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Original Paper

Water quality improvement effect from the installation of special-glaze-applied ceramics: Benten Pond, Ichikawa, Chiba, Japan

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Abstract

Objective**:** The special-glaze-applied ceramic pieces used in this study are bisque fired ceramics manufactured by applying a special glaze. It has been suggested that cyanobacteria, which are oxygen-producing photosynthetic bacteria, may proliferate when ceramic pieces are placed in soil or water. In this study, we installed ceramic pieces in the Benten Pond (Ichikawa, Chiba, Japan) and verified the effect of the pieces on water quality.

Method**:** Benten Pond is a closed water system with a little flow from upstream where a water inlet pipe is located, to downstream where a drainage channel is located. We placed three sandbags filled with approximately 6 kg of ceramic pieces in the water. We observed reddish mud and oil film around the water inlet pipe that were determined to be iron oxide biomats produced by iron bacteria, and ceramic pieces of 1 kg were scattered only on the west side of the pipe.

Result**:** The values of Biochemical Oxygen Demand (BOD)/Chemical Oxygen Demand (COD), which are indicators of water quality, were 1.8/4.2 mg/L when the ceramic pieces were placed (November 21, 2019), the values temporarily increased during summer and decreased to 1.2/3.5 mg/L after one year. An improvement was observed in suspended solids, which dropped from 17 mg/L to 12 mg/L. The previous value of total nitrogen was 14 mg/L, which was well above the standard value. Total nitrogen decreased during the summer and then approached the previous value. Although total phosphorus increased temporarily during summer from the previous value of 0.034 mg/L, the value was 0.030 mg/L after one year. The iron oxide biomats almost disappeared in the area where the ceramic pieces were placed, and the biomats remained unchanged in the area where the pieces were not placed. Adverse events associated with the installation of ceramic pieces were not observed.

Conclusion**:** The installation of ceramic pieces improved the water quality and reduced iron oxide biomats. Further studies regarding the involvement of cyanobacteria are required.

KEY WORDS**:** Special ceramic pieces, water quality improvement, cyanobacteria, iron oxide biomats, iron bacteria

Introduction

The special-glaze-applied ceramic pieces (referred to as "ceramic pieces") introduced in this paper were developed in 2000 by Shuichi Sugita (Noah Co., Ltd., Oita, Japan). Water treated with the ceramic pieces has been confirmed to have odor reduction and bactericidal effects [1]. Although the mechanism for the effects was unknown, studies have suggested that this could be due to the proliferation of cyanobacteria. Cyanobacteria are oxygen-producing photosynthetic bacteria and have existed for more than 2 billion years on Earth. A simple explanation for the improvement effects is that the dissolved oxygen concentration increases due to the oxygen produced. The resulting oxidation accounts for the odor reduction,

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bactericidal action and water purification effects. Studies to clarify the mechanism responsible for the special effects produced in the ceramic treated water will become more important. In this study, we placed ceramic pieces in Benten Pond (Ichikawa, Chiba, Japan) and verified the effect of the pieces on water quality.

Method

Materials and Methods

The ceramic pieces are bisque fired ceramics manufactured by the application of a special glaze. The glaze does not contain any harmful chemicals or chemical products. It is manufactured by mixing multiple plant embryo buds and sprouts found in Japan and fermented using special enzymes over a long period of time. The specially processed glaze (containing iron oxide) is then applied to the surface of the ceramic pieces (spherical and ellipsoid fine particles), the pieces are dried and subject to heat-treatment at a high temperature (1,300 °C for 1 hour)[1]. The ceramic pieces are spherical in shape, approximately 2.5 cm in diameter and weigh about 15 g each.

Three sandbags (48 cm x 62 cm, manufactured from polyethylene) filled with approximately 6 kg of ceramic pieces were prepared. The ceramic pieces were purchased from Noah Co., Ltd. The pieces were installed in Benten Pond located at Benten Pond Park, Ichikawa, Chiba, Japan. ***Figure 1*** shows the installation locations (three places). The area around Benten Pond is now a residential locality. There are no agricultural fields in the neighboring area. One water inlet pipe is present on the north side of the pond, which receives rainwater after rainfall, but the flow rate during clear weather is low and is less than 4 L/min. A part of the water inlet pipe goes through the soil. There is no contamination by domestic wastewater. There is an outflow channel to the south, and the flow rate here is also low.

Reddish mud and a filmy substance were observed on the water surface in the area around the water inlet pipe, which were determined to be iron oxide microbial mats (biomat)[2,3]. This is also known as "red water", "red sludge" or "red mud". However, "red mud" can sometimes refer to industrial waste, which should be differentiated.

The industrial waste "red mud" is produced when bauxite is refined into alumina (aluminum oxide) with the Bayer process[4,5]. In this process, bauxite is crushed and dissolved in sodium hydroxide to obtain sodium aluminate solution, from which aluminum hydroxide is precipitated, and aluminum is recovered. The dissolved residue is the sludge that is discharged during the process. This red mud contains mostly of the impurities in bauxite, and the red color originates from the primary component, hydrated iron oxide Fe (III). The primary component is iron oxide Fe (III) (Fe_2O_3) at about 40 %, the next highest are aluminum oxide (Al_2O_3) and silicon dioxide (SiO_2) at about 10 %, and other components include calcium oxide (CaO), titanium dioxide (TiO_2) and sodium oxide (Na_2O). The dumping of red mud into the ocean is a reality in Japan, with environmental consequences. Initiatives are being implemented to reduce the amount of waste dumped into the ocean.

Ceramic pieces (about 1 kg) were scattered on the soil (over an area of about 2 m in diameter) to the right (west) of the water inlet pipe to verify the effect of ceramic pieces on the iron oxide biomats. The water depth in this area ranged from 0 to 5 cm, and most of the ceramic pieces were almost

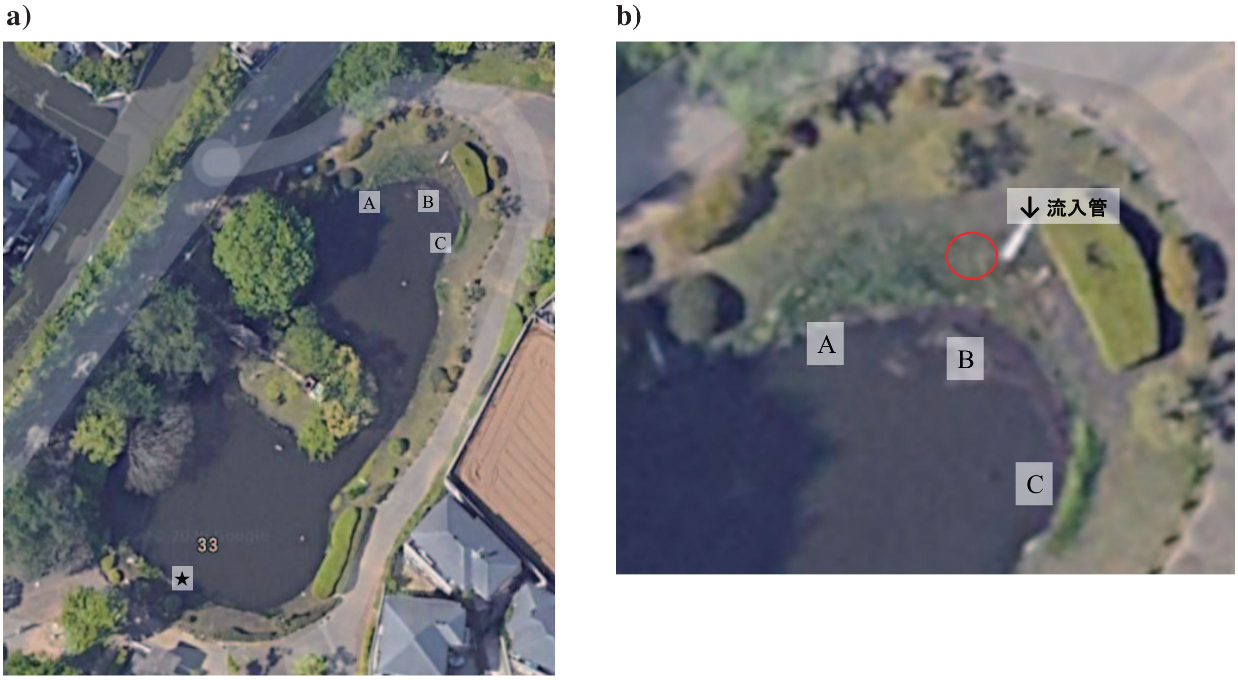

Fig. 1. **Aerial Photo of Benten Pond.**

a) Panoramic view. **b)** Magnified image Ceramic pieces are placed at locations A, B and C in the water. Sandbags (approximately 6 kg) are filled with ceramic pieces. About 1 kg of ceramic pieces were spread inside the red circle (an area of around 1 m in diameter). ★Water quality measurement site. Source: Google Aerial Photograph.

completely submerged. Also, a clay-like material (about 10 kg) made by pulverizing the ceramic pieces was spread on the periphery.

Water Quality Survey

A water quality survey was conducted at a fixed location every three months. The items measured were hydrogen ion concentration, temperature, biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids (SS), total nitrogen and total phosphorus. The water quality was tested by Chugai Technos Corporation (Hiroshima, Japan). The location for the measurement is shown in ***Fig. 1***. The measurement location is located near the outflow channel on the south side of Benten Pond.

Results

Changes in Appearance

The appearance of the water in Benten Pond is mostly transparent and odorless, and the pond is inhabited by carp, crucian carp, and Japanese pond turtles, with ducks and other migratory birds flying in and various insects living around the pond. One water inlet pipe is located to the north of the pond, and some of the rainwater after rainfall causes the water inflow volume to increase, but the average volume of water flowing into the pond is less than 5 L/min. The soil (radius of about 2 m) around the water inlet pipe (depth of water about 0 to 5 cm) had a reddish tinge, and an oil-like film was present in some areas of the water surface. These areas were also odorless. The films are iron oxide biomats formed from the proliferation of iron bacteria.

The appearance three months after the installation was the same as when the ceramic pieces were installed, a reddish tinge was observed in the soil, and a reddish film had formed on the water surface (***Fig. 2***).

The situation one year after installation is shown in ***Fig. 3***. A difference in the color tone of the soil and water surfaces in the left and right sides of the water inlet pipe was observed. The area on the left (east) was unchanged from 1 year ago: it had iron oxide biomats and a reddish tinge in the soil and film formation on the water surface was observed. On the other hand, the soil on the right (west) of the water inlet pipe where the ceramic pieces were placed was dark brown, and film formation on the water surface could not be observed. The iron oxide biomats had disappeared.

Offensive odor, abnormality or death of native organisms, pest outbreaks, red tide or blue-green algal blooms were not observed during the installation period.

Fig. 2. **Iron oxide biomat in the periphery of the inlet pipe (after three months).**

The soil in the periphery of the inlet pipe has a reddish tinge, and film-like structures are observed on the water surface. There is no significant difference in soil color on the left and right sides of the drainage pipe. Rainwater flows into the inlet pipe, and the inflow is temporary after rainfall. The left side of the pipe is untreated. Ceramic pieces are placed on the right side. The condition has not changed three months after the placement of ceramic pieces. Photographed on February 21, 2020.

a) **b)**

Fig. 3. **Iron oxide biomat in the periphery of the inlet pipe (after one year).**

a) In the periphery of the inlet pipe, the soil on the left side was reddish, while the soil on the right, where the ceramic pieces were spread, was dark brown. **b)** On the left side of the pipe, reddish mud and oil film-like structures are observed, unchanged from a year ago. Photographed on November 20, 2021.

Changes in Water Quality

Results of the water quality measurements are given in ***Table 1***. The temperature of the samples was kept constant (25～26°C) when conducting the water quality tests. Hydrogen-ion concentration was almost constant (pH 7.2), except for June when pH was 7.5.

Water quality at the start of the study was COD standard B (5.0 mg/L or less), which corresponds to Fishery Class 3 (for aquatic products such as carp and crucian carp inhabiting eutrophic lake-type waters), Industrial Water Class 1 (regular purification operation such as sedimentation is performed) and Agricultural Water, and the SS standard was C (15.0 mg/L or more), which corresponds to Industrial Water Class 2 (advanced water purification is performed by injecting chemicals or special water purification operations). The standard for total phosphorus was IV (0.05 ml/L or less), which corresponds to Fishery Class 2, while the value of total nitrogen was outside the standard (1 mg/L or more).

SOD was 1.8 mg/L when the ceramic pieces were placed (November 21) and increased on May 12 (2.0 mg/L) and August 17 (3.1 mg/L) when the temperature increased but was 1.2 mg/L one year later (November 20). Overall, SOD had improved from the previous year.

COD was 1.8 mg/L when the ceramic pieces were placed and increased on May 12 (4.5 mg/L) and August 17 (7.5 mg/L) when the temperature increased but was 3.5 mg/L one year later (November 20). Overall, COD had improved from the previous year.

SS improved to 12 mg/L after one year compared to 17 mg/L when the ceramic pieces were placed. The previous value of total nitrogen was 14 mg/L, which was significantly above the standard value. Total nitrogen decreased during summer and then approached the previous value. There were no findings of worsening conditions.

Total phosphorus was 0.034 mg/L when the ceramic pieces were placed and increased temporarily only on August 17 (0.057 mg/L), and the value was 0.030 mg/L after one year.

One year after the special ceramic pieces had been installed, COD standard was B (5.0 mg/L or less), which corresponds to Fishery Class 3, Industrial Water Class 1 and Agricultural Water, and SS standard was C (15 mg/L or more), which corresponds to Industrial Water Class 2, and there was no change in the category. The standard for total phosphorus improved to category III (0.03 ml/L or less) corresponding to Water Supply Class 3 (Water treated by advanced cleaning operations including pretreatment ("special type" means water treatment by special cleaning operation that can remove substances with a smell.)). The value of total nitrogen was outside the standard (1 mg/L or more) and remained unchanged.

Discussion

We installed ceramic pieces for this study in Benten Pond, a closed water system, and changes in water quality were examined over a year. The pond is small in size, and specific environmental water quality standards have not been established. Compared with the environmental quality standards for lakes (reservoir capacity of 10 million m^3 or more), the water quality of Benten Pond was characterized by the total nitrogen amount, which significantly exceeding the standard value. An improvement in BOD and COD was observed one year after the special ceramic pieces had been installed. The iron oxide biomats formed due to the iron bacteria found in the periphery of the inlet pipe were almost completely eliminated in the area where the ceramic pieces were installed.

The COD standard at the start was B (5.0 mg/L or less), which corresponds to Fishery Class 3, Industrial Water Class 1 and Agricultural Water, and the SS standard was C (15.0 mg/L or more), which corresponds to Industrial Water Class 2. The standard for total phosphorus was IV (0.05 mg/L or less), which corresponds to Fishery Class 2. The value of total nitrogen was outside the standard (1 mg/L or more).

One year after the ceramic pieces had been placed, COD standard was B (5.0 mg/L or less), which corresponds to Fishery Class 3, Industrial Water Class 1 and Agricultural Water, and SS standard was C (15 mg/L or more), which corresponds to Industrial Water Class 2, and there was no change in the category. The standard for total phosphorus

Table 1. Results of water quality test.

Date (Time course)			Nov 21, 2019 (0)	Feb 21, 2020 (3 months)	May 12, 2020 (6 months)	Aug 17, 2020 (9 months)	Nov 20, 2020 (1 year)
	Measurement limit	Unit					
pH			7.2	7.2	7.5	7.2	7.2
Temperature		°C	25	26	25	25	26
Biochemical oxygen demand (BOD)	0.5	mg/L	1.8	0.6	2.0	3.1	1.2
Chemical oxygen demand (COD)	0.5	mg/L	4.2	2.0	4.5	7.5	3.5
Suspended solids (SS)	1	mg/L	17	4	10	10	12
Total nitrogen	0.05	mg/L	14	14	12	11	13
Total phosphorus	0.003	mg/L	0.034	0.013	0.028	0.057	0.030

improved to category III (0.03 ml/L or less) corresponding to Water Supply Class 3. The value of total nitrogen was outside the standard (1 mg/L or more) and remained unchanged.

From the results obtained, it was determined that the placement of ceramic pieces improved the water quality of Benten Pond.

Inferred Action Mechanism of the Special Ceramic Pieces

Cyanobacteria breeding on the surface are presumed to play an important role in the action mechanism of the ceramic pieces [1]. Cyanobacteria can produce oxygen through fixation of nitrogen and carbon, which are present in water and soil, using the reaction of photo-amplifying enzymes. This reaction is significantly different from photosynthesis in common plants that use CO_2 and chlorophyll, and cyanobacteria can produce oxygen even under no-light or low light environments [6]. The nitrogen that is taken up has cyanophycin granules, which are non-ribosomal peptides made of arginine and aspartic acid, and is used as a nitrogen reserve [7]. Part of it is used as aminolevulinic acid (ALA) [8]. ALA is a basic substance that is essential for developing many life forms and is found in mitochondria and chloroplasts. It is a source of hemoglobin in animals and chlorophyll in plants.

While the mechanisms of cyanobacteria proliferation caused by the special ceramics are still under investigation, the release of trace elements may play a role. Certain trace elements, while toxic in high concentrations, are nevertheless necessary for metabolism. Safe amounts of metals contained in the special ceramics may be available for utilization by cyanobacteria and assist in their proliferation. Cyanobacteria require of a variety of metals as cofactors for electron transfer and other uses [9, 10]: metals utilized by cyanobacteria include iron, zinc, nickel, copper, cobalt, manganese and molybdenum. While they are present in small amounts, the special ceramics contain Fe_2O_3, ZnO, CuO and MnO in addition to their primary components [1].

BOD and COD values in Benten Pond improved, and the contribution of oxygen produced by cyanobacteria is considered to be the reason. The qualitative improvements in water quality and soil and the decrease in SS are presumed to be a result of these actions. Oxygen in water acts as a bactericidal agent against anaerobic bacteria and promotes the growth of aerobic bacteria. As a result, the ceramic pieces are presumed to have contributed to improving the water quality environment.

Potential of Cyanobacteria

Cyanobacteria breed extensively in the oceans, fresh water and soil. The taxonomy and scientific names of cyanobacteria have not been yet been definitively organized. Two genera of cyanobacteria, *Synechocystis* [11, 12] and *Cyanothece* [13], are major targets for study and contain important model species. Cyanobacteria is the only taxon of bacteria that performs oxygen-producing photosynthesis. They are prokaryotic organisms that do not have a nucleus enclosing the DNA inside the cells and are categorized as eubacteria. Cyanobacteria are also said to be the ancestor of chloroplasts that are present in higher plants. They can adapt to environments requiring salt tolerance [14] or a wide diurnal variation [15], and convert atmospheric CO_2 into sugar and cellulose.

The mechanisms of oxygen-producing photosynthesis in cyanobacteria are extremely diverse. Cyanobacteria use several types of chlorophyll and rhodopsin for various purposes and have shown a high degree of adaptability to breed even in harsh natural environments. As a result of their photosynthetic metabolism, they require more metals compared to other bacteria [16] and produce high amounts of reactive oxygen species, necessitating strong DNA repair mechanisms [17]. Although there are many unanswered questions about cyanobacteria, they have a unique hidden potential. In the environmental field, oxygen photosynthetic bacteria have been used for manure treatment and anti-odor measures at pig farms and wastewater treatment at food processing plants [18]. ALA is synthesized in the cyanobacteria cells. It is reported that the use of photosynthetic bacteria as a fertilizer in agriculture increased the yield and improved quality [19]. On the other hand, some species of cyanobacteria can cause red tide and blue-green algal blooms [20, 21]. There have been no cases of red tide or blue-green algal blooms in areas where ceramic pieces have been used.

Water Quality of Benten Pond

The total nitrogen concentration in water is set as an important factor for the conservation of the living environment. An environmental standard of 0.1 to 1.0 mg/L has been established for lakes and seas by type. The water quality of Benten Pond was characterized by a high total nitrogen concentration. There are no farmlands, orchards or pig farms around the pond at present. The area has an established sewage system and mixing of domestic wastewater is unlikely. A pear orchard was once present in this area, and the soil may still be affected by fertilizers used in the past. The possibility of nitrogen present in the soil having permeated underground and flowed into the pond cannot be denied.

Total nitrogen includes nitrate-nitrogen, nitrite-nitrogen, ammonia nitrogen and organic nitrogen. Daily life and agricultural activities often cause nitrate and nitrite nitrogen contamination. Untreated domestic wastewater can cause organic contamination and eutrophication. The degree of contamination varies depending on the treatment method used for domestic sewage, farming style, soil environment and weather conditions in the area. The area around Benten Pond is a residential locality, and there are no farmlands around the pond at present. Ammonia nitrogen is derived from ammonia formed by the hydrolysis of proteins discharged from kitchens and urea discharged from domestic wastewater. Judging from the residential environment around Benten Pond, with an established sewage system, a large amount of ammonia nitrogen is unlikely to be produced nor enter the pondwater. Although microorganisms decompose the nitrogen compounds, they also consume oxygen and cause BOD to increase.

Next, we will compare the water quality of Kasumigaura (Ibaraki, Japan) and Benten Pond. Kasumigaura (area 220.0 km^2) is the second largest lake in Japan after Lake Biwa (Shiga, Japan), and information on its water quality is readily available [22]. Since the 1970s, the Ministry of Land, Infrastructure, Transport and Tourism has been implementing water purification measures to counter large outbreaks of blue-green algae, offensive smells, and mass mortality of freshwater clams and cultivated carp, mainly in summer.

The COD (average of all water bodies) value in Kasumigaura reached a record high of 11.0 mg/L in 1979 but has decreased since 2010, reaching 6.9 mg/L in 2019. Since organic matter is produced by phytoplankton through photosynthesis in lakes, COD is higher than the inflowing rivers. Seasonal variation has also been observed, with COD increasing in summer and decreasing in winter. The variation is associated with the growth of phytoplankton.

Total nitrogen varies depending on the inflowing river. A higher value (> 6.0 mg/L) is observed in the inflow of the Kitaura river compared to the inflow of the Nishiura river (2.7 to 3.1 mg/L, ***Fig. 4***). The reason is considered to be the nitrogen content of fertilizers (compost) used in farmlands flowing into the rivers through groundwater over time. The reason for a lower total nitrogen concentration in the lake (1.1 mg/L in 2019) than in the inflowing rivers is because when nitrate ions, one of the components of nitrogen present in river water, enters the lake, they are converted into gaseous nitrogen by the action of denitrifying bacteria and released into the atmosphere (denitrification). Although nitrogen at the lake center shows little seasonal variation, a decrease in the summer and an increase in the winter are observed in some water bodies. The reason is that dissolved nitrogen increases when the water temperature is low.

The total phosphorus concentration indicates an upward trend until 2008, but has declined to 0.094 mg/L in 2019. In recent years, phosphorus concentration in the lake tends to be higher than that of the inflowing rivers. The reason is believed to be due to the seepage (elution) of phosphorus from the mud (bottom mud) that accumulates at the lake's bottom. Seasonal variation of total phosphorus indicates that it tends to increase in summer and decrease in winter. The elution of phosphorus from the bottom mud is believed to occur when the dissolved oxygen concentration at the lake's bottom is low (becomes anaerobic).

Benten Pond has lower COD, BOD, SS, and total phosphorus than Kasumigaura. It can be determined that the "water quality is not poor" for the pond except for total nitrogen. The seasonal variation of COD, total nitrogen and total phosphorus in Benten Pond is similar to that of Kasumigaura.

Iron Oxide Biomats

Iron bacteria are involved in the production of iron oxide biomats. Iron bacteria are widely present in soil and thrive in springs and soils that contain high levels of divalent iron (Fe (II)), as they use energy from the oxidation of water-soluble divalent iron ions (Fe^{2+}). Iron bacteria oxidize Fe (II) dissolved in water to form trivalent iron (Fe (III)) as a water-insoluble hydroxide or oxide, which is deposited and collected inside and outside the bacterial cells. A film of iron oxide is formed from Fe (III), and when the bacteria die, reddish-brown sediment is created and a biomat (microbial film) is deposited. The iron oxide film resembles an oil film in areas where there is no flow of water [23-25]. Iron bacteria include genera *Leptothrix* [26,27] and *Gallionella* [28,29].

Iron oxide biomats have also been observed near water inlets of paddy fields and leakage sites in concrete structures. Although the appearance of the landscape is spoiled, both bacteria and sediment are harmless at levels existing in nature. Treatments such as aeration and coagulative sedimentation are implemented to improve the visual appeal of the landscape [30]. Since these are methods used to remove dissolved iron by turning it into a solid as iron oxide, the solid must be regularly removed and disposed of as waste. Therefore, these are not simple methods that are suitable for use in springs, ponds, swamps and rivers.

In a typical river, most dissolved iron forms complexes with humic substances (HS) and flows downstream [31]. In addition to HS, other organic ligands that form complexes

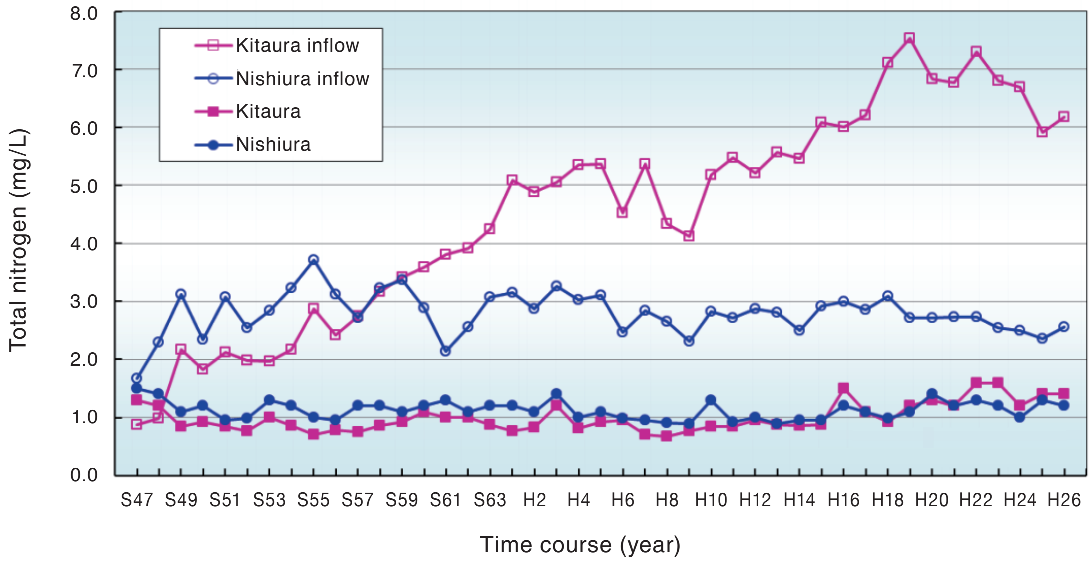

Fig. 4. **Water quality in Kasumigaura (Ibaraki Prefecture): Total nitrogen.**

Total nitrogen concentration in the Kitaura inflow river is exceptionally high compared to the lake water. Cited from reference 10).

with Fe (II) and Fe (III) in the water environment include siderophores[32, 33] produced by microorganisms such as bacteria and eumycetes, and exopolymeric substances (EPS) such as polysaccharides produced by bacteria and algae[34, 35]. Since soluble Fe (III) complexes are created if these substances are abundant, iron oxide biomats are less likely to form.

What is the effect of cyanobacteria proliferation, which performs oxygen-producing photosynthesis, in an environment with iron oxide biomats?

Iron oxidizing bacteria adapted to neutral pH are microaerophilic chemotrophs. A small minimum concentration of oxygen is necessary for oxidizing Fe (II) into Fe (III). However, under aerobic conditions at a neutral pH, abiotic oxidation of iron occurs spontaneously and outcompetes bacterial iron oxidation, which is a relatively slow process. Thus, a mostly deoxygenated environment is required for iron oxidizing bacteria to proliferate[36]. In natural environments they tend to exist at the boundary between aerobic and anaerobic environments[37]. As Benten Pond maintains a neutral pH while iron bacteria are able to proliferate, the oxygen concentration is likely low.

When the amount of dissolved oxygen is high, Fe (II) quickly oxidizes to Fe (III), and Fe (II) utilization by iron bacteria is expected to reduce. As a result, the growth of iron bacteria is similarly reduced. Many cyanobacteria are reported to take in inorganic dissolved iron Fe^{2+} and Fe^{3+} from water into the cells through transport proteins in the cell membrane[38]. Iron is vital to cyanobacteria metabolism, and plays an important role in the processes of photosynthesis and nitrogen fixation[39, 40]. The presence of cyanobacteria will provide additional competition for iron as well as increase oxygenation, further suppressing iron bacteria. In a study of freshwater Lake Taihu in China[41], areas of the lake that were dominated by cyanobacteria had significantly lower abundance of iron bacteria compared to typical environments. Dissolved iron was also decreased in these areas compared to parts of the lake dominated by aquatic plants. Under aerobic conditions, in the presence of a large quantity of dissolved oxygen, bacteria and algae increase, as do corrosive substances and soluble complexes.

It also affects iron oxide Fe (III), which is already sedimented. Some of the iron oxides deposited at the bottom of the water body by coagulation and sedimentation are reduced to Fe (II) in the anaerobic reduction layer and eluted as Fe^{2+} ions[42, 43]. Fe^{2+} released from the deposit directly into the water is oxidized relatively quickly, but it forms complexes such as organic complex Fe (III) on encountering organic macromolecule polymers (such as humic acid and siderophore) and dissolves. The iron supplied directly from the bottom of the water body and dissolved as organic complex iron forms an iron circulation system in water. These iron types are considered to be widely used by organisms other than iron bacteria. Microorganisms particularly tend to use the siderophore complex Fe (III).

Safety

The amount of trace elements in ceramic and ceramic-treated water is below the toxicity threshold level and is non-toxic. The ceramic pieces have been used for more than 25 years, and no side effects or hazards have been observed to date. The safety of the material is sufficiently guaranteed. Since the amount of each element produced by ceramic treatment is maintained at the optimum mixing ratio, ceramic treatment is assumed to have various beneficial effects such as increased physiological activity. No adverse events were observed, such as abnormal changes in the vegetation around the pond, an increase in the number of dead aquatic organisms, or the breeding of pests.

Study Limitations

There was a high concentration of total nitrogen in Benten Pond. However, since only total nitrogen was measured in this study, it was impossible to determine whether nitrate-nitrogen, nitrite-nitrogen or ammonia nitrogen was the cause. Since water from the pond was not tested for microbial composition, the presence and extent of cyanobacteria proliferation could not be verified.

Conclusion

Verification results of water quality improvement effect with the installation of ceramic pieces in Benten pond, Ichikawa, Chiba, showed an improvement in BOD, COD and a reduction in iron oxide biomat formation due to iron bacteria after one year, and no adverse events were observed. Although the underlying mechanism is unknown, it is inferred that the ceramic pieces have the effect of promoting the proliferation of oxygen-producing photosynthetic bacteria, and the phenomenon is awaiting further study.

Financial disclosure

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References

1) Hasegawa T, Sugiura S, Yonei Y. Cyanobacterium proliferative actions by special-glaze-applied ceramic pieces and their utilization. *Glycative Stress Res*. 2020; 7: 88-104.
2) Takahashi N, Segawa H, Tazaki K. The formation of microbial mats in drainpipes at landslide areas. *Journal of Japanese Association of Groundwater Hydrology*. 2007; 49: 115-137. (in Japanese)
3) Shimada T, Honda K. Observation on the reddish brown coloration by iron bacteria at biotope in the Aina Green Protection Area, Atsugi, Kanagawa, Japan. *Natural History Report of Kanagawa*. 2008; 29: 61-64. (in Japanese)
4) Takagi J, Honma K, Konno Y, et al. Concrete using industrial waste (red mud). *Report of Technical Study Group, Ministry of Construction*. 1975; 28: 89-95. (in Japanese)
5) Akashi K, Shiao S-J. Recent tendency of researches on utilization of red mud. *Journal of Japan Institute of Light Metals*. 1976; 26: 150-163. (in Japanese)
6) Gisriel C, Shen G, Kurashov V, et al. The structure of Photosystem I acclimated to far-red light illuminates an ecologically important acclimation process in photosynthesis. *Science Advances*. 2020; 6(6); eaay6415.
7) Berg H, Ziegler K, Piotukh K, et al. Biosynthesis of the cyanobacterial reserve polymer multi-L-arginyl-poly-L-aspartic acid (cyanophycin): Mechanism of the cyanophycin synthetase reaction studied with synthetic primers. *Eur J Biochem*. 2000; 267: 5561-5570.
8) Muramatsu M, Sonoike K, Hihara Y. Mechanism of downregulation of photosystem I content under high-light conditions in the cyanobacterium *Synechocystis sp*. PCC 6803. *Microbiology (Reading)*. 2009; 155: 989-996.
9) Facey JA, Apte SC, Mitrovic SM. A Review of the Effect of Trace metals on freshwater cyanobacterial growth and toxin production. *Toxins (Basel)*. 2019; 11(11): 643.
10) Bishop AB, Flynn SL, Warchola TJ, et al. Adsorption of biologically critical trace elements to the marine cyanobacterium *Synechococcus* sp. PCC 7002: Implications for marine trace metal cycling. *Chemical Geology*. 2019; 525: 28-36.
11) Ikeuchi M, Tabata S. Synechocystis sp. PCC 6803: A useful tool in the study of the genetics of cyanobacteria. *Photosynth Res*. 2001; 70: 73-83.
12) Branco Dos Santos F, Du W, Hellingwerf KJ. Synechocystis: Not just a plug-bug for CO_2, but a green E. coli [published correction appears in *Front Bioeng Biotechnol*. 2016; 4: 32]. *Front Bioeng Biotechnol*. 2014; 2: 36.
13) Welsh EA, Liberton M, Stöckel J, et al. The genome of *Cyanothece* 51142, a unicellular diazotrophic cyanobacterium important in the marine nitrogen cycle. *Proc Natl Acad Sci U S A*. 2008; 105: 15094-15099.
14) Kirsch F, Klähn S, Hagemann M. Salt-regulated accumulation of the compatible solutes sucrose and glucosylglycerol in cyanobacteria and its biotechnological potential. *Front Microbiol*. 2019; 10: 2139.
15) Saha R, Liu D, Hoynes-O'Connor A, et al. Diurnal regulation of cellular processes in the *Cyanobacterium synechocystis* sp. strain PCC 6803: Insights from transcriptomic, fluxomic, and physiological analyses. *mBio*. 2016; 7(3): e00464-16.
16) Shcolnick S, Keren N. Metal homeostasis in cyanobacteria and chloroplasts. Balancing benefits and risks to the photosynthetic apparatus. *Plant Physiol*. 2006; 141: 805-810.
17) Cassier-Chauvat C, Veaudor T, Chauvat F. Comparative genomics of DNA recombination and repair in cyanobacteria: Biotechnological implications. *Front Microbiol*. 2016; 7: 1809.
18) Kobayashi T. Environmental conservation with photosynthetic bacteria. Rural Culture Association, Tokyo, 1993. (in Japanese)
19) Sasaki Ken, Sasaki Kei, Takeno K. Application and future of photosynthetic bacteria in agriculture, environment and health. *Seibutu-Kougaku Kaishi*. 2016; 94: 146-156. (in Japanese)
20) Anderson DM, Cembella AD, Hallegraeff GM. Progress in understanding harmful algal blooms: Paradigm shifts and new technologies for research, monitoring, and management. *Ann Rev Mar Sci*. 2012; 4: 143-176.
21) Backer LC, Manassaram-Baptiste D, LePrell R, et al. Cyanobacteria and algae blooms: Review of health and environmental data from the Harmful Algal Bloom-Related Illness Surveillance System (HABISS) 2007-2011. *Toxins (Basel)*. 2015; 7: 1048-1064.
22) Ibaraki Kasumigaura Eviromental Science Center. Long-term changes in water quality in Kasumigaura. *Proceeding of the 10th Memorial Anniversary Research Results of Ibaraki Kasumigaura Eviromental Science Center*. 2016. (in Japanese) https://www.pref.ibaraki.jp/soshiki/seikatsukankyo/kasumigauraesc/04_kenkyu/shoukai/10th/documents/kankyou_10th_01.pdf
23) Tashiro Y, Tazaki K. The primitive stage of microbial mats comprizing iron hydroxides. *Earth Science (Chikyu Kagaku)*. 1999; 53: 29-37. (in Japanese)
24) Tazaki K, Asada R, Ikeda Y. Quick occurrence of Fe-rich biofilms on the water surface. *Journal of the Clay Science Society of Japan*. 2002; 42: 21-36. (in Japanese)
25) Maruyama K, Ando T, Iida M. Studies on clogged drainage pipes of groundwater drainage works in landslide areas. *Journal of Japan Landslide Society*. 2003; 39: 23-29. (in Japanese)
26) Satou K, Tazaki K. Accumulation of biogenic iron hydroxides by *Leptothrix ochracea* at neutral pH. *Environmental Conservation Engineering*. 2004; 33: 467-475. (in Japanese)
27) Suzuki T, Hashimoto H, Ishihara H, et al. Structural and spatial associations between Fe, O, and C in the network structure of the *Leptothrix ochracea* sheath surface. *Appl Environ Microbiol*. 2011; 77: 7873-7875.
28) Matsushita T. Studies on iron bacteria. I. Quantitative changes of the iron and organic matter in the media by the growth of *Gallionella ferruginea*. *The Journal of Hygienic Chemistry*. 1969; 15: 219-224. (in Japanese)
29) Suzuki T, Hashimoto H, Itadani A, et al. Silicon and phosphorus linkage with iron via oxygen in the amorphous matrix of *Gallionella ferruginea* stalks. *Appl Environ Microbiol*. 2012; 78: 236-241.
30) Koga H. Treatment Underground water (red water) from the abandoned coal mine and effective use of sediment. *Journal of the Mining and Metallurgical Institute of Japan*. 1978; 94: 641-645. (in Japanese)

31) Laglera LM, van den Berg CM. Evidence for geochemical control of iron by humic substances in seawater. *Limnol Oceanogr*. 2009; 54: 610-619.
32) Hider RC, Kong X. Chemistry and biology of siderophores. *Nat Prod Rep*. 2010; 27: 637-657.
33) Reid RT, Live DH, Faulkner DJ, et al. A siderophore from a marine bacterium with an exceptional ferric ion affinity constant. *Nature*. 1993; 366(6454): 455-458.
34) Hassler CS, Schoemann V. Bioavailability of organically bound Fe to model phytoplankton of the Southern Ocean. *Biogeosciences Discuss*. 2009; 6: 1677-1712.
35) Norman L, Worms IAM, Angles, E, et al. The role of bacterial and algal exopolymeric substances in iron chemistry. *Marine Chemistry*. 2015; 173: 148-161.
36) Rentz JA, Kraiya C, Luther GW 3rd, et al. Control of ferrous iron oxidation within circumneutral microbial iron mats by cellular activity and autocatalysis. *Environ Sci Technol*. 2007; 41: 6084-6089.
37) Erbs M, Spain J. Microbial iron metabolism in natural environments. *Microbial Diversity*. 2002; 1-19.
38) Morrissey J, Bowler C. Iron utilization in marine cyanobacteria and eukaryotic algae. *Frontiers in Microbiology*. 2012; 3: 43.
39) González A, Sevilla E, Bes MT, et al. Pivotal role of iron in the regulation of cyanobacterial electron transport. *Adv Microb Physiol*. 2016; 68: 169-217.
40) Jiang HB, Lou WJ, Ke WT, et al. New insights into iron acquisition by cyanobacteria: An essential role for ExbB-ExbD complex in inorganic iron uptake. *ISME J*. 2015; 9: 297-309.
41) Fan X, Ding S, Gong M, et al. Different influences of bacterial communities on Fe (III) reduction and phosphorus availability in sediments of the cyanobacteria- and macrophyte-dominated zones. *Front Microbiol*. 2018; 9: 2636.
42) Middelburg JJ, Levin LA. Coastal hypoxia and sediment biogeochemistry. *Biogeosciences*. 2009; 6: 1273-1293.
43) Thouvenot-Korppoo M, Lukkari K, Järvelä J, et al. Phosphorus release and sediment geochemistry in a low-salinity water bay of the Gulf of Finland. *Boreal Environmental Research*. 2015; 17: 237-252.