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Original article

Cyanobacterium proliferative actions by special-glaze-applied ceramic pieces and their utilization.

Takeo Hasegawa [1], Shinichi Sugiura [2], Yoshikazu Yonei [3]

1) Louis Pasteur Center for Medical Research, Kyoto, Japan
2) Faculty of Pharmaceutical Sciences, Doshisha Women's College of Liberal Arts, Kyoto, Japan
3) Anti-Aging Medical Research Center and Glycative Stress Research Center,
Graduate School of Life and Medical Sciences, Doshisha University, Kyoto, Japan

Abstract

This study reports outcomes of investigations of special-glaze-applied ceramic pieces and water which was treated with the ceramic pieces. The ceramic pieces were developed in 2000 and used for the purpose of environmental improvements. They were pieces of bisque fired ceramics manufactured with the application of a special glaze and were characterized by a porous structure. Investigations have confirmed diverse effects from the use of ceramic pieces such as water purification, bactericidal action and soil amelioration, although the mechanisms have not been identified. However, it has been clarified currently that cyanobacteria are proliferated in the ceramic periphery. Cyanobacteria produce oxygen via the fixation of nitrogen and carbon, which exist in water and soil. Cases with the use of ceramic pieces and the treated water indicated the effectiveness of odor decomposition, bactericidal activities, and the water quality improvement due to the reduction of biochemical oxygen demand (SOD). A theory could explain the mechanisms of these effects; that cyanobacteria of the ceramic enhances oxidation due to its oxygen production and induces these effects. Improvements of carbohydrate metabolism due to ceramic pieces were confirmed in C57BLKS/J Iar -+Lepr^{db} /+Lepr^{db}, type 2 diabetes animal models , with the ingestion of ceramic-treated water. Mechanisms for this finding have not been identified. Soil improvement effects were observed for the salt pollution caused by the tsunami of the Great East Japan earthquake in 2011 and the radioactive contamination following the Fukushima Nuclear Power Station accident. The ceramic pieces have diverse effects and further influences can exert global-scale significance for environmental improvements and increased agricultural productivities.

KEY WORDS: cyanobacteria, aminolevulinic acid, special-glaze-applied ceramic pieces, radioactive contamination, the Great East Japan Earthquake

Introduction

This research reports on special-glaze-applied ceramic pieces (referred to as ceramic pieces), which were developed in 2000 by Shuichi Sugita (Noah Co., Ltd., Oita, Japan). Case studies carried out on the use of ceramic pieces suggested that they had positive effects on water purification, bactericidal action and soil amelioration. Although the mechanisms underlying these activities are not known, recent analyses have suggested that the porous structure of the ceramic pieces might support the proliferation of cyanobacteria.

Cyanobacteria have been producing oxygen since the early days of Earth for more than two billion years and it is assumed that much of atmospheric oxygen can be attributed to the activities of cyanobacteria [1,2]. Cyanobacteria performs oxygenic photosynthesis, having both photosystem I (PS I) and photosystem II (PS II). In oxygenic photosynthesis, electron transports of the two systems of PS I and PS II are produced via cytochrome b6f complex [3,4]. PS I is a heterodimer, consisting of two proteins, which are approximately 80–85 kDa and have similar primary sequences. PS I is a complex of combined antenna pigment systems and photosynthetic reaction center systems. PS II consists of two proteins at 35–40 kDa. Oxygen concentration of the ceramic-treated water was high presumably provided by some source of oxygen production. This heightened oxygen level could contribute to

Corresponding to: Prof. Shinichi Sugiura
Faculty of Pharmaceutical Sciences, Doshisha Women's College of Liberal Arts
97-1 Minami-Hokodate Kodo, Kyotanabe, Kyoto 610-0395, Japan
TEL: +81-774-65-8627 FAX: +81-774-65-8627 Email: ssugiura@dwc.doshisha.ac.jp
Co-authors: Hasegawa T, hasegawa@louis-pasteur.or.jp;
Yonei Y, yyonei@mail.doshisha.ac.jp

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the odor reduction, bactericidal action and water purification of treated water that had been observed previously. However, further research of the ceramic pieces and the ceramic-treated water is needed to clarify the mechanisms responsible for its positive effects. The present study reports on the results of verification experiments employing the ceramic pieces and treated water.

Materials and methods

Special-glaze-applied ceramic pieces (ceramic pieces) in a variety of configurations (spherical objects, ellipsoid objects and fine particles) were coated with a special glaze manufactured from multiple specific plant embryo buds and sprouts that were fermented using special enzymes and then mixed with iron oxide. Pieces were dried and treated with heat at 1,300°C for one hour. Ingredients of ceramic pieces identified using X-ray fluorescence spectrometer analysis are shown in ***Table 1***. Most of the ceramic pieces were composed of oxides and main components were SiO_2, Fe_2O_2, Al_2O_3, CaO and K_2O. The elements that were dissolved into the water influenced the acceleration of chemical reaction (catalyst) due to dissolved minerals.

Cyanobacteria, identified by their blue-green color and unicellular, noncolonial histological profiles, were allowed to proliferate in the ceramic periphery. It is assumed that they contain natural minerals at the optimum ratio, which can induce physiological activities. It is generally confirmed that calcium and magnesium, which are essential minerals, change physiological activities even in an infinitesimal quantity. Functions of cells and tissues in humans and other living organisms are affected by content percentages of calcium and magnesium. The findings of the experimental results suggested mechanisms where ceramic pieces were soaked in water, an infinitesimal quantity of minerals were dissolved into water (a small quantity which was not detected using widely used measurement tools), proper environmental conditions were prepared for the cyanobacteria proliferation, and consequently, cell division and proliferation were promoted.

Table 1. Ingredients of special-glaze-applied ceramic pieces

Elements	X-ray spector	X-ray intensity (Kcps)	Content (%)
Na_2O	Na-KA	0.3644	0.3086
MgO	Mg-KA	0.6878	0.2197
Al_2O_3	Al-KA	34.9043	12.6799
SiO_2	Si-KA	176.3744	74.9525
P_2O_5	P-AK	0.7227	0.2232
SO_2	S-KA	2.0788	0.6499
Cl	Cl-KA	0.1593	0.0179
K_2O	K-KA	25.2004	0.8147
CaO	Ca-KA	28.6999	0.6090
TiO_2	Ti-KA	19.9868	0.8691
Cr_2O_3	Cr-KA	0.0298	0.0205
MnO	Mn-KA	0.0616	0.0216
Fe_2O_3	Fe-KA	109.5847	8.5001
CuO	Cu-KA	0.8446	0.0284
ZnO	Zn-KA	0.3205	0.0081
Br	Br-KA	0.4786	0.0049
SrO	Sr-KA	1.6997	0.0166
ZrO_2	Zr-KB1	0.7069	0.0230

Results

1) Odor decomposition

Odor was measured before and after spraying ceramic-pieces-treated water and cases which confirmed the effects of odor decomposition were as follows. Measurements were performed using an odor measurement device, Combustible Gas DetectorXP-3IIIR (New Cosmos Electric, Yodogawa-ku, Osaka, Japan).

In an excreta disposal treatment plant located in Kashiwara, Osaka, Japan, waste receptacles and disposal sites were sprayed one time with ceramic-treated water and odor level was measured ten minutes after the spray (***Fig. 1***). Smell was recognized to decrease significantly by 25% at every measurement spot ten minutes after the treated water spray. This reduction in odor lasted for several hours either when materials were washed with ceramic-treated water after usual washing, as well as spraying droplets or mist of treated water into the air. Its mechanisms are that the mist shower spray removed odor components, which were floating in the air, and that the bactericidal activities of ceramic-treated water reduced the number of harmful and putrefactive bacteria and offensive odor fermentation bacteria.

In a sewage disposal plant located in Aso, Kumamoto, Japan, three waste receptacle sites were sprayed with treated water and the odor level was measured 15 minutes after the spray (***Fig. 2***). Odor levels were lowered at site A from 200 to 62. These reduced levels compare favorably to those of ordinary households that range from approximately 60 to 100. By spraying treated water, odor levels of all sites were reduced by half. However, due to the process of wastewater injection, odor levels of the operational room returned to the level first observed when wastewater was introduced. The odor level at site A returned to approximately 170. Odor levels at site B and C indicated similar changes.

In a hog farm (Ubuyama Village, Kumamoto), three sites, the entrance, the side, and the inside sites of the swinery, were sprayed with ceramic-treated water and then, odor levels were measured (***Fig. 3***). The smell at all sites was reduced after the ceramic-treated water spray. Pig feces collection sites were not sprayed and there were no data on this site.

Another odor measurement was performed in a hog farm which was equipped with a filtration apparatus of a ceramic-pieces-filled tube (Hitoyoshi, Kumamoto), as shown in ***Fig. 4***. Compared with other hog farms located in proximity which were not equipped with a ceramic filtration apparatus, the odor level in the above hog farm was greatly reduced.

In a cattle farm (Ubuyama Village, Kumamoto), three sites, the entrance, the inside, and the entrance peripheral sites of the cattle farm were sprayed with ceramic-treated

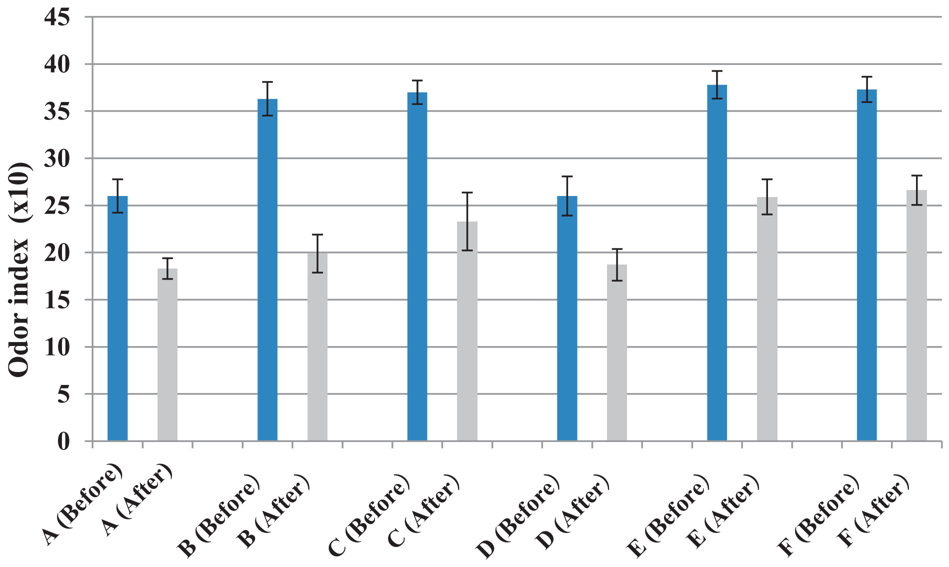

Fig. 1. **Odor decomposition of ceramic-treated water: An excreta disposal treatment plant (Kashiwara, Osaka, Japan)**

Results are shown in mean ± standard deviation. Before: before spraying. After: ten minutes after spraying. Measurement date: June the 10th, 2014 at 13:00. Method: ceramic-treated water was sprayed on the waste receptacle and disposal sites in the disposal treatment plant. Odor measurement device: XP-329IIIR. Measurement sites: Kashiwara City Aqua Center (A) Excrement receiving entrance, (B, C, D, and E) Four sites of excrement injection rooms, (F) Operational vehicle washing room.

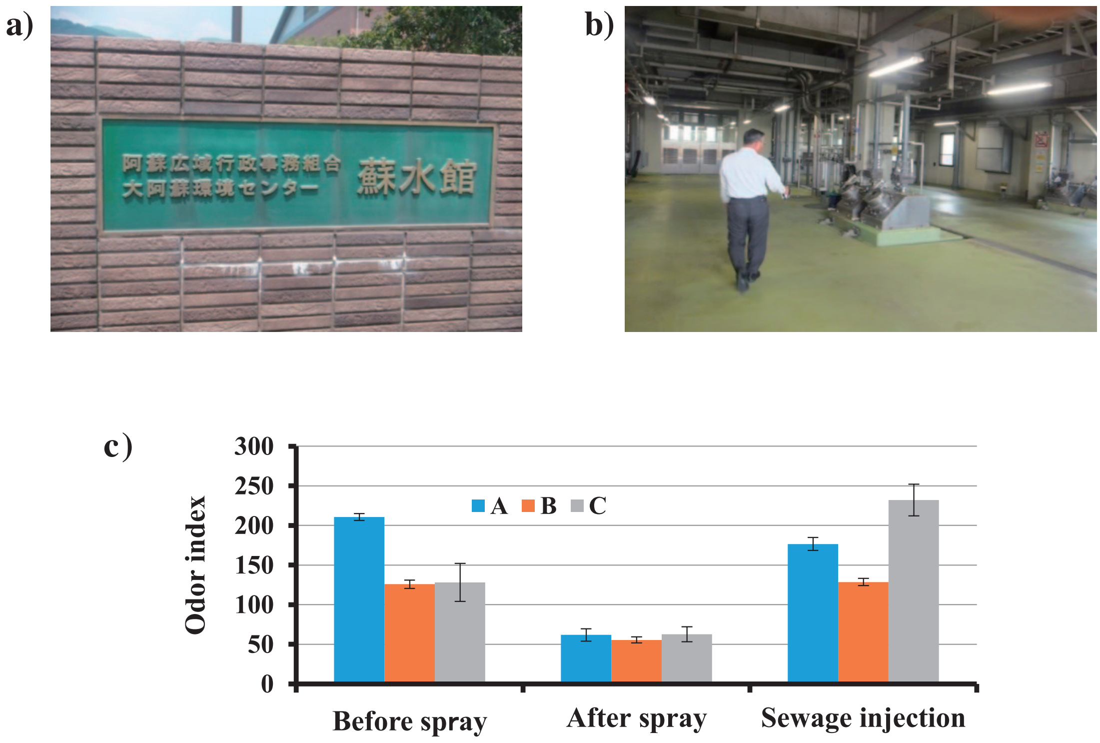

Fig. 2. **Odor decomposition of ceramic-treated water: A sewage disposal plant (Aso, Kumamoto, Japan)**

a) In front of the plant. **b)** Inside the plant. **c)** Alternation in odor levels. Results are shown in mean ± standard deviation. Ceramic-treated water spraying was performed at three waste holding sites at the sewage disposal plant, sites A, B, and C. Measurement date: August the 3rd, 2015 at 12:50. Measurement device: XP-329IIIR. Measurement site: Sosui-kan, Oaso environment center. (Aso integrated administration of a large region office work association).

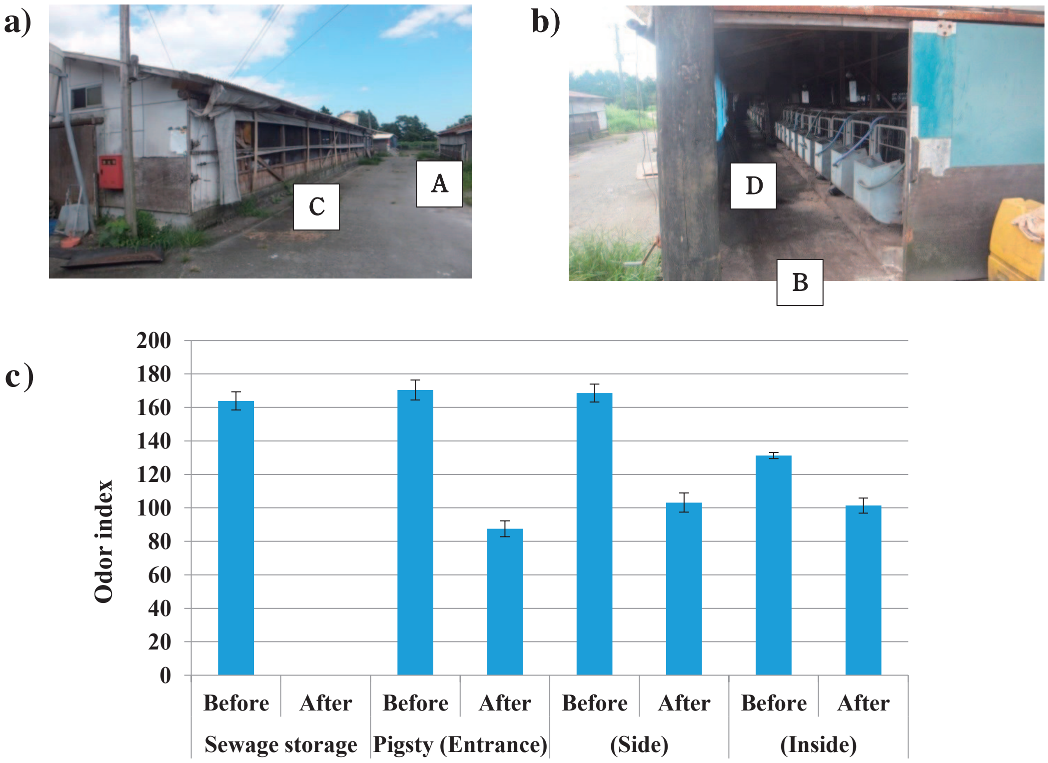

Fig. 3. **Odor decomposition of ceramic-treated water: A hog farm (Ubuyama Village, Kumamoto, Japan)**

a) A full view of the farm. **b)** The hog farm. **c)** Alternation in odor levels. Results are shown in mean ± standard deviation. Pictures show: A: cattle dung accumulation site. B: the entrance of the hog farm. C: the side of the farm. Measurement date: August the 23rd, 2015 at 11:00–16:30. Measurement site: Sakaguchi hog farm.

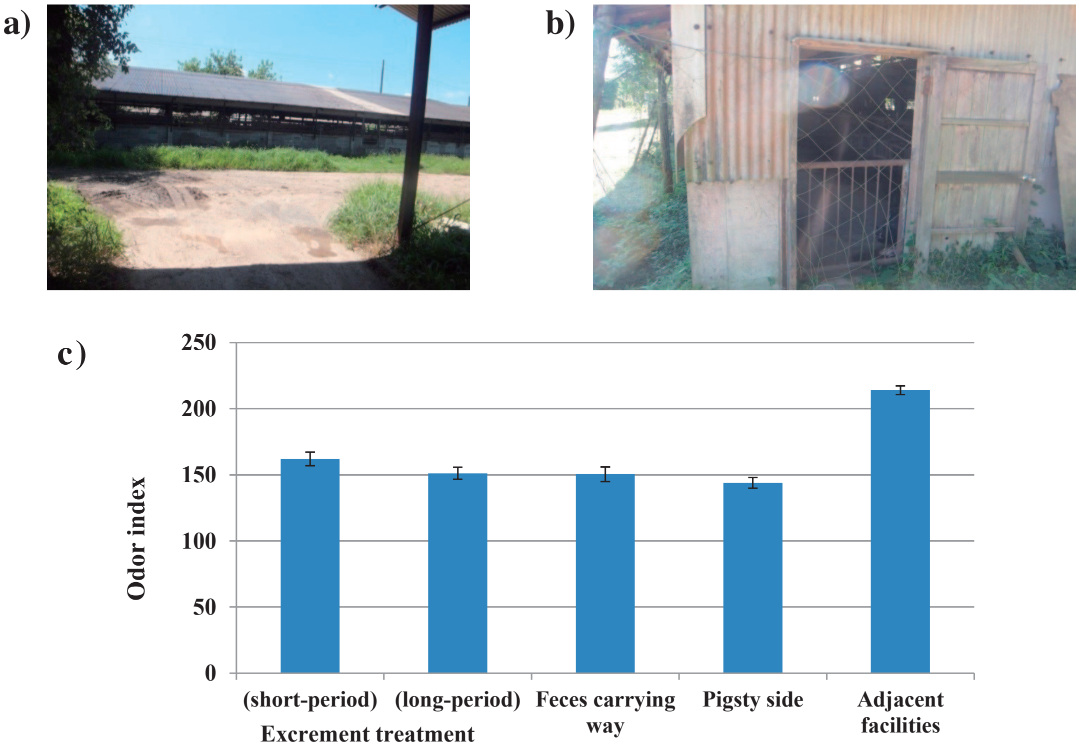

Fig. 4. **Odor decomposition of ceramic-treated water: A hog farm (Hitoyoshi, Kumamoto, Japan)**

a) A full view of the farm. **b)** The hog farm back door. **c)** Alternation in odor levels. Results are shown in mean ± standard deviation. This institute had been already equipped with a filtration apparatus of a ceramic-pieces-filled tube and indicated a significantly lower odor level. Measurement date: August the 23rd, 2015 at 11:00–16:30. Measurement device: XP-329IIIR. Measurement site: Kamata livestock hog farm.

water and odor levels were measured (*Fig.5*). The smell was reduced after the ceramic-treated water spray at every site.

An investigation was performed in an animal barn (Kyoto, Japan) for mouse bedding (*Fig. 6*). Spraying ceramic-treated water, the measurement sample was 33.7 g of mouse bedding material (the fifth day of breeding), and the amount of ceramic-treated water was 10 mL. Odor sampling and measurement were performed at 10-minute intervals. The first 10-minute measurement showed a significant effect, smell gradually reduced with time, and the odor level of the 60-minute measurement was reduced to approximately a third of the starting point. However, the smell of the mouse bedding was somewhat strong compared with that of room atmosphere. After that, spraying was continued day after day and then, the odor level of room atmosphere was reduced to as low as the level of a room where animals were not bred.

In a food processing plant (Ubuyama Village, Kumamoto), an investigation was conducted (*Fig. 7*). Reduction of the odor level was confirmed at every site. There was no problem from the perspective of food hygiene and safety.

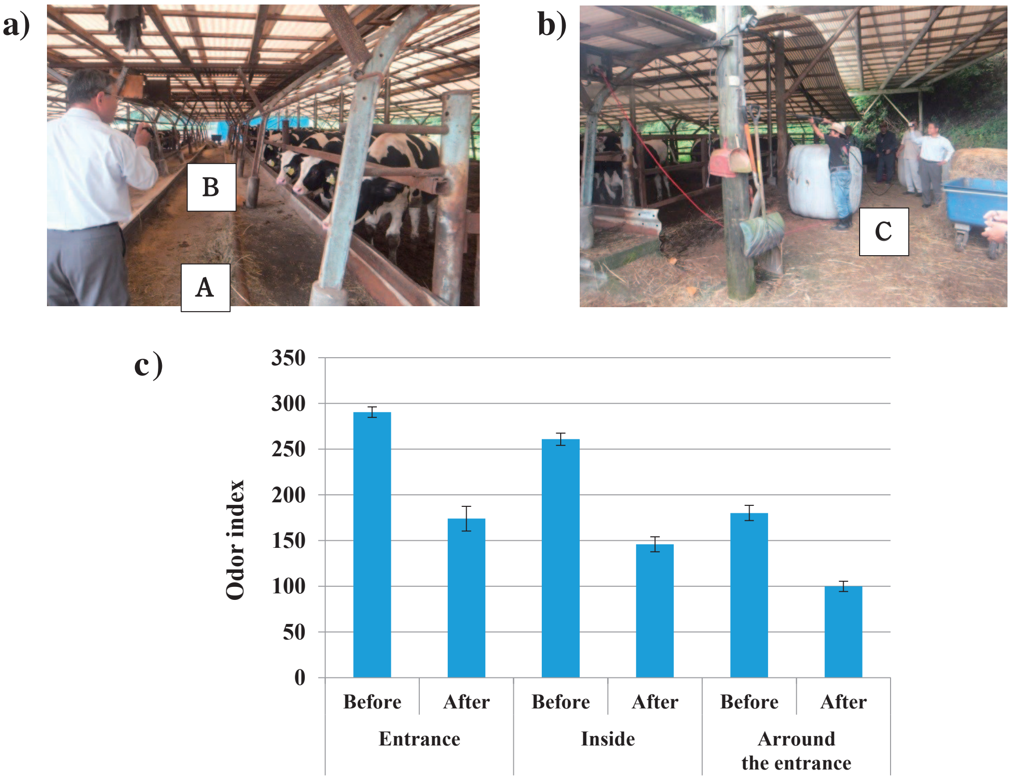

Fig.5. **Odor decomposition of ceramic-treated water: A cattle farm (Ubuyama Village, Kumamoto, Japan)**

a) Inside the cattle farm. **b)** Spraying operation. **c)** Alternation in odor levels. Results are shown in mean ± standard deviation. A: the entrance of the cattle farm. B: inside the farm. C: around the entrance of the farm. Measurement date: August the 23rd, 2015 at 11:00–16:30. Measurement site: Morimoto cattle farm.

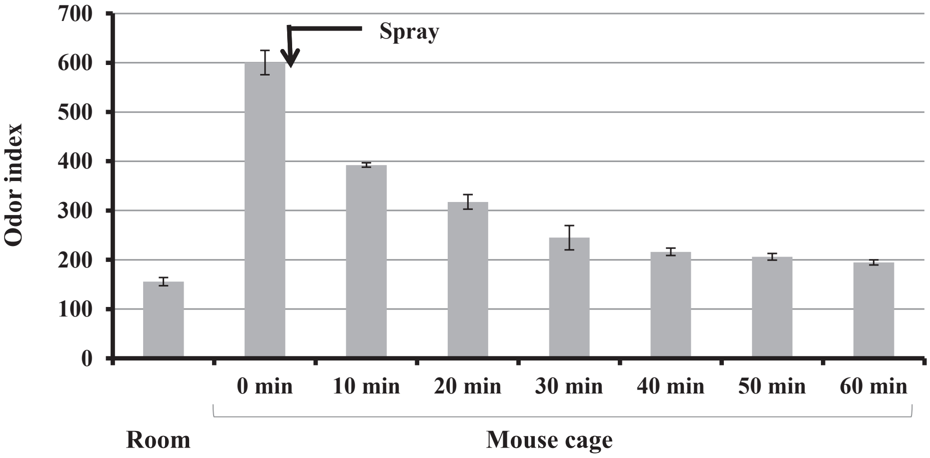

Fig.6. **Odor decomposition of ceramic-treated water: An animal barn (Kyoto, Japan)**

Results are shown in mean ± standard deviation. Measurement date: August the 4th, 2014. Odor measurement device: XP-329IIIR. Measurement site: Louis Pasteur Center for Medical Research, Animal Center.

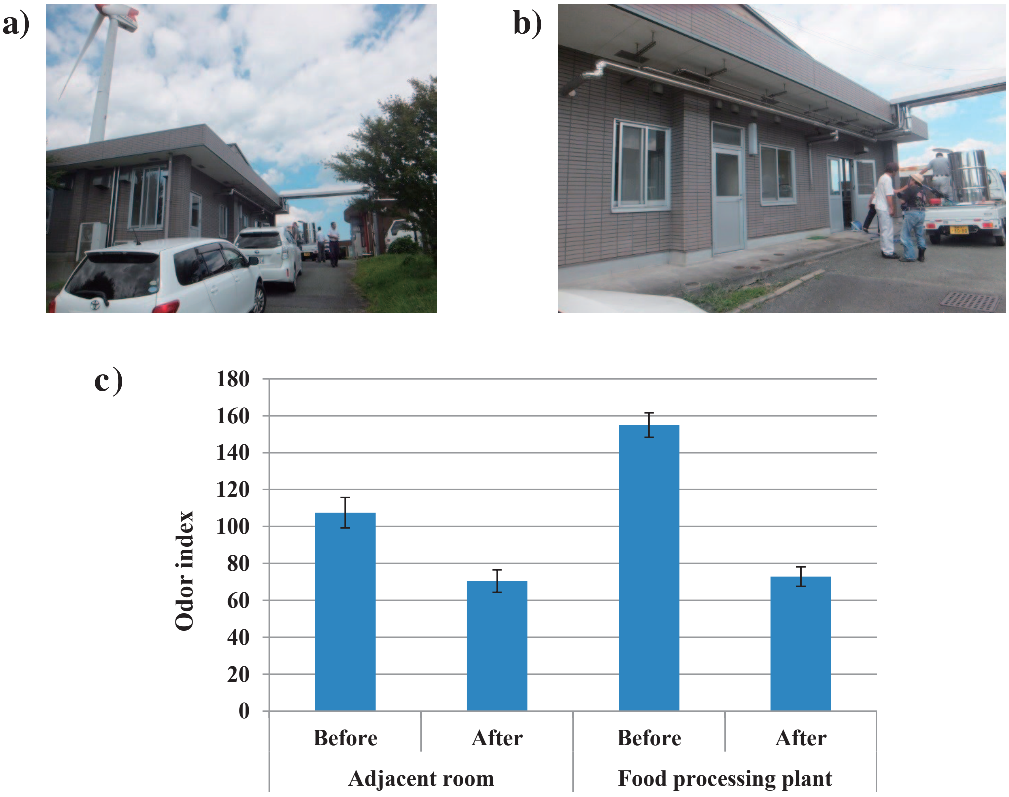

Fig. 7. **Odor decomposition of ceramic-treated water: A food processing plant (Ubuyama Village, Kumamoto, Japan)**

a) A full view of the plant. **b)** In front of the plant. **c)** Alternation in odor levels. Results are shown in mean ± standard deviation. Measurement date: August the 23rd, 2015 at 11:00 – 16:30. Measurement device: XP-329IIIR. Measurement site: Food processing plant of Ubuyama Farm,

2) Water purification

Study cases for the presence or absence of water quality improvement were as follows. Installing ceramic pieces in rivers and water supply and wastewater pipes, investigations were conducted.

In Tarumizu, Kagoshima, Japan, ceramic pieces, which were put in blue nets, were placed in the Honjo River (Igawa drainage system), as shown in *Fig. 8*. Rivers in this area were murky and smelled of animal feces and urine, as sludge water flowed out of stockbreeding farms located in the upper stream. Ten kg of ceramic pieces were put into multiple nets and the nets containing ceramic pieces were placed on the riverbed. Thirty minutes after the installation, the muddiness of the river disappeared, and the degree of transparency was improved. Simultaneously, smell, which had drifted, was mitigated.

In Midori River located in Saitama, Japan, which was contaminated by domestic wastewater, ceramic pieces were installed, and water quality tests were conducted (*Fig. 9*). Ten kg of ceramic pieces were spread over the outfall of the weir and another ten kg of ceramic pieces were placed on the basin of the weir of the river. One hour after the ceramic installation, black sludge turned into gray and the degree of transparency was improved. Furthermore, offensive odor was mitigated even 100 m downstream. Subsequently, due to overflow of detached sludge, water became muddy and then, the degree of transparency was reduced from 23 cm to 17 cm and dissolved oxygen (DO) was reduced from 5.3 mg/L to 4.1 mg/L. However, pH increased from 7.2 to 7.3. Chemical oxygen demand (COD) decreased from 22 mg/L to 17 mg/L. Biochemical oxygen demand (BOD) decreased from 35 mg/L to 28 mg/L. Suspended solids (SS) increased from 30 mg/L to 51 mg/L. Consequently, it was observed that the quality of the water was improved.

Figure 10 shows a sewage line located in Wakaba-cho, Nagasaki, Japan, where ceramic pieces were installed at two sites. Five kg of ceramic pieces, which were contained in nets, were placed at an exit of the sewage line and another 5 kg of ceramic pieces were placed at a sewer pipe, and then, a water quality investigation was performed downstream. Thirty minutes after the installation, detachment of sludge was confirmed from the wall. Offensive odor was reduced one hour later. As for water quality, both BOD and CDE were reduced by 50 %. Phosphor level was reduced by 30 % (*Table 2*).

Another ceramic investigation was conducted at a sushi restaurant in Watari-cho, Miyagi, Japan (*Fig. 11*). A filtration apparatus of a ceramic-pieces-filled tube was installed in a water supply pipe, and water quality of the sewer pipe was examined one month after the installation of filtration apparatus. Consequently, water purification and odor decomposition effects were confirmed (*Table 3*).

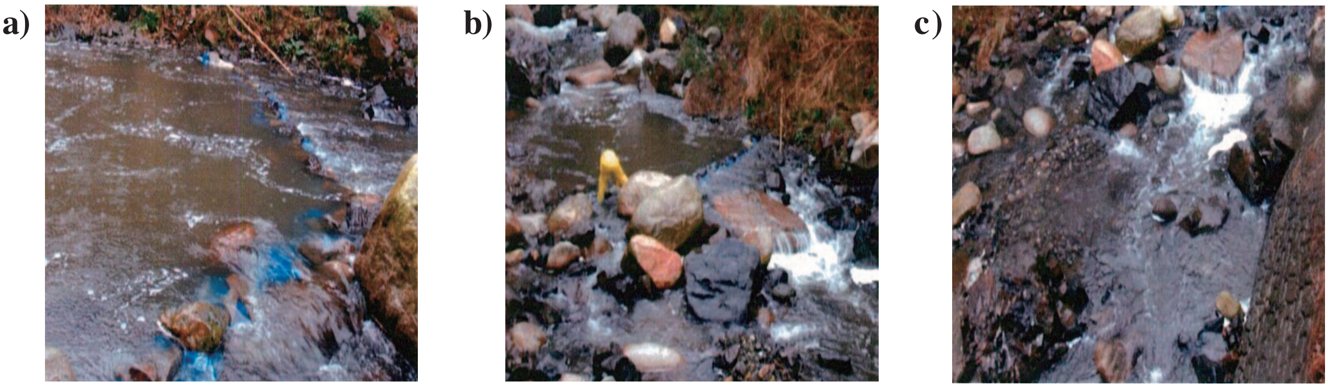

Fig. 8. **Water quality purification effects: A river (Tarumizu, Kagoshima, Japan)**

a) A full view of the river. **b)** Immediately after the installation of ceramic pieces. **c)** Thirty minutes after the installation. Installation site: Honjo River, Igawa drainage system.

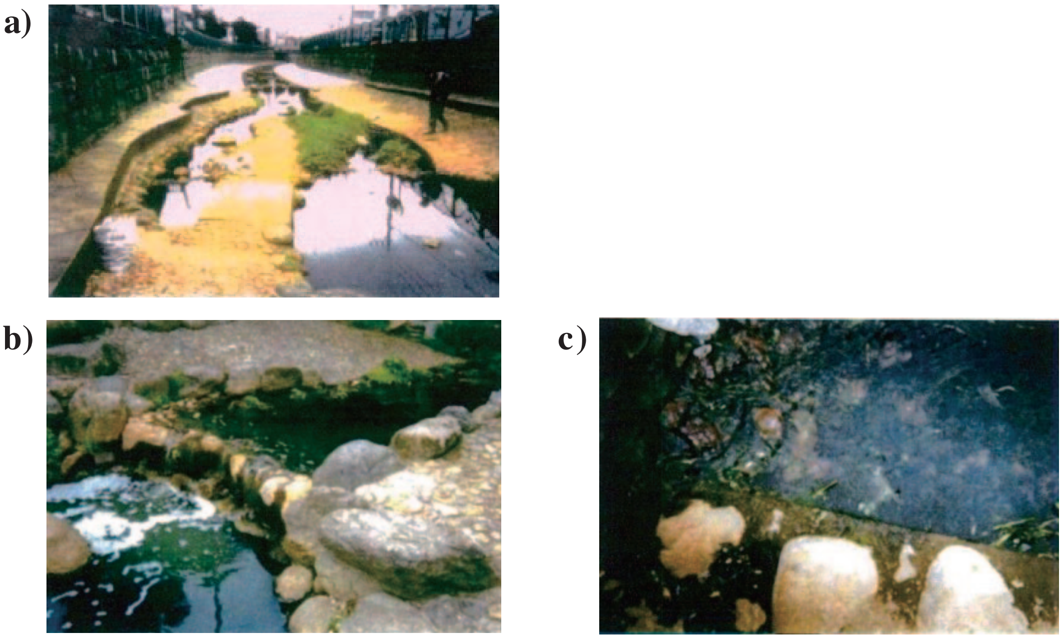

Fig. 9. **Water quality purification effects: A river (Saitama, Japan)**

a) Installation of ten kg of ceramic pieces over the weir and another ten kg on the waterfall basin. **b)** One hour later, black sludge turned into gray and the sludge was washed away. **c)** Two hours later, the color was changed to beige and offensive smell was mitigated over the installation site and 100 m downstream. River water purification was confirmed. Installation site: the Midori River in Saitama Prefecture. Installation personnel: Environment safeguard section of Saitama City Office. Installation date: March the 10th, 1999.

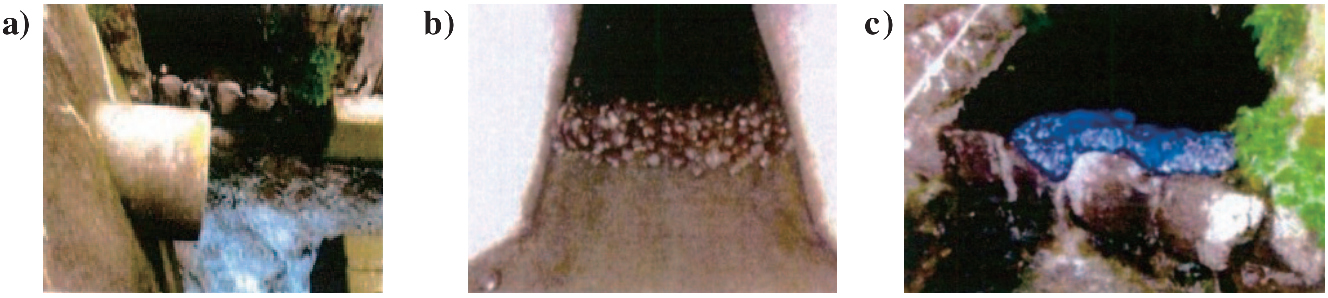

Fig. 10. **Water quality purification effects: A sewage line (Wakaba-cho, Nagasaki, Japan)**

a) Installation of a net filled with five kg of ceramic pieces at an exit of a sewer pipe. Limestone was placed as a base. **b)** Installation of five kg of ceramic pieces at a drainage canal. **c)** Installation of a net filled with five kg of ceramic pieces at a sewage exit. Measurement date: April the 6th, 2006. Installation personnel: Road maintenance division of Nagasaki City Office. Inspection institute: Nagasaki Food Hygiene Association. (registered number, # 77).

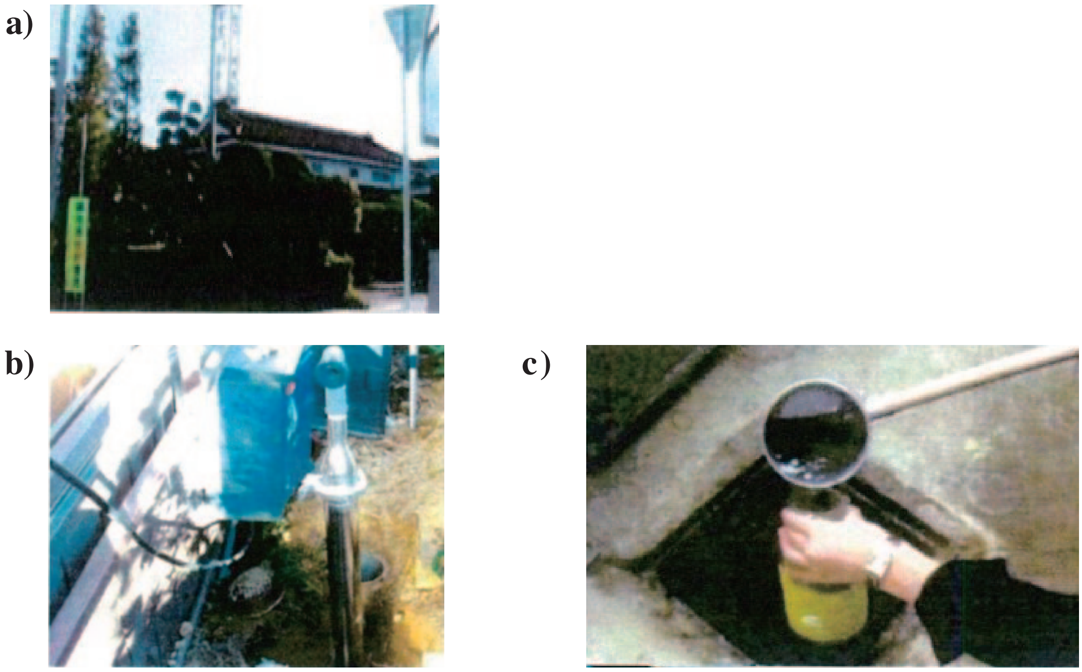

Fig. 11. **Water quality purification effects: A swage line (Watari-cho, Miyagi, Japan)**

a) A filtration apparatus of a ceramic-pieces-filled tube was equipped to a water supply pipe of a sushi restaurant (Hamazushi). **b)** Water supply system with the installation. **c)** Investigation of water after the installation. Measurement date: December the 14th, 2004. Installation personnel: Hamazushi. Inspection institution: foundational juridical person, Miyagi pollution and sanitation inspection center.

Table 2. Water purification effects: Sewage at Wakaba-cho, Nagasaki, Japan

Items	Before	After	% Change
pH	7.4	7.6	+
BOD (mg/L)	200	100	−50%
COD (mg/L)	240	120	−50%
SS (mg/L)	170	70	−58%
Nitrogen (mg/L)	21	12	−42%
Phosphorus (mg/L)	1.39	0.69	−31%

BOD, biochemical oxygen demand; COD, chemical oxygen demand; SS, suspended solid.

Table 3. Water purification effects: Sushi restaurant at Watari-cho, Miyagi, Japan.

Items	Before	1 month after
pH	5.8	6.3
BOD (mg/L)	600	170
COD (mg/L)	320	76
SS (mg/L)	2800	58
Hexane extracts (mg/L)	28	14

BOD, biochemical oxygen demand; COD, chemical oxygen demand; SS, suspended solid.

3) Bactericidal activities

Verification study results regarding bactericidal activities of ceramic pieces and ceramic-treated water are reported as follows:

Figure 12 shows ceramic pieces which were installed at a park site using water beautification for a decorative display (Mori-machi, Nagasaki, Japan). The irrigation canal (Seseragi Waterway) was filled with *Legionella pneumophila* and algae, when we started the investigation. The number of *Legionella pneumophila* (colony forming unit) reduced from 930 CFU/100mg/L in October the 4th, 2005, when ceramic pieces were installed, to 80 CFU/100mg/L in January the 13th, 2006. A reduction of 91% was achieved in three months, as confirmed. Furthermore, it was confirmed that algae were detached and odor was mitigated.

Bactericidal activities of ceramic-treated water were examined for putrefactive bacteria which were collected from putrid liquid of meat and raw fish. Putrefactive bacteria derived from bacteria floating in the air of a laboratory. Each putrid liquid sample from meat and raw fish was cultured in LB medium, which was prepared as follows: casein peptones 1.0%, yeast extract 0.5%, and sodium chloride 1.0% were mixed and diluted with distilled water. The culture solution was coated on a petri dish with an agar medium (addition of agar 1.5%). Putrefactive bacteria, which were sufficiently proliferated, were cultured in a LB medium which was prepared with ceramic-treated water and LB medium which was prepared with water. The number of bacteria in each culture solution was measured. Viable bacteria count was performed as follows: bacteria were cultured in each LB medium at 35°C for two days and then, serially diluted with physiological saline solution. After 0.5 ml of both diluted solutions 1×10^9/mL and 1×10^8/mL were dispensed into a dish, they were dispersed on a LB medium and cultured at 35°C for two days. Subsequently, the number of bacteria was measured by a hemocytometer. Measurement was performed by Pasuken Products laboratory (Kyoto, Japan). It was observed that the bactericidal effect of ceramic-treated water was 2.6 times higher than that of tap water, regarding putrefactive bacteria of raw fish (***Table 4***). There were no recognized bactericidal effects on meat. Similar experimental examinations regarding lactic acid bacterium did not indicate bactericidal effects. There were cases where bactericidal activities were not observed for aerobe and semi-anaerobic bacteria.

4) Effects on agricultural production

Verification study results regarding effects on agricultural production are reported as follows:

A case of ceramic-treated water was applied to rice cultivation in Mimata-cho, Miyazaki, Japan (***Fig. 13***). Plants of the family Gramineae grow with shoots growing around roots and branching off, which is called an offshoot. The use of ceramic-treated water enabled offshoots to double in number. Consequently, the number of seedlings were reduced compared with conventional farming methods. Rice yield with the use of ceramic treated water was 700 kg per *tan*. (1 *tan* = 300 *tsubo* = 991.74 m^2). Compared with an average rice yield of 400 g per *tan*, this was as more than 1.7 times that of the average. The quality of rice was improved with the increase in protein components and with higher than 80

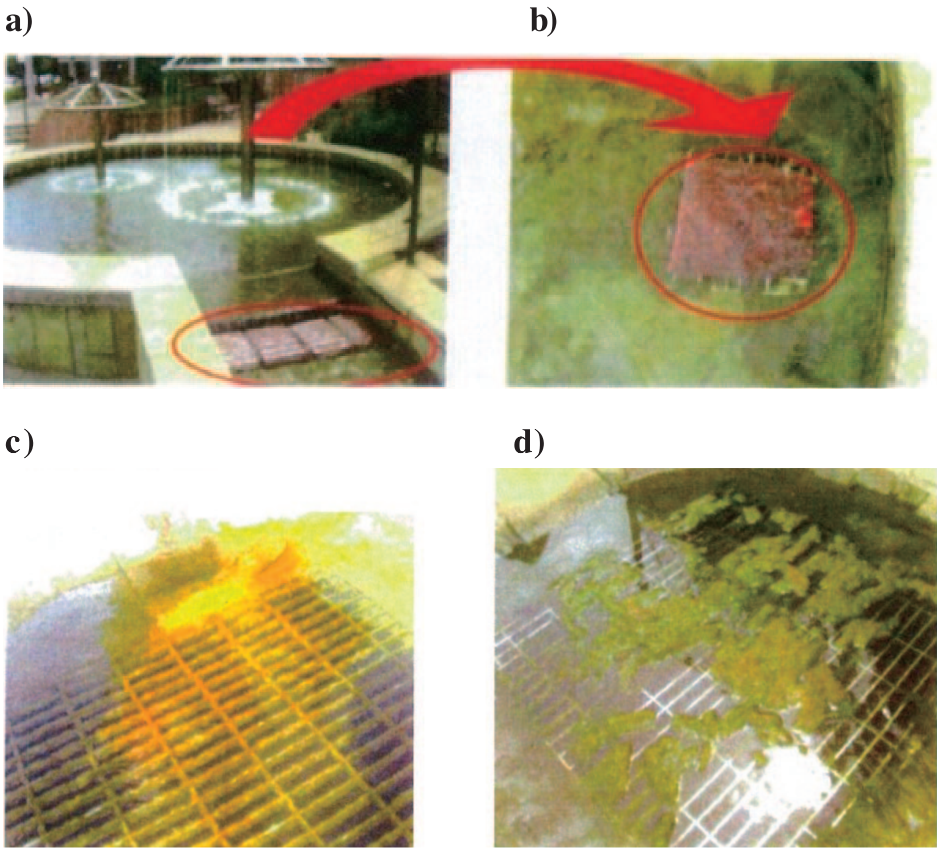

Fig. 12. **Bactericidal activities: A waterway in a park (Nagasaki, Japan)**

a) *Seseragi* Waterway with proliferated *Legionella pneumophila*. Installation of a hem sack filled with ceramic pieces at a fountain. **b)** Drainage. **c)** Algae attached to grating. **d)** Five days after the installation, the algae were detached and odor was improved. Measurement date: September the 13th, 2005. Installation personnel: Rivers Division of Nagasaki City Office. Measurement institution of the number of *Legionellae pneumophila*: Healthcare center of Nagasaki City Office.

Table 4. Bactericidal activities

Putrefactive bacteria	Sample water	The number of bacteria per mL	
		10^6 dilution	10^8 dilution
Meat-derived	Ceramic-treated water	unmeasurable	2.6×10^9
	Tap water	unmeasurable	1.0×10^9
Fish-derived	Ceramic-treated water	unmeasurable	1.8×10^9
	Tap water	unmeasurable	4.8×10^9

scores in all items. Therefore, it was confirmed that growing rice with ceramic-treated water led to quality improvement and increased yield.

Another application of ceramic-treated water on agricultural production was growing peach trees in Higashine, Yamagata, Japan (*Fig. 14*). Ceramic-treated water, which had a 20-fold dilution, was applied around the base roots once, followed by spraying 30-fold dilution on the leaves once. As a result, the yield of large-sized peaches was increased. The weight of a peach was increased by 100 g on average. Compared with conventional peaches, the flesh of a peach firmed up and the taste was improved. Furthermore, they were resistant to oxidizing and maintained their freshness for longer.

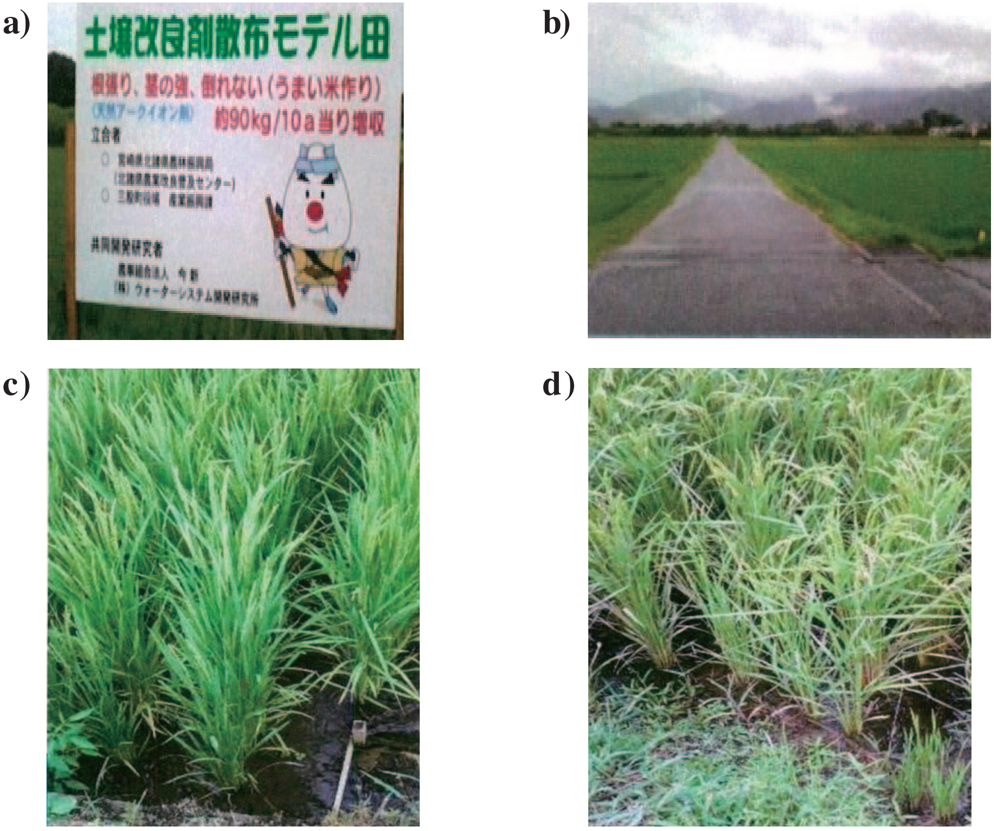

Fig. 13. **Application to agricultural products: Increase in agricultural harvest (Mimata-cho, Miyazaki, Japan)**

a) A signboard of the agricultural experimental station. **b)** A full view of the station. **c)** Experimental rice pad filled with ceramic-treated water (land area: 1 *chobu* = 9,917 m^2). An adjacent rice pad filled with ordinary water (land area: 1 *chobu*). Other conditions were equal in both pads. Experiment date: June–September 2010. Experiment site: Mimata-cho, Miyazaki Prefecture. Grower: Agricultural judicial person Imashin.

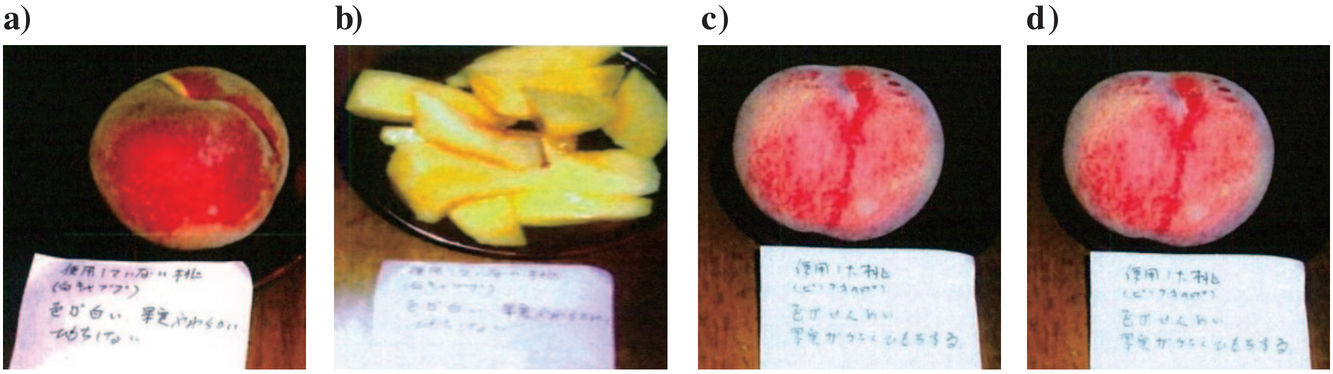

Fig. 14. **Application to agricultural products: Improvement of quality (Higashine, Yamagata, Japan)**

Fruits grown without ceramic-treated water were medium size; **a)** Flesh is soggy, **b)** Kept for a short time. Fruits sprayed at the part near the root with ceramic-treated water at 20-folded dilution three times a week for two months; **c)** Larger size of peach by 100 g on average, **d)** Kept for a long time. Experiment site: Zao Fruits. Measurement personnel: Saito Orchad.

5) Effects on pest control

The presence or absence of pest control effects by spraying ceramic-treated water was examined in an orange grove located in Miyazaki, Japan (***Fig. 15***). Ceramic-treated water at 20-fold dilution was sprayed on orange trees on the whole of the tree and again two days later; insect repelling effects were observed. Movements of harmful insects were observed. Specifically, prevention of mites was effective. Ceramic-treated water had an effect not as a pesticide, but as a repellent. Furthermore, it was confirmed two weeks after the spraying that the sugar concentration of peaches increased.

6) Inhibitory effects on hyperglycemia

C57BLKS/J Iar -+Lepr^{db}/+Lepr^{db} mice, type 2 diabetes animal models, were classified into two groups, a control group (n = 5) with the ingestion of water and a test group (n = 5) with the ingestion of ceramic-treated water. After the commencement of the experiment, blood glucose levels were measured once a week for six weeks, with a blood sample collection from caudal veins of mice and a use of a glucose sensor (Glucose PILOT, Aventir Biotech, LLC, Carlsbad, CA, USA), as shown in ***Fig. 16***. The test group demonstrated a significant inhibitory effect on hyperglycemia after one week, compared with the control group (significant difference between groups $p < 0.05$). It was indicated that the ingestion of ceramic-treated water improved carbohydrate metabolism and glycative stress was decreased.

7) Countermeasures for soil contamination resulting from the Great East Japan Earthquake

Application cases have been reported regarding the use of ceramic-treated water for the salt damage and radioactive contamination from the Great East Japan Earthquake as follows:

- **Amelioration in saline soil of farmlands**

The effectiveness of ceramic-treated water was examined in a strawberry farm which had salt damage in March the 11th, 2011 caused by the Great East Japan Earthquake (Watari-cho, Miyagi, Japan), as shown in ***Fig. 17***. After digging the

Fig. 15. **Pest control for agricultural products via ceramic-treated water (Miyazaki, Japan)**

a) Spraying ceramic-treated water at 20-folded dilution on the entirety of the fruit trees every day. **b)** Many harmful insects were observed before spraying. **c)** Three hours after spraying, insects moved away and kept away from trees. Experiment site: Orange grove.

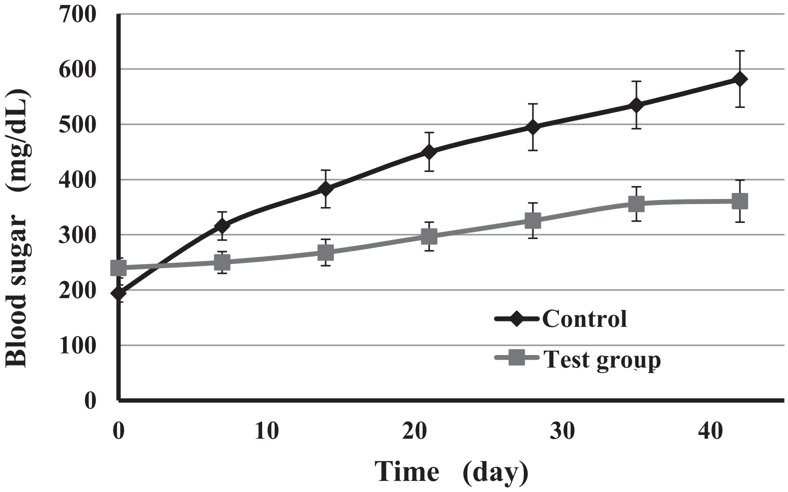

Fig. 16. **Inhibitory effects on hyperglycemia due to ceramic-treated water**

Results are shown in mean ± standard deviation. ◆ Control group: Ingestion of tap water (n = 5), ■ Test group: Ingestion of ceramic-treated water (n = 5), *$p < 0.05$, Students t-test. Animal model: C57BLKS/J Iar -+Lepr^{db}/+Lepr^{db} mice.

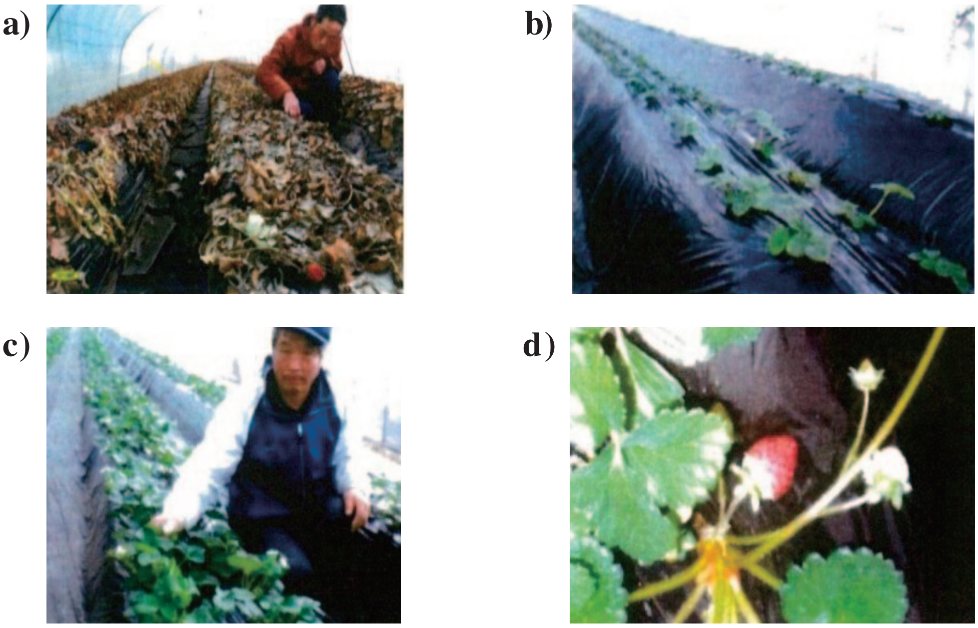

Fig. 17. **Remediation of soil contamination caused by the Great East Japan Earthquake: Amelioration of saline soil of farmland (Watari-cho, Miyagi, Japan)**

a) A strawberry farm after tsunami (land area: three *tan* = 2,975 m^2). **b)** Strawberries grown with conventional farming methods. **c)** Strawberries watered with ceramic-treated water. **d)** Young leaves were green and glossy. Measurement date: October 2011. Measurement institution: Azumi farm.

soil at 20 cm of depth, ceramic-treated water at 30-fold dilution was applied and water was settled. In the saline soil before the ceramic treatment, chlorine ions were 9,600 mg/kg dry weight and water content was 36.6%. Strawberries, which were sprayed with ceramic-treated water, grew faster and bore bigger berries than the average year.

- **Removal of radioactive substance**

In a playground located in Toyano, Fukushima, Japan, which had radioactive contamination caused by the nuclear power station accident in the Great East Japan Earthquake, effects of spraying fine ceramic powder were examined (***Fig. 18***). After measuring the radiation dosage of the soil, fine ceramic powder which had been processed from ceramic pieces was sprayed, and gamma-ray spectrum was measured using a high-purity germanium (Ge) semiconductor detector. Before the test, the soil showed 4,028 microsieverts per hour. However, after spraying, radiation dosage decreased to 1,276 microsieverts per hour. Radioactive cesium volume decreased to approximately one-hundredth of pre-application levels (***Table 5***).

Table 5. Decreased radioactive substances via fine ceramic pieces powder

Radioactive isotope	Soil before treatment (Bq/kg)	Soil after treatment (Bq/kg)
Iodine-131	undetectable	undetectable
Cesium-134	1.11×10^2	11.6
Cesium-137	1.45×10^2	11.4

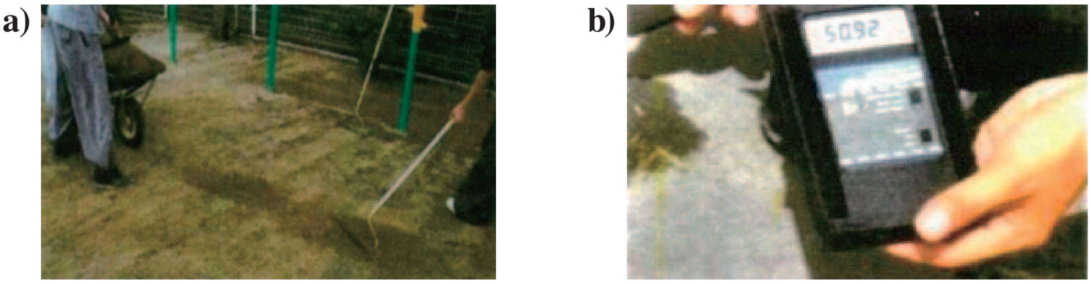

Fig. 18. **Remediation of soil contamination caused by the Great East Japan Earthquake: Treatment of radioactive contaminated soil (Toyano, Fukushima, Japan)**

a) Sampling playground. **b)** After measuring the contaminated soil level of the playground, fine ceramic powder was disseminated. One hour later, the soil was measured a second time. Measurement date: September the 1st, 2011 at 11:28–13:30. Measurement site: A kindergarten playground. Measurement device: High-purity germanium (Ge) semiconductor detector (gamma-ray spectrum measurement).

Discussion

Activities of ceramic-treated water

1) Order decomposition and bactericidal activity Odor was

Radicals such as O^+ and OH^- are generated via water molecular split. Oxygen radicals (O^-) bind hydrogen radicals (H^+) to generate hydrogen peroxide $(2H^+ + O_2^- = H_2O_2)$. Furthermore, cyanobacteria, which exists in nature, produces oxygen via physiological synthesis reaction. This produced oxygen binds hydrogen ions, and hydrogen peroxide (H_2O_2) is generated. Hydrogen peroxide (H_2O_2), which is generated in this manner, has bactericidal activity and order decomposition. Chlorine, which is used for sterilization, reacts with water and consequently, hydrogen chloride (HCL) and hypochlorite (HCLO) are produced. $(Cl_2 + H_2O = HCl + HClO)$. Thus, chorine is stabilized.

- **Deodorization of ammonia**

With hydrogen peroxide involved in ammonia decomposition (oxidation), nitrogen gas (N^2) and water, hydrogen oxide (H_2O) are produced, which is considered a mechanism of deodorization of ammonia $(2NH_3 + 3H_2O = N_2\uparrow + 6H_2O)$. It was confirmed that spraying ceramic-treated water had deodorization effects on ammonia in cattle sheds such as a hog farm, a cattle farm, and a poultry farm.

- **Deodorization of hydrogen sulfide**

Hydrogen sulfide is converted into sulfur via oxidation-reduction reaction with hydrogen peroxide $(H_2S + H_2O_2 = 2H_2O + S)$. Consequently, hydrogen sulfide odor is removed.

Commonly, toxic substances for odor decomposition and bactericidal activities require careful handling. However, the ceramic pieces and treated water do not contain any toxic substances. This ceramic is naturally-derived. Multiple natural plant embryo buds and sprouts are mixed and fermented to prepare the glaze. This glaze is applied to the surface of potteries, which is treated with heat at a thousand and several hundred degrees. The ceramic-treated water is refined via these special-glaze-applied ceramic pieces in the shape of a ball. This ceramic-treated water exerts odor eliminating and sterilizing effects, not containing any toxic substances. It was suggested that these effects were exerted via increased dissolved oxygen by these ceramic pieces. Measurement results of a mice-bedding case confirmed the odor eliminating effects of ceramic, observing that the degree of odor decreased from 600 to 194 in spraying the mice bedding which had been used. Continuous spraying day after day of ceramic-treated water indicated that the odor level of a mice-breeding room decreased to the level of a non-animal-breeding room.

In a restaurant district, odor level measurements have progressed at a sewer (food-derided sewage sludge) and effectiveness has been confirmed. The reductions in odor levels via spraying ceramic-treated water have been observed in diverse sites such as animal breeding (dogs and cats), and hog, cattle, and poultry farms. Further research on ceramic-treated water will be conducted to extend application range, examining physical and chemical properties, degrees of bioactivities for humans such as immunological activation, anti-allergic effects, anti-oxidative effects, blood glucose control, and inhibitory effects on hypertension. At present, academic research has started on mechanisms of ceramic-treated water for odor decomposition and pH control, types of bioactivates and toxicity tests. When ceramic-treated water is confirmed to perform odor decomposition, bactericidal action, and bioactivity without adverse effects, usefulness of ceramic-treated water can be applied to clinical treatments.

2) Effects of sterilization and antimicrobial activity

Bonding between oxygen radicals (O_2^-) and hydrogen ion (H^+) produces hydrogen peroxide (H_2O_2). Oxygen radical (O_2^-) is produced, gaining an electron in water (reduction).

$O_2 + e^- = O_2$ (Oxygen radical production)
$2H^+ + O_2^- = H_2O_2$ (hydrogen peroxide production)

Hydrogen peroxide, which is produced in this manner, has bactericidal activities and odor removal effects. On this principle, hydrogen peroxide is utilized as a skin disinfectant in medical fields. There is a possibility that minerals are involved as catalyzers in these reactions.

3) Rust prevention

Bonding between oxygen radicals (O_2^-) and hydrogen ion (H^+) produces hydrogen peroxide (H_2O_2). Iron (Fe) oxidizes in water (loss of two electrons) and is converted into iron ions (Fe^{2+}). Hydrogen peroxide (H_2O_2) reacts with iron ions, which leads to conversion of ferrous oxide (FeO) and water, hydrogen oxide (H_2O). Triirontetraoxide (Fe_3O_4), which is a stabilized oxide, is formed via bonding between ferrous oxide (FeO) and ferric oxide (Fe_2O_3). This triirontetraoxide (Fe_3O_4) prevents corrosion, forming a coating layer5). Oxygen (O_2) increases fuel output.

$H_2O + e^- = H^+ + OH^-$
$O_2 + e^- = O_2^-$
$2H + O_2^- = H_2O_2$
$Fe = Fe^{2+} + 2e^-$
$H_2O_2 + Fe^{2+} = FeO + H_2O$
$FeO + Fe_2O_3 = Fe_3O_4$

4) Water quality improvement

Ceramic-treated water has a chelating action (water molecules are isolated from water clusters. The water molecules of fine structure enter slits and exert stripping actions), and actions of flocculation and precipitation. Therefore, ceramic-treated water enables minute substance floating in water to be precipitated and removed for water purification, having the effect of the detachment of sludge and other attached materials on walls as well as the effect of washing them away. These types of effects could be applied to the removal of microscopically small substances (for example, radioactive isotope contamination in water) through the process of flocculation and precipitation. Ceramic pieces and treated water are also effective in water quality improvements through sludge removal of rivers, ponds and domestic wastewater.

5) Soil amelioration

Ceramic treated water contains minerals at a favorable ratio. Furthermore, after photosynthesis of cyanobacteria, which induces fixations of oxygen, nitrogen, and carbon, and bacterial activities, 5-aminolevulinic acid (ALA) is formed. By ALA, soil is provided with antibacterial effects and nutrition (fixed nitrogen and fixed carbon). Soil is improved by ALA, which is a precursor of heme, a vital substance for life 6). Heme, which is the end product, is provided to hemoprotein contained in the mitochondria electron transport chain complex, and functions as essential biomicromolecules for energy metabolism in cells. Provided with antibacterial effects, good nutrition, and a supply of oxygen, soil environments are enhanced to promote plant growth. Experimental data of the present study confirmed higher productivity of crops in the salt-damaged farm. It has been reported that photosynthetic bacteria have activities of removing salt from sea water 7). There is a possibility that cyanobacteria are involved in the saline soil case.

6) Effects on soil contaminated by radioactive substances

Diverse investigations have been conducted to reduce the amount of radioactive cesium in contaminated soil. Conventional measures are surface soil scraping 8), skim and burial ploughing 9), soil puddling with water 10), absorption and removal by plants (phytoremediation by *Helianthus annuus* and *Amaranthus*) 11), and heat treatment for removal of cesium 12). However, these conventional methods have problems, such as low economic efficiency, and difficulties in application to contaminated soil in a large scale. For example, the method of skim and burial ploughing is to remove 5 cm of contaminated top-layer soil, bring up the lower layer of 5 – 50 cm, and bury the contaminated soil beneath the lower layer 9).

There are two types of radioactive cesium which were deposited: one is water-soluble (medium, sulfate aerosol) and the other is insoluble particle (slag-like substance). A reason for the difficulty in removal is that most of the radioactive cesium is in the particle state.

The method of dissolution and separation is difficult, as the adhesiveness properties of cesium are different in soil depending on clay content and types of clay minerals. Adsorbing manners of cesium are diverse, such as adsorption to clay or silt (silt, clastics which is smaller than sand and coarser), adsorption to humic substances (mixture of plant-derived materials due to biodegradation), and colloidal distribution. For the method that plants absorb pollution from the soil, there is no recognized clear relationship between cesium concentration and variants of cesium moving to plants. There were cases with high cesium concentration in soil and low in plants and reverse cases, and the effectiveness has not been identified 13).

Reports of practical removal of radioactivity from the soil by photosynthetic bacteria 14-16) are remarkable. Photosynthetic bacteria produce extracellular polymeric substances (EPS) of negative charge on the surface, and have actions to draw metal cations including cesium 17). Furthermore, these bacteria capture cations via potassium pumps on their surface 14, 15). Consisting of polysaccharides (exopolysaccharides) and proteins, EPS is a fundamental component of biofilms, and plays a role in the defense against externality and the enclosure of mutualistic bacteria.

We expect that activities of cyanobacteria make a contribution to the decontamination of radioactive cesium, including the oxidative decompositions of fixed components (clay, silt, humic substance and colloid) via oxygen which are produced by cyanobacteria. We do not have information on further reactions, but we expect that radioactive cesium migrates after bacteria destruction and will be diffused and penetrate into deep layers of the soil.

The authors, Sasaki et al. emphasize cooperative activities with lactic acid bacteria 16). Lactic acid bacteria degrades organic matter in sludge and tidal flats, reduces COD and produces acetic acid, propionic acid and lactic acid 18). These organic acids are a source of nutrients for photosynthetic bacteria. Mechanisms for smooth decomposition and purification are suggested. In these ceramic pieces, there were no recognized bactericidal activities of cyanobacteria on lactic acid. Contrarily, there is a higher possibility that lactic acid is a mutualistic bacterium. There is no information of how soil bacteria will change, and further verification studies are required.

7) Agricultural production

Oxygen combined with ceramic material induces prevention of root decay, minerals at a favorable ratio, fixed nitrogen and fixed carbon as sources of nutrition, create cell proliferation activities of ALA which promote the growth of agricultural crops and activate functions of plant organs 19, 20). The activated functions enhance not only growth but also sugar content and taste. Therefore, delicious agricultural products are produced such as rice, fruits, and vegetables.

8) physiological activities

Ceramic-treated water contains oxygen, minerals at a favorable ratio, ALA, carbon, and nitrogen, which have bioactivations. We have confirmed influences on immunocompetence in an experiment with mice, the antitumor effects via activated immunocompetence, and the enhancement of physical strength of the sickly weak (unpublished reports). The present study confirmed inhibitory effects on hyperglycemia in mice experiments. This finding is extremely remarkable, observing that the ingestion of ceramic-treated water induced the improvements in carbohydrate metabolism and the reductions in glycative stress. However, the mechanisms have not been identified. At present, research is in the process of elucidating how the mechanisms of how these effects are exerted. Furthermore, there is a possibility of activating intestinal immunocompetence via improvement of intestinal flora, as it was observed that offensive odor was reduced in defecation in an animal experiment.

9) Effects on improvement in livestock breeding

Ceramic-treated water contains oxygen and minerals at a favorable ratio, ALA, carbon, and nitrogen, which have bioactivations. These effects elevate the relief of livestock due to decreased odor level, the sanitary maintenance control, and the functions of organs. Consequently, this leads to a decrease in fetal death incidents, an increase in milking amounts, a decrease in diseases (prevention), a high growth performance, and an improved quality of meat, which have been confirmed in livestock farms such as hogs, cattle, and poultry. Ceramic-treated water is effective for livestock environmental improvements.

Mechanism of ceramic pieces and ceramic-treated water

The mechanisms of ceramic pieces and ceramic-treated water were not identified in detail at first. Multiple theories were assumed, including the dissolution of several types of minerals into water, the diffusion into soil, the detachment of sludge via chelate effect, the actions of flocculation and precipitation of minute substances floating in water, and rust prevention via coating triiron tetraoxide (Fe_3O_4) layers on iron surfaces, as shown in ***Fig. 19***. The mineral mixing ratio of the ceramic material in the use of the ceramic pieces and ceramic-treated water was assumed to be optimal for homeostasis, as inducement of diseases in animals and plants and spontaneous remission are varied in increases and decreases, depending on the mixing ratios of minerals.

Thereafter, proliferation of cyanobacteria was detected in the ceramic surface. This finding could explain that the above effects, which we had experienced, could be induced by cyanobacteria. Cyanobacteria have activities to fix nitrogen and carbon in water and/or the atmosphere and produce oxygen via reactions of enzymes with light amplification. As "photosynthesis" is written in the figure, photosynthesis of cyanobacteria is greatly different from ordinary photosynthesis of a plant, which uses CO_2 and chlorophyll. Cyanobacteria can produce oxygen in an environment without light or with minimal light. Furthermore, cyanobacteria produces ALA, which is important for life to grow as an essential fundamental substance. This substance, ALA, exits in mitochondria and chloroplasts for life; materials of hemoglobin in animals, and materials of chlorophyll in plants.

It is assumed that these activities play roles on the water and soil improvement, the germicidal, antibacterial, deodorizing effects, rust preventions of iron, and increased crop yield.

The concentration of dissolved oxygen is high in ceramic-treated water, which provides favorable environments to, other than cyanobacteria, oxygen production bacteria (example: filamentous aerobic bacteria). Further examination is required to verify bacteria which proliferate in periphery of ceramics or ceramic pieces. The mechanisms of inhibitory effects on hyperglycemia have not been identified regarding the ingestion of ceramic-treated water in animal model experiments. This effect, which could lead to the reduction of glycative stress, is an intriguing research topic.

Expected duration span of ceramic-treated water

It might be thought that ceramic which is treated with heat at higher than 1,000 °C does not dissolve into water. As a matter of fact, the first and the second atomic layers of ceramic glaze on the surface, dissolve into water and an infinitesimal volume (an undetectable amount by an ordinary measurement method) of minerals are contained. The thickness of the glaze is 0.2 – 0.3 mm and the thickness of an atom is approximately 10^{-8} cm. Assuming that $2–3 \times 10^{-8}$ cm dissolves, it requires the following for all to dissolve:

$3 \times 10^{-2}/3 \times 10^{-8} = 10^{6}$ days = 1,000,000 days
= 1,000,000/365 days ≒ 2,740 years

When ceramic-treated water is boiled, the amount of trace elements which are dissolved from the glaze is doubled compared with room temperature water. It would take 1,370 years for the glaze to be completely dissolved by boiling. Regardless of whether boiled or non-boiled, it would take more than 1,000 years, which would mean that ceramics have a semi-permanent service life. Thus, ceramic-treated water can produce these beneficial effects via the maintenance of this optimal component ratio of trace elements.

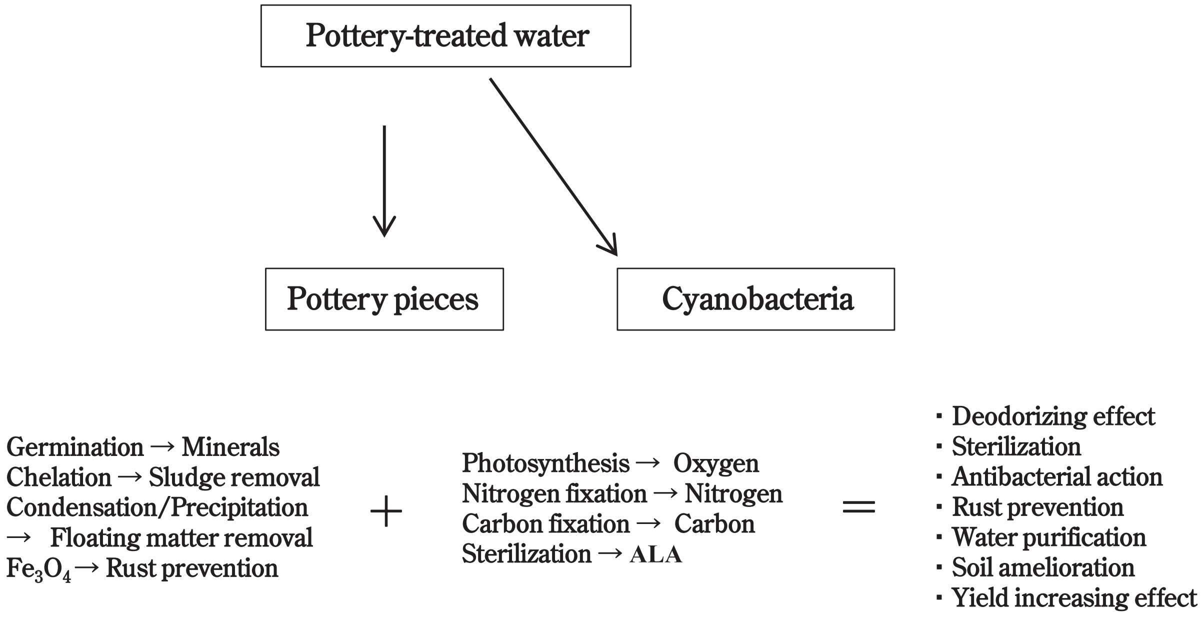

Fig.19. **Mechanisms of ceramic pieces and ceramic-treated water**

Safety

The amount of trace elements contained in ceramic and ceramic-treated water is less than the toxicity threshold. Ceramic materials have no toxicity. Over a 25-year length of usage, there are no-observed-adverse-effects and impairments. Safety is warranted by these materials. The present study suggested that the optimal ratio of components for the amounts of each element produced via ceramic treatment enables diversified and beneficial effects including an elevation of bioactivities.

Conclusion

The ceramic materials reported in the present study have a long service life (longer than several hundred years), have no observed toxicity, and present diverse effectiveness such as enhanced bioactivities, water quality improvements, odor decomposition, and bactericidal activities. There are no observed adverse effects or impairments. Improvements have been confirmed in farmland soil, livestock breeding, and meat quality. Their mechanisms, which were identified in detail, have started to be clarified by the findings of the cyanobacteria proliferations in water and soil where ceramic materials were installed. A reasonable possibility is that these diverse phenomena are based on the production of oxygen by cyanobacteria, including oxidation, sterilization, and deodorization. Ceramic and ceramic-treated water applications demonstrate favorable efficacy for the health and QOL maintenance of the aged, the sickly weak, and the sick. We hope that these effects will play a role of cost reduction in public medical expenses. Furthermore, there were possibilities that applications to soil were effective in the soil amelioration of agricultural land and the anaerobic digestion for soil. In addition to these diverse effects, ceramic fragments are expected to have global-scale significance for environmental improvements and increased agricultural productivities. At present, the presence of cyanobacteria is only morphologically identified in a procedure of using an optical microscope. Further investigations are required to thoroughly elucidate the activities of cyanobacteria in ceramic materials.

Acknowledgements

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