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REVIEW

Current status of water environment and their microbial biosensor techniques – Part I: Current data of water environment and recent studies on water quality investigations in Japan, and new possibility of microbial biosensor techniques

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Abstract

In my 2010 review, I addressed conventional water analysis and biosensing of organic pollutants in Japan between 1960s and 2000s. It is now timely to reexamine current analytical and biomonitoring approaches in view of the new challenges in assessing pollution, particularly in closed water bodies, as pollutants tend to accumulate in these endorheic basins. In the present review series, I presented current water environment and its microbial biosensors. In this part, I presented current data of the water quality of these water bodies in Japan and established the need to further develop microbial biosensor technologies to address and monitor water quality here.

Keywords Review · Water quality investigation · Water pollution · Organic pollution · Anthropogenic eutrophication · Toxification

Abbreviations

A2O	Anaerobic-Anoxic-Oxic
AHS	Aquatic humic substances (hydrophobic acids; HoA: contains humic & fulvic acids)
BaS	Bases (Hob and HiB)
BOD	Biochemical oxygen demand
COD	Chemical oxygen demand
DO	Dissolved oxygen
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
FY	Fiscal year
HABs	Harmful algal blooms
HiA	Hydrophilic acids
HiB	Hydrophilic base
HiF	HiA+BaS+HiN
HiN	Hydrophilic neutrals
HoA	Hydrophobic acids
HoB	Hydrophobic bases
HoN	Hydrophobic neutrals
LASs	Linear alkylbenzene sulfonates
NP	Nonylphenol
SAS	Standard activated sludge
SOM	Sediment organic matter
TN	Total nitrogen
TP	Total phosphorus
TDAA	Total dissolved amino acids
TDNS	Total dissolved neutral sugars
THMFP	Trihalomethane formation potential
TZn	Total zinc

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Overview

In the rapid economic growth around the 1960's (1955–1973) water pollution in Japan worsened because of the delay in

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construction of sewage and industrial wastewater treatment plants. Furthermore, pyrophosphate as an auxiliary agent contained in synthetic detergent increased the damage from water pollution [1, 2]. Figure 1a illustrates an instance of organic pollution by direct influx of wastewater and coexistence of eutrophication and toxification in water body. On the other hand, recent water pollution occurred indirectly by anthropogenic eutrophication due to influx of effluent containing nutrient salts after wastewater treatments (Fig. 1b). In closed water bodies, in particular, anthropogenic eutrophication causes water bloom (or algal bloom) and organic pollution by accumulation of dead algal bodies. Further, these pollutions occasionally produce toxic substances.

In this part of the present review series, status of main legislation, wastewater treatment, and environmental water monitoring are described, and the need to develop new microbial biosensors for actual water environments is discussed.

Current status of Japanese water environment

Main legislation concerning water pollutions in Japan

To solve the problem of water pollutions, the government executed a Water Pollution Control Law in 1971 [3]. This law regulates the discharge of water from factories and workplaces to public water bodies and the penetration of water into the underground. Further, by promoting implementation of domestic wastewater countermeasures, this law aims to prevent the pollution of public water bodies and groundwater. In 1993, the Basic Environment Law Article 16 was decreed, and then two types of the environmental standards were set for the protection of human health and for preservation of living environment [4]. The former standards currently include a total of 27 items as required monitoring items. The latter standards include the items concerning organic pollution, eutrophication, and aquatic toxicity, and these items are measured for each of three waterbodies consisting of rivers (total eight items excluding items concerning eutrophication), lakes (total 10 items), and closed seas (total 10 items).

Wastewater treatment

Standard method (SAS method) After the rapid economic growth, biodegradable organic matter is effectively removed from wastewater in the sewage and industrial wastewater treatment by employing a standard activated sludge (SAS) method. The removal rate of easily biodegradable organic matter by the SAS method is approximately 90%–95% at the biochemical oxygen demand (BOD) value [5]. The introduction rate of the sewage treatment system has risen year by year and reached 77.8% in FY 2016 (at fiscal year ended March; the data from Japan Sewage Works Association; JSWA). However, the SAS method fails to sufficiently remove nutrient salts such as nitrogen (N) and phosphorus (P), and the removal rates of total-nitrogen (TN) and total-phosphorus (TP) are from 20% to 30% and approximately 40%, respectively. Thus, causes of recent organic pollution in environmental water have been changing from biodegradable organic matter to nutrient salts. Figure 1b illustrates causes of water pollution in recent years. In most cases, the organic pollution occurs indirectly by anthropogenic eutrophication due to influx of effluent containing nutrient salts after the SAS treatment of wastewater from houses and industries. Because of this, the SAS treatment method was developed principally to remove biodegradable organic matter in wastewater; the method simply employed decomposer consisting of various microbial flora in the activated sludge. Therefore, in principle of the SAS method, it is impossible to remove nutrient salts efficiently.

Advanced method (A2O method) In the eutrophied water, nutrient salts act as inorganic nutrients to grow cyanobacteria (producer), so called "blue-green algae", which belong to phytoplankton. In recent years, the influence of nutrient salts on environmental waters has increased tremendously in urban areas because of increase of people who reside in such areas. For this reason, a superior treatment system that can effectively remove nutrients is being introduced in newer sewage treatment plants. In the superior treatment system, one of the most effective methods is an anaerobic-anoxic-oxic (A2O) method. In the A2O method, TN and TP are removed from 65% to 75% and 75% to 85%, respectively, and BOD is removed as satisfactorily as in the SAS method [5]. In fact, the introduction rate of the superior treatment system in the sewage treatment plant went from 4% (FY1995) to 45.6% in FY 2015 (data from Ministry of Land, Infrastructure, Transport and Tourism; MLIT) [6].

Application examples The Organization of Bureau of Sewage, Tokyo Metropolitan Government, has a total of 13 sewage plants. The water quality value of six indicators obtained from the influent or effluent in FY 2015 are shown as follows (averaged value of 13 sewage treatment plants); BOD: influent 151 mg L^{-1}, effluent 6 mg L^{-1} (removal rate 96%), chemical oxygen demand (COD; an index for total organic matter): 82 and 9 mg L^{-1} (89%), suspended solids (SS): 109 and 2 mg L^{-1} (98%), N content: 30.3 and 12.2 mg L^{-1} (60%), and P content: 3.3 and 1.0 mg L^{-1} (30%) [7]. Then, all water quality was below the Uniform National Effluent Standards value set by the Ministry of the Environment (MOE); BOD: 160 mg L^{-1} (day average 120 mg L^{-1}), COD: 160 mg L^{-1} (120 mg L^{-1}), SS: 20 mg L^{-1} (150 mg L^{-1}), N content: 120 mg L^{-1} (60 mg L^{-1}), and P content: 16 mg L^{-1} (8 mg L^{-1}) [8].

In addition, in the cases of Tokyo, approximately 80% of effluent flowing into environmental water is based on

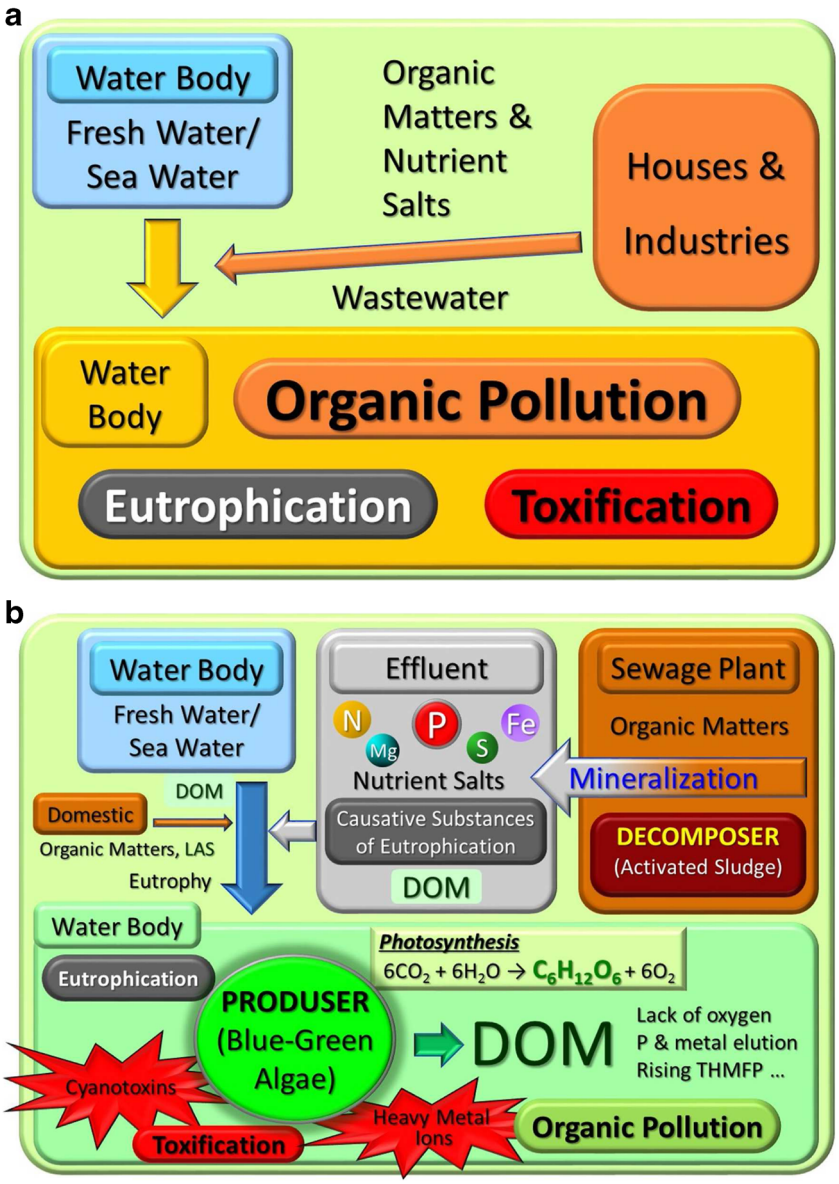

Fig. 1 Difference in cause of water pollution (a: 1960 and b: current). **(a)** Causes of water pollution in 1960. Organic pollution occurred directly by wastewater from houses and industries. **(b)** Causes of anthropogenic eutrophication in recent years. Organic pollution occurred indirectly by anthropogenic eutrophication due to influx of effluent after sewage treatment of wastewater from houses and industries

household wastewater, with 80% drainage volume, 77% COD, 79% N content, and 78% P content (in the case of industrial wastewater: with 6% drainage volume, 8% COD, 4% N content, and 6% P content). Further, the household wastewater consists of miscellaneous and toilet drainages, and among them, toilet drainage occupies 20% drainage volume, 26% COD, 75% N content, and 65% P content (Tokyo Metropolitan Government Bureau of Environment, 2005).

Environmental water monitoring

BOD · COD Contrary to such efforts for water pollution, the situation continues where improvement is not seen mainly in closed waters such as lakes. In Table 1, state of achievement of environmental water quality standards of BOD or COD value obtained during 41 y between FY 1974 and FY 2015 are compared. Here, FY 1974 was the year after the rapid economic growth in Japan (FY 1955–1973) [9, 10]. In general, organic pollution of river water estimates by BOD value and that of closed water estimates COD value. In Fig. 2 and Electronic Supplementary Material (ESM) Table S1, similar results for 36 y between FY 1979 and FY 2015 by annual averaged values were also obtained for river waters (–67% from 3.3 to 1.1 mg O_2 L^{-1} BOD), lake waters (-24% from 4.2 to 3.2 mg O_2 L^{-1} COD), and closed sea waters from inland seas and bays (+0.6% from 1.7 to 1.8 mg O_2 L^{-1} COD) [9, 10]. Then, the averaged values for 36 y (i.e., $n = 37$) were BOD_{av} 2.1 ± 0.7 mg O_2 L^{-1} for river waters, COD_{av} 3.5 ± 0.2 mg O_2 L^{-1} for lake waters, and COD_{av} 1.8 ± 0.1 mg O_2 L^{-1} for closed sea waters. As a result, water qualities of rivers have maintained good conditions because of the prevalence of sewage plants, the rainy climate, and the mountainous topography

Table 1 State of achievement of environmental water quality standards by BOD or COD value in Japanese public water [9, 10]

Fiscal year	River (BOD)	Lakes (COD)	Closed sea* (COD)	Total (Number of sampling points)
1974	51.3%	41.9%	70.7%	54.9% (1,927)
2015	95.8%	58.7%	81.1%	91.1% (3,340)
Improvement	+44.5%	+16.8%	+10.4%	+36.2% (+1,413)

*Inland seas; Seto Sea, Ariake Sea, Yatsushiro Sea, bays; Tokyo Bay, Ise Bay, and Osaka Bay.

creating many short rivers with steep slopes in Japan. Conversely, difficulty to improve the water quality of lake water is clearly shown.

TN · TP Anthropogenic eutrophication caused by nutrient salts such as nitrogen and phosphorus have induced organic pollution in closed water bodies after abnormal proliferation of algae due to accumulation of algae cell walls to water bottom. Therefore, estimates of both TN and TP values have continued in lake waters since 1985 and in closed sea waters since 1995. Figure 3a, b, and Tables 2 and 3 show the state of achievement of environmental water quality standards and TN or TP value; Table 2 shows the data from lake water obtained in FY 2015 and between FY 1985 and FY 2015, and Table 3 shows the data from closed sea water obtained in 2015 and between FY 1995 and FY 2015 [9, 10].

A total of 185 red tides occurred in the closed sea waters of Japan in 2015, 80 of which occurred in the Seto Inland Sea. In the Seto Inland Sea, 299 red tides occurred in 1976. This red tide can cause blue tides. This is because when the dead body of phytoplankton that formed the red tide is decomposed at the bottom of the water, the surroundings become oxygen deficient. The oxygen deficient state makes sulfur colloids, which cause the blue tide. Just as the red tide, it can cause mass death of fish and shellfish. However, these phenomena are greatly reduced in recent years.

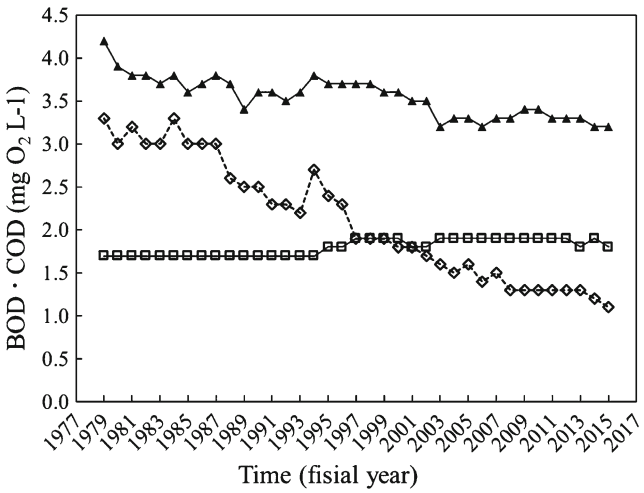

Fig. 2 State of BOD or COD value in Japanese public waters [10]. River water (◊), lake water (▲), and closed sea water (□)

Compared with the results obtained from closed sea waters, the results obtained from lake waters maintained relatively high values, and in particular the TN values were notably high. This indicates that eutrophication in the Japanese lakes continues constantly without dissolution. Further, such a trend in lakes was more severe in urban areas where the population concentrates. Since 1985, a total of 11 lakes such as Lake Biwa, Lake Kasumigaura, etc. have been labeled as designated lakes (including two brackish lakes) and are under the Law Concerning Special Measures for Conservation of Lake Water Quality. Under the Lake Water Quality Conservation Plan, special measures such as promotion of projects that contribute to sewage water quality, conservation, and regulation of loads such as factory wastewater have been implemented. As a result, although pollution loads flowing into the lakes have been reduced, environmental water quality standards have yet to be achieved in most designated lakes. Therefore, since 2006, the

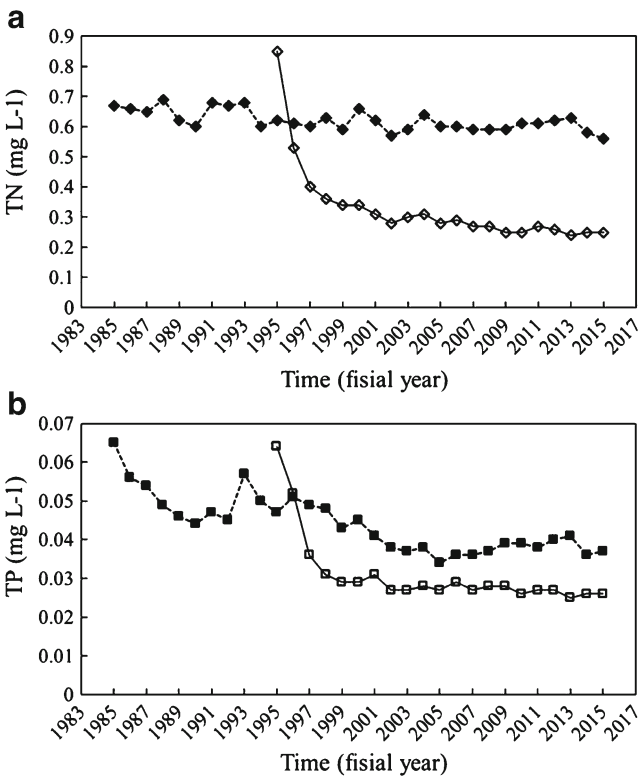

Fig. 3 State of nutrient salt concentration in Japanese closed waters [10]. **(a)** TN in lake water (♦), TN in closed sea water (◊); **(b)** TP in lake water (■), and TP in closed sea water (□)

Table 2 State of achievement of environmental water quality standards and TN or TP concentration in water of Japanese lakes [9, 10]

Fiscal year	TN			TP			Total	
	Achiev. rate (%)	Conc. (mg L^{-1})	Sampl. points (*n*)	Achiev. rate (%)	Conc. (mg L^{-1})	Sampl. points (*n*)	Achiev. rate (%)	Sampl. points (*n*)
2015	12.8	0.56	39	54.5	0.037	121	51.2	121
Total (1985–2015)*								
Average (*n* =31)	10.0	0.62		48.6	0.044		43.0	
SD (±)	3.6	0.03		5.9	0.007		6.5	

*The values are calculated from the values in Table S2a.

law was revised to improve water quality of the designated lakes, to reduce pollutant loads flowing into the lakes from so-called "surface sources" such as farmlands and urban areas. In addition, it also provided protection of vegetation surrounding the lake that contribute to improvement of lake water quality. The results are shown in Fig. 4 and Table 4. Averaged values during 10 y from FY 2006 to FY 2015 of each indicator were 5.8 ± 0.2 mg O_2 L^{-1} COD, 0.97 ± 0.03 mg L^{-1}, and 0.074 ± 0.005 mg L^{-1}, respectively. Figure 4 also shows the data obtained from the other lakes. Despite various efforts, the water quality of the designated lakes continues to be much lower than the other lakes (from FY 2006 to FY 2015; 3.2 ± 0.1 mg O_2 L^{-1} COD, 0.60 ± 0.02 mg L^{-1}, and 0.038 ± 0.002 mg L^{-1}) and there is no sign of improvement [10].

THMFP In 1995 in relation to organic pollution, THMFP was defined as an item for conservation of water quality in water bodies for water supply [11]. In Japan, the coverage of the water supply system is 100%; therefore, it is important to prevent water supply damage. Trihalomethane is generated by organic matter such as humic substances contained in raw water with chlorine injected in the process of water treatment. Trihalomethane is a compound in which three of the four hydrogen atoms of methane (CH_4) are replaced by halogen atoms such as chlorine and bromine, and is a carcinogenic substance. Four are representative substances, i.e., chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$), bromoform ($CHBr_3$), and dibromochloromethane ($CHBr_2Cl$). Environmental standard of the THMFP value is not currently set; however, its target value is provided depending on water temperature, e.g., THMFP value of 0.07 mg L^{-1} at a temperature exceeding 20 °C and not more than 25°C. The results of measurements are shown in Fig. 5 and ESM Table S3. Although the data obtained from the first 2 y in lake waters showed high averaged THMFP values, thereafter they kept stable values. The averaged THMFP value of 513 points in FY 2015 was 0.048 mg L^{-1} and an averaged THMFP value during 18 y from FY 1997 to FY 2015 was from 0.04 to 0.05 mg L^{-1} [10].

Environmental Water Quality Standard for Aquatic Organisms In 2003, Environmental Water Quality Standard to protect Aquatic Organisms was established in Japan, and consequently reduction of aquatic organisms and reduction of biodiversity were seen in various places [12]. It was recognized that standards considering the effects on aquatic organisms are necessary. The standard was already established in Europe and in the USA. At the first, total zinc was set as the standard, and as guideline values as required monitoring items, chloroform, phenol, and formaldehyde were also set to be monitored (currently, 4-t-octyl phenol, aniline, 2,4-dichlorophenol are newly added as required monitoring items). Currently, the standard value for the concentration of total zinc (TZn), nonylphenol (NP), or linear alkylbenzene sulfonates (LASs) are set for each type of the environmental waters in Japan.

Table 3 State of achievement of environmental water quality standards and TN or TP in water of Japanese closed seas [9, 10]

Fiscal year	TN			TP			Total	
	Achiev. rate (%)	Conc. (mg L^{-1})	Sampl. points (*n*)	Achiev. rate (%)	Conc. (mg L^{-1})	Sampl. points (*n*)	Achiev. rate (%)	Sampl. points (*n*)
2015	96.0	0.25	151	88.7	0.026	151	86.8	151
Total (1995–2015)*								
Average (*n* =21)	83.2	0.33		83.1	0.031		76.1	
SD (±)	16.0	0.13		12.1	0.009		15.1	

*The values are calculated from the values in Table S2b.

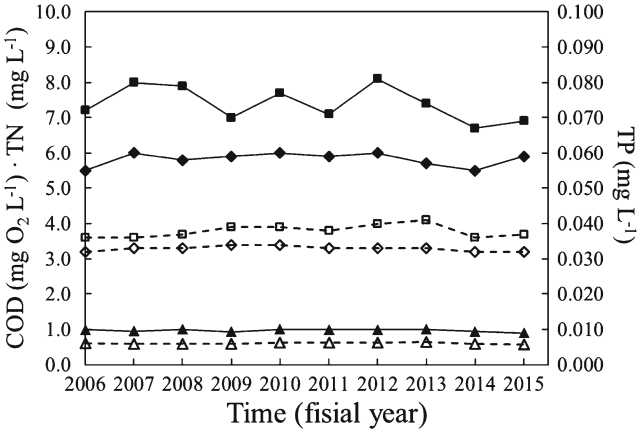

Fig. 4 Comparison of water quality between designated lake waters and the other lake waters [10]. COD (♦), TN (▲), and TP (■) in designated lake waters, and COD (◊), TN (△), and TP (□) in the other lake waters

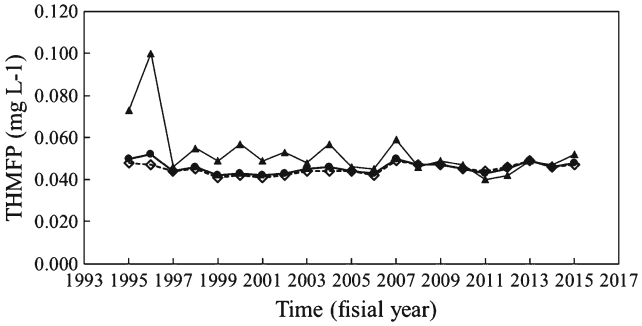

Fig. 5 State of THMFP value in Japanese river and lake waters [10]. River water (◊), lake water (▲), and average of river and lake waters (●)

TZn Zinc is an essential nutrient that can also be toxic if accumulated in excessive amounts [13]. Zinc is contained in foods, household goods, human excreta, household and industrial wastewaters, etc. and is discharged. The value of zinc concentration in the environmental water quality standard is below 0.03 mg L^{-1} in river or lake water and below 0.01 or 0.02 mg L^{-1} in closed sea of each type [12]. In FY 2015, the achievement of environmental water quality standards were 96.6% in river water (total 3655 points) and 100% in lake water (total 311 points) [10]. In the case of the closed sea water, TZn concentration was below 0.01 mg L^{-1} in 93.5% of the sampling points (total 944 points).

NP $C_{15}H_{24}O$ (NP) is artifact organic matter and is persistent in the aquatic environment, moderately bioaccumulative, and extremely toxic to aquatic organisms as well as nonylphenol ethoxylates (NPEs), NP is produced in large volumes, with uses that lead to widespread release to the aquatic environment. NPEs are nonionic surfactants and degrade to NP [14, 15]. The value of NP concentration in the environmental water quality standard depends on the waterbody type (from 0.6 to 2 μg L^{-1}) [12]. In FY 2015, collection of environmental water was conducted at 2359 points in the rivers, 201 points in the lakes, and 517 points in the closed sea areas. As a result, only 7 points (0.30%) in the rivers exceeded 0.6 μg L^{-1} and other points in both lakes and closed sea area did not exceed the value (the highest value was less than 0.3 μg L^{-1} in lake waters and less than 0.2 μg L^{-1} in closed sea waters).

LASs LASs are artificially synthesized organic compounds. Nonetheless, LASs have a property of decomposing readily and aerobically [16]. LASs are anionic surfactant having average chain length of 11.8, and major components of laundry detergent [17]. The value of LASs concentration in the environmental water quality standard depends on the waterbody type from 0.02 to 0.05 mg L^{-1} in river and lake waters, and from 0.006 to 0.01 mg L^{-1} in closed sea waters [12]. In FY 2015, collection of environmental water was conducted in

Table 4 State of environmental water quality of designated lakes in Japan [10]

(*n*)	Fiscal year	COD		TN		TP	
		Conc. (mg O2 L^{-1})	Sampl. points (*n*)	Conc. (mg L^{-1})	Sampl. points (*n*)	Conc. (mg L^{-1})	Sampl. points (*n*)
1	2006	5.5	43	0.99	36	0.072	39
2	2007	6.0	43	0.95	36	0.080	39
3	2008	5.8	46	1.00	39	0.079	42
4	2009	5.9	46	0.93	39	0.070	42
5	2010	6.0	46	1.00	39	0.077	42
6	2011	5.9	46	0.99	39	0.071	42
7	2012	6.0	46	0.99	39	0.081	42
8	2013	5.7	46	1.00	39	0.074	42
9	2014	5.5	46	0.94	39	0.067	42
10	2015	5.9	46	0.90	39	0.069	42
Average		5.8		0.97		0.074	
SD (±)		0.2		0.03		0.005	

2276 points in the rivers, 201 points in the lakes, and 546 points in the closed sea areas [10]. Iti showed that 72 points (3.2%) in the rivers and 1 point (0.5%) in the lakes exceeded 0.02 mg L^{-1} and 6 points (1.1%) in the lakes exceeded 0.006 mg L^{-1}. In addition, in FY 2015, three guideline values as required monitoring items were also measured and there was no excess of guideline values.

In FY 2015, the environmental standards related to human health protection were measured in a total of 26 items as required monitoring items; the sampling points were 544 to 950 points for rivers, 20 to 51 points for lakes, and 58 to 172 points for closed seas, respectively [10]. As a result, the guideline value had several points exceeding in the four items.

As shown here, current status of environmental water quality in Japanese water bodies was introduced in detail. Thus, the current status of water pollution is observed mainly in lake waters. In the next section, recent studies on water quality investigations are introduced and the possibility of microbial biosensor to adopt as a new tool for water quality investigations are discussed.

Recent studies on water quality investigations

New aspects

Recently, in closed waters such as lakes and reservoirs, the reasons why water pollution has not been resolved for long time are beginning to be understand. Advanced studies on organic pollution are comprehensively and intensively being carried out centered on the National Institute for Environmental Studies (NIES) ahead of the world in Japan. Here, Japanese studies on organic pollution are mainly introduced. The opportunity started from the study by Imai et al. in 2001 [18].

A steady increase in dissolved organic matter (DOM) has been observed particularly in several designated lakes, even though extensive measures have been implemented in order to reduce organic pollutant loadings from their catchment areas. For instance, in Lake Biwa, the largest lake in Japan, COD as a parameter for total organic matter has been gradually increasing in the surface water since 1984 (Shiga Prefecture, 1996) [19]. On the other hand, BOD as an indicator for easily biodegradable organic matter has remained virtually constant.

To investigate the cause of COD increase occurring in the lakes, Imai et al. improved the fractionation method of dissolved organic matter (DOM) in environmental waters, which was established by Leenheer in 1981 [20], and fractionated DOM using three kinds of resin adsorbents into five types, i.e., aquatic humic substances (AHS), hydrophobic neutrals (HoN), hydrophilic acids (HiA), bases (BaS), and hydrophilic neutrals (HiN). Here, AHS are typical recalcitrant DOM, account for 30%–80% of DOM as dissolved organic carbon (DOC), and constitute the largest fraction of natural organic matter in water [21]. BaS includes both hydrophilic and hydrophobic bases because hydrophobic base is only slightly contained in the environmental waters [22]. In some cases, the hydrophilic bases are represented by HiB. These DOM fractions gave rise to many findings concerning organic pollution.

In general, AHS as main substances of DOM has been studied ixtensively for environmental water analysis worldwide, and consists of two hydrophobic acids, i.e., humic acid and fulvic acid, which were standardized by International Humic Substances Society (IHSS) for the studies on DOM [23]. In recent years, DOM in environmental water has been measured spectrophotometrically, such as fluorescence or absorption [24, 25]. However, these methods are limited to investigate organic pollution in detail.

On the other hand, Imai et al. investigated organic pollution by employing the fractionated DOM, and then focused on Lake Kasumigaura, the second largest lake in Japan (hypertrophic lake; surface area: 171 km^2, average depth: 4 m, maximum depth: 7 m, water volume: 0.85 km^3, watershed: 2200 km^2). At first, fractionated DOM were characterized [18]. About AHS is described above. HoN contains pesticides, carbonyl compounds, and LAS. HiA contains carboxylic acids (fatty and hydroxyl acids) and sugar acids. BaS contains aromatic amines, protein, amino acids, and amino sugars. HiN contains oligosaccharides and polysaccharides. Next, the composition of the DOM contained in the environmental waters entering and leaving the Lake Kasumigaura was characterized. As a results, several important findings were newly obtained, i.e., it was found that not only AHS but also HiA were included in DOM in environmental water. Water that was artificially affected tends to contain HiA as well as AHS (domestic sewage, sewage-treatment-plant effluent, river water, lake water, etc.) and sewage treatment water was the closest to the water quality of Lake Kasumigaura among the environmental waters investigated in this study [26]. On the other hand, water requiring a long time to contact the soil tended to contain a lot of AHS (forest stream and plowed-field percolate). In addition, HoN containing LAS-like DOM was without exception included in artificially affected water [27].

Influences of fractionated DOM on the THMFP value

In Japan during the 1970s to early 1980s, studies on trihalomethane formation by humic substances in raw water were actively carried out, although isolation of humic substances occurred in the mid-1980s. Then, Imai et al. investigated the source of trihalomethane formation using the water of Lake Kasumigaura in 2003 [28]. Although the water of Lake Kasumigaura is used as water supply, eutrophication and organic pollution are concurrent because sewage treatment water also flows in. Therefore, THMFP-test of the water was important in ensuring safety as drinking water. Thus, the

THMFP values of AHS and hydrophilic fractions (HiF) (HiF=HiA+BaS+HiN) in a water were compared. As a results, the THMFP value of HiF (0.374 μmol THM L^{-1}) was higher than that of AHS (0.229 μmol THM L^{-1}) in spite of DOC concentration of HiF was about two times higher than that of AHS. This resulted in a reversal of the conventional theory that humic substances were causative material of trihalomethane formation.

Influences of DOC produced by phytoplankton on the water quality

In the case of lake water, the DOM is mainly supplied from two sources, i.e., allochthonous DOM derived from the catchment area and autochthonous DOM produced within lakes. The allochthonous DOM mainly consists of terrestrial humic substances and recalcitrant to bacterial degradation, whereas the autochthonous DOM are produced by dominant phytoplankton particularly in eutrophic lakes and changed to recalcitrant DOM by bacterial degradation [29, 30]. DOM excretion by phytoplankton depends on the light and temperature [31]. Water bloom is a phenomenon peculiar to anthropogenic eutrophication caused by abnormal proliferation of the blue-green algae *Microcystis aeruginosa*, which is the cause of algae in Japanese lakes. Choi et al. investigated the influences of UV irradiation on the algal DOM that was produced by *M. aeruginosa*, and biodegradability after UV treatment of the algal DOM [32].

As a result, no significant changes to the amount of DOC in the algal DOM were observed at any growth phase of *M. aeruginosa*. However, biodegradability of the algal DOM using bacteria collected from hyper-eutrophic pond, was changed by UV treatments and decreased by lessening the wavelength, i.e., no treatment > UBA > UVB. They also fractionated DOM into five fractions just as in the previous study [21]. The HiA and HiB fractions contained most frequently were produced by *M. aeruginosa* (HiA representing from 62.6% to 70.7% and HiB representing from 7.2% to 23.2% of the algal DOM). As the other fractions, HiN fractions were about 10%, and hydrophobic fractions, i.e., HoA and HoN, were less than 13%. The fluorescence at 270 (Ex)/350 nm (Em) to the algal DOM was decreased by UV treatments and then, amounts of several carboxylic acids (oxalic, formic, and acetic acids) contained in the HiA fractions increased. However, there was no difference in biodegradability of the HiA fractions between pre- and post-UV treatments. Biodegradability of HiB, which contains protein-like DOM, decreased significantly after UV treatment.

As in the other studies by them, it also found that the frequency of the water bloom caused by *M. aeruginosa* increases in sunshine hours [33], whereas the freshly produced DOC of phytoplankton origin can decomposed easily in a Japanese oligotrophic lake [34]. These studies showed an increase in the frequency of water bloom results in a sustained increase in recalcitrant DOM as a result, in combination with the influx of allochthonous DOM in Japan. Therefore, the algal DOM biosensor developments using biodegradable bacteria living in eutrophied water body are required.

Influences of microbial activity on the DOC in water quality

In lakes with high closedness occurring anthropogenic eutrophication, sewage treated effluent containing nutrient salts and recalcitrant DOM often flows a lot. The Lake Kasumigaura is not an exception; it was found that the water quality was the closest in the inflow water to the sewage treated water [18].

Kawai et al. studied effective COD removal by controlling the substrate degradability during the anaerobic digestion of recalcitrant wastewater [35]. The recalcitrant wastewater can then be anaerobically digested with labile synthetic wastewater by acclimatization of microbes to recalcitrant wastewater degradation; then the effective COD removal of recalcitrant wastewater can be maintained when the proportion of labile wastewater in the substrate is controlled. Such microbe degradable recalcitrant DOM should be isolated and applied to microbial biosensor developments.

Kawasaki et al. estimated molecule size of DOM. Over 35,000 Da of DOM could mainly come from microbial biopolymers. On the other hand, in sediment pore water, the increase in DOM concentrations with depth resulted in the increase of DOM with a size ranging to 2000 Da [36].

Next, Kawasaki et al. estimated bacterial contribution to DOM pool in eutrophied water [37]. Then, they performed incubation experiments using filtered waters and bacteria sampled from Lake Kasumigaura. Due to high bacterial activities, the total bacterial production accounted for 34% to 55% of the DOC consumption, and the DOC degradation was 12% to 15%. Furthermore, total dissolved amino acids (TDAA) and total dissolved neutral sugars (TDNS) contained in DOM constituents were preferentially degraded from 30% to 50%. These values of the total bacterial production were much higher than those estimated for the open ocean (20% to 30%) [38]. In conclusion, bacterially derived carbon was accumulated as recalcitrant DOM, and the role was more important in carbon cycles in freshwater environments than in open oceans. Thus, these results indicate that recent increases in recalcitrant DOC in various lakes could be attributed to bacterially derived carbon supplied from both sewage treated effluent and allochthonous phytoplankton. These studies showed the possibility of developing biodegradable DOM microbial biosensors by employing highly degradable bacteria isolated from such hypertrophic lakes.

Subsequently, Shimotori et al. investigated costal DOM. The costal DOM in water sampled from three locations in Japan, Tokyo Bay, Kashima Port, or Cape Inubo, had two

regions of the molecular weight, one was approximately 10^5 Da and another was 1000–2000 Da. By fluorescence and UV absorption measurements, it was found that the low molecule costal DOM had carbohydrates that are usually labile, and the total DOC concentrations were 0.80–1.38 mg carbon L^{-1} (mg C L^{-1}). On the other hand, the high molecule one accounted for 75%-80% of the total DOC and showed a humic-like fluorescence and UV absorption [39]. This study showed the costal water contained labile DOM, and therefore the possibility of developing sea DOM microbial biosensors.

It is known that dominant in freshwater is the beta-proteobacteria [40, 41] and in sea water is the alpha-proteobacteria [42], although most bacterial activities are similar between freshwater and sea bacteria [43]. In the cases of Lake Kasumigaura, beta-proteobacteria composed 84% and assimilated only a few types of amino acids or carbohydrates and preferred to assimilate organic acids [44]. Baldock et al. reported that DOM decomposition by bacteria was very different in freshwater and sea systems [45]. This result suggests that the chemical compositions of recalcitrant DOM could be different between oceans and lakes. In ocean, most of DOM is of phytoplankton origin, whereas in Lake Kasumigaura, various DOC sources contributed to DOM. The major components may be terrestrial humic substances, which are derived from decaying plant debris and are major components of organic matter in soil and freshwater environments.

Imai et al. reported that about 32% of DOC consisted of humic substances in Lake Kasumigaura [18], although the value is much lower in the open ocean, i.e., 0.7% to 2.4% (Opsahl and Benner) or ~10% (Meyers-Schultke and Hedges) [46, 47]. It is also known that microbes transform labile DOM into humic-like matter during cell growth and decline in seawater [48, 49]. As a results, heterotrophic bacteria play critical roles in carbon cycles in aquatic environments and chemical composition of recalcitrant DOM in environmental water depends on the autochthonous microbes and water sources.

Thus, in the case of microbes that form humic substances that can be isolated, there is a possibility of developing a microbial sensor that can monitor the process of forming humic substances from labile DOM. Such microbial biosensor development may be required for monitoring recalcitrant DOM deposited on the bottom of the water.

Influences of DOM on the elution of iron, manganese, and nutrient salts

In the above two sections, it was found that DOM produced by phytoplankton are partially altered to recalcitrant DOM by irradiation of UV in sun light, and microbial activities transform biodegradable DOM to recalcitrant DOM. In addition, also recalcitrant DOM such as humic substances flows from the outside. These recalcitrant DOM gradually accumulate onto the bottom of water and consume oxygen while accumulating with soil and sand, and lose solubility to water in sediments. Finally, sediments will undergo geothermal and pressure in the ground and over time they will turn into shale by diagenesis, and then, kerogen will be formed in the shale.

DOM accumulates through the sewage treatment water in the water source of the region where the population is dense, causing an increase in the COD value [18]. In addition, the accumulation of recalcitrant DOM onto the water bottom induces lack of oxygen due to oxidation of recalcitrant DOM and changes the bottom layer environment, as a result often has a large adverse effect on water quality depending on the water environment. Kristensen et al. investigated the influences of wind-induced resuspension occurring in a shallow eutrophic lake and found phosphorus release by resuspension of the sediment [50]. Nakazono et al. investigated the characteristics between dissolved oxygen (DO) condition and nutrient salts, iron, and manganese released from sediment during resuspension, which often occurs by winding up in a shallow lake like Lake Kasumigaura [51]. They found that the concentration of ammonium nitrogen (NH_4-N) and iron (Fe) increased after the resuspension of the sediment, whereas the release of manganese and phosphates was suppressed under the aerobic condition. Shimotori et al recently established a new method for sediment oxygen demand (SOD) measurement in lakes, and found that significant correlations between the amount of iron and phosphorus eluted to the overlying water and the SOD values [52]. In addition, Tada et al. described *M. aeruginosa* growth influences on manganese release from lake sediment [53]. In the large and deep Lake Biwa (surface area 675 km^2, maximum depth 104 m), Thottathil et al. recently investigated DOM behavior comprehensively [54]. The distribution and biodegradability of DOM in depth and season were measured relating to temperature, N and P concentrations, and chlorophyll-a distribution. They estimated the biodegradability of DOM in the hypolimnion and found that ~8% of the organic carbon degraded by hypolimnetic mineralization is transformed into humic substances.

As described above, heterotrophic microbes in the shallow eutrophic lake have high activity to aerobically decompose DOM, which induces anaerobic conditions at water bottom. By wind-induced resuspension, aerobic and anaerobic conditions are mixed and then, nutrient salts for cyanobacteria or algae such as iron, manganese, nitrogen, and phosphorus are released by elution from the sediments. In Japan, most of designated lakes are shallow (average 3.8 ± 2.5 m in seven lakes and maximum 8.1 ± 4.7 m in eight lakes) except for Lake Biwa and Lake Nojiri. Therefore, such elution of the nutrient salts produces water bloom caused by cyanobacteria or algae, and ultimately organic pollution occurs because of the large amount of algal DOM produced.

Influences of DOM on the cyanobacterial bloom

In a hypertrophic lake, water bloom occurs by cyanobacteria or algae. In the case of Japan, the dominant species causing water bloom is *M. aeruginosa* in general [55], although the dominant species is *Planktothrix agardhii* in Europe [56, 57]. In Lake Kasumigaura, *Microcystis* blooms have been observed every summer until 1986. However, they disappeared in 1987, and thereafter *Planktothrix* spp. were instead dominant for a long time, although eutrophic conditions continued [33]. Thus, Imai et al. investigated the reasons, and initially found that the limited conditions on iron were induced by complexation between iron and fulvic acid, which is main component of AHS (one of the fractionated DOM) in the Lake Kasumigaura [58]. The complex has higher binding force than the complex of siderophore produced by *M. aeruginosa*, and this result suggested the possibility that *M. aeruginosa* have higher auxotrophy for iron than *P. agardhii*, and therefore *P. agardhii* could be dominant species in Lake Kasumigaura. Subsequently, they also found that iron requirement or iron availability between *M. aeruginosa* and *P. agardhii* are different [59]. Further, it was also found that the complexation between AHS and iron might be influenced in both *M. aeruginosa* and *P. agardhii* growths [60]. As described above, the investigation is underway on the causes of cyanobacterial blooms. However, this might show that in a lake where the environs are surrounded by mountainous areas and the AHS component flows in a lot it may be harder to generate *Microcystis* blooms than a lake adjacent to an urban area and susceptible to artificial influences.

Influences of water bloom on the water quality

Wherever conditions of temperature, light, and nutrient status are conducive, surface waters may host increased growth of algae or cyanobacteria [61–63]. Thus, water bloom is caused by such conditions in both fresh water and the ocean, although it is hard to occur in Japanese rivers because of the short residence time. Where proliferation is dominated by a single (or a few) species, the phenomenon is referred to as an algal or cyanobacterial bloom. Problems associated with cyanobacteria are likely to increase in areas experiencing population growth with a lack of concomitant sewage treatment, and in regions with agricultural practices causing nutrient loss to water bodies through over-fertilization and erosion.

Cyanobacteria are a source of considerable annoyance in many situations [61]. The abundant growth of cyanobacteria in environmental water causes many water quality problems such as organic pollution, oxygen deficiency, elution of heavy metals and nutrient salts, and unpleasant smell. The emergence of strains containing toxins causes serious problems in water supply and the ecosystem. Therefore, cyanobacterial toxin, or "cyanotoxin", is of concern for both ecological and human health [64].

Other circumstances of Japan related to organic pollution

At one time in Japan, the water for washing rice was soiling the river water. Its components are rich in organic matters; 2,582 mg O_2 L^{-1} BOD, 1,181 mg O_2 L^{-1} COD, 1,955 mg L^{-1} SS, 84 mg L^{-1} TN, and 84 mg L^{-1} [65]. Currently, wash-free rice has become common, and has became one of major factors to lower the BOD value of river water. Also, with regard to sewage treatment effluents flowing into rivers and the like, study by DOM emission reduction and study on the odor of treated effluent are actively promoted by Urase et al. [66–68].

As Japan is a group of islands with many mountainous areas, the residence time of the river is shorter than that of continental. Therefore, rivers themselves are less susceptible to eutrophication and organic pollution. However, river water mixed with domestic wastewater, flood water from agricultural land, and groundwater flows into closed waters such as lakes and bays, causing eutrophication and organic pollution by nutrient salts and DOM. As an example, the Tama River is introduced as follows: this river reaches Tokyo Bay and during the high economic growth period was a death river where detergent bubbles give off a bad smell [69]. However, due to the establishment of sewage treatment facilities, we are maintaining a rich ecosystem now. In recent years, the population of sweetfish, *Plecoglossus altivelis*, called Ayu, which is known as fragrant fish, and its use as food is increasing (estimated 1.58 million Ayu in 2017) [70]. Ayu is amphidromous, native to East Asia, and feeds on adherent algae such as cyanobacteria and diatoms, and it can be said that abundant nutrient salts contained in sewage treated water maintain the population of Ayu.

The seaside environment in Japan also has unique circumstances that cause organic pollution. First of all, because Japan is a group of islands, there are many complex geographical features along the coastline, and as a result there are many closed bays. In such highly bayed terrains, although they are not as high as the designated lakes, nutrient salt and DOM flowing in from the land are easy to accumulate, so eutrophication and organic pollution are likely to continue in a state of simultaneous occurrence. In addition, because dam construction aimed at flood control and electrical power generation was actively carried out in Japan, the supply of soil and sand to the sea has drastically decreased. Resulting landfill in coastal areas has progressed, especially in urban areas where the population is dense, and thus many tidal lands and sand beaches were lost. Therefore, nutrients from the land and DOM flow into the closed sea area without being fixed to the aquatic organisms of the tidal lands, so eutrophication and organic pollution are more likely to occur. The result

continues to affect the surrounding water environment as red tide and blue tide as mentioned above. By artificial modification of these watersides, food poisoning of shellfish caused by red tide occurred occasionally in Japan. As one of solutions, the artificial tidal lands are also expanding in such bays [71].

The microbial biosensor has the ability to comprehensively measure various conditions in the environment [2, 72–74]. Such capability is not found in chemical or physicochemical analytical method. In facts, as I presented in this part of the review series, actual water environments require not only chemical or physicochemical analytical data, but also bioavailability data in ecological and biological aspects. In order to meet such demands, it is necessary to develop the biosensors.

Conclusion

I have written several review articles and a part of encyclopedia on microbial biosensors up to now [72–74]. During the term, I was concerned about the gaps between the direction of the microbial biosensor developments and the actual water environments, especially in the closed water bodies. Thus, following the previous review [2], in this part of the present review series, I introduced the current status of water environment and recent studies on water quality investigations in Japan. It was found that there is the necessity to estimate DOM behaviors, especially in the closed water bodies, to control organic pollution. For this purpose, biomonitoring using biological tools is suitable to evaluate ecological status in such water environments. Therefore, the microbial biosensor techniques must be applied to water quality control in the future. Such situation in Japan would be similarly occurring in other countries, especially in advanced countries, because our lifestyles are becoming common around the world. On the basis of these facts and for subsequent part [75], I presented the possibility of the microbial biosensor techniques to the current status of water environment.

Compliance with Ethical Standards

Conflict of interest The author declares that he has no competing interests.

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