its eastern shore, which consists of massive accumulations of lava. It is mouthless and has no inflowing streamlet, but there are several reasons supporting the view that the lake is connected with Syozi-ko and Nisino-umi by subterranean communications. The water is a beautiful blue, with a transparency of 16–18 m. According to D. Inaba,(6) this lake is the most oligotrophic of the five, the content of protein nitrogen in the winter of 1934, being reported as having been only 23 mg./l.

Syozi-ko, lying east of Motosu-ko, is the smallest of the five, its area being less than 1 km.^{2} Except in the south, its shore is surrounded by the Misaka mountains, where a gentle slope of lava-flow from Mt. Huzi projects into the lake in the form of a peninsula.

It is remarkable as has been pointed out by many authors that, throughout the year, below some 10 m., there is a sudden decrease in the oxygen content, and that, as Inaba has reported, in the winter of 1934 the protein nitrogen content was 109 mg./l., which is the highest value of the five lakes. Both of these facts prove that this lake has attained the most eutrophied stage of all the five.

Nisino-umi, called also Sai-ko, lying east of Syozi-ko, is separated from the latter by a gently sloping lava ridge, called "Aokigahara-marubi."

It is said that Nisino-umi and Syozi-ko were a large single lake, called "Seno-umi", until 863, when a lava flow divided this mother lake into two, as we find it to-day.

The lake, which is well aerated to depth, has a transparency of 10 m. Inaba's data of the protein nitrogen content of the water gives 35 mg./l. for the winter of 1934.

Kawaguti-ko, which lies due north of Mt. Huzi, has the lowest water level of the five lakes. The basin is divided into two main parts by a central islet, Uno-sima. According to D. Miyadi,(7) the maximum depth was 15.2 m. and the transparency 5.8 m., and according to Inaba's observations, in the winter of 1934, the protein nitrogen content was 34 mg./l.

The depth of Yamanaka-ko, the largest of the five lakes, is small for its area. In the south, it is bordered by a gentle slope of Mt. Huzi, while its northern shore is surrounded by the low Dosi mountains. These facts contribute to the agitation of the lake water by strong winds which are so common in this region, and consequently the water here does not stagnate to the extent that it does in Kawaguti-ko and in Syozi-ko. In the winter of 1934, the transparency was 5.4 m. and the protein nitrogen content 48 mg./l.

III. Studies of Mud.

Studies of mud are important for the following reasons:

First, they furnish information concerning the roles played by mud in supplying nutrient matter to the lake water.

---

(6) Inaba, *Jap. Journ. Limnol.*, **4** (1934), 101–114.

(7) Miyadi, *Japan. J. Zoology*, **4** (2) (1932), 82–100.

Secondly, since the mud consists of organic or inorganic debris that either has been formed in the lake or been carried into it at a certain time and precipitated there, they frequently indicate the condition of the lake in former times.

Thirdly, there is the problem of the changes that occur in mud, which can be answered only by a study of mud itself.

We may mention the names, B. W. Perfiliew,(8) Ch. S. Black,(9) D. S. Rawson,(10) R. T. Allgeier, W. H. Petersen, Ch. Juday and E. A. Birge,(11) W. Ohle,(12) G. Karsinkin and S. Kusnetzov,(13) all of whom have investigated mud along one or another of the lines just mentioned, resulting in the acquisition of much valuable knowledge. But, notwithstanding their importance, we have not yet succeeded in extracting sufficient knowledge from them to help us to solve the main problems, so that further studies are needed.

Seasonal Variation in the Intensity of the Physical, Chemical, and Biological Factors Affecting Mud Metabolism.

Separate studies of the physical, chemical, and biological factors that may affect metabolism in muds have been carried out repeatedly, but systematic investigation, wherein a number of these factors is treated at the same time in order to secure correct judgement on the correlations between them, has been very rare, as also studies for a protracted period of the variability in the intensity of any of these factors. The following is the result of observations made with the object of filling our present lack of knowledge in these respects, for which purpose, the writer visited Takasuka-numa eleven times in all, from November 1932 to December 1933, and examined the muds from the deepest part of the lake for their temperature, *p*H, bacteria, and macroscopic benthoic animals.

Methods.

Sampling and Measurement of Temperature. For sampling the muds, a Lenz's sampler was used, so devised that, by inserting metallic plates, the muds sampled could be divided horizontally into a few layers. The number of layers, thus divided, varied from six to eight, according to the thickness of the muds dredged.

The sampler has three holes, lying vertically in a line on one of its walls, each with a screwed lid. Immediately after the sampler was hauled up into the boat and its contents were divided into layers by metallic plates, the mud temperature was read in the following manner.

The upper hinged cover of the sampler is opened and the bulb of a thermometer is inserted directly into the mud, and its temperature determined, that is, of the surface mud.

---

(8) Perfiliew, *Verhandl. f. Internat. Vereinigung f. theoret. u. angew. Limnologie*, **4** (1929), 107-143.

(9) Black, *Transact. of the Wisconsin Acad. of Sciences, Arts and Letters*, **24** (1929), 127-133.

(10) Rawson, *Univ. of Toronto Studies, Public. of the Ontario Fisheries Research Laboratory*, **40** (1930), 1-183.

(11) Allgeier, Petersen, Juday and Birge, *Internat. Rev. d. gesamt. Hydrobiol. u. Hydrograph.*, **26** (1932), 444-461.

(12) Ohle, *Biochem. Z.*, **258** (1933), 420.

(13) Karsinkin and Kusnetzov, *Arbeiten der Limnologischen Station zu Kossino*, **13-14** (1931), 47-68.

The values of the intermediate layers are determined by merely removing the lids of the holes above mentioned, and inserting the thermometer bulb into the mud through them.

Finally, the temperature of the deepest mud is determined, first, by opening the bottom of the sampler and emptying out the mud of the deepest layer into a receiving vessel, after which the thermometer is introduced.

Determination of the *p*H-value. The sample was carried cold to the laboratory and kept in an ice-chest. 1 c.c. of the thoroughly kneaded mud is mixed well with 2 c.c. of distilled water for one minute, after which a knife-tipful of quinhydrone is added and the mixture stirred cautiously for half a minute. A platinum electrode is then inserted, and the *p*H determined potentiometrically in three minutes, referring to a saturated calomel cell.

Since it frequently happens that the room temperature, at which the *p*H-value is determined in the laboratory, differs from that in the mud as it lay in the lake bottom, naturally, the *p*H-value thus estimated differs from the value in its natural condition. The writer, therefore, studied, as a separate subject, the relation between the *p*H-value of the muds and the temperature, and constructed a *p*H-temperature curve, from which the observed values could be corrected into natural values.(14) Table 5 gives the values thus corrected.

Bacteriological Examination. The muds were transferred into a number of sterilized Petri-dishes and kept cold. The work of counting the number of bacteria by the plate culture method was completed in 24 hours. The method is mainly after E. J. Russel(15). As counting medium, mannitol-agar-asparagine was used, and its *p*H regulated to 7.6 by caustic soda.

1 c.c. of the mud is shaken with 99 c.c. of sterile salt solution, containing NaCl and $MgSO_4$, after which 1 c.c. of what is suspended in the solution is diluted with the same sterile salt solution. 1 c.c. of the latter suspended matter is further diluted with another 99 c.c. of salt solution. From the suspension thus prepared, 1 c.c. is transferred into each of five Petri-dishes, into which the sterile counting medium is poured molten. After ten days incubation at 20°C., the number of colonies developed is counted.

Examination of Macroscopic Benthoic Animals. Each layer of the sampled mud, divided as already mentioned, was sieved by shaking in water through wire netting with meshes of 1 mm. *Corethra* and Orthocladiinae larvae, which represent the benthoic fauna of this lake, were then picked out from the residue. The number of individuals found in each divided layer and their sum are given in Table 7. The sum, multiplied by 44, is conventionally converted into a value per 1 m.^{2}, seeing that the sampler dredges mud (given in the same table) covering a bottom area of 225 cm.^{2} at a time.

Results and Discussions.

Temperature. The lowest value observed was 5.3°C, which was found on February 11, 1932, in the 1st layer. The highest value, which was observed on September 17, was 17.2°C in the 3rd layer in the deepest part of the lake, and 19.3°C. in the 1st layer at 4.7 m. depth. Thus the mud temperature, which varies from 5.3°C. to 17.2°C. throughout the year in the deepest part of Takasuka-numa, seems to attain its maximum during the period from the end of September to the middle of October.

It is also noteworthy that, during the period from December 1932 to the following February, the temperature of the muds increased from its surface towards its deeper parts, indicating that heat flows from the muds

---

(14) Sugawara, *Bull. Chem. Soc. Japan*, **9** (1934), 446.

(15) Russel, "Soil Condition and Plant Growth," 6th edition, 579, London, (1932).

into the lake water during this season, while during the period from March to the end of September, matters are reversed, the mud temperature increasing from the deeper parts towards the surface.

There are, therefore, two periods in which the flow of heat occurs in mud in opposite directions, between which are two others, where the flow is minimized.

These points will be better understood if we compare the temperature of the muds with that of the bottom water, which is given in the bottom row in the table. But here we meet a difficulty. On March 30 and on April 17, the temperature of the bottom water was lower than that of the muds. Had the temperature relation been reversed, that is, had the temperature of the water been higher than that of the muds, the circumstance could easily be explained. This difficulty should be settled by further studies.

The presence of temperature gradients along the vertical line and its seasonal reversal in sense are of special importance to mud metabolism. They may, on the one hand, affect the migration of benthos either from the surface layer towards the deeper layer or in reverse direction; they may also lead to increase in the vicissitudes of the microflora in different parts of the muds. On the other hand, they may affect the equilibrium, purely physical and chemical, in muds.

For example, they could control the diffusion of dissolved or colloidal dispersed matter through muds by capillary action, which matter could sometimes diffuse into the overlying bottom water, or, contrary to it, they may, another time, be carried back into the deeper muds, and accumulated there to a higher concentration, and thus give rise to unequal distribution of matter in various parts of the mud.

Table 4. Temperature of Muds in °C. (1932–33)

Depth	Dec. 28	Feb. 11	Mar. 30		Apr. 17	May 7	June 11	Aug. 11	Sept. 17		Oct. 22	Dec. 3
			(a)	(b)					(a)	(b)		
↓	5.5	5.3	9.1	9.5	10.3	13.4	14.5	—	16 6	19.3	17.0	9.4
	5.6	5.4	9.1	—	9.8	12.8	13.7	—	16.6	18 1	17.1	9.7
	5.9	5.5	9.0	9.0	9.6	12.5	13 8	—	17.2	17.8	17.0	10.3
	6.1		8.3	8.6	9.1	12.4	14.0	—	16.4			10.9
	4.6	5.0	8.0		8.7	14.1	15.8	15.9	17.3		16.5	8.8

The pH-value. (1) No marked seasonal or vertical fluctuation in pH could be noticed. It varied from 5.70 to 7.54; the former value was recorded on September 17 in the top layer and the latter on February 11 in the 4th layer.

(2) It is notable that the top layer has always a smaller pH than the deeper ones, which is assumed to be due to the following two causes:

First, during the period of circulation, some constituents of the surface muds are oxidized into acidic substances by the oxygen dissolved in the overlying water, whereby the pH of the mud is diminished.

Secondly, decrease in pH can also result secondarily. Very often the amount of mud in the top layer is not sufficient to fill the Petri-dish, leaving an air space in it, which is never the case with the other layers, where a larger quantity of mud is always available. Thus, while awaiting its turn to be examined, the mud of the top layer, being exposed to atmospheric oxygen, is oxidized, thus resulting in decrease of its *p*H. The smaller *p*H shown by the top layer mud during the period of stagnation, can be explained by these two reasons.

(3) It is also notable that *p*H tended to diminish from winter to summer, where it attains its minimum, and increases towards autumn. On February 11, the *p*H-values in almost all the layers were exceptionally small, of which no adequate explanation has yet been made.

(4) In comparing the distribution of benthoic animals with the data of *p*H, it is remarkable that, in winter and spring, when the density of *Corethra* larva is larger, the variability in *p*H in the different layers of mud is larger than in summer, when the larval density is greatly reduced.

Table 5. The *p*H of muds (1932–33).

Depth	Nov. 12	Dec. 28	Feb. 11	Mar. 30	Apr. 17	May 7	June 11	Aug. 11	Sept. 17	Oct. 22	Dec. 5
I	6.40	7.05	6.67	6.29	6.44	6.92	6.57	6.62	5.70	—	6.87
II	6.72	7.07	6.92	6.70	7.07	7.08	6.85	6.71	6.77	6.74	6.96
III	6.73	7.03	6.77	7.07	6.92	6.94	6.85	6.73	6.87	6.92	6.97
IV	6.54	7.23	7.54	6.96	7.13	6.92	6.82	6.75	6.90	6.89	6.97
V	6.53	7.05	6.85	7.01	6.71	6.88	6.77	6.82	6.91	6.99	6.91
VI	6.26	7.09	6.93	7.00	7.02	6.84	6.81	6.84	6.91	7.02	6.94
VII		6.98	6.95	6.95	6.85		6.79	6.81	6.92	6.98	7.00
VIII		7.03					6.80	6.85			6.85

Table. 6. Variation in bacterial number (millions per 1 c.c.) (1932–33).

Depth	Nov. 12	Dec. 28	Feb. 11	Mar. 30	Apr. 17	May 7	June 11	Aug. 11	Sept. 17	Oct. 22	Dec. 5
I	0.54	1.40	1.13	1.05	0.49	0.82	.	0 77	0.53	0.60	0.68
II			1.14	1.02	1.00	1.35	1.18	0.78	0.51	0.56	0.90
III	0.67	1.27	1.03	1.00	1.30	1.24	1.16	0.91	0.47	0.54	0.52
IV			0.98	0.65	0.87	1.21	1.02	1.00	0.58	0.42	0.45
V	0.63	1.52	1.42	0.91	1.05	1.03	1.16	0.85	0.59	0.60	0.57
VI	0.53		1.35	1.08	0.79	0.86	0.85	0.17	0.36	0.58	0.58
VII		1.32	1.33	0.51	0.85		0.44	0.11		0.64	0.50
VIII		0.98			0.74		0.60				0.57

Further, it is worth mentioning that the layer with a considerably larger *p*H is always that in which more *Corethra* larvae are counted.

On February 11, contrary to the general relation above, *Corethra* larvae were most abundant in the 3rd layer, while maximum *p*H was seen in the 4th layer. This discordance, however, can be explained by the fact that, on that day, the determination of benthos and that of *p*H were carried out on separate samples. In that case, should the thickness of the muds dredged by one operation differ from that of the next, there is the contingency, notwithstanding all precautions, hence remote, that a particular zone of the layers in one dredging may not exactly correspond to that in the layer of another.

Bacteriological Examination. (1) The number of bacteria reached maximum on December 28, 1932, in the 5th layer and minimum on August 11, the following year, in the 7th layer. In a year, the number varies between the two extremes of 100,000 and 1,500,000.

(2) On the other hand, there is a tendency for the number to grow somewhat towards winter and spring and diminish towards summer and autumn a variation in the number of bacteria that seems to be intimately related to the distribution of *Corethra* and Orthocladiinae larvae. As a matter of fact, on December 28, 1932, when the benthoic animals were most abundant, the number of bacteria attained also its maximum. Thence until May 7, the following year, there was only a barely discernible decrease.

Now, in June, especially in the 6th and 7th layers, a marked decrease was seen in the bacterial number, which extended towards the top layer and continued until September 17.

On August 11, when, in the 7th and 8th layers, no individuals of *Corethra* or Orthocladiinae larvae could be found, the number of bacteria fell to the order of 100,000.

In September, the bacteria began to increase in number.

(3) On comparing the data for December 3 with those in the previous December, it was found that the former values were much smaller than those in the latter. This difference can easily be explained when we consider that, in the previous December, the observation was made later in the season by practically one month than that in the following year, so that, during that interval of one month, a sudden increase in number of bacteria was quite possible.

The Examination of Macroscopic Benthoic Animals. From the observation of benthoic animals, the following results were obtained.

(1) During the greater part of the year, the muds at all the depths examined were inhabited by a considerable number of *Corethra* or Orthocladiinae larvae, by whose movements the muds at different depths can be mixed, affording chances for the products of mud metabolism to be carried into the lake water, and for certain substances in the overlying water to be brought into the deeper parts of the muds.

The greater part of the bacteria mentioned in the previous article may have entered the muds by this latter course. Thus their number greatly declined in August, when the number of *Corethra* larva was also reduced.

(2) The number of *Corethra* larva increased from the end of

December 1932, to the following February, after which there could be found no marked change until May, when the number began to decrease until September 17, when it attained its minimum value and began to increase towards winter.

Thus the number of *Corethra* larva in Takasuka-numa varies between 1190 and 8700 per m.^{2}

Table 7. Variation in the distribution of macroscopic benthoic animals (1932–33).

Depth	Dec. 28				Feb. 11		Mar. 30		Apr. 17		May 7	
	(a)		(b)									
	Cor.	Orth	Cor.	Orth	Cor.	Orth.	Cor.	Orth.	Cor.	Orth.	Cor.	Orth.
I	1	0	5	1	9	0	0	0	4	1	6	2
II	8	0	13	0	35	1	5	0	20	5	8	1
III	12	0	30	0	53	1	44	2	34	6	15	0
IV	35	1	31	2	35	1	29	4	66	7	19	4
V	50	8	33	1	3	1	23	1	24	1	15	2
VI	8	1	9	1	5	2	2	5	8	0	6	0
VII	0	1					1	1	2	0		
VIII	5	0										
Sum	119	11	121	5	198	6	104	13	158	20	69	7
×44	5240	480	5300	220	8700	260	4600	570	6950	880	3040	310

Depth	June 11		Aug. 11		Sept. 17		Oct. 22		Dec. 3	
	Cor.	Orth.	Cor.	Orth.	Cor.	Orth.	Cor.	Orth.	Cor.	Orth.
I	—	—	3	0	8	0	3	0	2	0
II	6	1	7	0	5	0	3	0	4	0
III	29	0	13	0	5	1	7	1	5	0
IV	20	0	8	0	7	0	9	0	14	0
V	19	1	0	0	1	2	34	0	8	0
VI	2	0	0	0	1	1	17	0	16	1
VII	1	1	0	0			6	1	15	0
VIII									1	0
Sum	77	3	31	0	27	4	79	2	65	1
×44	3390	130	1370	0	1190	180	3480	90	2860	44

Cor. = *Corethra* larva. Orth. = Orthocladiinae larva.

(3) As to the vertical distribution of the animals, the following points were established. The zone that showed maximum density of animals is in the 5th layer on December 28, 1932, later shifting upwards to the top layer, in September.

The sense of shift thenceforth reversed, and the zone of maximum density suddenly dropped to the 5th layer in October and to the 6th in December.

Thus the zone of maximum density of benthos, which shifts upward first, does so downward in the latter half of the year, forming an annual cycle.

Finally, no individual was found on December 28, 1932, in the 7th layer, and on March 30 in the top layer, and on August 11 in the 6, 7. and 8th layers.

(4) Orthocladiinae larvae were too scanty to offer a clue to any reliable relationship, although they showed that their number increased from December to April, after which it decreased. In August, the animal disappeared completely from all mud depths, after which it began to increase in number again.

The Chemical Composition of Muds.

Determination of the chemical composition of the muds of a lake is indispensable to systematic study of lake metabolism. On the one hand, it provides knowledge of the transitions the lake has undergone, while, on the other, it gives us some knowledge of the metabolism that has occurred, or is occurring, in the muds themselves.

Many examples of chemical analyses of lake deposits have been reported. In the older examples of such analyses, attention was paid more to the mineral constituents than to the organic ones, the knowledge of which latter, however, is more important to the present problem.

The greater part of recent studies, in which the organic constituents have been treated, are of little help to us, because the objects of their research differ from ours.

Thus, for example, W. Ohle(12) determined the organic constituents of the bottom deposits of the "kleines Ukleisee" from the top layer, every 1 m. vertically down to 11 m. This study, the purpose of which was to obtain support for the "lignin theory" of Fr. Fischer, contributed much to the knowledge of the geological transition of the lake itself and of its neibourhood, but not so much to that of lake metabolism.

The same could be said of many other studies, which were undertaken for the purpose of elucidating the mechanisms of petroleum formation.

The only example of organic analysis of lake muds performed from the view-point of pure limnology, is that of S. Kusnetzov,(16) who estimated the sugar, pentosan and hemicellulose, cellulose, lignin and humus, wax and pitch, fatty acids, etc., of samples from two different layers that were 0–5 cm. and 25–40 cm. from the surface of the muds in Lakes Beloje, and 0–5 cm. and 10–15 cm. from the same in Glubokoje, both near Moscow.

The following analysis by the writer was started as an indispensable item in a programme for studying lake metabolism; it brought about some results worth mentioning.

These results, together with those of Kusnetzov, help us in contributing data for geochemistry a long neglected branch of science.

---

(16) Kusnetzov, *Verhandl. d. Internat. Vereinigung f. theoret. u. angew. Limnologie*, **7** (1935), 571-582.

Methods.

The muds that were analysed were dredged on February 11, 1933, from the deepest part of Takasuka-numa. They were 17.5 cm. thick, divided into seven layers, each 2.0 cm. thick, except the bottom layer, which was 5.5 cm. thick. A part of each of the seven kinds of samples thus prepared, was used in determining the bacteria, as shown in the preceeding article, while the remainder was used for the chemical analyses, which were made as follows.

First, the density of the sample is determined. Then, by drying at 110°C, the water content of the muds is determined. With the dried sample, the ignition loss and its mineral constituents are determined.

Determination of the Mineral Constituents. *Determination of silicon, aluminium, and iron.* For determining the mineral composition, about 0.5 g. of the dried preparation is fused with carbonate mixture, and its silica content determined after the usual method. The filtrate from the silicic acid is made up to 50 c.c., 8 c.c. of which are used for precipitating the aluminium and iron. The precipitated iron and aluminium are dissolved in a small quantity of hydrochloric acid and then diluted to 50 c.c., 20 c.c. of which solution are evaporated to dryness. To the residue, after cooling, are added 5 c.c. of conc. hydrochloric acid. The residue immediately dissolved into a yellow solution, the content of iron in which is determined by the iodometric method of Blackwood-Stirling.(17)

From another 8 c.c. of the 50 c.c. filtrate from the silicic acid, iron and aluminium are precipitated as mixtures of their hydroxides, ignited and then weighed as a mixture of ferric oxide and aluminium oxide.

The content of aluminium in the dried preparation is calculated as alumina by reducing the iodometric iron value from the total weight of iron and aluminium oxides.

Determination of manganese. 20 c.c. of the filtrate from the silicic acid are used. Hydrochloric acid gas is driven off by evaporation until the residue remains slightly moist. 30 c.c. of water are added, and the residue is dissolved by warming. 0.5 c.c. of colourless concentrated nitric acid and a sufficient quantity of 1% silver nitrate solution are added to precipitate the chlorine ions. 2–3 c.c. in excess of silver nitrate solution are then added and the solution filtered. The precipitate is washed with about 50 c.c. of dilute nitric acid. The filtrate is now evaporated down to about 10 c.c., to which is added 5 c.c. of 10% ammonium persulphate solution. After warming for ten minutes on a water bath, it is diluted with water to 30 c.c. and then compared with standard potassium permanganate solution, 1 c.c. of which contains 0.02 mg. of manganese.

Determination of calcium. From the 50 c.c. filtrate from the silicic acid, 20 c.c. are taken, and after separating the iron and the aluminium, the filtrate is neutralized with hydrochloric acid, using methyl red as indicator, and 5 c.c. of ammonium sulphide solution added. After setting aside for 24 hours, the precipitate is filtered off and the filtrate boiled for some time. The precipitate formed during the boiling is filtered off. The filtrate is acidified with hydrochloric acid, evaporated to less than 10 c.c., and neutralized with ammonia and diluted to 30 c.c. To the resulting solution is added, in turns, 5 c.c. of 0.025 N oxalic acid and 0.5 c.c. of 0.1 N hydrochloric acid. The liquid is then neutralized by the addition of 0.2 N ammonia, a drop being added at a time, with 5 minutes' interval after each addition. Neutralisation is continued until the *p*H of the liquid is 7.8. A further 0.6 c.c. of ammonium oxalate is then added. After two days, the precipitate is filtered off, washed with saturated ammonium oxalate solution, which is regulated to a *p*H of 7.8, and then ignited in a small platinum crucible.

The ammonium sulphide solution is prepared as follows: Concentrated ammonia is diluted with an equal volume of water, and hydrogen sulphide gas is passed through the solution to saturation. The resulting solution is diluted with the same volume of half-diluted ammonia.

---

(17) Blackwood and Stirling, *Biochem. J.*, **26** (1932), 353.

Determination of phosphorus.(18) 2 c.c. of the 50 c.c. filtrate from the silicic acid, are diluted to 10 c.c. after which 1.2 c.c. of 60% perchloric acid, 1 c.c. of 5% ammonium molybdate solution, and 0.5 c.c. of 1 aminonaphthol-(2)-4-sulphonic acid solution are added, and water to make the whole 15 c.c.

A standard containing 1 mg. of P_2O_5 per 1 c.c. and a quantity of ferric chloride equivalent to the iron content of the sample solution is prepared at the same time and in the same way.

The contents of the flask are gently shaken between each addition, and finally mixed by inverting and shaking. The colours are read after five minutes in a Duboscq colorimeter.

The 0.2% aminonaphtholsulphonic acid is prepared as follows: 0.5 g. of the acid, 40 g. of sodium bisulphite, and 6 g. of crystalline sodium sulphite are dissolved by shaking with enough water to make 250 c.c. If the solution does not filter clear, it should be left overnight and again filtered. A fresh solution should be prepared every two weeks.

Determination of the Organic Constituents. Separate portions of the dry preparation are used for determining carbon and hydrogen, total nitrogen, ammoniacal nitrogen, and for the complete organic anaylsis.

(a) Carbon and hydrogen are determined by microanalysis after Pregl, for which the writer is much indebted to Dr. Shu Furukawa.

(b) For the determination of total nitrogen, 0.1 g. of the material is taken in a small Kjeldahl flask. 1 c.c. of concentrated sulphuric acid, three drops of 5% copper sulphate solution, and 0.05 g. of potassium persulphate are added successively, and the flask is heated for a few hours until the liquid is completely decolourised, when the ammonia content of the liquid is measured in the usual way.

(c) For the determination of ammoniacal nitrogen, 0.5 g. of the dry material is suspended in 30 cc. of distilled water, to which is then added 0.5 c.c. of 1% soda solution, which is sufficient to bring the *p*H of the liquid to that point where the bromothymol-blue turns blue. Through the suspended matter, air is bubbled for some 40 minutes. The air, here used, is previously freed from traces of ammonia by concentrated sulphuric acid. The ammonia driven off, is absorbed by 10 c.c. of 1/70 N hydrochloric acid solution and estimated by the usual method.

(d) The complete organic analysis is carried out mainly after S. A. Waksman,(19) on two or more of 2 g. portions of the dry material.

The procedure is as follows: The material is first extracted with ether in Soxhlet for 20 hours. For this process, 100 c.c. of ether are used, 50 c.c. being added at the start, and the remaining 50 c.c. later. The ether-soluble portion of the material is then determined by evaporating the ether, then drying the residue in a weighing bottle at 90°C. until a constant weight is attained. The residue, left after the ether treatment, is separated from its container, the cylindrical filter paper, by cutting the latter and washing out the contents into a beaker. Water is added to 100 c.c. and the suspended matter is shaken overnight, after which the liquid is filtered and washed with 100 c.c. distilled water for 10 hours. The filtrate from the cold water digestion is condensed into 50 c.c., of which 5 c.c. is used for determining the reducing sugars.

The residue from the cold-water treatment is now digested with 100 c.c. of hot water for two hours in a water bath. The liquid is filtered off and the residue washed with hot water until the filtrate is colourless. The filtrates are combined and evaporated to a small volume, in order to determine the reducing sugar contained. The residue is dried and then extracted with 100 c.c. of boiling alcohol for 5 hours. The filtrates are combined and evaporated, in weighed dishes, to a constant weight at 90°C.

The residue is dried and extracted with 100 c.c. of 2% hydrochloric acid for 5 hours. The hot extract is filtered through filter paper that has been previously dried and weighed, the residue being washed into the filter and treated with distilled water until free from acid. The combined filtrate and washings are analysed for total nitrogen and total reducing sugar (an aliquot portion of the solution having been

---

(18) King, *Biochem. J.*, **26** (1932), 294.

(19) Waksman and Tenney, *Soil Science*, **24** (1927), 275–283.

neutralised with 40% sodium hydroxide solution). To get the hemicellulose content of the mud, the total reducing sugar is next multiplied by 0.9.

The residue left on the paper after the treatment with dilute acid is dried for 24 hours at 70°C. and weighed. About a 0.3–0.6 g. portion is taken from the dried residue and treated with 5 c.c. of 80% sulphuric acid. The acid is allowed to act in the cold for 2 hours and then 75 c.c. of distilled water are added, and the content is boiled for 5 hours under a reflux condenser. The extract is now filtered through a small dried and weighed quantitative filter paper. The residue is washed thoroughly with distilled water, dried at 70°C., and weighed and finally divided into two portions, one of which is ignited to determine the ash and the other is used to determine the total nitrogen. The weight of the residue after the sulphuric acid treatment, minus the sum of the weight of the ash, and of the total nitrogen which has been multiplied by 6.25, gives the lignin content of the 0.3–0.6 g. portion left after the treatment with dilute hydrochloric acid. From this value, the percentage of lignin in the initial substance is further calculated.

The filtrate from the 80% sulphuric acid solution is analysed for sugar, an aliquot portion having been neutralised previously with 40% caustic soda solution. The reducing sugar multiplied by 0.9 gives the cellulose content in the portion.

The determination of reducing sugar is carried out mostly according to Ch. S. Hanes.⁽²⁰⁾

Results and Discussions.

When we compare the data of analysis (Tables 8 and 10) with those of other lakes, whether in Japan or abroad, we easily find a number of facts of special interest, whose description, however, will be here dispensed with, owing to risk of straying from the main object.

Vertical Variations in Mineral Constituents. The mineral constituents may be classified into three groups, according to their modes of vertical variation.

Table 8. Vertical variation in the chemical composition of the mud from Takasuka-numa. (A)

Depth	I	II	III	IV	V	VI	VII
Density	1.250	1.260		1.260	1.263	1.280	1.285
Weight Decrease (%) (110°C)	79.61	78.97		78.50	78.08	76.08	75.60
Loss on ignition (%)	18.69	18.94	18.94	19.08	18.83	18.12	17.95
SiO_2 (%)	47.48	47.79	47.73	48.03	47.93	46.88	46.29
Fe_2O_3 (%)	14.30	14.37	14.23	14.46	14.84	15.51	16.22
Al_2O_3 (%)	13.63	14.29	14.60	15.29	14.49	14.08	13.63
MnO (%)	0.64	0.60	0.61	0.61	0.66	0.74	0.76
CaO (%)	2.06	1.98	1.83	1.71	1.58	1.58	1.86
P_2O_5 (%)	0.17	0.16	0.14	0.23	0.17	0.18	0.15

Group. I. Silica and alumina belong to this group. Their percentages grow from the top layer towards the fourth, where they attain

---

(20) Hanes, *Biochem. J.*, **23** (1929), 99.

their maxima, after which they diminish with depth. The loss on ignition also varies in parallel with the foregoing.

Group II. The iron and manganese oxides belong to this group. Their percentages grow evenly from the top towards the deepest layer, except two anomalous values, which are found for iron in the third layer and for manganese in the top layer. This anomaly in manganese is possible of satisfactory explanation, as will be seen later.

Now, although a part of the iron and manganese may come as components of dead plankton, it is, at any rate, insignificant, while the remaining major part probably comes as ingredients of the clayey substances that are carried into the lake by influent waters.

Notwithstanding this, the mode of vertical variation in the iron and manganese oxides differs entirely from that of alumina and silica—a point on which a satisfactory explanation is desired.

First we shall examine the ratio of ferric oxide to silica (Table 9). It is remarkable that the ratio keeps constant from the top to the fourth layer, its value lying between 0.301 and 0.298, while the ratio increases further, downwards, to 0.350 in the seventh layer. The same may be said of manganese. The ratio of manganese oxide to silica, except in the top layer, varies between 0.0126 and 0.0127 to the fourth layer, after which it begins to increase downwards until it is 0.0164 in the seventh layer, whence we may conclude that, in muds of about 8 cm. thickness from the surface, the four elements, silicon, aluminium, iron, and manganese are distributed quite uniformly.

Table 9. The ratios of iron, aluminium and manganese to silicon as their oxides at various depths of the mud of Takasuka-numa.

Depth	I	II	III	IV	V	VI	VII
Fe_2O_3	0.301	0.301	0.298	0.301	0.310	0.331	0.350
Al_2O_3	0.286	0.298	0.300	0.318	0.302	0.300	0.294
MnO	0.0134	0.0126	0.0128	0.0127	0.0136	0.0157	0.0164

Conditions differ, however, at depths below 8 cm. Here the ratios of iron and manganese to silicon are no longer constant; they increase with depth.

In contrast to the foregoing, the ratio of aluminium to silicon seems to decrease at the seventh layer, although the decrease is much smaller in amount compared with the increase in iron or manganese. Moreover, accuracy in analysis in the case of aluminium falls below those of the other elements in question. Thus, when we take these circumstances into consideration, we see that it is better, instead of stressing the matter of decrease, to conclude, as just mentioned, that the ratio of alumina to silica is practically constant through all the depths that were examined.

Explanation I. The simplest explanation possible from the foregoing examination is that the ratios of iron and manganese relative to the elements of the first group that were all carried into the lake, varied gradually in decreasing proportion from the time when the deepest muds deposited until the muds of the fifth layer settled. Then the variation in the ratio stopped; further deposition occurred and continued at the fixed ratio until the present.

Explanation II. There is another possible explanation, in which the variation in the ratios of iron and manganese to silicon and aluminium are assumed to have continued to occur up to the present time, and the uniform distribution of the four elements in question in the upper layers of mud is assumed to have been brought about secondarily by the micro-organisms present. In fact, the upper four layers (8 cm.) are densely inhabited by *Corethra* and Orthocladiinae larvae (Table 7), by the movements of which the upper and lower muds are mixed, resulting in uniform distribution of the elements in question, while the layers below 8 cm., which are inhabited by fewer of these insect larvae, are subject to less mixing, hence retaining the previous mean ratios.

Explanation III. In the preceding two explanations, we assumed that the ratios of iron and manganese to silicon and aluminium had varied during deposition of the mud layers in question.

We shall now examine this assumption. As a matter of fact, the time during which the mud layers in question had been deposited, is easily estimated from the result given in page 00 as three years, at least, but not much longer. That during such a short period of time, there should have occurred a considerable change in the ratios of the four elements in question that were carried into the lake by influent waters, is rather improbable. Therefore, in the third explanation, we must try to suppose that the ratio of the iron, manganese and silicon that were carried into the lake water remained constant during the time in question, and further suppose that the apparent change in the ratios occurred secondarily, in the course of settling, or after they had already settled on the bottom.

In order to follow this reasoning, it is necessary to note a certain peculiar property that distinguishes iron and manganese from silicon and aluminium.

The first to be mentioned is the oxidation-reduction property of iron and manganese. These elements are, indeed, deposited in higher oxidized forms, after which, owing to anaerobiosis in the deeper water layers or at the bottom, they are reduced partly into soluble or easily diffusible forms. When this occurs to any considerable extent, and if this occurrence grows more intense year by year, the result is nothing but a vertical decrease in the ratios in question towards the surface, that is, the ratio has a smaller value in the recently precipitated muds than in the older ones, notwithstanding the fact that during the time in question, there was no considerable change in the ratios of these elements that were carried into the lake water.

Now this idea, that the reduction of iron and manganese has been growing more intense during the time in question, seems to be supported by the data of organic analysis. Here the ratios of the various organic constituents of planktonic origin to silicon are also found to increase on the whole towards the surface, which can be explained by saying that the deposited quantity of organic debris has been increasing, that is, the lake has become more eutrophic and consequently the reductive changes in the lake have grown more intense.

It must be added that, in the present case also, two different ways are conceivable in which the lake became more eutrophic. When we stress the importance of the mixing action of benthoic animals, eutrophic action cannot be considered to have continued, not at least, at the same rate until the present, otherwise we have to agree that eutrophication stopped at the time when the fifth layer muds had settled.

Unfortunately, our knowledge at present is still too meagre to enable us to say which of the various explanations suggested above is correct.

Group III. Calcium oxide belongs to this group. Although its percentage decreases evenly towards depth, it suddenly begins to grow from the seventh layer.

Calcium differs from other elements in the way in which it is carried to the bottom. In fact, the water of Takasuka-numa, like in many other Japanese lakes, is so poor in calcium that the concentration of this ion is much less than that from which the calcium carbonate is precipitated, so that if calcium be carried to the bottom, it must occur in one of the following two ways.

Table 10. Vertical variation in the chemical composition of the mud from Takasuka-numa. (B)

Depth	I	II	III	IV	V	VI	VI
Carbon (%)		6.06			6.03		5.81
Hydrogen (%)							1.50
Total Nitrogen (%)	0.719	0.852	0.770	0.702	0.700	0.660	0.638
Ammoniacal nitrogen (%)	0.005	0.007	0.005	0.002	0.004	0.006	0.007
C/N		7.11			8.62		9.10
Ether soluble fraction (%)	0.43	0.47	0.65	0.65	0.65	0.50	0.48
Alcohol soluble fraction (%)	0.59	0.59	0.61	0.62	0.51	0.56	0.50
Reducing sugars soluble in cold water (%)	0.24	0.24	0.19	0.24	0.23	0.21	0.18
Reducing sugars soluble in hot water (%)	0.23	0.24	0.24	0.21	0.25	0.21	0.20
Reducing sugars soluble in cold or hot water (%)	0.47	0.48	0.43	0.45	0.48	0.42	0.38
Hemicelluloses (%)	3.37	3.44	3.65	3.09	3.68	3.54	4.16
Cellulose (%)	0.77	0.86	0.82	0.79	0.92	0.90	1.09
Lignin (%)	3.60	3.86	3.65	3.54	3.82	3.40	3.74
Ash (%)	43.8	43.8	44.1	43.3	41.7	42.3	42.9

(1) Calcium, which is assimilated first as a body substance of planktonic organisms, is next deposited as a constituent of organic debris.

(2) When certain flocculent precipitates are formed in the lake water, for example, the coagulum of ferric hydroxide or that of planktonic debris, these coagulations tend to adsorb calcium ion from the water and to carry it to the bottom.

Of these two ways here assumed, the former accords with the view of the lake having become more eutrophic. As eutrophication progresses, the deposited amount of planktonic debris increases, with the result that much more calcium is carried down to the bottom, so that the percentage of calcium oxide is larger in the later deposits, namely, in the upper layer. But it is difficult to explain the sudden increase in the percentage in the deepest layer by this assumption, and this point can be interpreted more easily by means of the latter explanation.

These are points on which further researches, it is hoped, will be made.

Finally, the quantity of phosphorus being too small to give precise analytical results, it is not possible to say in what group it should be placed.

Vertical Variations in Organic Constituents. The results of organic analysis cannot be regarded as sufficiently accurate, there being many difficulties in the method itself, and not enough samples being at our disposal, but the main features can be seen at a glance.

(a) The total nitrogen decreases with depth.

(b) The total carbon seems to decrease in the same way.

(c) The carbon-nitrogen ratio increases from 7.11 in the 2nd layer to 9.10 in the 7th layer.

(d) The amount of ether-soluble fraction is large in the middle layers, but smaller in the upper and deeper layers.

(e) The alcohol-soluble fraction behaves similarly but less evidently.

(f) The amounts of the sugars soluble in cold and in hot water do not greatly differ. Their totals tend to decrease from the 5th layer with depth.

(g) Hemicelluloses, cellulose, and lignin show no marked variation except in the 7th layer, where their percentages are evidently larger.

In summarising the results, we conclude that nitrogeneous and ether- or alcohol-soluble matters are distributed rather densely in the upper layers, and that the celluloses and lignin are in the deeper layers.

As one of the most probable causes that contribute to the development of such distribution of organic substances, we might mention the contingency of the lake becoming more eutrophic during the deposition in question. Thus the composition of muds has varied in such a way that the upper layers, that deposited later, are more planktonic, that is, they are richer in nitrogeneous and in ether- or alcohol-soluble substances. Therefore the celluloses and lignin, mainly of littoral or allochtonic origin, became smaller in their relative proportions to these substances.

Another cause, not less important to the question of distribution of matter, is the decomposition of the celluloses or lignin in the lake bottom, which is believed to become more intense the more the lake becomes eutrophic. Consequently, the proportion of these substances that have been left undecomposed will be reduced to that extent.

The effect also of the distribution of the benthoic animals and other microorganisms and that of their excrements cannot be overlooked. And since it is obvious that the substances composing the bodies of these organisms and their excrements are rich in nitrogen, and are often of alcohol- or ether-soluble nature, it is quite conceivable that the distribution of these organisms in the muds affects directly the chemical composition of such muds. But, to what extent do these factors affect them? This is a question that, it is hoped, will be solved by further studies.

The Relative Ease with which Iron and Manganese Return to the Lake Water, and its Geochemical Significance.

It is interesting to find that the ratio of iron to manganese is smaller in the lake water than in the muds, the ratio varying between 21.9 and 24.2 in the muds, as easily calculated from the data given in Table 9, while that in the lake water is, for example, 3.57 at the surface and 4.50 at a depth of 2 m., calculated as follows.

The content of iron is 0.87 mg./l., reckoned as Fe_3O_3 at the surface, and 1.19 mg./l. at a depth of 2 m., being each the mean value of the result of six observations made in June 1935–January 1936. The manganese content, also calculated similarly, is 0.23 mg./l. at the surface and 0.25 mg./l. (MnO) at a depth of 2 m. Therefore, the ratio is 3.57 and 4.50 at the two depths. It might be added that this ratio decreases in the deeper water zone (v. Tables 12 and 13).

This larger value of the ratio of iron to manganese in the mud appears to be due merely to the fact that the iron and manganese that were carried into the lake from outside, were already in that larger ratio when they were precipitated to the bottom, without any change in their relative ratio when they entered the lake, so that there is nothing odd in finding a larger ratio in the mud compared with that in the water. However, the actual situation is too complicated to be explained by such a simple cause alone.

The writer will now mention the following two characteristic properties of iron and manganese, one oxido-reductive and the other circulatory, which, no doubt, contribute much to the uneven distribution of iron and manganese, with a ratio in the mud different to that in the hydrosphere.

First, both elements have a circulatory property. As already mentioned, the iron and manganese that have settled on the bottom do not remain there long; a large part of them is transformed into the soluble state by anaerobic decomposition, after which they are returned to the water body.

Now, it is quite conceivable that this "returning" does not occur in a manner equally suitable to both elements; the manganese seems to be better fitted for this process, owing to its oxidation-reduction potential, ($Mn^{++} \leftrightarrow Mn^{++++}$), which is higher than that of iron ($Fe^{++} \leftrightarrow Fe^{+++}$).

Therefore, when the supply of oxygen to the deeper water is hindered and an anaerobic condition begins to be set up at the bottom, as it does in early spring, the oxidation-reduction potential on the spot is gradually lowered to that point where reduction of manganese is made possible, after which the manganese will be immediately reduced, while the iron oxide remains unaffected, because of its lower reduction potential, so that, of the elements in question, manganese returns first into the hydrosphere.

Further, even when the oxidation-reduction potential on the spot falls to that point where the iron can be reduced to the ferrous state, resulting in the actual reduction of the iron, the ferrous iron thus formed, is immediately reoxidized by the higher oxide of manganese, so long as the latter is present in its vicinity. Thus manganese always precedes iron in returning into the water zone.

The result of these behaviours of the elements consists in nothing more than a larger accumulation of manganese in the water, with simultaneous decrease in the ratio of manganese to iron in the mud.

This view is directly supported by the results of studying deposition. As a matter of fact, as one of the items in the study, which will be given precisely later, the writer determined the iron and manganese contents of the water in a collecting bottle that was kept suspended at depth.

The result, which is given in Table 11, shows that the concentrations of these elements in the examined water were always larger than that in the surrounding water. (v. Tables 12 and 13).

These facts indicate that the larger part of the iron and manganese in the bottle water came, at least, from the deposit that had dropped into the bottle. Moreover, the fact that the ratio of iron to manganese is smaller in the water than in the deposit (Table 15), proves conclusively that redissolution by reduction takes place more readily in manganese than in iron.

Table 11. Iron and manganese contents of the bottle water.

	Oct. 20 (1935)	Nov. 11 (1935)	Jan. 19 (1936)
Fe_2O_3 mg./l.	39.4	23.7	5.10
MnO mg./l.	12.5	22.4	0.95
Fe_2O_3/MnO	3.15	1.06	5.37

Table 12. The iron content of the waters of Takasuka-numa during June 23, 1935—January 19, 1936. (reckoned as Fe_2O_3 mg./l.)

Depth (m.)	June 23	July 24	Sept. 29	Oct. 20	Nov. 11	Dec. 15	Jan. 19
0	0.75	0.44		0.80	1.76	0.79	0.66
1		0.50	0.16	1.30	1.83		0.53
2	1.11	0.83	0.86	1.20	2.60		0.56
3	0.75	0.89	3.02	0.73	1.81	2.01	0.57
4	1.05	5.04	6.52	1.39	1.87	0.93	0.83
5	4.00	14.4	17.7	21.8	2.47		
5.5	6.75		36.9	27.9	1.75	5.87	

Table 13. The manganese content of the waters of Takasuka-numa during June 23, 1935—January 19, 1936. (reckoned as MnO mg./l.)

Depth (m.)	June 23	July 24	Sept. 29	Oct. 20	Nov. 11	Dec. 15	Jan. 19
0	0.06	0.02		0.40	0.37		0.24
1	0.05	0.01	0.11	0.85	0.50		0.14
2	0.07	0.11	0.18	0.23	0.68		0.16
3	0.07	1.11	0.45	0.70	0.45	0.18	0.20
4	0.86	0.82	1.05	0.95	0.69	0.24	0.13
5	11.2	11.2	10.3	10.6	0.84		
5.5	13.8		18.6	12.8	0.67	0.59	

Going a step further, it is expected that the ratio of iron to manganese in the lake water of Takasuka-numa has decreased to a considerable extent with lapse of time.

As said above, a part of the manganese and iron that had once deposited at the lake bottom returns into the hydrosphere with a ratio richer in manganese. The elements, thus returned to the hydrosphere, are now carried up by the circulation currents toward the upper layers, where they are oxidized and again precipitated to the bottom. The precipitated elements are then subjected to a second fractionation by reductive redissolution, with a much larger proportion of manganese.

Thus the repetition of such a cycle is expected to lead to a yearly decrease in the ratio of iron to manganese in lake water, also that in the deposits, and consequently in mud the ratio may have been gradually increasing from depth upward.

Actually, the vertical variation in the ratio in muds seems to accord generally with this view. Indeed, with the exception of the surface value, whose deviation is comprehensible to a certain extent when we take into consideration the season in which the mud was sampled, the ratio decreases evenly from the deepest layer of the oldest deposition toward the surface that was deposited recently.

It is also of some interest to examine the ratio of iron to manganese in the earth that constitutes the land surrounding the lake, which is regarded as the most important source of the iron and manganese in Takasuka-numa.

For this purpose, the writer analyzed two kinds of soil samples, the one from the northern bank and the other from the path that runs along the western beach, the result of which is given in Table 14.

Table 14. Chemical composition of the soil surrounding Takasuka-numa

	Sample from the northern bank	Sample from the path along the west beach
SiO_2 (%)	56.64	60.61
Fe_2O_3 (%)	6.55	6.31
Al_2O_3 (%)	19.92	17.40
MnO (%)	0.13	0.10
Fe_2O_3/MnO	50.4	63.1

In both samples, the ratio of iron to manganese varied between 50.4–63.1, which is very large compared with that of the lake mud.

From this result, it is also very probable that if iron and manganese were supplied from the surrounding land soils, reductive redissolution played an important part, resulting in the relative concentration of manganese.

Next, it is worth mentioning that the concentration of manganese in the lake water seems to contribute to the geochemical separation of manganese from iron.

In the vadose zone of the earth's crust, iron is always accompanied with manganese. This is comprehensible when we consider the geochemical history of these elements.

Indeed, we have learned that manganese is found in juvenile rocks only in the state of manganous compounds, while the higher oxides of manganese, such as pyrolusite, occur as vadose through the oxidation of manganous compounds by atmospheric oxygen, after the latter have been dissolved in water and brought up to the surface of the earth.

The same may be said of iron. Since, it is then very natural that, in that condition wherein iron is precipitated as its higher oxide, manganese also is precipitated as such, we can easily understand that in the vadose zone of the earth's crust, iron is always accompanied with manganese.

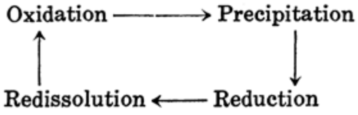
Now, in vadose minerals, there is another relation between manganese and iron which cannot be overlooked.

We usually find vadose minerals to consist mainly of iron with a little manganese, or mainly of manganese with a little iron, it being very seldom that we find minerals in which the ratio of iron to manganese had not such an extreme value.

The geochemical process above mentioned is unsuited for explaining the relationship, because the process, although it brings about the mixing of the two elements, does not cause their separation, so that it is necessary to search for another geochemical process that makes the segregation of manganese and iron possible.

As such a process, there is the returning of the manganese and iron by anaerobic decomposition of the organic detritus in the lake bottom that was fully discussed in the foregoing articles.

Now this process could not have existed until that time when life first appeared on the earth, since when there began a cyclic process for manganese and iron.



It is useless to repeat that, by the repetition of this cyclic process, manganese and iron gradually separated from each other, resulting in increase in the relative concentration of manganese in lake water.

It is not difficut to imagine that, as the destiny of such a lake, segregation of wad would occur, slightly contaminated with iron. On the other hand, it is also not so rare to find limonite contaminated with a slight admixture of manganese.

Thus we are led to the conclusion that the concentration of manganese in lake water, which is caused by the difference in the oxidation-reduction potential between iron and manganese, plays, at any rate, one of the most important parts in the geochemical fractionation of the two elements in question.

Mud Deposition.

The annual deposition of mud has been estimated repeatedly by a number of investigators. For example, A. Reissinger gave it as 11.8–20.7 mm. in Niedersonthofensee during 1914–32. But no estimate of its annual variation has ever been reported.

In order to obtain some information in this respect, the writer carried out the following observations in Takasuka-numa for twenty months, and established the variation in the amount of deposition and the chemical nature of the deposits.

Methods.

From four to six wide-mouthed glass bottles were fixed in a wooden frame, and then suspended at a depth of 1 m. above the deepest bottom of the lake. At intervals of about 21–67 days, they were hauled up, and their contents examined.

The sample of the deposits for chemical analysis was prepared as follows: 0.2 g. of the deposit, dried at 110°C., was digested with 2 c.c. of boiling concentrated hydrochloric acid in a small decomposition flask for about two hours: then 0.05 g. of potassium chlorate and 1 c.c. of the hydrochloric acid were added. After digestion for two hours more, 2 c.c. of the acid were again added, and the digestion was continued for another two hours. The liquid was filtered and washed and the filtrate condensed to 50 c.c.

The sample solution, thus prepared, was divided into sufficient portions for the determination of iron, aluminium, and manganese. Further treatment, together with the determinations of loss on ignition and total nitrogen were carried out as already described in a previous paragraph.

Results and Discussions.

Seasonal Variations in the Amount of Deposition (v. Table 15). Owing, unfortunately, to accidental loss of the equipment, for two periods, one between November 11 and December 25, 1935, and the other between June 21 and September 13, 1936, data are unavailable, although there is little doubt that in the former of these two periods there occurred much deposition, as proved by the following facts.

Despite the heavy deposition that occurred during October 20 – November 11, not only a large amount of solid matter remained in the lake waters on November 11, but its amount had increased from October to November, as will be seen from the determination of the residue on evaporation (v. Table 17 and Fig. 3).

Table 15. Annual variation in deposition in Takasuka-numa, and in its chemical composition. (1935–37)

Date (days)		June 23—July 24 (31)	July 24—Sept. 29 (67)	Sept. 29—Oct. 20 (21)	Oct. 20—Nov. 11 (22)	Dec. 25—Jan. 19 (24)	Jan. 19—Feb. 28 (40)	Feb. 28—Mar. 26 (27)	Mar. 26—Apr. 29 (34)
Deposition during the whole period	(mg./cm^2.)	131	306	160	230	18.5	39.3	35.5	153
Deposition per day	(mg./cm^2.)	4.2	4.6	7.6	10.5	0.77	0.98	1.32	4.50
Loss on Ignition	(%)	19.57	19.84	19.61	19.17	17.92	15.30	15.15	14.40
	(mg.)	0.828	0.907	1.490	1.970	0.138	0.150	0.200	0.648
Total nitrogen	(%)	0.782	0.814	0.498	0.736	0.782	0.675	0.661	0.622
	(mg.)	0.033	0.037	0.061	0.077	0.006	0.007	0.009	0.028
Fe_2O_3	(%)	12.2	11.2	10.9	11.3	12.5	9.96	10.61	8.85
	(mg.)	0.52	0.54	0.83	1.17	0.10	0.10	0.14	0.40
Al_2O_3	(%)						8.47	9.21	8.82
	(mg.)						0.083	0.122	0.397
MnO	(%)	0.46	0.63	0.33	0.94	0.70	1.03	0.98	1.12
	(mg.)	0.019	0.029	0.025	0.099	0.005	0.010	0.013	0.050

Date (days)		Apr. 29—May 25 (26)	May 25—June 21 (27)	Sept. 13—Oct. 4 (21)	Oct. 4—Oct. 25 (21)	Oct. 25—Nov. 25 (31)	Nov. 25—Dec. 26 (31)	Dec. 26—Jan. 31 (36)	Jan. 31—Mar. 5 (33)
Deposition during the whole period	(mg./cm.^{2})	78.0	95.4	258	133	31.1	78.1	43.9	76.9
Deposition per day	(mg./cm^2.)	3.00	3.09	12.29	6.34	10.05	2.52	1.22	2.33
Loss on Ignition	(%)	14.95	15.47	20.06		16.16			
	(mg.)	0.449	0.478	2.467		1.625			
Total nitrogen	(%)	0.698	0.718	0.952		0.718			
	(mg.)	0.021	0.022	0.117		0.072			
Fe_2O_3	(%)	9.35	9.04	11.54		11.78			
	(mg.)	0.28	0.28	1.42		1.18			
Al_2O_3	(%)	8.67	8.05	8.75		13.39			
	(mg.)	0.260	0.249	1.075		1.345			
MnO	(%)	0.56	0.54	0.64		1.44			
	(mg.)	0.017	0.017	0.078		0.145			

It is quite natural to suppose from the foregoing that, in the succeeding period, November 11 – December 25, still greater deposition occurred, the amount of which the writer estimated as follows:

First, it was assumed that the whole autumnal deposition did not differ much in amount in the two years 1935 and 1936, so that the amount of deposition for the period September 14 – December 24, 1935, was assumed to have been the same as that for the period September 14 – December 24, 1936. Consequently, 8.1 g. was obtained as the amount of deposition per day during the period, November 11 – December 24, 1935, and the gap in this was filled with a blank rectangle (v. Figs. 3 and 4).

A distinct parallel will now be seen between the variation in deposition and that in the mean values of the residue on evaporation in the lake water from various layers, ranging from the surface to the bottom, as given in Fig. 3. For the same reason, the second gap between June 21 and September 13 of the following year should have been filled with a moderate quantity of deposits, the deposition curve running similarly to that in the preceding year.

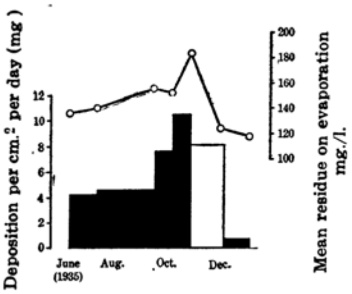


Fig. 3. Seasonal variation in the deposition in Takasukanuma and also in the mean residue on evaporation of the lake water (June 23, 1935—January 19, 1936.)

The deposition throughout the one and a half years actually varied, as follows:

The amount of deposition that began to increase at the beginning of autumn 1935, reached maximum during the period from the middle of November to that of December, after which it suddenly diminished to minimum from the end of December to the middle of January, the following year.

This condition lasted until the beginning of spring, when deposition increased to some extent, after which it again slightly declined during the succeeding time intervals, namely, April 19 – May 25 and May 25 – June 21.

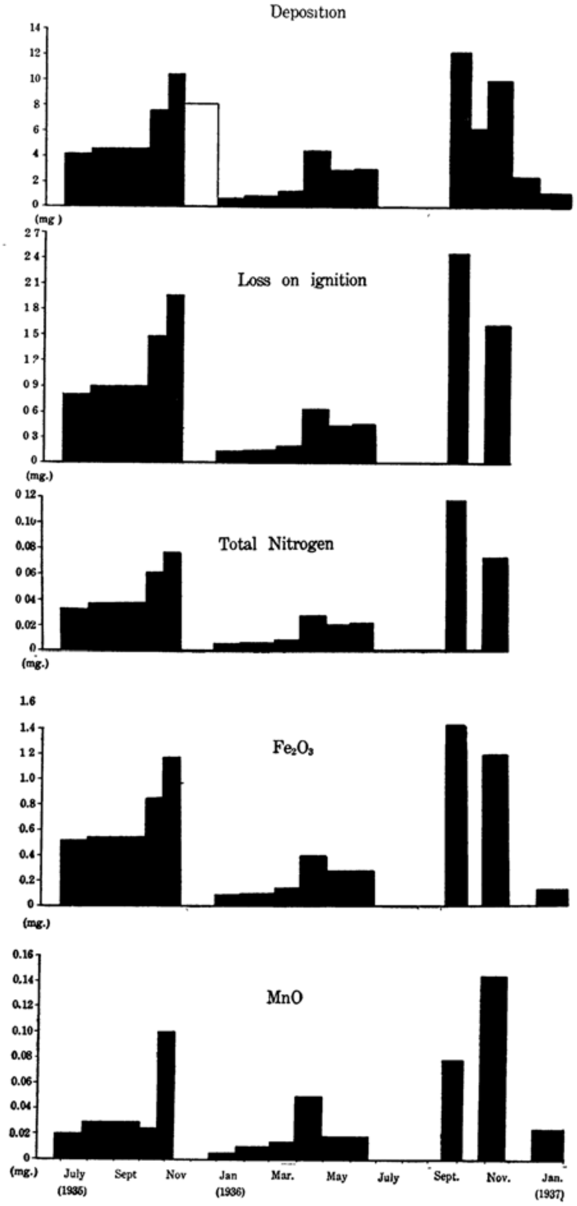


Fig. 4. Annual variation in the deposition in Takasuka-numa, and also in its chemical composition.

The last mentioned interval seems to have been followed by moderate deposition that continued till the end of summer.

Then, as it did in the previous year, a heavy autumnal deposition followed, which continued till the end of November, making it evident that the annual variation in deposition in Takasuka-numa has two maxima, a distinct one in autumn and another one less distinct in spring, with a minimum in winter and a moderate (medium) deposition period ranging from middle spring to the end of summer.

These maximum and minimum will hereafter be called the 'autumnal vernal maximum', and the 'winter minimum', respectively.

In certain years, these maximum and minimum may come earlier and in others, later, as the result of meteorological conditions, etc., whence it is comprehensible that, in 1936, the autumnal maximum seemed to have begun and ended somewhat earlier than in the previous year.

It is also remarkable that the autumnal maximum in 1936 differed from that in the previous year, the development of the two peaks in the graph being supposed to be the effect of meteorological conditions.

At any rate, the diurnal deposition per cm.^{2} varied between the two extereme values of 0.77 mg. and 12.29 mg. in dry weight, the deposition for the whole year having amounted to 1.66 g. which would fill the bottom to a thickness of 5.88 cm., assuming that the density and the water content of the deposit were as large as those of the bottom mud.

It is also notable that this amount of deposition for the whole year surpassed all the values hitherto reported for various lakes.

Chemical Composition of Deposits and its Seasonal Variations. (a) The loss on ignition was larger in the samples from the end of June to the beginning of November, the value varying between 19.17 and 20.06%, being smaller in the samples from the end of January to the end of June (14.40–15.47%), and medium in the samples from November to the middle of January (16.16–17.92%). It is remarkable that the variation in its values occurred continually in order of magnitude evenly and smoothly.

(b) The percentage of total nitrogen was found to vary between the two extreme values of 0.498 and 0.952%, without any regularity in its seasonal variations.

(c) The percentage of iron reckoned as Fe_2O_3 varied between 8.85 and 12.5%. It might be mentioned that the values from the end of January to the middle of June were smaller, while those for the remaining half of the year were generally larger.

(d) With the single exception of a very large value, 13.39%, for the period October 25 – November 25, 1936, the percentage of aluminium, as alumina, did not vary much, lying between 8.47–9.21%.

(e) The percentage of manganese, as manganese oxide, varied between the two extreme values of 0.33 and 1.44%; no regularity was found.

It is also of interest to compare the seasonal variation in the deposited amounts of these chemical components, which are represented graphically in Fig. 4.

At a glance, the graphs for iron, the total nitrogen and the loss on ignition are seen to be shaped similarly to that for the total deposition, while the graph for manganese differs, indicating that the precipitation of manganese in the autumnal maximum was delayed compared with those of iron and other components; the maximal point consequently appeared later.

This is easily explained by the difference in the oxidation-reduction potential between iron and manganese. As mentioned above, the potential is higher in manganese than in iron. At the end of autumn, when circulation of the water increases considerably, the potential in the deeper water gradually begins to rise, and a condition is reached in which the reduction of manganese proceeds easily, which, however, is difficult for ferric iron. Thus in such a season, in deeper water, the supply and accumulation of manganous salts still continue, and the manganese, thus accumulated, is carried upward by the circulation current, to undergo continuous oxidation and precipitation. Thus, as shown by the present result, complete precipitation of manganese follows that of iron.

Factors Controlling Deposition. The following may be mentioned as the factors that control the amount and the character of the deposits.

(a) The circulation of water, which is caused by cooling and by wind. On circulation, the water carries the substances accumulated at depth to the upper water and exposes them to further changes, while it whirls up the deposit that is settled in shallower bottoms, carrying it into depths.

(b) The amount of suspended substances that are carried into the lake by influent waters.

(c) The production of organisms, whose debris are partly deposited as essential constituents of the mud.

(d) The temperature of the deeper waters, which controls the chemical changes that occur in the deposits after they have settled in the suspended bottles.

When the present result of the foregoing studies in the amounts deposited is examined, it is easily found that the effects of the factors (a) and (d) are self-evident, while those of (c) and (e) are not so apparent.

The effects were most obvious in the autumnal maximum. In fact, there is no doubt that the main cause of the heavy deposition was the circulation of water by cooling.

By the circulation of water, a large amount of iron, that was accumulated in the tropholytic zone in the preceding period of stagnation, was carried towards the upper water zones, rich in oxygen. Then there followed the oxidation of ferrous iron to ferric, which, in its turn, formed a flocculent hydroxide and precipitated to the bottom.

It will easily be seen, therefore, that the percentage of iron in the deposit was quite large.

At the same time, the fact cannot be overlooked that, during this interval, deposition of aluminium and total nitrogen increased to a smaller extent than iron, although they were still in fairly large proportions.

Such a circumstance cannot be explained without assuming that the organic debris, as well as the clayey substances that had once settled on the shallower zone of the bottom, were whirled up into the water by the force of the water movement, and after being carried toward the middle of the lake were deposited there. Such transference of deposits, no doubt, occurs during the period in question to a considerable extent.

The effect of temperature of the bottom water is also evident. As will be seen from the preceding paragraph (Table 4) and also from the following Fig. 12, the temperature attains its maximum in the middle of autumn, so that in this season, the chemical changes that occur in the suspended bottle are expected to be accelerated considerably.

Among the changes that could occur, fermentative decomposition of organic substances may be mentioned as the first, as was clearly proved by studies on the dissolved gases of lake waters, to be given later.

Owing to decomposition, a considerable portion of the organic components of the deposit was consumed before the bottle was hauled up, so that the percentages of these components should be reduced, with consequent increase in those of the other components. Naturally, such a reduction is also conceivable in the case of iron, which would follow the above organic decomposition. But the escape of iron from the deposit, that is, the decrease in percentage of iron will often be insignificant compared with those of the organic components, hence, what follows is nothing but increase in the percentage of iron relative to those of the organic components. It is highly probable that the larger percentage of iron in the autumnal deposits, is due, partly, to just such circumstances.

It must also not be overlooked that, in the development of autumnal deposition, the preceding season of summer stagnation plays an important part.

The large amount of iron and manganese in the tropholytic zone, which forms a storage for the autumnal deposition, had been formed during the period of summer stangantion. Were this period absent, there would be no large precipitation in autumn.

This is the case in the season that embraces the beginning of winter to the beginning of spring, when, despite the continuous circulation of water between the upper and deeper layers, there occurs only a slight deposition, owing to the fact that the greater part of the substances stored were deposited almost completely during the preceding seasons, leaving little to settle.

IV. The Density of Lake Water.

General Description.

So far as the writer's knowledge goes, examples of density determination, in which the density of the lake water was measured directly, are very rare. The only example available being that of A. Hamberg,(21) who measured the density of the water at Lake Walloxen, a small lake near Upsala, in April 1909, when the lake lay covered by ice, and found that the lake water had a density of 1.0003 at a depth of 1 m. and 1.0005 at a depth of 7.5 m.

Little is gained by merely asserting that the density of water plays an important part in problems dealing with the circulation or the

---

(21) Hamberg, *Petermanns Mitteilungen*, **57**, II (1911).

stability of lake water. It is odd to find that, notwithstanding its importance, our knowledge of the density of lake water is so meagre.

The writer puts it down to the fact that, for directly measuring the density of lake water, special and painstaking care is needed, demanding considerable time.

Heretofore, when investigators desired to compute the density of lake water at a certain depth, they usually chose the easiest way, namely, they first measured the water temperature at a depth, next they ressort to a table for the value of the density of pure water corresponding to the temperature observed, and assumed this value for the actual density at that depth.

A discussion based on a value of density obtained in such a manner can never be hoped to go very far.

At the same time, it is well known that, at present, the method of measuring the temperature of deep water has been improved to such an extent that it is not a difficult task to attain an accuracy of one hundredths of a degree, which is equivalent in the case of density of water to one two-millionths.

When we reflect on the actual situation of our knowledge as just outlined, we are at once confronted with certain problems demanding solution, for example.

(1) What is the difference in density of lake water and that of pure water?

(2) What is the actual variation in the density of lake water with depth?

(3) What is the actual variation in the density of lake water with season?

(4) How does the density of water vary in the various kinds of lakes?

(5) What is the degree of accuracy in the value of density as computed by the customary method just mentioned.

These questions led the writer to undertake the following measurements: (1) Four series of determinations of the water density of Takasuka-numa were made between September and December, 1935, at six to eight different depths. (2) A similar determination was made in Kizaki-ko and Nakatuna-ko, both in Sinano Province, the following summer, the results and discussions of which will now follow.

Methods.

The bottles containing the sampled waters were tightly stoppered to prevent bubbles and other troublesome gas effects, both from inside and out, that are likely to cause changes in the density of the water.

The determination of density was completed within 24 hours after the water was sampled, although in the case of samples, the density of which was likely to alter, they were determined within a few hours.

For the determination, a Sprengel's pycnometer was used, the temperature of the thermostat being controlled at 20.0°C. in the case of Takasuka-numa, and at 17.9°C. in the cases of Nakatuna-ko and Kizaki-ko the variation in temperature being kept within 0.05°, whereby the error in observation of the water density was kept within 0.00001. But in the observations of September 29 and October 20, these conditions were not yet adequately fulfilled.

From these values at 20.0°C. or at 17.9°C., and from the result of temperature determination, the actual density on the spot was computed on the assumption that the coefficient of cubical expansion of the examined water was the same as that of pure water.

The values at 20.0°C. or at 17.9°C. are given in column (a), while those at the respective temperature on the spot, which were thus computed, are shown in column (b) in Table 16, 19 and 20. The densities of pure water at the corresponding temperatures are also given in (c).

The Result from Takasuka-numa.

The result of the determination in Takasuka-numa is given in column (a) in Table 16, which when compared with the data of hydrographical observation (Table 17), clearly shows that on September 29, the stratification in water density was parallel with those of a few of the chemical constituents.

Here the value of the water density varied in the different water layers in the order of 1/10,000, and, barring the two exceptions of density decrease, one at a depth of 2 m. and the other at a depth of 5.5 m., the density on the whole increased from the surface towards the bottom.

Although, in appearance, these two exceptions are identical, they are quite different in nature, which may be clearly shown when these values are nevertheless reduced to those at the respective temperatures on the spot, assuming that the coefficient of the cubical expansion of the lake water is identical with that of pure water. In fact, when we examine the reduced values given in column (b), it will readily be seen that the former decrease in density at a depth of 2 m. has disappeared, while the latter, at 5.5 m., is still present.

This point is quite important, because the latter decrease runs counter to the law of stability, whereas the former does not.

It must be added that in the present case the water was not treated previously in any way whatsoever, and that the determination was carried out on it as a whole, including suspended matter, such as planktons, organic debris, etc.

Therefore, it is not certain what part of the difference in density that was observed is owing to the presence of suspended matter, and what part to the water itself?

In order to make this point clear, the writer, on October 20, conducted the determination in two different ways, the one on water not previously treated and the other on water that was filtered.

For filtration, filter paper "Toyo" brand No. 5 was used, and precautions were taken to avoid risk of changes in density of the water during filtration owing to oxidation by atmospheric oxygen, etc.

This time again, waters that were not filtered showed decrease in density at 20.0°C. for two different depths, the one from 1 to 3 m., and the other at a depth of 5.5 m.; and, as in the previous observation, when the observed values of density are reduced to those at temperatures on the spot, decrease at the upper depth disappears, while that at the bottom persists.

Table 16. Densities of the lake waters of Takasuka-numa (1935).

Date of sampling	Sept. 29				Oct. 20			
Date of determin.					Oct. 20			
	not filtered				not filtered			
Depth (m.)	a	b	(°C)	c	a	b	(°C)	c
0	0.9984	0.9980	(21.9°)	0.99782	0.9983	0.9985	(18.8°)	0.99847
1	0.9985	0.9982	(21.3°)	0.99795	0.9985	0.9988	(18.5°)	0.99852
2	0.9983	0.9985	(19.2°)	0.99839			(18.3°)	0.99856
3	0.9985	0.9987	(18.8°)	0.99847	0.9984	0.9988	(18.2°)	0.99858
4	0.9987	0.9990	(18.5°)	0.99852	0.9984	0.9988	(18.1°)	0.99860
5	0.9987	0.9992	(17.3°)	0.99875	0.9985	0.9989	(17.3°)	0.99875
5.5	0.9985	0.9991	(16.6°)	0.99887	0.9982	0.9988	(16.9°)	0.99882
5.7								

Date of sampling	Oct. 20				Nov. 11			
Date of determin.	Oct. 21		Oct. 22		Nov. 13			
	filtered		filtered		not filtered			
Depth (m.)	a	b	a	b	a	b	(°C)	c
0	0.9983	0.9985	0.9983	0.9985	0.99827	0.99934	(13.8°)	0.99930
1			0.9983	0.9986	0.99826	0.99931	(13.9°)	0.99928
2			0.9984	0.9988	0.99828	0.99933	(13.9°)	0.99928
3			0.9985	0.9988	0.99827	0.99932	(13.9°)	0.99928
4	0.9983	0.9987	0.9985	0.9988	0.99827	0.99933	(13.85°)	0.99929
5	0.9984	0.9989			0.99831	0.99937	(13.85°)	0.99929
5.5	0.9982	0.9988			0.99827	0.99934	(13.75°)	0.99930
5.7							(13.2°)	0.99937

Date of sampling	Nov. 11				Dec. 15			
Date of determin.	Nov. 11		Nov. 12					
	filtered		filtered		filtered			
Depth (m.)	a	b	a	b	a*	b	(°C)	c
0								
1								
2								
3								
4			0.99829	0.99935				
5	0.99828	0.99934	0.99828	0 99934	1.00008*	1.00039	(6.68°)	0.99994
5.5			0.99826	0.99933	1.00003*	1.00034	(6.67°)	0.99994
5.7	0.99823	0.99937	0.99823	0.99937				

a .... densities at 20.0°C.
b .... densities reduced to the temperatures of waters which are given in parenthesis.
c .... densities of pure water at the temperatures of waters.
a*.... densities at 11.0°C.

Table 17. Residue on evaporation and the potassium permanganate consumption of the lake waters of Takasuka-numa in the autumn 1935.

Depth (m.)	Sept. 29		Oct. 20		Nov. 11		Dec. 15	
	Resid. on evap. (mg./l.)	Cons. of $KMnO_4$ (mg./l.)	Resid. on evap.	Cons. of $KMnO_4$	Resid. on evap.	Cons. of $KMnO_4$	Resid. on evap.	Cons. of $KMnO_4$
0	127	18.3	120	33.4	190	19.7	126	20.6
1	127	18.6	118	22.6	180	20.3	123	20.1
2	126	17.1	123	19.5	181	19.4	117	19.6
3	126	16.1	119	19.1	133	18.0	125	18.1
4	121	15.2	124	19.2	173	20.5	122	21.9
5	201	23.6	203	44.7	178	19.2	124	20.7
5.2							128	16.6
5.5	262	55.4	260	51.3	247	20.7		

It is worthy of note that the filtered water usually showed a smaller density than before filtration, the amount of decrease, particularly at a depth of 1 m., was larger, with the result that the irregular stratification of water density found in unfiltered water at the 1–3 m. zone, disappeared in the filtered ones, even at 20.0°C.

It is therefore concluded that of the two kinds of irregular distribution of density, the one found in the upper water zone was caused by unequal distribution of suspended matter in the various layers of this zone. In fact, the consumption of potassium permanganate shows that a smaller amount of reducible substances was always (not only on October 20 but also on September 29) present in the water at a depth of 2 m. than in that at a depth of 1 m., where the amount of decrease in density by filtration was specially larger.

The nature of the decrease in density at the bottom differs also at this point. Since it persisted even after filtration, it was shown that the decrease was not connected, at any rate, with the suspended matter, which can be filtered away by filter paper.

On November 11, although the water had practically circulated completely, as was shown by the smaller content of oxygen and by the larger residue on evaporation, circulation was not yet complete at the bottom, for which reason, the density of the water had become uniform at every depth tested, except in the deepest layers, so far as its value in the order of one ten-thousandths was concerned, although it showed a certain variation in the order of one hundred-thousandths, and notwithstanding which, it is notable that the decrease in density at 5.5 m. was still evident.

Noteworthy also is the fact that, on December 15,(22) when the circulation was supposed to have been completed, a faint decrease in density was still evident, so that of these two anomalous stratifications of water density that were found at 20.0°C., that in the upper water

---

(22) It is necessary to add that the density was measured this time at 11.0 °C.

zone was caused by uneven distribution of the suspended matter, and it disappeared in the actual distribution of water temperature, while that in the bottom still remained, the reason of which will be discussed later as a separate subject.

Comparison of Results of the Two Different Methods for the Determination of Water Density, the Customary Method and the Writer's Computation Method.

A comparison will now be made of the value of the density itself, and of the gradient of the vertical variation in density.

(a) The Density Value. This comparison is merely that between the writer's values (a) and those of pure water (c) at corresponding temperatures.

The difference between these two values varied between 0.00000 and 0.0005.

The former value was found at a depth of 5.7 m. on November 11, and the latter 5 m. on September 29.

It is worth mentioning that this difference was somewhat larger during the stagnation period, especially at depths below the thermocline.

At any rate, these results lead to the conclusion that the customary method of determining the density of lake water is followed by an error that is, at least, too small by 0.00000–0.0005.

Now, a difference in density of 0.0005 in pure water through variation in temperature, corresponds to a variation of 2.5°, which means that the customary method of determining the density of lake water often merely yields a result which is equivalent to that given by rough measurement of water temperature with an error of as large as 2.5°.

Consequently, if we continue the customary method in determining the density of lake water, we may frequently disregard, in our measurements of temperature, without hesitation, observation errors up to 2.5°.

(b) Gradient of Vertical Variation in Water Density. The gradient of vertical variation in the water density constitutes one of the most important factors that determine the stability, that is, the resistance to the mixing of water. It is expressed by $\frac{d\rho}{dz}$ where ρ represents the water density and z the depth.

In Table 18, its values from the customary density determination are compared with those from the writer's computation method.

During the period of complete circulation, vertical variation in the water density is determined only by water temperature, because both dissolved and suspended matter are distributed uniformly in all the water layers, except the bottom one, where a decrease in denstiy persists, so that during this season, the gradient of vertical variation in water density, as calculated from the values of density by the customary method does not differ much from that of the writer's method.

Table 18. Gradient of vertical variation in the water density in Takasuka-numa (1935).

Depth (m.)	Sept. 29		Oct. 20 (filtered)	
	a	b	a	b
0 > 1	2×10^{-6}	1.3×10^{-6}	1×10^{-6}	0.5×10^{-6}
1 > 2	3× ,,	4.4× ,,	2× ,,	0.4× ,,
2 > 3	2× ,,	0.8× ,,	0× ,,	0.2× ,,
3 > 4	3× ,,	0.5× ,,	0× ,,	0.2× ,,
4 > 5	2× ,,	2.3× ,,	1× ,,	1.5× ,,
5 > 5.5	(–)1× ,,	1.2× ,,	(–)1× ,,	0.7× ,,

$\frac{d\rho}{dz}$ (c. g. s. units)

a .... $\frac{d\rho}{dz}$, from the writer's computation method.

b .... $\frac{d\rho}{dz}$, from the customary method.

In contrast to the foregoing, however, when the water stagnates and the distribution of both dissolved and suspended matter is not uniform, the water density is no longer a simple function of temperature, with the result that the two kinds of gradient of vertical variation in water density as calculated by the two methods, differ considerably, the maximum difference being as much as 0.00025.

It is scarcely necessary to add that the value of the change in vertical variation in water density found by the customary method is meaningless at the bottom, where the actual change in density was found to be quite the reverse.

Decrease in Density at the Bottom.

The most important result from Takasuka-numa was the discovery of reversed stratification in the density of lake water at the bottom.

Since the water density usually increases with increase in the amount of solid matter dissolved in it, it will easily be understood that the density of lake water increases from the surface towards depth parallel with the increase in the amount of the residue on evaporation. Against this general relation, sudden decrease in density at the bottom is followed by a further increase in the amount of residue after evaporation, which precludes the possibility of the decrease being due to the lighter water with a smaller content of solid matter rising up from the bottom, so that we must look elsewhere for the cause of the decrease.

The question now arises whether there is any substance in sufficient amount in the bottom water that causes a decrease to show in the density of water.

As such a substance we could mention quite a number, namely, diethylamine, trimethylamine, the anilines, and the alkaline soaps of stearic and palmitic acids, as well as the wellknown examples of alcohol, ammonia etc., although it is difficult to suppose that any of these substances is present in the bottom water in sufficient concentration to cause a decrease in the water density observed.

The effect of dissolved gases is also conceivable. According to the studies of K. Ångstrom⁽²³⁾ and of J. Horiuti,⁽²⁴⁾ the density of a liquid is reduced by the absorption of a gas whose density in the condensed state is smaller than that of the solvent, whence it is quite probable that the density of lake water is lowered by the dissolution of hydrogen and other gases of the hydrocarbon series that have densities less than water when they are condensed. Moreover, these gases are the products of fermentation at the bottom, which are accumulated there especially in large quantities during the stagnation period, when the decrease in density of the bottom water is also large.

As shown in the following paragraphs, it is possible to compute the effects of the various gases dissolved upon the density of the lake water, and to prove that the decrease in question is caused partly by the unequal distribution of these gases in different water layers.

However, as will be shown later, the decrease in density caused by unequal distribution of gases is only one-tenth that of the total, so that search must be made for other causes of the decrease.

From one of the studies begun in this connection, it is probable that the decrease in the density is partly due to changes in the state of aggregation of the dissolved colloidal substances, such as hydroxide of iron, the details of which will be published in a separate paper.

It will also be seen that it is not certain whether the two causes, the unequal distribution of dissolved gases and the variation in the state of aggregation of dissolved substances, are sufficient to explain the phenomena completely. It is hoped to carry out studies in this respect still further.

Significance of the Decrease in Water Density at the Bottom in the Return of the Deposited Matter into the Water Body.

Although, as just mentioned, the cause of the density decrease in the bottom water of Takasuka-numa is not yet clearly known, it can at any rate, be said to be true, that certain changes are going on incessantly at the bottom or in the bottom water resulting in decrease in the density of water there.

Since a condition in which a heavy body lies upon a lighter one is unstable, granting, for sake of argument, that such a condition did at one time exist, it cannot continue.

Decrease in the density at the bottom is an example of such an unstable state, and if such a distribution of density is once formed, the probability is that it will soon be disturbed by mixing of the upper with the lower water.

---

(23) Ångstrom, *Ann. Physik*, **15** (1882), 297–308.

(24) Horiuti, *Sci. Papers Inst. Phys. Chem. Research* (Tokyo), **17** (1931–32), 126–256, 257–264.

Nevertheless, we do find such a decrease, which tells nothing beyond the fact that the change which brings about decrease in the density at the bottom water is of a kind that occurs vigourously and constantly, rendering possible the persistence of such an anomalous distribution of density.

Going a step further, the writer thinks it very probable that such a change, which acts in diminishing the water density, is taking place not only in the lake bottom, where its existence is readily found, but also in other places, where it is unnoticed.

At the same time, the writer believes that there is more intense mixing of upper and lower waters in the lastnamed places, by which permanent establishment of density decrease is hindered. He, therefore, concludes that changes that bring about decrease of water density at the bottom, as well as that mixing up of the waters that follow, are of world-wide occurrence.

The mixing of the waters in the bottom, which is accepted as of wide occurrence, has special significance in lake metabolism.

As repeated above, the circulation or mixing of waters in the upper and lower layers plays an important role, giving back the products of anaerobic decompositions at the bottom to the upper water zones.

It has been accepted for a long time that the two main factors, which control the circulation of water are wind and temperature.

Wind gives rise to a current in the lake water, and this leads to mixing of the upper and lower waters. The conditions favourable for this mixing of water by wind are found during such seasons as when the surface water is frequently cooled by cold weather or by fall of temperature in the night. This is the period of circulation that one finds in Takasuka-numa practically through the greater part of the year, from the end of summer to the beginning of the next summer, while the remainder of the year constitutes the period of stagnation.

During the latter season, that is, in summer, the surface water is warmed from day to day, and its cooling, even if it does occur sometimes can scarcely cause the water to circulate, which condition leads to the development of marked stratifications of various kinds chemical, physical, and biological. These are the general features attending the circulation of water in a lake.

Now the changes that are responsible for the decrease in density at the bottom, as found in Takasuka-numa, and which is believed to occur also widely in other lakes, is important, seeing that it forms the third means by which the water is circulated, in addition to two preceding ones, which have to do with the wind and the cooling of the surface water.

As just stated, decrease in water density at the bottom is followed by mixing of water of decreased density with the upper water, whereby a large part of the accumulated matter in the bottom water is enabled to be carried into the upper water zones.

This process of carrying back the bottom substances up into the upper layer is extremely important, since it occurs in the bottom water during periods of stagnation when other means of circulation are virtually unavailable. Moreover it occurs more actively in this season than in others.

Densities of the Waters of Nakatuna-ko and Kizaki-ko.

The densities of the waters of Nakatuna-ko and Kizaki-ko were determined during the summer of 1936 at the Kizaki Station of the Imperial Fisheries Experimental Institute.

The result is given in Tables 19 and 20. As was the case in the previous table for Takasuka-numa, the values of the water density at 17.9°C. are given in column (a) and those that were reduced to temperatures on the spot in (b), while the densities of pure water at the respective temperatures are shown in column (c).

Table 19. Densities of the lake waters of Nakatuna-ko.
(August 17–19, 1936.)

Depth (m.)	(a)	(b)	Water temp. °C.	(c)	Residue on evap. mg./l.
0	0.99862	0.99713	(24.8)	0.99713	
1					22.6
4					38.0
5	0.99863				37.5
10	0.99863	0.99964	(10.9)	0.99964	39.3
11	0.99863	0.99976	(9.6)	0.99976	
11.5	0.99866	0.99981	(9.4)	0.99978	
12.0	0.99865	0.99982	(9.1)	0.99980	
12.4	0.99866	0.99983	(9.1)	0.99980	
12.6	0.99867				
12.7	0.99867	0.99985	(9.0)	0.99981	39.3
12.8	0.99867	0.99985	(9.0)	0.99981	81.8
13.0	0.99867	0.99985	(9.0)	0.99981	94.1
13.1	0.99869	0.99987	(9.0)	0.99981	

Table 20. Densities of the lake waters of Kizaki-ko.
(August 7–9, 1936.)

Depth (m.)	(a)	(b)	Water temp. °C.	(c)
24	0.99864	0.99997	(6.1)	0.99996
25	0.99864	0.99998	(6.0)	0.99997
26	0.99864	0.99998	(5.9)	0.99997
28	0.99864	0.99999	(5.7)	0.99998
28.3	0.99865	1.00000	(5.7)	0.99998
28.6	0.99865	1.00000	(5.7)	0.99998
28.7	0.99864	0.99999	(5.7)	0.99998
28.8	0.99865	1.00000	(5.7)	0.99998

The water density of Lake Kizaki-ko exceeded that of pure water only by 0.00002 at the most. It showed a tendency to increase slightly with depth between the 24.0–28.1 m. levels, with an exceptional slight decrease at 28.7 m. But it will be going too far to attribute this decrease of water density to the same causes that were proposed in the case of Takasuka-numa. This point should be carefully investigated.

At layers ranging from the surface to a depth of 11 m., the density of the waters of Lake Nakatuna-ko practically did not differ from those of pure water. The difference, which first appeared at 11.5 m., grew larger with depth, the maximum difference, 0.00006, being found at the bottom, 13.1 m.

The difference between the gradients of vertical changes in the water density by the direct and the computation methods did not exceed 0.000002, one of the lake bottoms on which the matter was tested being that of Nakatuna-ko.

Effect of Dissolved Gases on the Density of Lake Water.

The Calculation. In this chapter, the writer will discuss the effect of dissolved gases on densities of the water of Takasuka-numa, Nakatuna-ko, and Kizaki-ko, using Ångtrom's dilatation coefficients of water for the absorption of a few of the common gases,(23) and Horiuti's formula in referring to the density change in a liquid caused by the dissolution of gas,(24) and the writer's own data of analysis in referring to the dissolved gases in the waters of these lakes. (v. Tables 32, 33 and 35)

Horiuti's formula is

$$D-D_0=g\frac{P_s\gamma}{RT}(d-\delta D_0)$$

where D_0 and D are respectively the density of the solvent and that of the solution, d is the normal density, i.e., the density of the solute at N. T. P. g is its molecular volume; P_S is the partial pressure of the solute gas, γ is Ostwald's solubility of the solute gas, and δ is Ångstrom's value of the dilatation coefficient of water for the absorption of gas.

The values of δ, d, and $d-\delta D_0$ for the five gases that are to be dealt with in the following paragraphs, are given in Table 21.

Table 21.

	δ	d	$d-\delta D_0$
O_2	1.13×10^{-3}	1.43×10^{-3}	$+0.30\times10^{-3}$
N_2	$1.44\times$,,	$1.26\times$,,	$-0.18\times$,,
CH_4	$1.23\times$,,	$0.566\times$,,	$-0.66\times$,,
H_2	$1.06\times$,,	$0.071\times$,,	$-0.99\times$,,
CO_2	$1.27\times$,,	$1.96\times$,,	$+0.69\times$,,

The values of δ for oxygen, nitrogen, hydrogen, and carbon dioxide are quoted from Ångstrom, while for methane, since its value is lacking, the arithmetical mean of the values of the four gases is adopted as its δ.

If it is desired to calculate the change in the density of the lake waters in question, due to the dissolution of the gases, by means of this formula, and to obtain results with an accuracy of the same order as in the data of the density of the lake waters observed above, the accuracy of the values that are to be inserted in the formula need not exceed two figures.

The writer adopted $R = 8.2$, $g = 22400$; for T, 286 was chosen as the middle temperature between 6–20°C., for D_0, density 1.00 of pure water, free from dissolved gases, was taken. Finally for $P_S\gamma$ was chosen the volume of a gas in one c.c. of water, converted into N. T. P.

Two Kinds of Gases, the One Decreasing and the Other Increasing the Density of Water by Dissolution. According to its particular behaviour towards change in density, gases may be divided into two groups.

The one, to which $d - \delta D_0$ is positive, tends to increase the density of water by dissolution. To this group belong such gases as oxygen and carbon dioxide, whose molecules are rather large. The other, to which belong gases of smaller molecular weights, such as nitrogen, hydrogen, and methane, $d - \delta D_0$ has a negative sign, and their dissolution brings about a decrease in water density. It is also notable that the smaller the molecule of the gas in question, the larger the decrease in the water density.

Thus the diminishing effect upon the density of water is greatest in methane and hydrogen.

Now, upon comparing the various behaviours of gases towards the density of water with the distribution of these gases in the lake waters, it is interesting to find that, excepting carbon dioxide, the greater part of which is dissolved in the combined or in the partially combined state, all the other gases are distributed in such a way that the deeper lying waters are influenced by these gases to become lighter than the overlying waters.

As already mentioned, oxygen is a gas whose dissolution brings about an increase in the density of water, and the distribution of this gas in lake water is such that it is rich in the upper and middle layers, but poor and often absent, from the deeper layers.

After all, oxygen is so distributed as to make the lake water heavier in the upper zones than in the deeper ones.

In contrast to this, nitrogen, methane, and hydrogen, whose dissolution results in decrease in the density of the water, are distributed in such a way that they are all poor in the upper water zones, getting richer towards the bottom.

Thus the distribution of the latter gases in lake waters is such that the deeper waters tend to become lighter than the upper waters.

Such a general relation in the distribution of dissolved gases in lake waters is not only obtained in Takasuka-numa, but in almost every lake in the temperate zone.

It must be added that such a distribution of the gases helps the mixing of waters between the upper and lower layers, and plays one of the most important roles in the circulation of matter in a lake, a role that is especially important, for during the stagnation period, such a distribution develops to the highest degree when circulation by other means is interrupted, the mixing of the waters being thus made much easier than if distribution were otherwise.

To this general relation, carbon dioxide seems to be an exception.

In effect, this gas, which is distributed abundantly in the deeper layers, but less so in the upper layers, increases the density of the water by dissolution, and seems to make the water density in deeper waters greater, much more so than in the upper waters. However, as stated above, the manner of dissolution of the gas in lake water differs from that in pure water; in the former it is dissolved partly in the combined state and partly in the partially combined one. Therefore it is a question, what proportion of the amount given by analysis acts in increasing the density of lake water, as in the case of pure water. This is a point which, it is hoped, will be elucidated by further studies.

The Results from Takasuka-numa. The change in the density of water $\varDelta D = D - D_0$ (in the following descriptions $\varDelta D$ will be used in place of $D - D_0$), which was calculated by means of Horiuti's formula, will be found in Table 22.

On comparing the values for various seasons and depths, we find that

(1) $\varDelta D$ for oxygen varied between $0.00000 \sim +0.000023$ during the period of observation (September 1935 – October 1936).

The gradient of the change in density due to the dissolved gas with depth, $\frac{d(\varDelta D)}{dz}$ (m. was taken as the unit for the measure of depth) attained its maximum (absolute value), 0.000013, on July 1936 at a depth between 2 and 3 m.

(2) $\varDelta D$ for nitrogen lay between -0.000017 and -0.000036, and the maximum of its $\frac{d(\varDelta D)}{dz}$, 0.000006, was found on January 19 at a depth between 3 and 4 m.

(3) $\varDelta D$ for methane varied between 0.00000 and -0.000086. The maximum of its $\frac{d(\varDelta D)}{dz}$ was found to be 0.000064 and 0.000077 on October 20, 1935, at a depth between 4 and 5 m., and on October 4, the following year, at a depth between 5 and 6 m.

(4) $\varDelta D$ for hydrogen varied between 0.00000 and -0.000042, and the maximum of its $\frac{d(\varDelta D)}{dz}$ was found to be 0.000008 on July 25, 1936, at a depth between 5 and 5.5 m., and on October 4, the same year, at a depth between 5 and 5.5 m.

Summarizing the result, it may be said that, of the maximum values of the gradient of the change in water density for the four gases, that due to methane was the largest in its absolute value, those for oxygen hydrogen, and nitrogen following in the order named.

For the water depth, where the dissolved gases were completely analysed and the $\varDelta D$ for these gases were all completely calculated, the sum of these $\varDelta D$, namely, the total change in the water density is also given as $\Sigma \varDelta D$.

With the exception of the depth of 0 m., in December, where $\Sigma \varDelta D$ had a positive sign, it is remarkable that $\Sigma \varDelta D$ was always negative, that is, the total effect of the dissolved gases was to cause the density to decrease in all seasons and at all depths.

The maximum for the absolute value of $\Sigma\Delta D$ was found on October 20, 1935, at a depth of 5.5 m. and on October 4, the following year, at a depth of 6 m.

When compared seasonally, $\Sigma\Delta D$ increased from September until October, when it attained its maximum, after which it began to diminish.

Finally $\Sigma\Delta D$ had the smallest values below 0.0000007 (except the deeper layers) in December, while it showed 0.000004 in November at a depth of 5–5.5 m., and in September 0.000025 at a depth of 4–5.5 m.

Thus it seems that $\Sigma\Delta D$ had its maximum value in the deepest layer in October 1936, at a depth 5–5.5 m., namely 0.00014.

The Results from Nakatuna-ko and Kizaki-ko. (v. Tables 23 and 24). The values of ΔD as well as those of $\Sigma\Delta D$, for any of the gases in question which we obtained for these lakes, lay always within the range of the two extreme values found in Takasuka-numa.

$\Sigma\Delta D$ was always found to be negative, and its maximum value (in absolute value) was found at a depth of 25 m. in Kizaki-ko, and at a depth of 12.5 m. in Nakatuna-ko.

Table 22. The change in the water density in Takasuka-numa due to the dissolved gases ($\Delta D\times10^5=(D-D_0)\times10^5$).

		0 m.	1 m.	2 m.	3 m.	4 m.	4.7 m.	5 m.	5.2 m.	5.5 m.	6 m.
Sept. 29 1935	Oxygen	+1.5		+0.6	+0.5	0		0	0	0	
	Nitrogen	-2.1				-2.1					
	Methane	0				-0.1					
	Hydrogen	0				-0.1					
	$\Sigma\Delta D\times10^5$	-0.6				-2.3					
Oct. 20	Oxygen	+1.5		+0.9	+0.8	+0.9		0		0	
	Nitrogen	-2.2		-2.2						-1.8	
	Methane	0		0	-0.4	-0.4		-6.8		-6.3	
	Hydrogen	0		0						-4.2	
	$\Sigma\Delta D\times10^5$	-0.7		-1.3						-12.3	
Nov. 11	Oxygen	+1.5		+1.4	+1.4	+1.4		+1.4		+1.3	
	Nitrogen	-2.2		-2.3	-2.4	-2.3		-2.3		-2 3	
	Methane	0		0	-0.1	-0.1		-0.2		-0.2	
	Hydrogen	0		0	-0.1	0		-0.1		-0.1	
	$\Sigma\Delta D\times10^5$	-0.7		-0.9	-1.1	-1.1		-1.2		-1.3	
Dec. 15	Oxygen	+2.3	+2.2	+2.1	+2.2	+2.1			+1.1		
	Nitrogen	-1.7		-2.8		-2.8		-2.8	-2.8		
	Methane	0		0		0		0	0		
	Hydrogen	0		0		0		0	0		
	$\Sigma\Delta D\times10^5$	+0.6		-0.7		-0.7			-1.7		
Jan. 19 1936	Oxygen	+2.3	+2.3	+2.2	+2.3	+2.2		+2.1			
	Nitrogen	-3.2	-3.2	-3.0	-3.0	-3.6		-3.2			
	Methane	-0.1	-0.1	-0.2	-0.3	-0.2		-0.1			
	Hydrogen	0	-0.1	0	0	0		-0.1			
	$\Sigma\Delta D\times10^5$	-1.0	-1.1	-1.0	-1.0	-1.6		-1.3			

Table 22.—(*Continued*).

		0 m.	1 m.	2 m.	3 m.	4 m.	4.7 m.	5 m.	5.2 m.	5.5 m.	6 m.
Feb. 28	Oxygen	+2.2		+2.2		+2.2		+2.2			
	Nitrogen	−3.0		−3.0		−3.0		−2.9			
	Methane	0		−0.1		−0.3		−0.1			
	Hydrogen	−0.2		0		0		−0.2			
	$\Sigma\Delta D\times10^5$	−1.0		−0.9		−1.1		−1.0			
Mar. 28	Oxygen	+1.9	+1.9	+1.9	+1.9	+1.9					
	Nitrogen	−2.6		−2.7		−2.7	−2.7				
	Methane	−0.1		−0.1		0	−0.1				
	Hydrogen	−0.1		0		−0.1	−0.1				
	$\Sigma\Delta D\times10^5$	−0.9		−0.9		−0.9					
Apr. 29	Oxygen	+2.0	+2.2	+1.8		+0.9		+0.3			
	Nitrogen	−2.3		−2.3	−2.3	−2.4		−2.4		−2.6	
	Methane	−0.1		−0.1	−0.1	−0.1		−0.1		−0.1	
	Hydrogen	−0.2		−0.2	−0.4	−0.2		−0.2		−0.2	
	$\Sigma\Delta D\times10^5$	−0.6		−0.8		−1.8		−2.4			
May 25	Oxygen	+1.7	+1.7	+1.5	+1.0	+0.4		0		0	
	Nitrogen	−2.0		−2.2	−2.2	−2.4		−2.5		−2.5	
	Methane	0		−0.1	−0.1	0		−0.1		−0.1	
	Hydrogen	−0.2		−0.1	−0.1	−0.1		−0.1		−0.1	
	$\Sigma\Delta D\times10^5$	−0.5		−0.9	−1.4	−2.1		−2.7		−2.7	
June 21	Oxygen	+1.7	+1.7	+1.6	+1.0	+0.1		0		0	
	Nitrogen	−2.0	−2.0	−2.1	−2.2	−2.3		−2.4		−2.4	
	Methane	0	−0.1	0	−0.1	−0.1		−0.5		−0.9	
	Hydrogen	0	0	−0.1	0	0		0		−0.2	
	$\Sigma\Delta D\times10^5$	−0.3	−0.4	−0.6	−1.3	−2.3		−2.9		−3.5	
July 25	Oxygen	+1.8	+1.8	+1.4	+0.1	+0.1		0		0	
	Nitrogen	−1.9	−1.9	−1.9	−2.1	−2.3		−2.4		−2.6	
	Methane	−0.1	−0.1	−0.1	−0.2	−0.8		−2.3		−1.5	
	Hydrogen	0	−0.1	−0.1	0	−0.1		−0.3		−0.7	
	$\Sigma\Delta D\times10^5$	−0.2	−0.3	−0.7	−2.2	−3.1		−5.0		−4.8	
Sept. 13	Oxygen	+1.6		+1.5	+0.4	0		0		0	0
	Nitrogen	−1.9		−1.7	−1.8	−2.0		−2.2			−2.3
	Methane	−0.1		−0.4	−0.3	−2.3		−4.8			−5.1
	Hydrogen	0		0	−0.1	−0.1		−0.8			−0.8
	$\Sigma\Delta D\times10^5$	−0.4		−0.6	−1.8	−4.4		−7.8			−8.2
Oct. 4	Oxygen	+1.0		+0.9	+0.7	+0.6		+0.6		0	0
	Nitrogen	−2.2		−2.0	−1.8	−1.9		−2.0		−2.1	−2.2
	Methane	0		−0.3	−1.0	−0.8		−0.9		−6.8	−8.6
	Hydrogen	0		−0.1	−0.2	−0.1		−0.1		−0.5	−0.8
	$\Sigma\Delta D\times10^5$	−1.2		−1.5	−2.3	−2.2		−2.4		−9.4	−11.6
Oct. 25	Oxygen	+1.1	+1.2	+1.1	+1.1	+1.1		+1.0		+0.9	
	Nitrogen	−2.3	−2.2	−2.2	−2.2	−2.3		−2.2		−2.1	
	Methane	0	0	0	0	−0.1		−0.1		−0.4	
	Hydrogen	0	0	0	0	0		0		0	
	$\Sigma\Delta D\times10^5$	−1.2	−1.0	−1.1	−1.1	−1.3		−1.3		−1.6	

Table 23. Changes in the water density in Nakatuna-ko due to dissolved gases. (August 17–19, 1936.) $\Delta D \times 10^5$.

	0 m.	2 m.	4 m.	6 m.	7 m.	8 m.	9 m.	10 m.	11 m.	11.5 m.	12 m.	12.5 m.
Oxygen	+1.5	+1.6	+1.6	+2.0	+1.3	+1.0	+0.5	+0.1	+0.1		+0.1	0
Nitrogen	−1.8	−1.8	−1.8	−2.0		−2.4		−2.5	−2.5	−2.4	−2.6	−2.5
Methane	−0.1	−0.1	−0.1	−0.1		−0.1		−0.1	−0.8	−2.0	−2.7	−4.3
Hydrogen	0	0	0	0		0		0	0	0	−0.4	−0.6
$\Sigma\Delta D \times 10^5$	−0.4	−0.3	−0.3	−0.1		−1.5		−2.5	−3.2		−5.6	−7.4

Table 24. Changes in the water density in Kizaki-ko due to dissolved gases. (August 7–9, 1936.) $\Delta D \times 10^5$.

	0 m.	1 m.	2 m.	5 m.	10 m.	15 m.	20 m.	25 m.	28.8 m.	28.9 m.
Oxygen	+1.5	+1.6	+1.6	+1.8	+1.9	+1.8	+1.4	+0.8		+0.2
Nitrogen	−1.9			−2.1	−2.5	−2.7	−2.7	−2.7	−2.7	
Methane	0					0	−0.1	−0.2	−0.3	
Hydrogen	0					0	0	0	0	
$\Sigma\Delta D \times 10^5$	−0.4					−0.9	−1.4	−2.1		

The maximum values of $\frac{d(\Sigma\Delta D)}{dz}$, 0.000036 and 0.000010, were found between the depths of 12 to 12.5 m. in the case of Nakatuna-ko, and between 20–25 m. in that of Kizaki-ko respectively.

Decrease in Density of the Bottom Water of Takasuka-numa, and the Dissolved Gases. It has been shown that in Takasuka-numa, the four gases, oxygen, nitrogen, hydrogen, and methane, are so distributed that the deeper-lying waters tend to become lighter than those lying above them. Consequently, the total decrease in density due to the dissolved gases, and the gradient of its vertical change, increase with depth, while both attained their maxima in October 20, after which they became smaller with time.

Parallelism is evident when we compare these results with data covering measurements of the densities of the waters of the lake, as already reported. Thus the density decrease in the deepest water reported can be explained partly by the dissolution of gases, but partly, because, as it will be shown in the following, the amount of decrease due to the dissolved gases did not exceed 10 per cent of the total decrease observed. For example, the difference in water density, actually observed between depths of 5 and 5.5 m., was −0.0002 in September and October, and −0.00003 in November and −0.00005 in December, while the corresponding density change by the dissolved gases was merely −0.000001, for example in November. Consequently, the observed difference in the water density always rises higher by one figure to the density change due to the dissolved gases.

When this point is taken into consideration, we are led to the conclusion that a part of the decrease in the density of the bottom water of Takasuka-numa was naturally caused by the dissolved gases, although this part, presumably did not exceed 10 per cent of the total decrease actually observed.

Moreover, there are, in a lake, various factors, operating to increase the density of water, but more so in deeper water than in the upper, such as the unequal distribution of dissolved solid matter in the upper and deeper waters.

The observed decrease in density at the bottom is then nothing but a balancing of the actual density decrease and the accepted density increase.

We must, therefore, assume an actual decrease in density that is greater in magnitude than the apparent decrease, and we must also assume the presence of a cause that is more conductive to density decrease.

V. The Dissolved Gases in the Lake Waters, and their Significance in Lake Metabolism.

General Description.

As an example of an early study of the dissolved gases in lake water, we might mention that of A. Delebecque.(25) In the summer of 1894, he determined the content of oxygen by the method of Mohr, at various layers of Lakes Mont-Cenis, Bourget, Genève, and Nantua; and in the following year, he and M. A. Le Royer boiled the sample water of Lake Genève and collected the dissolved gases. The oxygen was absorbed by alkaline pyrogallol solution and the unabsorbed gas reported as nitrogen.

These analyses, done at that time with the utmost care, have to-day only a historical significance.

During 1905–8, following A. Delebecque, E. A. Birge and Ch. Juday(26) studied some Wisconsin lakes. The dissolved gases were also collected by boiling, and analyzed after Hempel.

Stimulated by these studies, similar investigations were made repeatedly of the dissolved oxygen in lake waters, for which a simple method of determination, such as that of Winkler, had been devised, the data from numerous lakes all over the world, in which practically all possible types of lake are represented, filling a bulky volume.

In contrast to this, no studies have since been reported on other gases, of which may be mentioned nitrogen, hydrogen, methane, etc. In the circumstances, the old results of Birge and Juday are still the only data available for discussing these gases in lake water.

The strides made by limnology during the past thirty years is so striking that repetitions and extensions of determination of the latter

---

(25) Delebecque, "Les Lacs Français," (1898).

(26) Birge and Juday, Wisconsin Geological and Natural History Survey, *Bull.*, **22** (1911), Scientific Series No. 7.

gases have assumed greater importance than before, while more accurate results by improved method of analysis or of procedure are greatly to be desired. It is also desired that continued observations for longer periods than heretofore be done on a particular lake for the purpose of obtaining a right understanding of the seasonal variations in the gases in different water layers, for only then the significance of the gases in "lake metabolism" can be hoped to be elucidated.

Indeed, if we regard a lake as life of a certain kind, as we did in the general introduction to this paper, lake water is nothing but the fluid that circulates through the body which sustains that life in this case, the lake. Therefore, as physiologists study the metabolism that takes place in a particular organ or in the whole body by analyzing the blood gases, analyses of the dissolved gases in lake water will no doubt supply us with the necessary knowledge of the metabolism that is taking place or has taken place in a lake or in a particular part of it.

For this reason, the following studies were made as an indispensable part of the programm of the writer's studies on "lake metabolism".

This study may be divided into three parts.

(1) A study of the five lakes at the foot of Mt. Huzi, the object of which was to compare the distribution of gases (especially in deeper layers) between lakes of different types.

(2) For the purpose of obtaining precise knowledge of the vertical distributions of gases in summer, the three chain-lakes of the Nisina-group in Nagano Prefecture, were examined.

(3) The study of Lake Takasuka-numa, Saitama Prefecture, was for the purpose of obtaining records of the annual variation in the proportions of these gases at different water layers.

In these studies, which were made during 1935–36, parallel with the examinations of the dissolved gases, nitrogen, hydrogen, methane, and total carbonic acid (free, partially, and totally combined), as well as oxygen, the determination of *p*H, temperature, transparency, and the potassium permanganate consumption, and also the iron and manganese contents of the lake water were carried out. The results will be given in the following Tables and Figures.

Methods.

The water was sampled with either a Kitahara or Eckman's bottle. The latter bottle was found unsuitable for sampling waters at intervals of less than a few score cm., because, in turning the bottle, a considerable amount of the surrounding water is stirred. Therefore in comparing water layers at such small intervals, for example, in the deeper layers of Nakatuna-ko, Kitahara's water bottle was sunk slowly and pulled up very slowly the first 1–2 m., allowing the surrounding waters to remain undisturbed.

The water temperature was read from a reversible thermometer attached to the Eckman bottle.

The *p*H of the waters was determined by the colorimetric method, using phenol-red and bromo-thymol-blue.

Oxygen was determined in two different ways, by Winkler's iodometric method, and by "the carbon dioxide method" to be described later. The values in the following tables are those by the former method.

Nitrogen, methane, and hydrogen were expelled by means of carbon dioxide, and then analysed as usual after Hempel.

The total carbonic acid was determined by the method of Petterson, modified by L. W. Winkler,(27) and further improved in certain points by the writer and his co-workers.

The Carbon Dioxide Method of Winkler, Modified by the Writer. The usual method for expelling the dissolved gases from water is that by boiling, which, however, has a number of drawbacks, chief among which is the difficulty of transporting the apparatus and the cumbrous work of setting it up and handling it on the field, besides the need for a source of heat.

For these reasons, the writer used the method of expulsion by means of carbon dioxide bubbles, after Winkler, with modification in certain points.

The procedure. About 10 g. of crushed marble are placed in a glass bottle, whose capacity (about 500 c.c.) is determined exactly beforehand. The bottle is then filled with the sample water and furnished with a glass funnel (b) to which about 15 c.c. of 25% hydrochloric acid are added. Next, the gas burette (a) is fitted to the funnel with a connecting stop-cock (g), which is previously filled with water and the cock closed after connection.

The burette is filled with 10% caustic soda solution, to which 50 gm. of Rochelle salt have been added per litre. Capillary tube (e) as well as head funnel (c) are also filled with the same solution. While this is being done, a part of the hydrochloric acid in the funnel (b) is made to gradually flow into bottle (A), yet no marked reaction occurs. Stop-cock (g) is then opened. Violent reaction first occurs, and bubbles of the evolved carbon dioxide ascend, carrying along with them other gases from the sample water through the liquid in the burette.

During the passage of the gas bubbles through the caustic soda solution, the carbon dioxide as well as the other gases that are soluble in alkali, are completely absorbed, leaving only insoluble gases in the burette. During this process, to facilitate evolution of gas, the level of the liquid in the reservoir (f) is kept slightly lower than the level in the burette.

For complete absorption, fresh alkali solution is made to flow down from the head funnel along the wall of the burette by opening cock (d). In the cold season, when the evolution of carbon dioxide is frequently very feeble, the bottle (A) is warmed by hot water (k). Expulsion of the dissolved gases is usually completed in 40 minutes. After closing cock (g), the burette is disconnected at (J) and rocked backward and forward several times in order to secure complete dissolution of the soluble gases. The burette is set up again vertically, left standing for a while, and the readings of the volume of the unabsorbed gas, the temperature, and the atmospheric pressure are taken. Pyrogallol solution and fresh caustic soda solution are next made to flow down the burette from the head funnel for the absorption of oxygen. After treatment with alkaline pyrogallol solution, the volume of the unabsorbed gas, as well as that of oxygen, is determined.

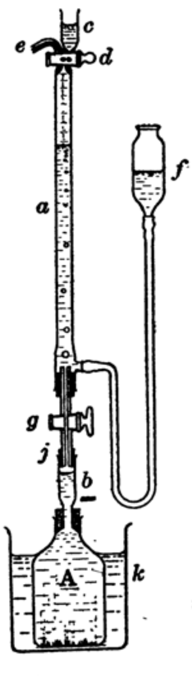


Fig. 5.

Errors Liable in the Carbon Dioxide Method. The errors inherent to the present method may be divided into the following two: the error in method, which can occur only through some defect in the method, and the errors in observation, which, for example, may occur in reading the scale, etc., and which may be + or —. The writer examined these two kinds of errors separately.

Errors in method. At room temperature, air was continually bubbled through 15 l. of distilled water for a few hours until the water was saturated with air. The water was then transferred into three bottles by a syphon, and the dissolved oxygen in each bottle was determined in the following three different ways.

---

(27) Winkler, "Trink- und Brauch-wasser in Lung-Berls chemisch-technischen Untersuchungsmethoden", 7. Auflage, Band I, 573.

First, the gas in one of the three bottles was determined by Winkler's carbon dioxide method, that in the second bottle by the usual boiling method as modified by the writer, and that in the remaining bottle by the iodometric method, the results of which are given in Table 25.

When we compare the values of oxygen with one another, it is found at once that the value obtained from the carbon dioxide method is the largest, that from the boiling method the least, and that from the iodometric method intermediate, a result that was expected from the beginning. Of the three, the iodometric method is the most direct and consequently the most reliable. That the value from the boiling method was smaller than that from the iodometric method, is easily explained as being due to the incomplete removal of the dissolved gases from the examined water by the boiling method. On the other hand, that the carbon dioxide method showed a larger value than the iodometric method, is probably due to the fact that the dissolved oxygen in the caustic soda solution, used for the absorption of carbon dioxide, is also partly expelled and is added to the gases from the examined water, leading to the erroneously larger value observed. An error of this kind also occurs with other gases dissolved. Thus the value of the unabsorbed gas that was left over after treatment with alkaline pyrogllol solution, is always smaller with the boiling method than with the carbon dioxide method.

Table 25. Comparison of the three different methods for determining the dissolved gases in water.

	Oxygen (c.c./l.)		Unabsorbed gas (c.c./l.)	
	Result	Mean observation error	Result	Mean observation error
a ··· The carbon dioxide method	7.69 7.70 7.62 7.68		14.84 14.91 14.70 14.84	
	7.67	±0.03	14.82	±0.06
b ··· The boiling method	7.28 7.34 7.24		14.83 14.61 14.63	
	7.29	±0.04	14.69	±0.09
c ··· The iodometric method	7.524 7.530 7.534			
	7.529	±0.007	0.13	
The difference between the values from a and b	0.38			
The difference between the values from a and c	0.14			
The difference between the values from b and c	0.24			

Table 26. The error in sampling the water and in its transference.

		Oxygen (c.c./l.)	Unabsorbed gas (c.c./l.)
An example with Kizaki-ko (August 8, 1936.)	(a)	6.10	15.40
	(b)	6.10	15.40
Another example with Nakatuna-ko (August 19, 1936.)	(a)	0.20	17.29
	(b)	0.20	17.40

Next, quantitatively, the oxygen value from the carbon dioxide method was found to be larger by 0.14 c.c. than that from the iodometric method, which is taken here as the standard, while the value by the boiling method is smaller by 0.24 c.c. than that by standard method. Besides, as referred to the standard, the two values deviate in opposite sense, the magnitude of the deviation by the carbon dioxide method being about half that by the boiling method. Thus it is concluded that the value from the carbon dioxide method is nearer to the right value than that from the boiling method.

It is also remarkable that in both methods, the difference between the values of the unabsorbed gases that were left after the removal of oxygen, does not exceed 0.13 c.c., which is only one-third the difference between the oxygen values, 0.36 c.c., given above.

It would seem more natural if the above values by both of the methods under examination, had differed by double the difference between the oxygen values, when it is considered that the volume of the unabsorbed gases is close to double that of oxygen.

But, as already said, actually this is not the case, the reason for which probably is that the treatment after expulsion of the dissolved gas, such as that with pyrogallol solution, affects the oxygen differently to what it does the unabsorbed gases. At any rate, the small difference in the values of the unabsorbed gases by the two methods, (the true value being assumed to lie between these two values) is better suited to the determination of the gases in question, because there is no simple and accurate method of determining the unabsorbed gases, as the iodometric method does for oxygen.

Next, we must discuss the errors in method to which both methods are subject. As just said, the true value for the unabsorbed gases lies between those given by the two methods, but the question is, at what point between the two does the correct one lie?

Assuming that the true value is the mean of those by the two methods, we get 0.07 c.c., but if we assume that the values by the two methods deviate from the true in the same proportion as they did from the value by the iodometric method in the case of oxygen, the error by the carbon dioxide method works out to 0.05 c.c., and that by the boiling method to 0.09 c.c.

Whichever assumption we may make, the errors from method in both cases, namely, the deviation in the values of the unabsorbed gases from the true value, do not exceed 0.1 c.c. Consequently, should we wish to carry out the determination of these gases where an observation error of 0.1 is of no serious consequence, then the observed values may be taken as true without correction.

Summarising the foregoing, the writer concludes that:

(1) The carbon dioxide method proved to be equal to the boiling method, judged from the point of error in method, in determining the dissolved gases of lake water other than oxygen.

(2) In analysis, where an error of 0.1 is admitted, the value by the carbon dioxide method can be taken as true, without any correction.

Errors in observation. Upon comparing, next, the observational errors peculiar to the various methods, it is obvious that the mean error is least in the iodometric method, intermediate in the carbon dioxide method, and largest in the boiling method.

In addition, certain advantages make the carbon dioxide method more appropriate for field work.

(1) By using from five to six sets of apparatus, simultaneous determination can be carried out by a single worker.

(2) The apparatus being simple, in the event of damage, it could be set up again with the aid of readily accessible substitutions and makeshifts, which, however, is fairly difficult with the boiling method.

An Examination in the Error in Sampling the Water and in Transferring it into the Measuring Bottles. (Table 26.) This error could be minimized with sufficient care. The two pairs of examples in the table will suffice. Each pair consists of the results of analyses on two different samples obtained from the same depth, at two succeeding samplings. The following results are mostly the mean values of a duplicate analysis.

Determination of Carbon Monoxide. The oxygen was absorbed with the alkaline pyrogallol solution from the gas that was expelled by the carbon dioxide method, and the unabsorbed gas charged into a storage tube (Fig. 6) for sending it to the laboratory for analysis. The analysis was carried out by the usual Hempel method; the procedure, by which the determination of carbon monoxide was made, was found to be worth discussing. It has been repeatedly reported that the absorption method by ammoniacal cuprous chloride solution gives an unusually large value to this gas. For this reason, even where the presence of carbon monoxide was proved by the cuprous chloride method, the writer tried also the spectroscopical test with the use of diluted blood on the same sample. The test proved that there was no trace of the gas in any of the samples that had been shown to contain considerable amounts of it by the cuprous method. Therefore, in later analyses, the samples were tested first for the gas by the blood method, and if its absence was shown as was usual in the writer's case, the treatment with cuprous chloride solution was omitted.

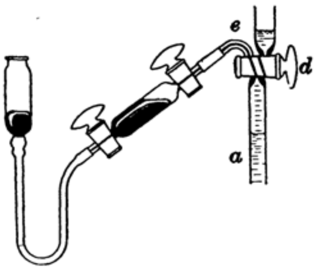


Fig. 6.

The Boiling Method. The original method of Treadwell(28) has two drawbacks.

(1) It was often found difficult to secure air-tight connection between the leading tube (C) and the gas burette (B).

(2) Some errors arise in the reading of the volume of the expelled gas owing to the fact that the dissolved gases are eliminated by a number of expulsions into the measuring burette, which are measured, and the sum of the readings is taken as the volume of the total gas obtained.

S. Sintani, the writer's co-worker, improved these two defects by means of the following modification, and succeeded in securing more accurate results.

First, for remedying the second defect, besides the burette (B_1), which corresponds to burette (B) in the original method, a second burette (B_2) was installed. The gas, which was driven into burette (B_1), in several instalments, was collected into the measuring burette (B_2), where the total volume was measured after it was ascertained that no more gas remained in the boiled flask.

In order to improve the first point, (R) was furnished with a side-tube, and the parts (I), (R), (K) and (C) were filled with mercury before the boiling was started.

The procedure is as follows.

A round flask of about 250 c.c. capacity, whose exact volume when level with the etched line at the neck, was previously determined, is filled with the sample water.

Into a rubber stopper, a glass tube (R) is inserted through a hole in it, until a small opening blown near the fused bottom of the tube, appears just below the bottom of the rubber stopper. The stopper is then pushed down into the flask to the etched line. While this is being done, a part of the water in the flask will flow into the tube, which is then raised, so that the side hole is buried in the rubber. In this way, the apparatus is made air-tight, after which the flask is connected with the other parts of the apparatus, as shown in Fig. 7.

By raising (I), and by releasing the screw cock (p), the mercury is made to flow into (R), (K) and (C). The cock (q) is closed so that the connection between (q) and the connecting tube (C) is cut, while the tube (B_1) is connected with the exterior by the three-way cock (s). The reservoir (N_1) is then raised and the burette (B_1), as well as the capillary to the cock (s), are filled with mercury. By giving the cocks (s), (r), and (t) a suitable turn, and by raising the reservoir (N_2), the burette as well as the connecting capillaries (rs) and (rt) are filled with water.

The screw-cock (p) is now opened and the reservoir (I) is lowered. The mercury then flows back into the reservoir, when there develops a vacuum in the bulb (K). When the top of the mercury reaches just above the cock (p), the latter is closed, and the reservoir (I) is dipped into boiling water to warm the mercury in it.

The tube (R) is now pushed into the flask until the small opening reaches below

---

(28) Treadwell, "Lehrbuch der Analytischen Chemie" II (1923), 638.

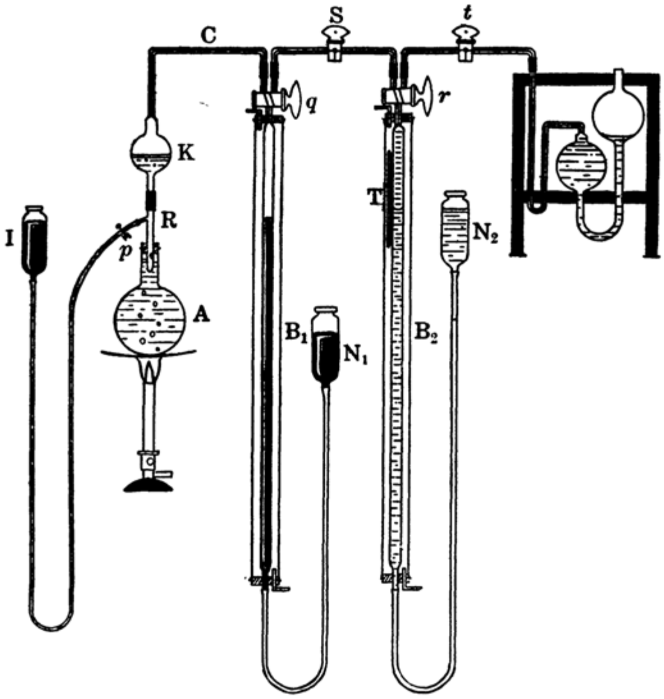


Fig. 7.

the lower surface of the rubber stopper. Evolution of gas begins immediately. The reservoir (N_1) is lowered, and the cock is turned so that the bulb (K) and the burette (B_1) are in connection. The flask (A) is heated to boiling as soon as possible. Here, it is necessary to regulate the level of the mercury in the reservoir (N_1), to prevent the water in the flask from flowing over into the receiving burette (B_1). When a sufficient amount of gas has been driven into the burette (B_1), the stopcock (q) is closed and the gas is transferred into the burette (B_2). After this operation is repeated several times, the reservoir (I) is raised and the screw-cock (p) opened. Hot mercury is poured into the flask, whereby the level of water in the bulb (K) is elevated and the remaining gas is completely transferred into the receiving burett (B_1) and finally into the measuring burette (B_2). As soon as the evolved gas is completely transferred into the burette (B_2) and brought into equilibrium with the surrounding temperature, its volume is read. The gas is then analyzed.

Determination of Total Carbon Dioxide. The apparatus used for this determination is the same as that for the boiling method. In place of mercury, the reservoir (I) is charged with concentrated hydrochloric acid. About 0.01 g. of aluminium wire is placed into the flask, after which the flask is filled with the sample water and stoppered.

The apparatus is set up in the same way as in the "boiling method". The air in the bulb (K), which is drawn out into the burette (B_1) by lowering the reservoir (N_1), is then expelled through the three-way cock (s), whose operation is repeated several times. Then, when the bulb is exhausted of air, the glass tube (R) is pushed into the flask. Evolution of gas immediately begins. The screw-cock (p) is now opened, and hydrocholoric acid is made to flow into the flask (A). A lively decomposition of carbonates occurs with the evolution of hydrogen gas which, in turn, is caused by the action of the acid upon the aluminium. When the evolution of gas is subdued, the flask is heated to boiling, and the remaining carbon dioxide expelled.

After reading the volume of the gas that has been collected in the burette (B_2), the gas is led into the Orsat tube, and the absorbed carbon dioxide determind.

Study of the Five Lakes at the Foot of Mt. Huzi.

The determination was carried out during August 19–22, 1935. Tables 27–31 show the results of observations, from which the following parallelism could be established.

Parallelism in the Degree of Eutrophication and that of Nitrogen Saturation. In the paper previously cited⁽⁶⁾ D. Inaba pointed out the existence of a parallelism in the amounts of plankton and of protein nitrogen in his observation of the five lakes in the winter of 1934, where the amount of plankton and the degree of eutrophication (the latter, judged from the amount of protein nitrogen), which are the largest in Syozi-ko, decrease in the order of Yamanaka-ko, Kawaguti-ko, Nisino-umi, Motosu-ko.

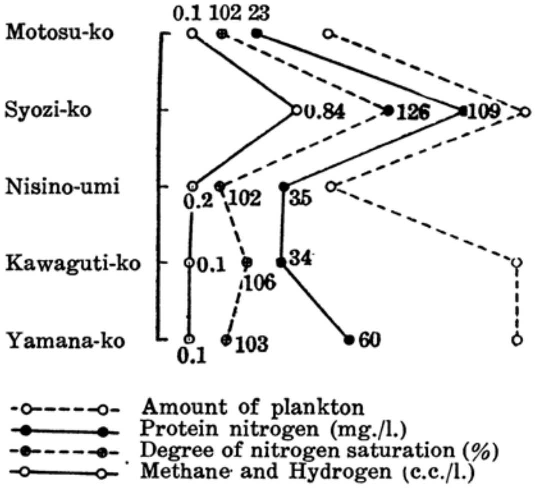


Fig. 8.

Table 27. Motosu-ko.

August 19, 1935 (1.30-4.00 P.M.), Transparency 12.5 m., Atmospheric pressure 688 mm.

Depth m.	Temp. °C.	pH	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.
0	22.8	6.9	5.63	101.8					
1	22.8								
2									
5	21.6	7.0	5.60	99.3					
10	21.5	7.0	5.60	99.7					
15	14.3	6.9							
20	9.8	6.8	7.79	107.8					
30	6.9								
115		6.5			15.12	15.0	102	0.03	0.11
120	6.1	6.4	6.67	84.5	15.03				

Table 28. Syozi-ko.

August 20, 1935 (5.40-9.00 A.M.), Transparency 5.0 m., Atmospheric pressure 690 mm.

Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.	C_nH_{2n} c.c./l.
0	22.5	7.4	5.77	103.6						
2			5.90							
5	22.3	7.4	6.00	107.5						
10	20.5	7.4	5.82	100.6						
12	15,4	6.4	2.98	46.5	16.17	15.3	126	0.4	0.4	0.1

Table 29. Nisino-umi.

August 20, 1935 (2.20-5.00 P.M.), Transparency 10.5 m., Atmospheric pressure 690 mm.

Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c /l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.
0	23.0	7.3	5.58	101.1					
5	21.7	7.4	5.72	101.2					
10	14.7		8.18	126.6					
20	12.0	7.3	8.10	117.6					
30	5.5	7.0	7.00	88.2	15.53	15.4	102	0	0.2
40	5.2	6.8	6.50	80.3					
52	5.2	6.8	6.44	79.6					
70	5.2	6.5			15.53				

Table 30. Kawaguti-ko.

August 21, 1935 (9.55 A.M.-1.50 P.M.), Transparency 5.0 m., Atmospheric pressure 696 mm.

Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.
0	23.1	7.9	5.86	105.6					
2	23.1	7.9	5.92	106.6					
5	22.8	7.9	5.89	105.3					
8	20.8	6.7	4.01	69.2					
9	17.2	6.4	0.20	3.2					
10	16.4	6.4	0.10	1.9					
11	15.0	6.4	0.11	1.7					
11.5	15.0				13.26	13.1	106	0	0.1

Table 31. Yamanaka-ko.

August 22, 1935 (8-10 A.M.), Transparency 5.5 m., Atmospheric pressure 686 mm.

Depth m.	Temp. C.	pH	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.
0	23.7	7.9	5.63	104.0					
2	23.3	7.9	5.72	104.8					
5	21.7	7.9	5.60	99.8					
6	21.5	7.7							
7	21.2	7.1							
8	20.9	6.9							
9	20.4	6.7	1.09	19.0					
10	20.1	6.5	1.21	20.6					
10.5	20.0	6.5	0.89	15.3	11.53	11.5	103	0.1	0

It was also seen from the writer's observation that the distribution of nitrogen and other gases in the deep waters varies in a similar order. In fact, the degree of nitrogen saturation in the deepest waters was always more than 100% in all the five lakes, and its maximum value, 126%, was found in Syozi-ko, which is the most eutrophic, while in the other lakes, the degree of saturation of this gas decreased in the above order of eutrophication. Thus it was 106% and 103% in Kawaguti-ko and Yamanaka-ko, both moderately eutrophic, and 102% in Nisino-umi and Motosu-ko, the least eutrophic.

The conclusion is that the degree of eutrophication of a lake runs parallel with that of saturation of nitrogen gas in the deepest water in summer.

Such a conclusion indicates that, although the greater part of the nitrogen in the deepest water might have come from the atmosphere, a considerable part was no doubt supplied by the denitrifying decomposition going on in the bottom water or in the bottom deposits.

Since, in an eutrophic lake, the quantity of organic debris deposited is large, much more decomposition is expected to occur, whence it is quite natural to find there a large production of free nitrogen as was found above. This point will be referred to again.

Methane and Hydrogen. The two other members of the unabsorbed gas family after treatment with alkaline pyrogallol solution, were methane and hydrogen. The sum of the volumes of these gases per 1 litre of water was 0.1 c.c. in Motosu-ko, 0.2 c.c. in Nisino-umi, 0.1 c.c. in Yamanaka-ko, 0.1 c.c. in Kawaguti-ko, and 0.9 c.c. in Syozi-ko. We find here, as in the case of nitrogen, the largest methane and hydrogen content in the most eutrophic lake, the cause of which can also be ascribed to the intense decomposition in the bottom.

Since the values approximate the observational error, precise comparison and discussion of the values in the remaining four lakes are omitted.

Study of the Nisina Lake Group in Nagano Prefecture.

A study was carried out during August 7–19, 1936, for the purpose of establishing precise vertical distributions of the gases that are not absorbed by alkaline pyrogallol solution.

In Lake Kizaki, 11 different water layers were examined with intervals of 1–5 m. in the upper layers and with that of 0.3–0.5 m. in the deeper layers. The samples from the surface and from depths below 20 m. were completely analyzed, the results of which showed that the samples from the intermediate layers must have had oxygen and nitrogen up to 99%. In the case of these intermediate layers, therefore, the collected gas was merely treated with alkaline pyrogallol solution, the unabsorbed gas being treated as nitrogen.(29)

In Nakatuna-ko, 14 different layers were examined with an interval of 1–2 m., except in the deeper layers, where the interval was reduced to 0.5–0.2 m.

Owing to weather conditions and to difficulties in obtaining various articles necessary in prosecuting these studies, determinations in Aoki-ko were not so reliable as in the case of the other two lakes.

The Vertical Variation in the Degree of Saturation of Nitrogen Gas.

(1) The amount of the dissolved nitrogen gas, which increased evenly with depth, suddenly decreased, although slightly, at the bottom. (The data from Aoki-ko does not follow this general tendency, to which, however, little importance can be attached, because of less accuracy in the observation of this lake.)

(2) The degree of nitrogen saturation was always above 100%, except in Nakatuna-ko at depths of 0, 2 and 4 m., where the degree was either 100% or just below it.

(3) Maximum saturation lay always slightly below thermocline. In the case of Nakatuna-ko, a second maximum was found at a depth of 12 m.

Such vertical distribution of nitrogen may be very common in many lakes during the period of summer stagnation, and the way in which such a distribution develops, is easily conceivable. As is usually the case with oxygen, from autumn to winter, abundant nitrogen is absorbed from the air and supplied to the water in all the layers during this period of circulation, while from the following spring to summer, the waters are daily warmed from the surface. This warming occurs in two different ways: (a) by the direct absorption of radiation and (b) by the mixing of the warmed water with the underlying cooler water.

In the latter way, the deepest water, which lies beyond the effective limit of heat radiation, can also be gradually warmed.

At any rate, from the rise in temperature of the water, decrease in solubility of nitrogen gas as well as those of other gases in water, results. Consequently, the degree of gas saturation in water grows larger, often exceeding 100%.

---

(29) Here and in the following descriptions, the term "dissolved nitrogen" means dissolved nitrogen and argon.

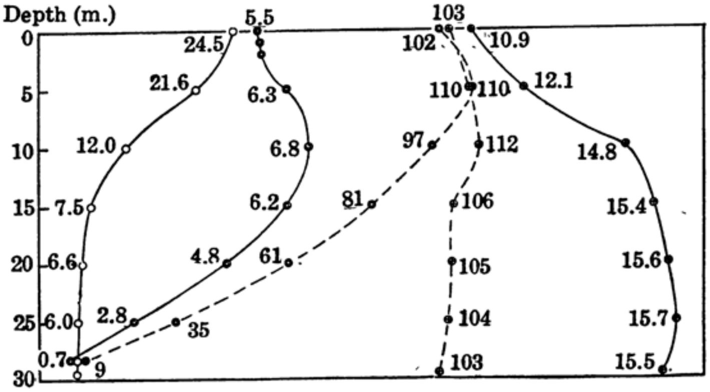


Fig. 9. Kizaki-ko (August 7-9, 1936.)

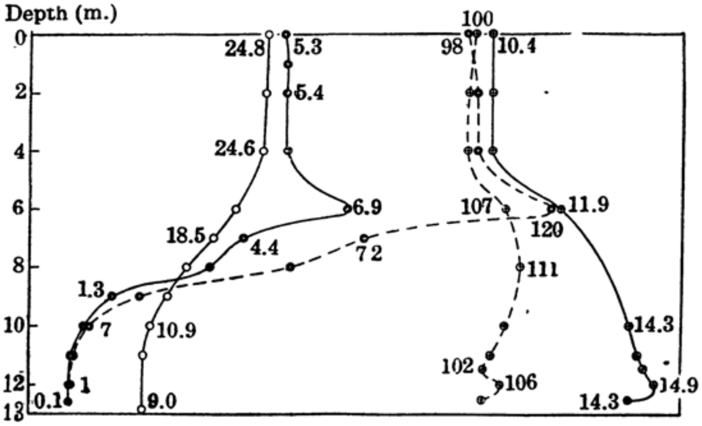


Fig. 10. Nakatuna-ko (August 17-19, 1936.)

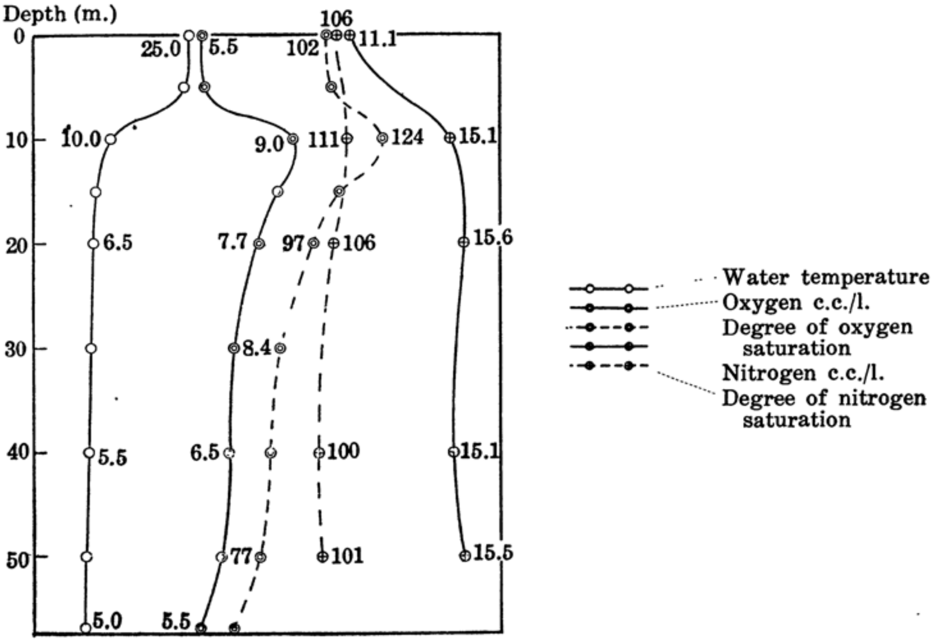


Fig. 11. Aoki-ko (August 18, 1936.)

Table 32. Kizaki-ko.

August 7-9, 1936.

Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.
0	24.5	6.9	5.51	101.6	10.94	10.9	103	0	0
1		6.9	5.49		11.20				
2		6.9	5.70		11.13				
5	21.6	6.8	6.29	110.1	12.12	(12.1)	110		
10	12.0	6.5	6.76	97.4	14.80	(14.8)	112		
15	7.5	6.3	6.22	80.5	15.40	15.4	106	0	0
20	6.6	6.2	4.81	61.0	15.80	15.6	105	0.2	0
25	6.0	6.0	2.81	35.1	15.97	15.7	104	0.3	0
28	5.7				16.01		103		
28.8	5.7				16.03	15.5		0.5	0
28.9	5.7	6.0	0.74	9.2	15.82				

Table 33. Nakatuna-ko.

August 17-19, 1936.

Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.
0	24.8	6.8	5.29	98.1	10.62	10.4	100	0.2	0
2	24.7	6.8	5.40	100.2	10.55	10.3	99	0.2	0
4	24.6	6.8	5.41	100.2	10.37	10.3	98	0.1	0
6	20.9	7.0	6.91	119.5	12.08	11.9	107	0.1	0
7	18.5	6.4	4.35	71.8					
8	14.9	6.1	3.60	55.2	13.81	13.7	111	0.1	0
9	12.4	6.1	1.27	18.5					
10	10.9	6.0	0.49	6.9	14.37	14.3	107	0.1	0
11	9.6	6.1	0.22	3.0	15.59	14.3	104	1.3	0
11.5	9.4				17.35	14.2	102	3.1	0
12.0	9.1	6.3	0.21	2.9	19.73	14.9	106	4.2	0.4
12.5	9.1	6.4	0.11	1.4	21.47	14.3	101	6.8	0.6
12.7	9.0				20.83	13.9	100	6.1	0.8
12.8	9.0	6.4	0.10	1.3					

Table 34. Aoki-ko.

August 18, 1936.

Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %
0	25.0	6.8	5.47	102.1	11.12	(11.1)	106
5	24.0	7.0	5.63	102 9			
10	10.0	7.4	8.99	123.8	15.10	(15.1)	111
15	7.2	7.0	8.37	107.8			
20	6.5	6.6	7.70	97.3	15.64	(15.6)	106
30	5.9	6.4	6.73	83.9			
40	5.5	6.3	6.54	80.6	15.12	(15.1)	100
50	5.0	6.2	6.30	76.7	15.51	(15.5)	101
57	5.0	6.2	5.48	66 7			

This is the case with the metalimnion where, although the warming effect from the above two causes is still sufficiently strong, escape of the nitrogen, in excess, is greatly hindered. In the surface layers, however, the nitrogen excess has a better chance of escaping from the water into the atmosphere, so that the degree of saturation is kept at 100% or near it, notwithstanding that the warming effect is greatest in these layers.

On the other hand, in the deeper layers, where the temperature elevation of the water is small, no marked supersaturation was found.

Thus in summer, the degree of nitrogen saturation that starts from the surface value of 100% or near it, increases towards the bottom and attains maximum at a layer slightly below thermocline, from which it decreases towards the bottom, as established.

A brief discussion is also necessary regarding the origin of the second increase in the degree of nitrogen saturation at the bottom, such as was found in a layer near the bottom of Nakatuna-ko.

This increase, no doubt, has no relation to the factors above considered, but owes its origin to the denitrifying decomposition that occurs at the bottom, as was suspected to be the case in Syozi-ko, and was proved conclusively in Takasuka-numa, as will be referred to later.

Methane and Hydrogen. These two gases, besides oxygen and nitrogen, represent the two other dissolved gases from these lakes, which were abundant in water layers below 20 m. in Kizaki-ko and in those below 11 m. in Nakatuna-ko. The content of these gases in water layers less deep, never exceeded 0.2 c.c. per litre.

The parallelism that was pointed out as existing between the content of these gases at the bottom water and the eutrophication of lakes in the case of the five lakes at the foot of Mt. Huzi, was here also found true; the content of these gases in the bottom water greatly exceeded that in Nakatuna-ko, which is more eutrophic than Kizaki-ko.

Comparison of Oxygen with Nitrogen. The difference in form of the curves of oxygen and nitrogen is worth mentioning.

Oxygen starts also from the surface value of its degree of saturation, 100% or near it, and increases with depth, as was found in the case of nitrogen. But, in contrast to nitrogen, it attains maximum at a less deep layer, lying slightly above the thermocline. Oxygen also differs from nitrogen in that the degree of saturation, as well as the absolute amount, begins to decrease suddenly and evenly from the maximum layer towards the bottom.

No doubt, this difference in the distribution of these gases is due solely to differences in the chemical nature of these gases, seeing that, physically, the two are similar.

Chemically, however, the two differ distinctly from each other. Owing to its activity, oxygen is easily consumed in the decompositions that go on in the water, while it is produced in the assimilation process by phytoplankton in sufficiently illuminated water layers, so that the oxygen content of the water is controlled by these two kinds of chemical changes as well as by its exchange, namely, the supply from the air and its escape from the water, which occurs at the surface of the water. Nitrogen, on the contrary, is practically inert, the content of this gas in the water being virtually determined solely by the gaseous exchange that takes place at the surface. It is true, of course, as said above, that in many lakes nitrogen evolves in the bottom layer, and that the fixation of this gas in the water is also possible. However, the effect of these two changes is insignificant on the content of this gas in the waters of the upper and middle layers of a lake of medium size or larger.

Then, by accurately tracing the distribution of nitrogen, it may be possible to obtain some knowledge of the physical factors that control the distribution of the dissolved gases in the water. On the other hand, we may find in the oxygen curve the total effect of these factors, physical as well as chemical, that is, assimilative production and consumption in decomposition. Therefore, by comparing the distribution of oxygen with that of nitrogen, we may obtain the most reliable knowledge of the net effect of assimilation and dissimilation upon the oxygen in lake waters. Further discussion in these respects will be reserved until sufficient data are obtained.

The Study of Takasuka-numa.

Observations were begun in September 1935, and continued to the end of October the following year. (v. Table 35)

Water Temperature. (v. Fig. 12) The circulation of water, which began towards the end of summer in 1935, proceeded to such an extent that, on October 20, the water layers, ranging from the surface to the depth of 4 m., had practically the same temperature, while on November 11, the temperature difference between the surface and the bottom layers was only 0.6°.

On January 14, the water began to freeze, and continued till January 19, when the lake was covered almost completely by ice, leaving uncovered only a small area in the south-eastern corner of the lake. The ice, which was 33 mm. thick on the east coast, and 45 mm. at a point 50 m. distant, melted away on February 23. From this time onward, the water was gradually warmed, but had not yet been sufficiently stabilized, the isothermal, therefore, still running vertically.

Toward May, the temperature gradient grew quite steep, the temperature difference between the surface and the bottom layers being more than 15°C, the deeper waters having completely stagnated. This stagnation continued till the beginning of autumn, when a sudden mixing of waters occurred and a distribution of temperature, similar to that in the previous year, again set in.

Nitrogen. (v. Figs. 13 and 14) *Degree of saturation.* The nitrogen in the water was found to have increased in all the water layers at every observation from September 1935 to the following January.

This increase, needless to say, was brought about by supply of the gas from the atmosphere in order to satisfy the increased solubility of this gas in the waters caused by lowering of the temperature.

Table 35. Takasuka-numa.

	Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c c./l.	Total carbon dioxide c.c./l.
September 29, 1935. Weather fine Air temp. 23.0°C. (9.20 A.M.) Transparency 1.70 m.	0	21.9	6.7	5.23	85.6	12.35	12.4	106	0	0	
	1	21.3	6.7								13.2
	2	19.2	6.4	3.34	34.6						
	3	18.8	6.4	1.58	24.3						
	4	18.5	6.4	0.19	2.9	12.82	12.4	98	0.2	0.2	
	5	17.3	6.4	0	0						50.4
	5.5	16.6	6.5	0	0	30.9					81.8
October 20 Weather fine Air temp. 16.7°C. (9.30 A.M.) Transparency 0.80 m.	0	18.8	6.7	5.20	80.0	12.82	12.8	102	0	0	23.8
	1	18.5	6.7								24.2
	2	18.3	6.6	3.34	50.8	12.61	12.6	99	0	0	25.8
	3	18.2	6.6	2.98	45.3	13.13			0.6		23.8
	4	18.1	6.6	3.10	44.7	13.40			0.6	0	
	5	17.3	6.6	0	0	13.70			11.2		20.0
	5.5	16.9	6.6	0	0	25.40	10.5	80	10.1	4.5	
November 11 Weather fine Air temp. 12.0°C. (9.40 A.M.) Transparency 1.10 m.	0	13.8	6.7	5.27	73.0		12.7	92	0	0	23.4
	1	13.9	6.7	5.73	79.6						24.2
	2	13.9	6.7	5.00	69.5	13.27	13.3	96	0	0	24.7
	3	13.9	6.7	4.97	68.9	13.60	13.4	97	0.1	0.1	25.1
	4	13.85	6.7	5.14	71.3						
	5	13.85	6.7	5.15	71.5	13.67	13.3	96	0.3	0.1	20.2
	5.5	13.75	6.7	4.67	64.5	13.72	13.3	96	0.3	0.1	21.9
December 15 Weather fine Air temp. 4.4°C. (9.30 A.M.)	0	6.8	7.3	5.40	63.5						20.3
	1	6.8	7.2	4.93	58.0						
	2	6.7	7.2	5.37	62.9	16.17	16.2	100	0	0	18.0
	3	6.7	7.2	4.96	58.2						18.1
	4	6.7	7.2	4.82	56.5	16.20	16.2	101	0	0	20.3
	5	6.7	7.2			16.13	16.1	100	0	0	22.3
	5.5	6.7	7.2			16.26	16.3	101	0	0	27.8

Table 35.—(*Continued*)

	Depth m.	Temp. °C.	*p*H.	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.	Total carbon dioxide c.c./l.
December 26 Weather fine Air temp. 7.7°C. (11.30 A.M.) Transparency 1.90 m.	0	5.1	7.1								
	1	4.95	7.1								
	2	4.95	7.1								
	3	4.9	7.1								
	4	4.9	7.1								
	5	4.9	7.1								
January 19, 1936. Weather fine Air temp. 0.35°C. (9.30 A.M.) Transparency 2.03 m.	0	0.85	7.2	8.04	80.7	17.81	17.6	95	0.2	0	20.0
	1	3.4	7.1	7.90	85.2	17.80	17.6	101	0.1	0.1	28.5
	2	3.4	7.1	7.66	82.6	17.70	17.4	100	0.3	0	30.0
	3	3.5	7.2	7.88	85.3	17.80	17.4	100	0.4	0	27.3
	4	3.6	7.1	7.75	84.2	18.13	17.8	103	0.3	0	32.3
	4.5	3.7									
	5	3.7	7.0	7.37	80.1	17.81	17.6	102	0.1	0.1	18.3
February 28 Weather fine Air temp. 2.7°C. (9.30 A.M.) Transparency 2.48 m.	0	4.0	7.0	7.71	84.4	17.31	17.1	99	0	0.2	14.6
	1										
	2	4.0	7.0	7.63	83.5	17.32	17.1	100	0.2	0	14.4
	3	4.0	7.0								
	4	4.0	7.0	7.65	83.8	17.54	17.1	100	0.4	0	15.8
	5	4.0	7.0	7.55	82.7	17.03	16.6	97	0.2	0.2	17.0
March 28 Weather fine Transparency 2.00 m.	0	8.7	7.0	6.78	83.6	15.72	15.4	100	0.2	0.1	16.3
	1	8.6	7.0	6.72	82.8						
	2	8.6	7.0	6.73	82.9	15.61	15.5	101	0.1	0	15.9
	3	8.6	7.0	6.76	83.1						
	4	8.7	7.0	6.53	80.3	15.80	15.7	102	0	0.1	15.4
	4.7	8.7	7.0			15.88	15.7	102	0.1	0.1	15.5

Table 35.—(*Continued*)

	Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.	Total carbon dioxide c.c./l.
April 29 Weather cloudy Air temp. 13.2°C. (9.30 A.M.) Transparency 1.40 m.	0	16.4	7.3	6.90	101.0	13.61	13.2	100	0.2	0.2	32.1
	1	16.4	7.3	7.54	110.4						18.6
	2	16.35	7.3	6.42	93.9	13.60	13.2	100	0.2	0.2	17.4
	3	16.35	7.3			13.78	13.3	101	0.2	0.4	18.4
	4	15.65	6.7	3.29	47.3	14.24	13.9	104	0.1	0.2	
	5	15.6	6.6	1.01	14.5	14.44	14.2	107	0.1	0.2	
	5.5	15.6	6.6			14.57	14.3	107	0.1	0.2	25.2
May 25 Weather fine Air temp. 17.3°C (9.20 A.M.) Transparency 1.26 m.	0	19.9	7.1	5.92	89.6	11.91	11.7	95	0	0.2	13.5
	1	19.3	7.1	5.96	92.3						13.5
	2	19.2	7.1	5.09	78.8	12.79	12.6	101	0.1	0.1	15.3
	3	17.8	6.7	3.44	51.7	13.20	13.0	101	0.1	0.1	20.5
	3.5	15.8	6.4								
	4	14.7	6.6	0.13	1.7	14.03	13.9	102	0	0.1	24.2
	5	13.3	6.8	0.09	1.2	14.24	14.5	104	0.2	0.1	
	5.5	13.0	6.9	0	0	14.87	14.7	104	0.1	0.1	28.3
June 21 Weather fine Air temp. 27.5°C. (9.20 A.M.) Transparency 2.55 m.	0	25.4	7.7	5.89	103.0	11.74	11.7	105	0	0	12.8
	1	25.2	7.7	5.93	103.0	11.93	11.8	105	0.1	0	13.7
	2	24.7	7.5	5.71	98.1	12.61	12.5	110	0	0.1	18.3
	3	20.2	6.6	3.62	57.0	13.00	12.9	105	0.1	0	17.4
	3.5		6.5								
	4	16.55	6.4	0.18	2.7	13.68	13.6	104	0.1	0	23.2
	4.5		6.6								
	5	14.1	6.9	0	0	14.73	13.9	101	0.8	0	42.2
	5.5	13.7	7.1	0	0	15.78	14.1	102	1.5	0.2	57.8
	5.8		7.1								
July 25 Weather fine Air temp. 32.0°C (9.30 A.M.) Transparency 1.35 m.	0	28.7	7.5	6.25	115.7	11.24	11.1	105	0.1	0	13.4
	1	28.2	7.5	6.45	118.6	11.29	11.1	104	0.1	0.1	13.9
	2	25.9	6.7	4.86	85.5	11.28	11.1	100	0.2	0.1	16.1
	2.5	22.9	6.3								
	3	20.7	6.3	0.22	3.8	13.27	12.4	102	0.3	0	22.4
	4	16.85	6.5	0.20	3.0	14.62	13.3	102	1.3	0.1	36.1
	5	15.0	6.9	0	0	17.56	13.8	102	3.6	0.3	53.0
	5.5	14.6	6.9	0	0	18.43	15.4	113	2.3	0.7	57.9

Table 35.—(*Continued*)

	Depth m.	Temp. °C.	*p*H	Oxygen c.c./l.	Oxygen %	Unabsorbed gases c.c./l.	Nitrogen c.c./l.	Nitrogen %	Methane c.c./l.	Hydrogen c.c./l.	Total carbon dioxide c.c./l.
September 13 Weather cloudy Air temp. 29.6°C. (1.30 P.M.) Transparency 1.68 m.	0	31.0	7.6	5.41	104.6	10.98	10.8	105	0.2	0	17.0
	1	28 95	7.5								
	2	28.5	7.5	5.09	93.9	10.49	9.8	96	0.7	0	20.7
	2.25		7.1								
	2.35		6.6								
	2.5		6.5								
	3	26.2	6.4	1.36	24.1	11.21	10.7	101	0.4	0.1	26.9
	3.5	23.35	6.2								
	4	20.2	6.4	0	0	15.41	11.7	95	3.6	0.1	61.2
	5	16.8	6.5	0	0	21.65	12.8	98	7.6	0.8	76.7
	5.5	15.75	6.5	0	0						96.5
	6	15.6	6.5			25.59	13.1	98	8.1	0.8	
	6.1	15.6	6.5								
October 4 Weather fine Air temp. 19.6°C. (9.00 A.M.) Transparency 1.55 m.	0	20.2	6.6	3.47	54.8	12.60	12.6	102	0		26 1
	1	20.1	6.6								
	2	19.9	6.6	3.28	51.5	11.96	11.4	92	0.5	0.1	
	3	19.7	6.6	2.56	40.1	12.20	10.4	83	1.6	0.2	28 2
	4	19.5	6.6	2.17	33.7	12.37	11.1	89	1.2	0.1	27.2
	5	18.7	6.6	2.04	31.3	16.83	11.4	90	1.4	0.1	55.7
	5.5	16.6	6.6	0	0	23.33	12.0	92	10.8	0.5	113.7
	6	15.85	6.6	0	0	27.01	12.7		13.6	0.8	117.8
October 25 Weather cloudy Air temp. 12.5°C. (9.20 A.M.) Transparency 1.26 m.	0	14.9	6.7	3.92	55.7	13.24	13.2	98	0	0	25.7
	1	14.85	6.7	4.03	57.1	13.02	13.0	96	0	0	25.7
	2	14.8	6.7	3.80	53.8	12.96	13.0	96	0	0	25.5
	3		6.7	3.71	52.6	12.95	13.0	96	0	0	26.6
	4			3.69	52.3	13.27	13.2	98	0.1	0	26.6
	5	14.8	6.6	3.50	49.6	13.01	12.9	95	0.1	0	27.7
	5.5	14.75	6.6	3.17	44.9	12.97	12.3	91	0.7	0	

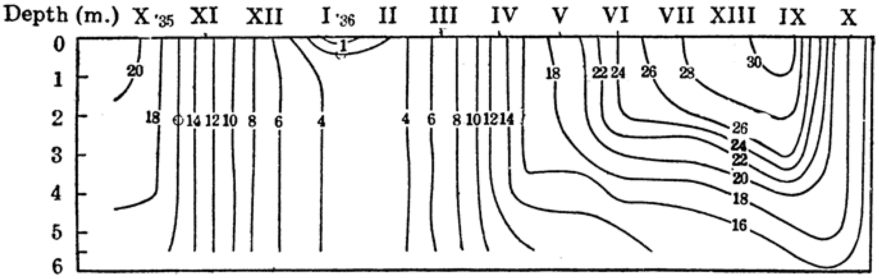


Fig. 12. Annual variation in the isothermals in Takasuka-numa.

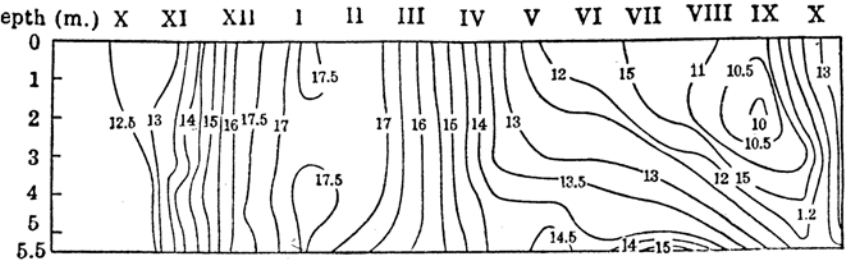


Fig. 13. Annual variation in the nitrogen content of the lake waters of Takasuka-numa (c.c./l.).

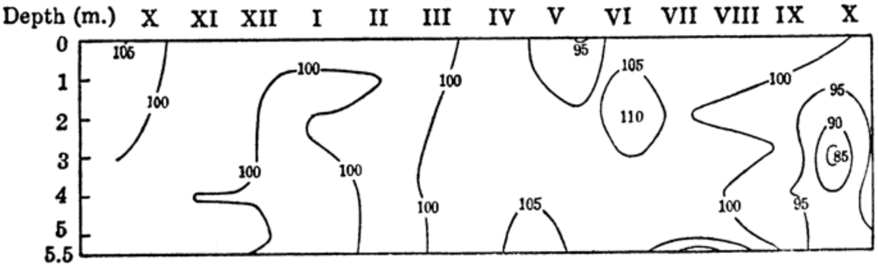


Fig. 14. Annual variation in the degree of saturation of nitrogen in the lake waters of Takasuka-numa.

What is noteworthy is that, this supply of nitrogen from the air was satisfied very quickly, as is evident from the fact that degree of nitrogen saturation lay always between 96 and 100% with two exceptions, one at a depth of 5.5 m. on October 20 and the other at the surface on November 11.

At any rate, the above period which covers the season from the end of summer to the middle of the following January, is a period when the waters are more or less in a state of undersaturation with respect to nitrogen, this gas being continually absorbed by the water from the atmosphere. For this reason, this period may be called the "period of nitrogen absorption", which completes the annual cycle with the "period of nitrogen escape" that follows. In the latter period, owing to the warming of the water, the excess of nitrogen is expelled from the water into the atmosphere.

From the foregoing observations in the Nisina Lake Group, it was ascertained that this escape of nitrogen into the atmosphere is hindered considerably, and that in every one of the cases examined, a maximum value of more than 110% in the degree of saturation of nitrogen gas was observed. Now in the case of Takasuka-numa, owing to the smallness of the lake, the water is easily mixed, as a consequence of which the escape of nitrogen from the surface is greatly facilitated. In this lake, therefore, it is very rare to find a value of more than 105% for the degree of saturation of nitrogen gas.

The value of 110%, found at a depth of 2 m. on June 21, and 113%, at a depth of 5.5 m. on July 25, represent the only two exceptions. The latter was, no doubt, due to the evolution of nitrogen caused by anaerobic decomposition of organic debris at the bottom, which will be discussed later, while the former was due to too rapid temperature rise in a particular layer that did not allow an immediate effect on the nitrogen percentage.

At all events, the general conclusion is that, in a shallow lake, as in the case of Takasuka-numa, ingress or egress of nitrogen to or from the hydrosphere takes place so soon that the nitrogen contents of the waters are always suited to the temperature variation of the waters.

This point will be found to be far more important when we consider that that is the case also for other gaseous constitutents of the lake water, for example, oxygen, which is similar to nitrogen in nature, as will be discussed later.

Nitrogen gas evolution at the bottom, and its effect on the gas content of the waters. Along with the study of the dissolved gases in lake water, direct measurements of the gas evolution at the bottom were made and an analytical study of them was carried out in the following way.

A glass bell-jar was suspended at about 50 cm. above the bottom in the deepest part of the lake, which, after a certain period, was hauled up, and the volume of the gas accumulated in it measured and its chemical composition examined, with results as given in Table 36.

The gas, which had accumulated during the period from May 25 to June 21, 1936, and which amounted to 221 c.c. per day/m.^{2}, was shown to be composed mainly of nitrogen, its percentage being 95.77%.

There are many reasons for supposing that this proportion of the gas is not what it was at the time the gas was evolved.

First, it is very likely that a certain chemical change occurs in some of the constituents of the evolved gas mixture while in the bell-jar. Second, when the evolved gas bubbles rise up toward the jar, it is also likely that gaseous exchanges occur between the gas bubbles and the water. Thus, both these causes might bring about a measurable change in the composition of the evolved gas.

It is also conceivable that the change in the gaseous composition cannot be so large as to make it difficult to surmise the chemical composition of the gas at its beginning from that of the gas after storage in the bell-jar.

Then, as shown by the composition of the stored gas during the period from May 25 to June 21, it is highly probable that considerable

Table 36. Volume of the gas, accumulated in the suspended glass bell-jar and its composition.

	May 25 - June 21		September 13 - October 4	
	Composition %	Accumulated amount c.c. per day/m^2.	Composition %	Accumulated amount c.c. per day/m^2.
Nitrogen	95.77	211.7	10.34	144
Methane	2.60	5.7	80.83	1129
Hydrogen	0.05	0.1	6.44	90
Carbon dioxide	0.24	0.5	1.12	16
Oxygen	1.36	3.0	0.80	11
C_nH_{2n}	0	0	0.47	7
		——(+		——(+
		221.0		1397

	October 4 - October 25		October 25 - November 25	
	Composition %	Accumulated amount c.c. per day/m^2.	Composition %	Accumulated amount c.c. per day/m^2.
Nitrogen	20.27	112	32.69	14.4
Methane	73.66	408	61.30	27.1
Hydrogen	4.31	24	3.82	1.7
Carbon dioxide	0.88	5	0.23	0.1
Oxygen	0.34	2	1.02	0.5
C_nH_{2n}	0.54	3	0.95	0.5
		——(+		——(+
		554		44.3

evolution of nitrogen occurred with the slightest evolution of other gases at the bottom, a result that agree well with the foregoing results of observation on the dissolved gases at the bottom water during the time in question, that is, early summer.

Indeed, the amount of nitrogen in deeper waters was found to increase during two periods, namely, the one just preceding and the other succeeding the period in which the above measurement of gas evolution was made. Thus, in the former period, from April 29 to May 25, the nitrogen value increased from 14.2 c.c. at 5 m. and 14.3 c.c. at 5.5 m. to 14.5 c.c. and 14.7 c.c. at corresponding depths, while in the latter period from June 21 to July 25, it also increased from 14.1 c.c. at 5.5 m. to 15.4 c.c. at the same depth.

Such an increase in nitrogen value in the deeper water layers can be explained only by evolution of this gas in the bottom water or in the mud, because the season now in question is that in which nitrogen escapes, hence one would naturally expect to find a decrease of this gas in the deeper waters, which, however, is contrary to the present case.

These circumstances indicate that the formation of nitrogen at the bottom began as early as the end of April and continued to the end of July, at least, with considerable intensity.

The fact also that no increase in nitrogen was found during May 25 to June 21, does not contradict the above direct observation of nitrogen evolution, it being easily understood if we assume that, during this period, there were circumstances owing to which a more rapid escape of the gas took place.

The conclusion, therefore, is that, in Takasuka-numa, evolution of nitrogen occurs to such an extent that its content in deeper waters often increases notwithstanding its tendency to decrease, owing to rise of temperature in the water layers in question.

Evolution of nitrogen and the bottom temperature. As shown in the foregoing paragraphs, the temperature of the bottom water is minimum in January, whence it begins to increase towards autumn, attaining maximum at the end of September or in the beginning of October.

Now, if the denitrification at the bottom is accelerated by rise in temperature, which is usually the case in ordinary chemical changes, the amount of gas evolved should increase from spring towards autumn, and attain maximum between September to October.

The writer's result is contrary to this expectation. The amount of evolved nitrogen decreased from 212 c.c. per day/m.^{2} (May – June, when the bottom water was still cold) to 144 c.c. (September 13 – October 4), and further to 112 c.c. (October 4 – 25), when the bottom water was quite warm.

The evolution of nitrogen by denitrification at the bottom is thus apparently independent of the temperature of the bottom water.

As the causes of such behaviour in denitrification at the lake bottom, the following three are conceivable:

(1) Smallness of the temperature coefficient of the reaction velocity of denitrification.

(2) The optimal temperature of denitrification is comparatively low.

(3) The existence of a certain factor that acts against rise of temperature upon denitrification.

As to (1) and (2), the writer lacks sufficient information to enable him to deal further with the problem, while (3) may be claimed to represent, at least, one of the most important causes of the observed phenomenon, and which the writer attributes to diminution in the nitrate reserve in the water.

As is well known, pre-existence of nitrate or nitrite is an indispensable condition for the evolution of nitrogen gas. Besides these, ammonium salts also part with their nitrogen when they come in contact with one of the intermediate products of nitrate reduction, nitrite, under favourable conditions.

Now, since in the case of the deeper waters of a lake, the main or often unique source of these salts, is organic deposits, evolution of nitrogen in the lake occurs in the following stages:

$$\boxed{\text{Organic deposits}} \rightarrow \boxed{\text{Ammonia}} \xrightarrow{\text{aerobiosis}} \boxed{\text{Nitrate}} \xrightarrow{\text{anaerobiosis}} \boxed{\text{Nitrogen gas}}$$

Of these three steps in the process, nitrate production occurs more in aerobiosis, while for reduction of nitrate, which in turn leads to the final evolution of nitrogen, an anaerobic condition is required.

Thus for the evolution of nitrogen gas, two different conditions, namely, aerobic and anaerobic, are necessary. In the former condition, much nitrate is formed and stored in deeper waters, and when the condition turns to anaerobic, then reduction of the nitrate begins, resulting in the formation of nitrogen.

That such a change from aerobiosis into anaerobiosis did occur during the period between April 29 and May 25, has been conclusively shown by the oxygen value of the deeper waters. In fact, on April 29, there was still sufficient oxygen, even in the deepest water, whereas on May 25, the water at a depth of 5.5 m. was already completely deprived of its oxygen, this depletion of oxygen extending thereafter into less deeper water zones.

It is, therefore, only natural that we found a heavy production of nitrogen, as much as 214 c.c. per day/m.^{2} in May – June, following the turning period.

When such an anaerobiosis had continued for a certain period, it is also natural that the stored nitrate should be gradually consumed and the production of nitrogen eventually checked, which will thus be admitted as the most probable reason for the nitrogen evolution being checked towards autumn, despite the rise in temperature of the bottom water.

To the writer's regret, there is no result of a simultaneous determination of ammonia and nitrate contents of lake waters that should support his views, although it is fortunate that his views are supported by Yoshimura's observation in 1930, provided it can be admitted that the bottom metabolism took place that year in the same order that it did in 1936.

The following quotations are from Yoshimura's paper(29) (in Japanese) that relates to the present discussion.

"In May, the water began to stagnate and, at a depth of 5 m., a sudden increase in ammonia was observed. This increase in ammonia lasted to the end of August, when the accumulated ammonia suddenly vanished......"

"With the beginning of the period of stagnation, the nitrate content of the waters increased actively for a while, but this did not last long, and the nitrate content began to decrease. The decrease was more evident in autumn, the reason for which is not yet certain......"

"Also, the organic nitrogen in the deeper waters increased slightly at the beginning of the period of stagnation, but it decreased to that value, that is customary in the period of stagnation."

When these observations of Yoshimura are compared with the writer's views, we find many points in agreement, so that the former are regarded as supporting the latter view.

It is also worth while comparing the writer's result with that of the

---

(29) Yoshimura, *The Geographical Review of Japan*, **6** (1931), 733-736.

laboratory experiment that was carried out by R. J. Allegeier, W. H. Peterson, Ch. Juday, and E. A. Birge.⁽³⁰⁾

These authors filled a number of glass bottles with samples of the bottom deposits of Lake Mendota, Wisconsin, to which was added a sufficient amount of water. The bottles were left to ferment at different temperatures, and the evolved gases examined after a lapse of about 100 and 200 days.

Of the results of this experiment, the most important to the present problem is that the percentage of nitrogen in the fermented gas in the bottle at lower temperature exceeded that of higher temperature.

Such a result can no doubt be understood only by assuming that denitrification itself is suited to a lower temperature, or that it is interfered with by a certain external force, for example, by the effect of methane or hydrogen fermentations, which are greatly accelerated by rise in temperature owing to their larger temperature coefficients.

Another result, also worth mentioning here, is that within the range of temperature examined (7°–55°C), the percentage of nitrogen in the fermented gas was smaller on the 200–210th day than on the 112th day. This observation agrees with the writer's view, according to which at least one of the limiting factors of nitrogen production is attributed to diminution in the nitrate supply.

To conclude, the reason that in Takasuka-numa, the evolution of nitrogen declines towards autumn, despite temperature elevation in the deeper waters, is explained partly by interruption in the nitrate supply and partly by assuming that denitrification had adapted itself to a lower temperature, the exact cause being yet obscure.

Methane and Hydrogen. (v. Fig. 15) *Methane and hydrogen accumulation.* Contrary to the case of nitrogen evolution, in a bell-jar suspended at the bottom, fermentations of methane and hydrogen are accelerated by rise in temperature.

The amounts of both these gases, accumulated in the suspended bell-jar, increased from their small values in early summer (2.6 c.c. per day/m.^{2} of methane and 0.1 c.c. of hydrogen) to the enormous values in the period, September 13 – October 4, of 1129 c.c. of the former gas and 90 c.c. of the latter, while fermentation had declined, the amount of the evolved methane and hydrogen having dropped to 408 c.c. and 24 c.c. between October 4 – 25, and further to 27 c.c. and 1.7 c.c. in the succeeding period until November 25.

The contents of methane and hydrogen in the lake waters varied similarly. In all the waters, neither of these two gases were found to any measureable extent until May 25. On June 21, small amounts of these gases were found first at depths of 5 and 5.5 m., with the simultaneous disappearance of oxygen. Thereafter, their contents in deeper waters increased evenly towards autumn, and on October 4 they reached the maximum values of 10.8 c.c. of methane and 0.5 c.c. of hydrogen at a depth of 5.5 m. and 13.6 c.c. and 0.8 c.c. at 6 m. Then, owing to circulation of the water, the stored gases in deeper waters were dispersed and,

---

(30) Allgeier, Peterson, Juday, Birge, *Internat. Rev. d. gesamten Hydrobiologie und Hydrographie*, **26** (1932), 444-461.

due to the accompanying temperature drop and to the supply of oxygen to the deepest water, bottom fermentation was greatly hindered. From this time onwards, no considerable amount of these gases was found in any of the water layers. This was the same as in the previous autumn.

Methane and hydrogen fermentations and temperature. One of the outstanding features in the present study of methane and hydrogen is that the daily amount of methane evolution reached in autumn to nearly 500-fold of that in early summer.

Such a huge increase in gas production cannot easily be explained merely by rise in temperature in the bottom water.

Three facters that are supposed also to contribute more or less to the huge acceleration of methane and hydrogen production towards autumn will now be discussed.

(1) Anaerobiotic condition in the deeper waters. The fermentation of the two gases in question being anaerobic, it is inhibited by the presence of oxygen, so that it is rather natural to find both these changes greatly inhibited when the supply of oxygen to the bottom is still sufficient, as in the period of circulation and at the beginning of the stagnation period. It is also natural to find gas evolution suddenly accelerated as soon as the water stagnates for a certain period and the bottom oxygen is completely consumed.

(2) Bacteria that oxidize methane and hydrogen. Next, the role played by bacteria that oxidize methane and hydrogen, will be considered.

Recently, Kuznetzov(31) and his co-workers proved the existence of such bacteria in the water of Lake Beloje, near Moscow, and tried to explain the limnological characteristics of that lake.

It is very likely that such bacteria are not restricted to any particular lake, such as Beloje, but are widely distributed in all lakes where the evolution of these gases occurs.

In the water layers of a lake, where the evolved methane or hydrogen easily comes in contact with the dissolved oxygen, we find the most appropriate breeding ground for these bacteria. In such a layer of water, the oxygen, owing to the action of bacteria, acts on the gases in question as soon as they are evolved, so long as the supply of oxygen continues there.

This will exactly be the case with the bottom water in the period of circulation or at the beginning of the period of stagnation, so that in these periods, no marked storage of dissolved methan or hydrogen in the bottom water can be expected, even if these gases were produced in considerable quantities.

In contrast to this, when the stagnation of water continues for some time and the oxygen in the deeper waters is completely consumed, bacterial decomposition of the evolved gases is hindered and the gases are dissolved (unaltered) in the water or are accumulated in the collecting jar as they are evolved.

For these reasons, the apparent evolution of these gases, as computed from the amounts accumulated in the collecting jar, is very much larger in the advanced season, compared with that in early summer.

---

(31) Kuznetzov, *Verhandl. d. Internat. Vereinigung f. theoret. und angew. Limnologie*, **7** (1935), 562-582.

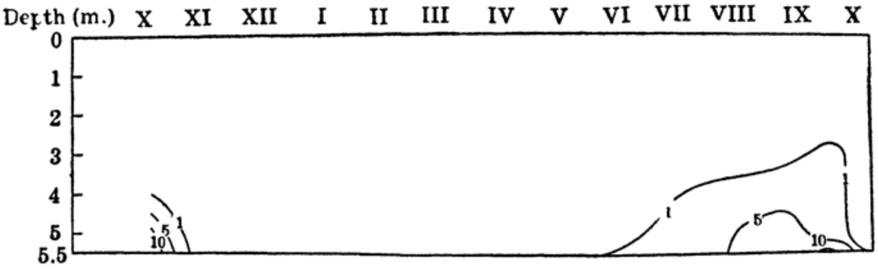


Fig. 15. Annual variation in the content of methane of the lake waters of Takasuka-numa (c.c./l.)

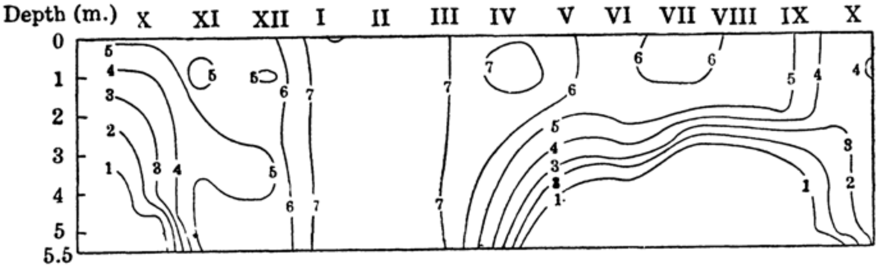


Fig. 16. Annual variation in the oxygen content of the lake waters of Takasuka-numa. (c.c./l.)

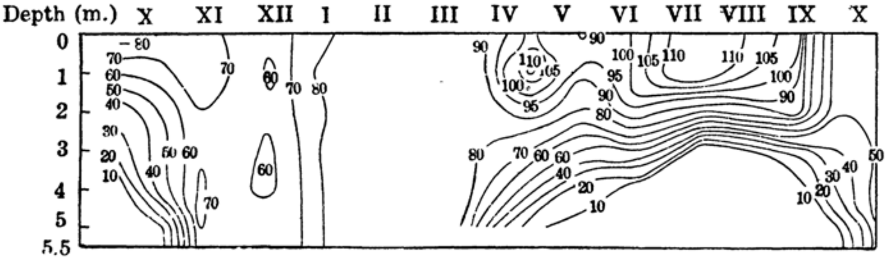


Fig. 17. Annual variation in the degree of oxygen saturation in the lake waters of Takasuka-numa (c.c./l.)

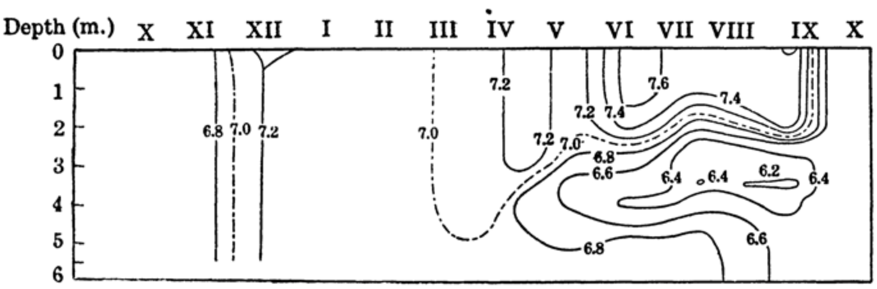


Fig. 18. Annual variation in the *p*H value of the lake waters of Takasuka-numa

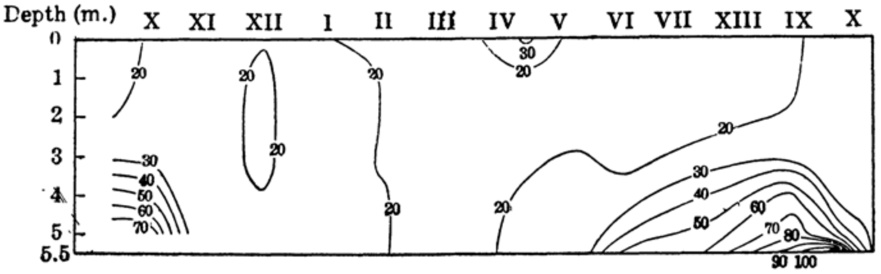


Fig. 19. Annual variation in the content of total carbon dioxide in the lake waters of Takasuka-numa (c.c./l.)

(3) Direct and indirect effects of rising bubbles. When the bottom fermentation has advanced to a certain extent, and the evolved gases have dissolved sufficiently into the waters in the neighourhood, the gases begin to escape in the form of bubbles rising towards the upper water surface. In this case, oxygen as well as other gases, that are dissolved in the waters lying in the way of the rising bubbles, are very likely to be driven out by them.

On the one hand, such a process seems to secure directly a sooner establishment of anaerobic conditions in deeper waters, while, on the other, it hinders the ingress of oxygen from the upper water zones into the deeper ones, as the result of which, anaerobiosis, once established at the bottom, is indirectly extended quickly in range and increased in intensity.

The author therefore holds that the huge increase in the accumulation of methane and hydrogen towards autumn was owing to the following three causes:

(1) Rise in temperature of the bottom water.

(2) Decomposition oxidation of the evolved methane and hydrogen by bacteria in the earlier period, and its cessation in the later period.

(3) Setting up of anaerobic conditions in the deeper waters, which in their turn, are maintained by the stagnation of water as well as by the rising bubbles of the evolved gases.

Oxygen. (v. Figs. 16 and 17) *Degree of oxygen saturation.* Although the saturation of oxygen varied between 0 and 118.6%, generally speaking, the value was usually below 100%, except those in the upper water layers during the four or five months from early summer to early autumn. Then throughout the year, excepting these months, oxygen must have been continually absorbed from the air into the hydrosphere in order to make up for its lack in the waters. Seeing that the surface water was markedly undersaturated, this absorption must have been intense, whence the bulk of the oxygen that is absorbed into the lake water per annum per unit surface must reach an enormous value in Takasuka-numa, thus differing from many other lakes, where the degree of oxygen saturation never drops to any great extent.

The roles of iron and manganese as oxygen carriers. This large quantity of absorbed oxygen is at the end consumed for the oxidative decomposition of organic matter that was either formed in the lake or had been carried into it.

But we cannot overlook the fact that, in the course of the oxidative consumption of oxygen, this gas, at any rate, does not directly act upon organic matter in general, but is frequently first combined with ferrous iron or with manganous manganese, after which the oxygen is transferred to less oxidizable organic substances. In this case, it is worth mentioning that the oxidation-reduction propert of these elements in question plays an important role in facilitating such oxygen-transference.

Such is also the case with the flocculation and precipitation of oxidized iron and manganese, by which the deeper lying organic matter are made accessible to oxygen, resulting in oxidation.

Such relation between the absorbed oxygen and the amount of iron and manganese in the lake water, is very evident in the autumnal observations.

In the autumn of 1935, the oxygen content in the waters was found to have increased in harmony with increase in solubility due to lowering of the water temperature, but saturation was found to have decreased every time it was observed from September to December.

Thus the latter value dropped gradually, for example, at the surface layer, from 85.6% on September 29 to a minimum of 63.5% on December 15.

When we compare these data with those of nitrogen, and are reminded of the above conclusion that the absorption velocity of oxygen by water can never differ much from that of nitrogen, and that, in the case of nitrogen, absorption occurs so easily that it adapts itself to the autumnal lowering of temperature of the lake waters, we know that the small autumnal value of oxygen saturation does not arise from any interruption in the supply of this gas, but from its heavy consumption by a huge quantity of oxygen-consuming substances present in the water.

The amount of these oxygen-consuming substances in the waters was greatly reduced towards the end of December, as shown by a sudden increase in oxygen saturation in the observation of January 19, despite the still continued lowering of the temperature of the lake water.

From the variation in the iron and manganese contents in the waters that ran parallel with that of the degree of oxygen saturation, the writer concludes that these two elements are nothing but substances that play, at least, one of the main roles in the consumption of oxygen by the autumnal waters.

In fact, as shown in Tables 12 and 13, the contents of iron and manganese in the upper waters increased continually from September to November, corresponding to the decreasing variation in oxygen saturation, after which the iron and manganese contents began to decrease to their minimum values on January 19, also corresponding to the sudden increase in oxygen saturation as well as in its absolute content in the water. It might be scarcely worth repeating that the direct determination of iron and manganese by the suspending bottle method (Table 15) also gave a result that pointed to a similar variation in iron and manganese deposition. The circulatory property of iron and manganese is also significant in the matter of oxygen supply to the deeper waters.

As shown in the foregoing paragraphs, these elements precipitate upon combining with oxygen, and when they part with this oxygen to combine with other oxidizable substances at depths, they change into the soluble state, whence they are carried back by the circulation current into the upper water zone.

In this way, owing to this circulatory property of these elements, the transport of oxygen from the upper aerated zone to the deeper oxygen deficient zone is effected by these carriers, so long as there remain substances to receive the oxygen in the deeper waters.

Thus the role of iron and manganese as oxygen carriers is accomplished by their circulatory property.

It will also be of some interest to compute the amount of oxygen that is carried downwards by the elements in question per annum, per unit area, and to estimate roughly the percentage of the organic deposite that are likely to be oxidized completely by this computed amount of oxygen.

In the previous determination of deposition, what was lacking in observation was made up as shown in Fig. 4, the percentages of iron and manganese in the deposit for this period being assumed equal to 10.78% and 0.74% respectively, the mean values of 12 observations during the whole period. Then the deposited iron during a period of one year, June 24, 1935 – June 24, 1936, was computed to be 0.179 g. (as Fe_2O_3), and the corresponding manganese to be 0.0122 g. (as MnO). The loss in oxygen that occurred in the reductions of Fe_2O_3 to FeO and of MnO_2 to MnO, are respectively 10.02% and 18.45%, referred to the mother oxides, whence the amount of oxygen that was freed by the above deposited iron and manganese, must amount to 17.9 mg. and 2.6 mg. respectively, and to a total of 20.5 mg. per annum/cm.^{2}.

On the other hand, the annual deposition of organic matter is roughly estimated as follows. The total deposition per annum is computed to be 1.66 g. as given in the foregoing paragraph. The mean percentage of carbon in this deposit is assumed to be 6.00% and the mean composition of the organic deposit is taken to be practically the same as that of cellulose. These assumptions are admissible from the result of analysis of the mud, given above. The annual deposition of organic matters is then equivalent to about 99.6 mg. of cellulose, which requires 124 mg. of oxygen for its complete combustion, so that the oxygen transported by manganese and iron amounts to 16.5% of this cellulose requirement.

But since the actual supply of oxygen is not restricted to that by iron and manganese, the actual amount of organic deposits that are destined to oxidative decomposition would considerably exceed this estimate.

At any rate, it is noteworthy that more than 16.5% of the deposited organic matter is decomposed into carbon dioxide, again to be assimilated.

Mention should be made here that the expenses of the present studies were partly defrayed by grants from the Imperial Academy and from the Ministry of Education, and that this publication was accomplished through financial aid from the Sakamoto-hôkôkai.

Also the writer is deeply indebted to Professor Y. Shibata for the ardent encouragement he gave in the present studies, and for his kind advice in the publication of the results.

Thanks are also due to Dr. K. Kikuchi of the Tokyo Teachers College for Women, Dr. S. Yoshimura of the Tokyo Bunrika University, Messrs. M. Kawajiri and K. Hata of the Kizaki Station of the Imperial Fisheries Experimental Institute, and to the co-workers in the writer's laboratory for the kind help and facilities rendered him.

Chemical Laboratory, Tokyo Higher School.