Characteristics of Microbial Mats in Fukushima prefecture, after the 2011 off the Pacific coast of Tohoku Earthquake on March 11, 2011.

博 士 論 文

Characteristics of Microbial Mats in Fukushima prefecture, after the 2011 off the Pacific coast of Tohoku Earthquake on March 11, 2011.

(和題：2011 年 3 月 11 日に発生した東北地方太平洋沖地震後の福島県内のバイオマットの特徴)

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Characteristics of Microbial Mats in Fukushima prefecture, after the 2011 off the Pacific coast of Tohoku Earthquake on March 11, 2011.

By

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Abstract

The characteristics of microbial mats formed after "The 2011 off the Pacific coast of Tohoku Earthquake" (3.11 Earthquake) were clarified by studies of their living environment, chemical compositions and microscopic observation at following four study sites. White microbial mats formed in flooded paddy fields in Karasuzaki, Kashima-ku, Minamisoma city, Fukushima prefecture were damaged by salt water of Tsunami after the 3.11 Earthquake. These microbial mats consist largely of diatom with mineralized halite and gypsum. The radionuclides of Cs were detected in microbial mats by using a Ge semiconductor analyzer. Particular elements concentrated in microbial mats were detected by semi-quantitative analysis and their elemental content maps were obtained by using a SEM-EDX. The white microbial mats formed by *Nitzschia* sp. was resistant or adapted to the brackish water appeared in the environment change caused by the tsunami. Microbial mats were continuously formed even in dring and wetting cycles. Reddish brown microbial mats were formed by iron oxidizing bacteria, *Leptothrix ochracea*, in the canal of the spring in Azumagaoka park, Haramachi-ku, Minamisoma city, Fukushima prefecture. Iron concentrated in microbial mats was detected by using WD-XRD and SEM-EDX. Based on the elemental content maps using SEM-EDX, it is suggested that the microorganisms forming microbial mats of Karasuzaki and Azumagaoka park are able to adsorb trace elements such as Sr, U, Th. Hot spring water suddenly gushed out from a house foundation located in Uchigo-Takasakamachi, Iwaki city, Fukushima prefecture after the 3.11 Earthquake. At this site, we observed reddish brown microbial mats, formed by iron oxidizing bacteria, *Leptothrix ochracea*. Similarly, very hot spring water gushed out from a shaft of old coal mining at Izumi-Tamatsuyu, Iwaki city after the aftershock happened in 11th April 2011. At this site, we observed that native sulfur and gypsum were formed in sulfur-turf develop in sulfide-containing hot springs water. Differences of microbial mats formed in Izumi-Tamatsuyu and Uchigo-Takasakamachi was

arose by the segregation of microorganisms due to the changes of redox environment and water chemistry in the course of hot spring water flows. The change of the chemistry of hot spring water was caused by the concentrations of particular elements in microbial mats. In these four study sites, it could be concluded that the bacteria inhabit adopting each growing environment selectively forming microbial mats and they concentrate particular elements. Biomineralization, and the process for concentration of particular elements by microbial mats were easy to maintenance, and sustainability can be expected. These process are potentially useful for the environmental remediation at the place where was damaged by 3.11 Earthquake.

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Chapter 1　Introduction

1-1 The 2011 off the Pacific coast of Tohoku Earthquake

"The 2011 off the Pacific coast of Tohoku Earthquake" (henceforth 3.11 Earthquake) occurred on March 11, 2011. Its seismic center was in offing of the Sanriku coast, at a depth of about 24 kilometers. The magnitude of the earthquake was M9.0. This magnitude is extrodinally large, because destruction of three earthquake source faults occurred continuously (Japan Meteorological Agency, 2011). In this earthquake, it recorded a seismic intensity of 6-upper at the maximum in Fukushima prefecture, Japan. Tsunami was occurred at the Pacific coast of the Tohoku district. The structures or farmland suffered serious damage by this tsunami. Above all, radioactive materials were scattered on wide area by nuclear reactor damage and a hydrogen explosion because of power supply loss by tsunami at Fukushima Daiichi Nuclear Power Plant (FDNPP). Therefore double damage of a tsunami and radioactive contamination occurs at the coast area of Fukushima (Kokubun et al., 2013; Tazaki et al., 2014a). The height of more than 9.3m for this tsunami was observed at tide station of Soma near Minamisoma city, Fukushima, Japan(Japan Meteorological Agency, 2012). Minamisoma city is one of the study areas. However, a geological survey company observed that the point with an altitude of 10.5 m was flooded 1.5m depth, flooded range of tsunami has exceeded in Soma (Tazaki et al., 2014a). The total amount of radioactive materials released by the FDNPP accident is said to be 11.64EBq (11.64×10^{18}Bq), and those materials such as ^{134}Cs, ^{137}Cs, ^{90}Sr, and ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu have 2 years or more of the half-life. Meanwhile, since Pu has heavy atomic weight and it is non-volatile in addition to its limited released amount, the scattering areas are limited. Only a little quantity of Pu was observed around FDNPP, and it is regarded not so problematic (Iwamoto et al., 2014). There is the farmland at where decontamination and salt removal of Pu were not progressing after the 3.11 Earthquake in a part of the coast of Fukushima prefecture.

After one month of the 3.11 Earthquake, the inland aftershock of M7.0 occurred in the shallow seismic center of about 5km in depth at Hamadori, Fukushima prefecture on April 11, 2011(henceforth 4.11 aftershock). The 4.11 aftershock was the biggest inland aftershock which occurred at an active fault after the 3.11 Earthquake, and it was suggested the relation with the earthquake which occurs at an ocean trench (Ishiyama et al., 2011; Sugito et al., 2011; Japan Meteorological

Agency, 2012). After the 4.11 aftershock, hot spring waters gushed out in two locations at Iwaki-Yumoto hot springs area, Iwaki city, Fukushima prefecture. The hot spring water gushed out from the foundation and drainpipes of a house in Uchigo-Takasakamachi, Iwaki city. The other hot spring water gushed out from the shafts of old coal mine in Izumi-Tamatsuyu, Iwaki city. These hot springs continue gushing out in January, 2014, and the hot spring waters were drained into gutter and river.

1-2 About microbial mats.

The microorganism is bearing the important role in material recycling of an ecosystem through disassembly of an organic matter, fixation of nitrogen, and collection of trace heavy metal elements. The microorganism inhabits not only the place that normal creatures inhabit but also under the conditions of low temperature below the freezing point, a high temperature, the strong acid and the strong alkali environment. These microorganisms formed aggregate which was often called "microbial mats" in environment with the water including those of rivers, lakes, ocean, volcanos and hot springs etc. The aggregates formed by a microorganism have various structures, such as films, tunicas, mats, and terraces (Tazaki, 2010). Microorganism in the microbial mats collects various elements and minerals, which was called biomineralization.

Microbial mats formed in drainpipes at landslide areas, and well pipes, deteriorate the function of pipes. Generation of hydrogen sulfide by reducing bacteria, and change in electric potential by iron microbial mats, those phenomena corrode concrete or metal. In addition, microorganism was related to formation of sediments in hot springs and weathering of rocks. On the other hand, biomineralization, and decomposition of organic matter by microorganisms have been used for water purification such as coagulation sedimentation of organic matter and removing iron component. In addition, environmental cleanup of the soil and the groundwater polluted by hardly decomposable organic compound by using microorganisms are paid attention as "bioremediation". "biostimulation" is one of bioremediation, it is environmental cleanup and environmental remediation by activating indigenous microorganisms.

It is become clear that various microorganisms live even in farmland flooded by a tsunami and polluted by radioactive materials, and in the hot spring water gushed out by an earthquake at Fukushima prefecture (Shimojima et al., 2014b; Tazaki et al, 2013a; Tazaki et al, 2013b; Tazaki et al, 2014a, b).

1-3 The purpose of this study

Various damage such as the collapse of the buildings and ground deformation, slope failure, tsunami, and radioactive contamination by the FDNPP accident occurred by the 3.11 Earthquake. There are many studies such as mechanism of an earthquake and tsunami occurrence, displacement of a fault caused by the earthquake, mechanism of the hot spring water gushed out, and behavior of groundwater. There are reports such as diffusion and the pollution situation of a radioactive material by FDNPP accident, and experiments such as method of physical and chemical decontamination, volume reduction of contaminated soil after the 3.11 Earthquake. On the other hand, there are little scientific data and studies for microbial mats in the tsunami deposit, and microorganisms which live in hot spring water gushed out by earthquake, and biostimulation using by activating indigenous microorganisms.

Rubble by this earthquake was removed, but the salt and tsunami deposits at the farmland were not yet completely removed. There is not a method to stop a spout of the hot spring water. The contaminated hot sprong water is discharged into a gutter and the river without being processed, and the water environment in the hot spring area in Iwaki city chaged.

Restoration for gameged area is not developed even more than 4 years after the earthquake. Continuous environmental restoration for a long period of time is also expected even now. The environmental cleanup with the microbial mats has so small. Therefore, sustainable and effective techniques are expected for the revival and environmental restoration. Therefore, bioremediation is sutable for long-term and sustaineable restration of environment damaged and contaminated by earthquake and the related accident, because bioremediation need not heavy mentenance and special techniques.

In this study, the paddy field microbial mats at Krasuzaki, Kashima-ku, Minamisoma city, Fukushima prefecture that were damaged by the tsunami of the 3.11 Earthquake. The four places in this area were studied. The white microbial mats were formed in the paddy field damaged by the tsunami at Krasuzaki, Kashima-ku, Minamisoma city, Fukushima refecture after the 3.11 Earthquake. The reddish brown microbial mats is guessed to be formed in Azumagaoka park Haramachi-ku, Minamisoma city, Fukushima prefecture from before the 3.11

Earthquake. Reddish brown microbial mats and the white microbial mats were formed in the hot spring water of two locations in Uchigo-Takasakamachi and Izumitamatsuyu, Iwaki city, Fukushima prefecture after the 4.11 aftershock.

The characteristics and environment of the microbial mats were discussed from chemical constitutions, biomat origin minerals, electron microscopic observations, water chemistry, a dose of radiation, and radioactive nuclide analysis. From these results, an association between microbial mats and geological feature and the water chemistry, collection of the particular element including the radionuclide, environment change after the biomat formation, and the possibility of the environmental restoration by biomineralization, were discussed.

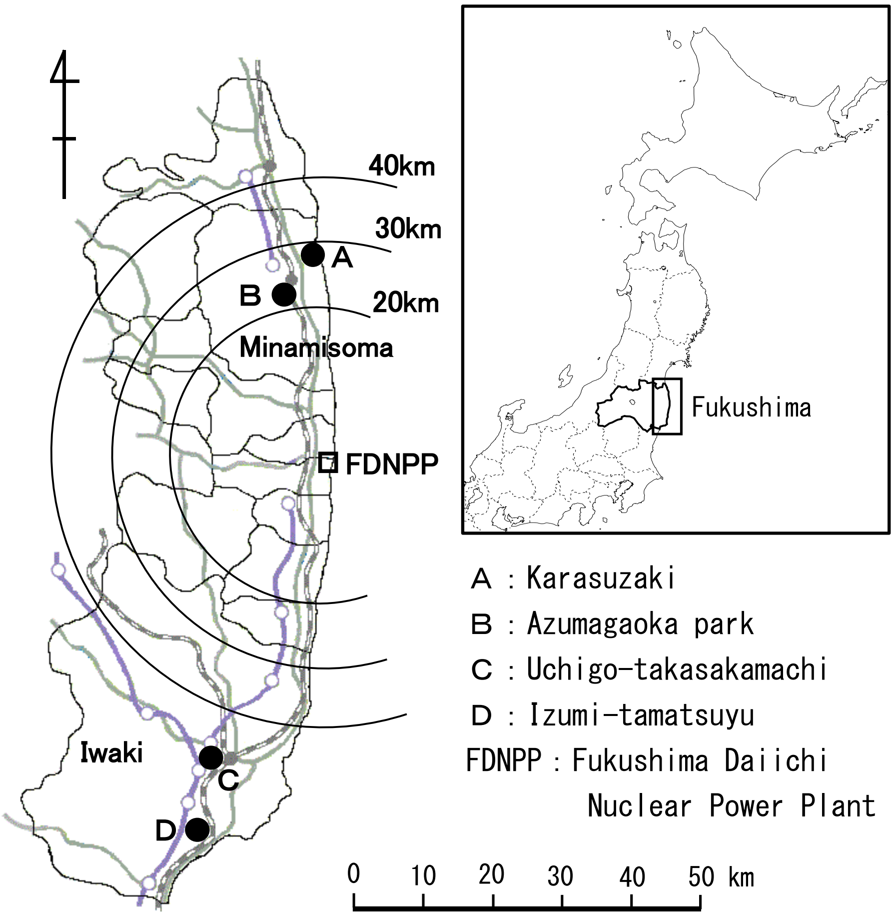

Fig. 1-1. Location map at Fukushima, Japan

A: Karasuzaki Kashima-ku Minamisoma city

B: Azumagaoka Park Haramachi-ku Minamisoma city

C: Uchigo-Takasakamachi Iwaki city

D: Izumi-Tamatsuyu Iwaki city

FDNPP: Fukushima Daiichi Nucear Power Plant

Chapter 2 Characteristics of microbial mats at Krasuzaki, Kashima-ku, Minamisoma city, Fukushima prefecture.

2-1 Background and purpose of this study

The study site is located in Karasuzaki, Kashima-ku, Minamisoma city in the northeastern part of Fukushima prefecture (Fig. 2-1). Due to the earthquake on March 11, 2011, the Pacific coastal area in the Tohoku region including Minamisoma city was hit by a giant tsunami, so that many paddy fields and housing areas were submerged in seawater. In Minamisoma city, the tsunami had a height of over 10 m. The tsunami had a height from 3 to 5 m in approximately one hour from the earthquake occurrence. Immediately after that, however, the highest tsunami rushed toward the coast at Karasuzaki. This highest tsunami realized gigantic crest of over 5 m even though it was far from the coast. By this highest tsunami, a location at 10.5 m altitude and approximately 800 m away from the coast was also submerged to a depth of even 1.5 m. Even two hours after the first attack of tsunami, its backwash was seen on the coast, and this brought about submergence damage for a long period of time (Tazaki et al., 2014a). Minamisoma city suffered submergence due to the tsunami up to approximately 3 km inland. Most areas along rivers were submerged up to 1 to 2 km inland (Geospatial Information Authority of Japan, 2011). Samples were collected belongs to Karasuzaki area with the widest submergence in Minamisoma city.

This sampling site is a flat land that was used as a paddy field, near the Karasuzaki fishing port, approximately 29 km to the north from the FDNPP. In this site, sand was observed as tsunami deposit in a wide area. White microbial mats were formed on the surface of the former paddy field, and puddles were also seen in dips in the vicinity. Although rubble was removed from there, its recovery the paddy field has not been achieved as of October 2014 and it is still a wasteland.

I studied the microbial mat formation environment and an elemental concentration action and the possibility of adsorption and immobilization of radionuclides by microbial mats in Karasuzaki area. The microbial mat formation process and their sustainability were examined through continuous observation.

2-2 Topography of investigation place and a geological feature

A study area was the flat land used as paddy field near the Karasuzaki fishing port established around the estuary on Mano-gawa River. There is the sedimentation area of a gravel, sand, and mud of the Holocene epoch in the Quaternary period in this area. The beach ridges were formed on the part that an alluvial plain faces the sea, and small lagoons and the wetlands were formed behind beach ridge (Yanagisawa et al., 1996; Kubo et al., 1990). Karasuzaki fishing port is a kind of the lagoon, and study area was a marsh from a geographic characteristic. In the study area, silt and clay, and mixes sand distributed. The sea sands of the tsunami deposit covered surface of the paddy field after the 3.11 Earthquake in Karasuzaki.

2-3 Materials and methods

2-3-1 Sample collection

Microbial mats samples collected on September 15, 2012 and October 14, 2013. Samples of microbial mats were divided in surface part and sands of reverse side by cutter knife, and each part were investigates by using various techniques.

About the water, water temperature, pH, electric conductivity (EC), and oxidation-reduction potential (ORP) in field were measured by using HORIBA B-211 Compact pH Meter (pH meter), HORIBA B-173 Compact Conductivity Meter (EC meter), and CUSTOM ORP6041 (ORP meter) on site, respectively. Water samples collected in polyethylene bottles, and were performed chemical analyses of water in the laboratory. The standard hydrogen electrode potential (Eh) was calculated from the oxidation-reduction potential (ORP) by the following relation.

$$Eh\ (mV) = ORP + 206 - 0.7 \times (t - 25)$$

t : water temperature (°C)

Air radiation dose was measured at height of 1m from a ground level. Radiation dose of microbial mats were measured by sample contact technique on site. Measurements of β (γ)-ray radiation used ALOKA GM survey meter TGS-136 (cpm), and measurements of γ-ray + X-ray radiation used ECOTEST TERRA-P+ (μSv/h) were performed. At GM survey meter, 1000cpm was equivalent to 1.52μSv/h, and there was an error of ± 20 to 30 %. At TERRA-P, there were ± (25+2/10H) of error.

2-3-2 Chemical analyses of water

Inorganic ion ingredient (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}) and EC(mS/m) of water samples were analyzed. Chemical analyses of water were performed using an ion chromatograph (Metrom, 883Basic IC Plus). Chemical analyses of water samples were performed by an official analysis based on JIS K 0102 or a water supply test method. Ion chromatograph was developed for the analysis of the inorganic ion and it was used in many environment-related fields, (Goto, 2011). Chemical analyses of water were performed by NSS Co., Ltd. (Tsubame city, Niigata prefecture).

2-3-3 Radionuclide analysis by the Ge semiconductor detector

The radionuclides of microbial mats and sand samples were analyzed on June 5, 2013 and October 19, 2013 by Ge semiconductor (CANBERRA GC2520) and ^{131}I, ^{40}K, and ^{134}Cs, ^{137}Cs (Bq/Kg-dry) were detected. Radionuclide analyses were performed by Yamato Environmental Analysis Co., Ltd. (Kawakita town, Ishikawa prefecture).

2-3-4 X-ray powder diffraction measurement (XRD)

XRD measurements for samples were performed using RIGAKU RINT 2200 with Cu Kα radiation. The $\theta/2\theta$ scanning technique was adopted over the $2\theta = 2 - 60^\circ$ with a scan step of 0.02°. The applied acceleration voltage and current were 40 kV and 30 mA, respectively.

2-3-5 Wavelength dispersive X-ray flourescence analysis (WD-XRF)

Chemical compositions of the samples were determined by wavelength dispersive X-ray fluorescence analyzer (WD-XRF ; SHIMADZU XRF1700) using Rh-Kα ray, which operated at an accelerating voltage of 30kV, under vacuum condition, and FP bulk qualitative and sem-quantitative analysis technique were used. WD-XRF analysis were performed by Yamato Environmental Analysis Co., Ltd. (Kawakita town, Ishikawa prefecture).

2-3-6 Scanning electron microscopy and energy dispersive X-ray analysis (SEM-EDX)

The scanning electron microscopy equipped with energy dispersive X-ray analyzer (SEM-EDX ; HITACHI S-3400N and HORIBA, EMAX X-act) was used for micro morphological observations, semi-quantitative analyses, and elemental content maps of the samples under 15kV accelerating voltage and current 70-80μA, with an analytical time of 1,000 seconds, and an area of 10mm x 10mm on the carbon double tape with Pt coating. The energy dispersive X-ray analyzer (HORIBA, EMAX X-act) used in this study could be analyzed even small amount of element and displayed on the elemental content maps. Even in the cases that samples include unexpected elements and microparticles, the apparatus can distinguish them (HORIBA, 2010; Burgess et al, 2013; Shozugawa, 2014). The semi-quantitative analyses were performed. The peak separation software was used for analyses of coexist elements with similar fluorescence energies, and a value of zero is indicated in the case that lists of fluorescence energy are overlapped.

2-4 Results

2-4-1 Characteristics of sampling site

On the sampling on September 15, 2013, a white sheet-like substance thinly covering the surface of the tsunami deposit was observed, and puddles were also formed in the places (Fig. 2-3A). The electric conductivity (EC) of the puddles exceeded the measuring range of the EC meter, therefore the measurement was impossible in the field(Table2-1). This result showed the water of a puddles were influenced of the tsunami so to be brackish.

Sheet-like white, gray, or black substances (microbial mats) covered the surface of soil in the paddy field to a depth of several millimeters. In a dried place, they showed cracks and some parts peeled up, which could be easily peeled away (Fig. 2-3B). When oxydol was put on a removed white microbial mat, it intensely bubbled to indicate the presence of microorganisms and an organic substance. On the black or reddish brown backside, its black part was rapidly oxidized to turn brown (Fig.2-3C). The dried white microbial mats could be easily peeled off the ground surface. They were elastic and could not be easily teared even when bent (Fig.2-3D). On September 15, 2012, the air dose was 0.44 μS/h and the dose on the microbial mat surface was 0.6 μS/h (Table2-2).

Several observations suggested that the former paddy field repeated drying and moisturization and increased cracks and peeled-up parts on the white microbial mats. At that point, the air dose was 100 to 170 cpm (0.10 μSv/h) and the dose on the microbial mat surface was 85 to 140 cpm (0.13 to 0.16 μSv/h). In contrast, the dose of the sand under the microbial mats was 92 cpm (0.13 μSv/h), which was lower than that of the surface. At a depth of 10 cm, the dose decreased down to 50 to 60 cpm (0.13 μSv/h) (Table2-2). In 2014, the old dried microbial mats further peeled up and new microbial mats were formed in the crevice including the moisture.

In radiation dose measurement at the site, the dose measured on the microbial mat surface indicated a somewhat higher value than the air dose and the dose of the sand under the microbial mats. When the doses of samples obtained on October 14, 2013 were also measured in Kanazawa city on October 16, 2015, the microbial mat surface indicated 140 cpm and the sand under it indicated 100 cpm while the air dose was 50 cpm. Besides, when the doses of samples obtained on January 11, 2014 were measured in on January 14, 2014, the microbial mat surface indicated 100 cpm and the sand under it indicated 70 cpm while the air dose was 70 to 80 cpm (Table 2-2). Both measurement results indicated somewhat higher doses of the microbial mat surfaces than others.

2-4-2 Water chemistry

It was found that much amount of Na^+, Cl^-, Mg^{2+}, SO_4^{2-} dissolved in the water by the chemical analyses of water, which could be attributed to sea water carried by the tsunami even 2.5 years after the 3.11 Earthquake. Because the water chemistry showed Na+-Cl- type (Table2-1). The measurement results in the field shwed that the water temperature was 21-22°C, pH 7.5-7.8, , oxidation-reduction potential ORP 78-88 mV, Eh 287-296 mV, salinity 2.3-3.1% and the electric conductivity (EC) measured in the laboratory was 3480-4520 mS/m (table 2-1).

2-4-3 Radionuclide analysis

Samples of white microbial mats and sea sand in Karasuzaki collected on September 15, 2012 and October 14, 2013, and the radionuclides of these samples were analyzed on June 5, 2013 and October 19, 2013.

Ge semiconductor analyses of microbial mats in Karasuzaki showed the radiations of ^{134}Cs (400 Bq/Kg-dry), ^{137}Cs (890 Bq/Kg-dry) and traces radiation of ^{40}K (140 Bq/Kg) and sea sand showed the radiations of ^{134}Cs (340 Bq/Kg-dry), ^{137}Cs (810 Bq/Kg-dry) and traces rasiation of ^{40}K (230 Bq/Kg). The isotope ^{131}I was not detected in microbial mats and sand, because the half-life time of ^{131}I is reratively short (8.04 days) (Table2-2). The ratio of ^{134}Cs/^{137}Cs of microbial mats was about 1/2 at 2.5 years after the FDNPP accident. This fact is consistent with the half-life time of ^{134}Cs (2.1 years). The ratio of ^{134}Cs/^{137}Cs just after the FDNPP accident was approximately 1.0 (Kawada et al., 2012), and it is harmonized with the result of Ge semiconductor analyses. The radiations of ^{134}Cs and ^{137}Cs for microbial mats was higher than those of sea sand. Radiation dose of the samples was measured for GM survey meter in Kanazawa, detected radiation on microbial mats surface was 140cpm, that at the sand of reverse side of the microbial mats was100cpm, and the air dose was50cpm on October 16, 2013.

The microorganisms concentrates the several elements and mineralizes in the cell. There is a kind of microorganisms which concentrates uranium, and the uptake of uranium is not performed through the metabolism of the living body, and most is physicochemical adsorption to a cell substance (Sakaguchi, 1996). Tazaki, 2013 and Tazaki et al., 2013 shows the fact that the radionuclide deposited and was fixed on the diatom and foraminifer with structure that the specific surface area is wide, and many holes. Ge semiconductor analyses results shows that a radionuclide concentrated in microbial mats.

In addition, Ge semiconductor analyses of microbial mats at the same point detected radiation of ^{134}Cs (2,000-2,200 Bq/Kg-dry), ^{137}Cs (4200-4300 Bq/Kg-dry) on June 5, 2013 (Tazaki et al., 2014a, b). The result of October 19, 2013 analysis showed 1/5 in comparison with the result of June 5, 2013 analysis.

2-4-4 XRD analysis

The obtained XRD data for the microbial mats are shown in Fig.2-3A and B. The results show that the sample contains both minerals and amorphous materials. The microbial mats surface data show the broad peak of amorphous background at 15-30 degree. The microbial mats also contained minerals, such as halite (d = 2.82Å, 1.99Å and 1.63Å), gypsum (d = 7.63Å, 4.28Å, 3.07Å, 2.87Å and 2.68Å) and quartz (d = 3.34Å) (Fig.2-3A).

On the other hand, the data indicate that the sand of reverse side of the microbial mats contains mainly minerals in the sandy paddy field, quartz (d = 4.26Å, 3.34Å and 1.82Å), feldspars (d = 3.35Å and 3.18Å), and halite (d = 2.82Å and 1.99Å) associated with trace of 7 Å clay minerals without high background (Fig.2-3B).

Halite were recognized at both of microbial mats and sand of the tsunami deposits. This fact may indicate these materials had influence of the seawater. However, minerals contained in these two samples were slightly different.

2-4-5 WD-XRF analysis

The chemical compositions of microbial mats surface and sea sand collected in Karasuzaki was analyzed by WD-XRF analyzer (Table2-3). Na and Cl are major elements of these samples. The origin of these element is sea water carried by the tsunami.

Na (6.88 wt%) and Cl (13.5wt%) were detected in microbial mats sample. On the other hand, the sand sample included 6.88wt% of Na and 8.31wt% of Cl. The quantities of Na and Cl in the microbial mats were higher than those of sand. Mg, P, and Br are included in the microbial mats and sand samples. Mg, P, and Br are also the major ingredients of sea water. Their contents for the microbial mats were higher than those for sand sample.

2-4-6 Scanning electron microscopy

In the SEM observation of the surface of white microbial mat and the sea sand directly, the outermost surface of the white microbial mats were covered at *Nitzschia* sp., and the other types of diatom were not found in microbial mats (Fig.2-3A). Its shell has a linear lanceolate shape, and the shell length is about 15-50 μm of a medium-sized species. The shell width is about 4-5 μm, and a medium part between plates is strong, and its spot mark is also large.

The *Nitzschia* sp. is a common ragweed widely distributed in freshwater such as lakes and rivers. They have a resistance up to medium rotten water (Kojima et al. Ed., 1995). Both *Nitzschia* filiformis and *Nitzschia* scalpeliformis in this genus are widely distributed from fresh water to seawater, but they are epiphytic species often found in brackish waters such as estuarine region and they have a tolerance up to medium rot waters in brackish water (Kojima et al. Ed., 1995). In addition, the water chemistry in Karasuzaki on October 14, 2013 is pH(7.5-7.7), EC(3480-4520 mS/m), ORP(78-88 mV), (Eh 287-296 mV), water temperature(21-22℃) and a brackish water environment with salinity(2-3%), meeting almost the habitat environment of this genus.

In the white microbial mat, it was confirmed that *Cyanophyceae, Nostocales* and *Anabaena genera* were deposited on the surface of the *Nitzschia* genus in a mesh-like shape (Fig.2-3B). This genus appears in eutrophic lakes and ponds, sometimes causes outbreak to form "algal water blooms" and distributed in the wide temperate ranges. Among these genera, many lumps of *Anabaena circinalis* and *Anabaena flosaquae* forma flosaquae irregularly intertwine with each other to lead the floating life. Between the diatom shells, fine mineral particles and inorganic materials with particle size of 30-50 μm are scattered (Fig2-3C, D). On the other hand, in the sea sand just beneath the microbial mat, fine particle mineral, clay mineral and inorganic material are often confirmed, but microorganisms such as a diatom are scarce (Fig2-3E, F).

2-4-7 SEM-EDX analysis

The chemical composition of microbial mats in Karasuzaki obtained by semi-quantitative analyses of SEM-EDX analyses (Table2-3).

The Si abundant detected in the microbial mats sample was 19.45-26.62wt%, and the other elements such as Mg,Al,Cl,Ca,S and Zn were also detected in the sample. Furthermore, it was suggested a possibility that trace radionuclides of Th and U were included in the microbial mats (Table2-3).

It was found that Si derives from diatoms by SEM observation and an elemental analysis. It was also found that calcium sulfate and sodium chloride derived from seawater (tsunami) were formed to halite and gypsum by the microorganisms from XRD and elemental analysis.

The elemental content maps of diatom and the inorganic particle show the Si were mainly included in the diatom part. Mg, Al and Fe were detected in the inorganic particle, and it is considered from clay minerals or feldspars. Ca thoughted to be gypsum was detected on the map lower right part of the sample (Fig2-4). The elemental content maps of diatom and the micrococcus show that Si were detected in the diatom part, and the dotted distribution at outskirts part may be Cl of seawater origin (Fig.2-5). The trace elements such as Sr were detected in diatom, and it is suggested that trace elements are collected by diatom (Fig.2-4, 2-5).

2-5 Discussion

2-5-1 Biomineralization

In the present study, it was found that the white microbial mats formed in the paddy field were mainly made up of *Nitzschia* sp., and tsunami-derived halite and gypsum were concentrated in the microbial mats. Besides, the results of radionuclide analysis suggested a possibility that the microbial mats adsorb radioactive materials by the involvement of microorganisms. The concentration of halite and gypsum was also seen on the surface of a paddy field damaged by the tsunami in the coastal area of Miyagi prefecture, and halite and gypsum was formed by ion exchange and the chemical interactions of soil and seawater (Kokubun et al., 2013 ; Nanzyo, 2012). In this study, halite and gypsum were formed on the white microbial mat at the tsunami deposit surface in Karasuzaki. Comparing the XRD results of the microbial mats and the sand of tsunami deposit, gypsum was not confirmed with sand. In XRF analysis, the elements constituting halite (Na, Cl) are abundant at the microbial mats compared with the sand of tsunami deposits, and Ca included in gypsum is a little in the microbial mats, but S is abundant. It is suggested that gypsum is formed abundantly in microbial mats. These results show that a microorganism participates in formation of the minerals.

2-5-2 Radioactive materials and microbial mats

By the combined use of an analytical electron microscope, Ge semiconductor detector, and GM survey meter, the present study revealed the presence and distribution of long-half-time radionuclides in the white microbial mats. However, after two years and six months from the nuclear plant accident, radionuclides of iodine and potassium from the FDNPP were hardly detected. The doses of ^{134}Cs and ^{137}Cs were higher on the white microbial mats than those of sea sand. This indicates the fact that radionuclides are deposited and immobilized on porous diatoms with a greater specific surface area to be concentrated. Another radionuclide analysis was conducted at the same location on June 5, 2013 (Tazaki et al., 2014a). Comparison of both analysis results indicates that the doses of ^{134}Cs and ^{137}Cs both decreased to approximately 20%. The physical half-life of ^{134}Cs is 2.06 years and that of ^{137}Cs is 30.3 years. Therefore, the results obtained in this study do not correspond to the physical half-lives. This difference is considered to be attributed to adsorption, absorption, immobilization, and mineralization by microorganisms, which suggests a possibility that absorbing and immobilizing radioactive materials inside microorganisms and shielding them by a mineralization allow prevention of diffusion of the radioactive materials. It is reported that the ambient dose decreased when paddy field soil contaminated by radioactive materials was covered with diatom earth produced in Noto (Tazaki et al., 2015b). There also shown the fact that porous diatom earth with a large surface area has a high radiation shielding effect (Odano et al., 2012). Diatom earth thus has a radiation shielding ability, and this suggests that microbial mats made up of diatoms also shield radioactive radioactivity of materials.

XRF chemical composition analysis performed on the topmost white microbial mat and sea sand under it. The microbial mat contained higher amounts of elements affected by tsunami than the sea sand. The weight loss of the microbial mat by heat treatment is greater than the sea sand. This difference depends on the difference in the organic substance contents and microbe contents and also indicates that seawater components were concentrated by microorganisms. The presence of Sr in microbial mats is confirmed by a XRF analysis, and it is shown that Sr is concentrating on a diatom from the results of the elemental content maps. This indicates that Sr is deposited and immobilized on porous diatoms with a greater specific surface area to be concentrated.

2-5-3 Living environment of microorganisms

Even two and a half years after the tsunami, the water puddles of the paddy fields in Karasuzaki still shows EC 3480-4520mS/m, and salt concentration of 2.3-3.1%. Also, the agricultural soils of Miyagi prefecture that were hit by the tsunami had EC of 200-2900mS/m in April 2011, and 100-1410mS/m in June 2011, and standing water in the drainage had 2700mS/m. On the other hand, the agricultural soils that were not hit by the tsunami had EC of about 8mS/m (Kitagawa et al., 2012). EC of the agricultural fields in Soma city Fukushima prefecture was 1200-2400mS/m, with the content of the sodium chloride reached 2.9-5.7%, and some tsunami deposits contained about twice as much of sodium chloride of the sea water (3%) (Goto et al., 2011). The result of EC measurement in Karasuzaki shows 1.2-1.6 times as much of that of the standing water within Miyagi prefecture after 3 months, and it has been revealed that the influence of tsunami is still strong in the area. Foraminifers of species that is characteristic in the closed-off sections of inner bays with a greater salinity variation were observed in the sand that is tsunami deposit. These foraminifers are a species living in a different environment from outer sea. Therefore, most of the foraminifers are considered to have not been carried by the tsunami but have newly grown in a brackish environment created with tsunami seawater left in the paddy field (Nemoto et al., 2014). Further, foraminifers were observed there even after four years and a half from the tsunami occurrence. This demonstrated that foraminifers repeated the alternative generations in Karasuzaki (Nemoto et al., 2015). On the other hand, an electron micrograph of diatoms of multiple species in a paddy field with no tsunami damages in Baba, Haramachi-ku, Minamisoma city was reported (Tazaki et al., 2013a). Besides, an electron micrograph of diatoms of multiple species attached to the fibers of a petroleum product sunk in an agricultural waterway in Baba, Minamisoma city was reported (Tazaki et al., 2015a). Thus, diatoms of multiple species should multiply proliferously in nature under normal circumstances. However, in the microbial mats in Karasuzaki, *Nitzschia* sp. was the majority while other species were observed marginally. This fact could be considered as follows. In Karasuzaki, due to the tsunami, only species resistant or adapted to seawater in native microorganisms living in the paddy field multiplied proliferously to form the white microbial mats. Further, this is backed up by the fact that environmentally-adapted species in foraminifers carried by the tsunami also newly grew there. These results may become the index on estimating environment and the paleoenvironment before and after the tsunami event in a tsunami deposit distribution area.

5-3-4 Formaition of microbial mats

As a result of continuous observation in the study site, it suggested that biofilms formed on oil slicks in puddles left after the tsunami, and they were dried to form white microbial mats with a thickness from 2 to 3 mm duaring one year. Tsunami-derived halite and gypsum were observed in the white microbial mats, and also radionuclides absorbed in them were also observed. The soil in the former paddy field is composed of an upper layer of tsunami-derived sand and a lower layer of aggregated black clay in the reduced state. Two years after that, the white microbial mats had cracks due to drying and peeled up in places. Three years later, new microbial mats and green microbial mats mainly made up of green algae were formed between the peeled-up old microbial mats (Fig. 2-6). This demonstrated that even though drying thus hindered the formation of the microbial mats, re-moisturization allowed the sustainable formation of the microbial mats. Besides, this indicates a possibility that the sustainable formation of the microbial mats allows re-concentrating tsunami-derived halite, gypsum, and radionuclides contained in the soil.

It seems that environmentally-adapted limited species of native microorganisms multiplied proliferously due to the water chemistry environment change caused by the tsunami to form the microbial mats to concentrate specific minerals and elements. Besides, the elemental content maps indicate a possibility that trace elements and radionuclides are adsorbed and immobilized by the microorganisms that form the microbial mats.

2-6 Conclusion

Chemical analyses of water on the puddles in the study site showed that they were a brackish environment with salt concentrations from 2 to 3%, and they are still under the influence of the tsunami.

In the former paddy field affected by the tsunami, the white microbial mats mainly made up of diatoms were formed and tsunami-derived halite and gypsum were concentrated in the microbial mats by biomineralization.

The radionuclide analysis revealed that ^{134}Cs and ^{137}Cs were concentrated in the white microbial mats. This suggests that because of their wide specific surface areas, radionuclides were deposited and immobilized on porous diatoms.

The radionuclides in the white microbial mats decreased to about 20% during one year, and this suggests that radiation was shielded by the diatoms that formed the microbial mats.

Even if the microbial mats are dried, re-moisturization by rain, snow, or the like allows their sustainable formation. This suggests a possibility that the sustainable microbial mat formation allows re-concentrating tsunami-derived halite, gypsum, and containing radionuclides in the soil.

The study site was a freshwater environment before the earthquake because the site was used as a paddy field. Due to a water chemistry environment change caused by the tsunami, microorganisms resistant or adapted to salt water limitedly multiplied proliferously to form the microbial mats. Environmentally-adapted species in foraminifers that were carried by the tsunami newly grew and repeated alternation of generations. These results may become the index on estimating environment and the paleoenvironment before and after the tsunami event in a tsunami deposit distribution area. These results may become the index on estimating environment and the paleoenvironment before and after the tsunami.

Table 2-1 Water chemistry in Karasuzaki

Sampling 2013/10/14

Point name		Karasuzaki No.1	Karasuzaki No.2
Microbial mats		Whaite microbial mats	Whaite microbial mats
Characteristics of the water		Na^{+}-Cl^{-} type	Na^{+}-Cl^{-} type
		Brackish water(Tsunami)	Brackish water(Tsunami)
Ion equivalent (meq/L)	Na^{+}	313.18	430.63
	K^{+}	6.39	8.18
	Ca^{2+}	14.47	19.96
	Mg^{2+}	71.59	98.75
	Cl^{-}	394.89	535.93
	HCO_3^{-}	2.29	2.29
	SO_4^{2-}	39.56	52.05
	NO_3^{-}	＞0.08	＞0.08
pH		7.5	7.7
EC(mS/m)	field	<410	<560
	Laboratory	3480	4520
ORP (mV)	E (field)	+ 78	+ 88
	Eh (Reduced value)	+ 287	+ 296
Oxidation-reduction state		Oxidative	Oxidative
Salinity (%)		2.3	3.1
Water Temperature (℃)		21	22

Table 2-2 Radiation dose in Karasuzaki

Date	Sample	Radiation dose	
		cpm	μSv/h
2012/9/15	Air dose		0.44
	Microbial mat surface		0.60
2013/10/14	Air dose	100-170	0.10
	Microbial mat surface	85	0.12
	Microbial mat surface	120	0.16
	Microbial mat surface	140	0.16
	Under microbial mat	92	0.13
	under 10㎝ (sand)	50-60	0.13
2013/10/16 (Kanazawa)	Air dose	50	
	Microbial mat	140	
	Sand	100	
2014/1/11	Air dose	120	0.13
	Microbial mat	160	0.13
	Microbial mat	120	0.13
	Sand	100-130	0.12
2014/1/14 (Kanazawa)	Air dose	70-80	
	Microbial mat surface	100	
	Microbial mat back	70	

Table.2-3 Ge semicondacture analyses of microbial mats and sand

This Table was quoted the part from Tazaki et al. (2014a),

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Unit (Bq/kg-dry)

Sampring date Analysis date	2012/9/15 2013/6/5		2013/10/14 2013/10/19	
Sample	White microbial mats surface	Sea sand underneath the white microbial	White microbial mats surface	Sea sand underneath the white microbial
I 131	N.D.	N.D.	N.D.	N.D.
Cs 134	2200	2500	400	340
Cs 137	4200	5500	890	810
K 40	N.D.	N.D.	140	230

N.D.; not detected

Table.2-4 WD-XRF analyses of White microbial mats and sea sand

Unit : wt(%)

element	White microbial mats	Brown sand present underneath the white microbial mats
Na	6.880	4.490
Mg	4.270	2.990
Al	13.700	17.100
Si	22.700	38.400
P	0.341	0.129
S	4.180	2.140
Cl	13.500	8.310
K	2.460	2.750
Ca	4.510	6.500
Ti	0.976	1.460
V	—	—
Mn	0.387	0.403
Fe	10.200	12.100
Co	—	—
Cr	0.030	0.066
Zn	0.042	0.036
Br	0.072	0.021
Rb	—	—
Sr	0.052	0.064
Zr	—	—
W	—	—
Ignition loss (600 ℃)	15.700	3.000

ND : not detected

Table.2-5 SEM-EDX analyses of White microbial mats

Unit : wt(%)

element		diaton and paddy soils Fig.2-4	diaton and bacteria Fig.2-5
Mg	K	2.12	3.03
Al	K	6.10	1.28
Si	K	26.62	19.45
S	K	0.51	2.95
Cl	K	2.32	9.05
K	K	1.59	0.47
Ca	K	0.70	1.57
Mn	K	0.15	0.00
Fe	K	11.16	2.00
Zn	L	2.04	9.97
Ag	L	0.00	0.00
I	L	0.18	0.19
Cs	L	0.00	0.00
Ba	L	0.92	0.23
Ce	L	0.00	0.14
Nd	L	0.12	0.00
Th	M	1.63	2.05
U	M	0.02	0.15
Np	M	1.01	0.93
Pu	M	0.00	0.08
O	K	42.79	43.22

0.00 : Analysis value shows zero

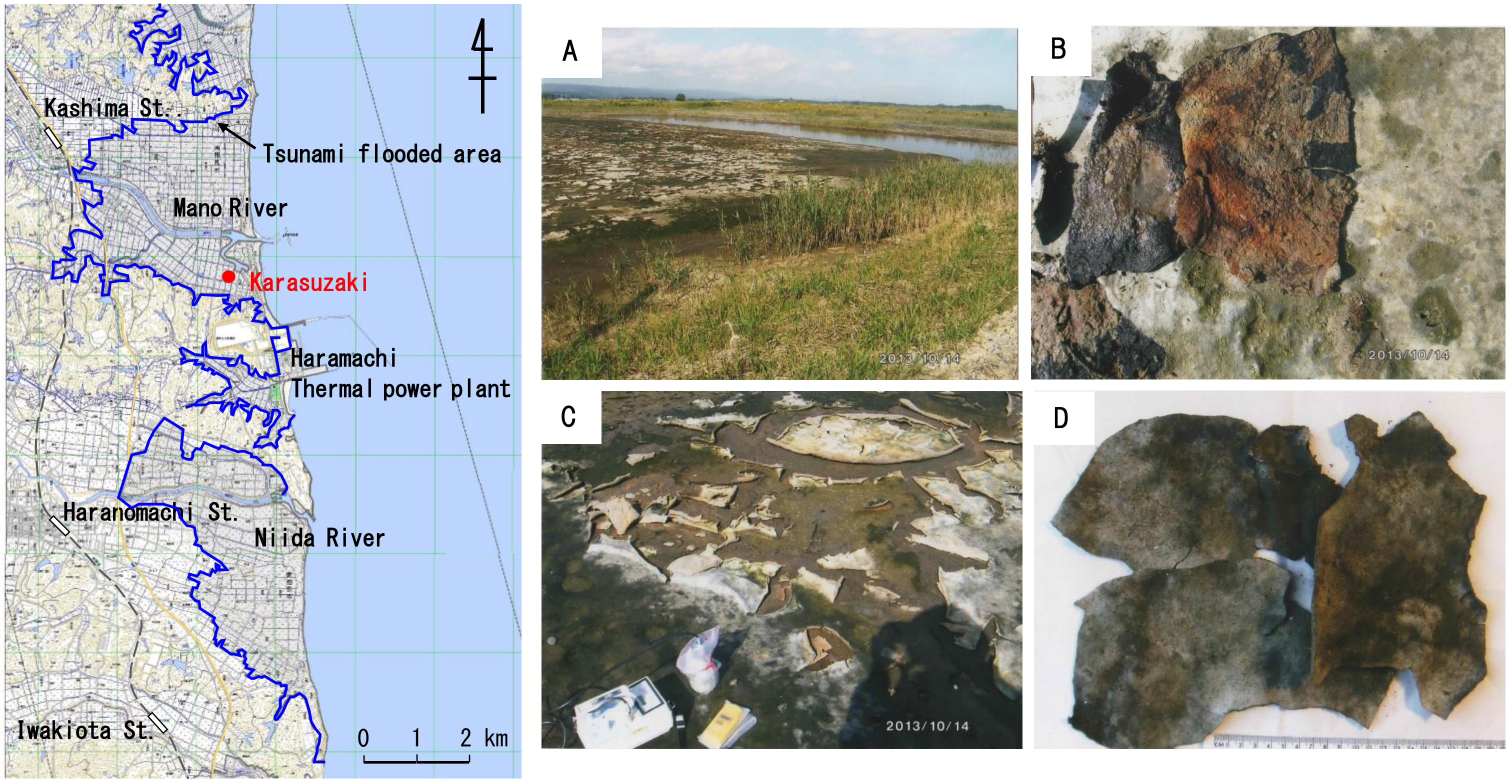

Fig.2-1. Location map of the white microbial mats were formd in paddy field in Karasuzaki, Kashima-ku, Minamisoma, Fukushima, Japan

A: The greater part of the paddy field has been affected by Tsunami flooding.

B: White thin microbial mats occurred on the top surface of paddy field.

C: Brown sea sand present underneath the white microbial mats.

D: The white microbial mats about 0.5cm thick.

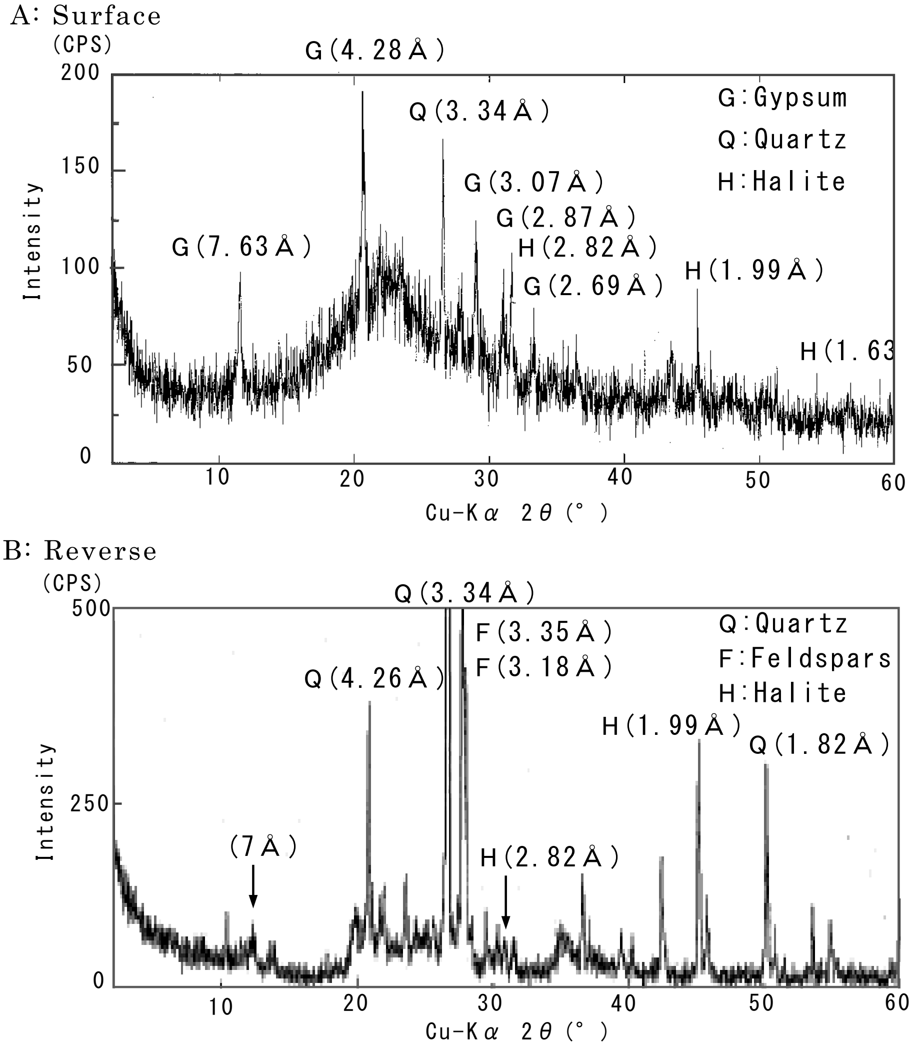

Fig. 2-2. X-ray diffraction pattern of the white microbial mats and sand.

A: Surface of the white microbial mats.

B: Sand present underneath the white microbial mats.

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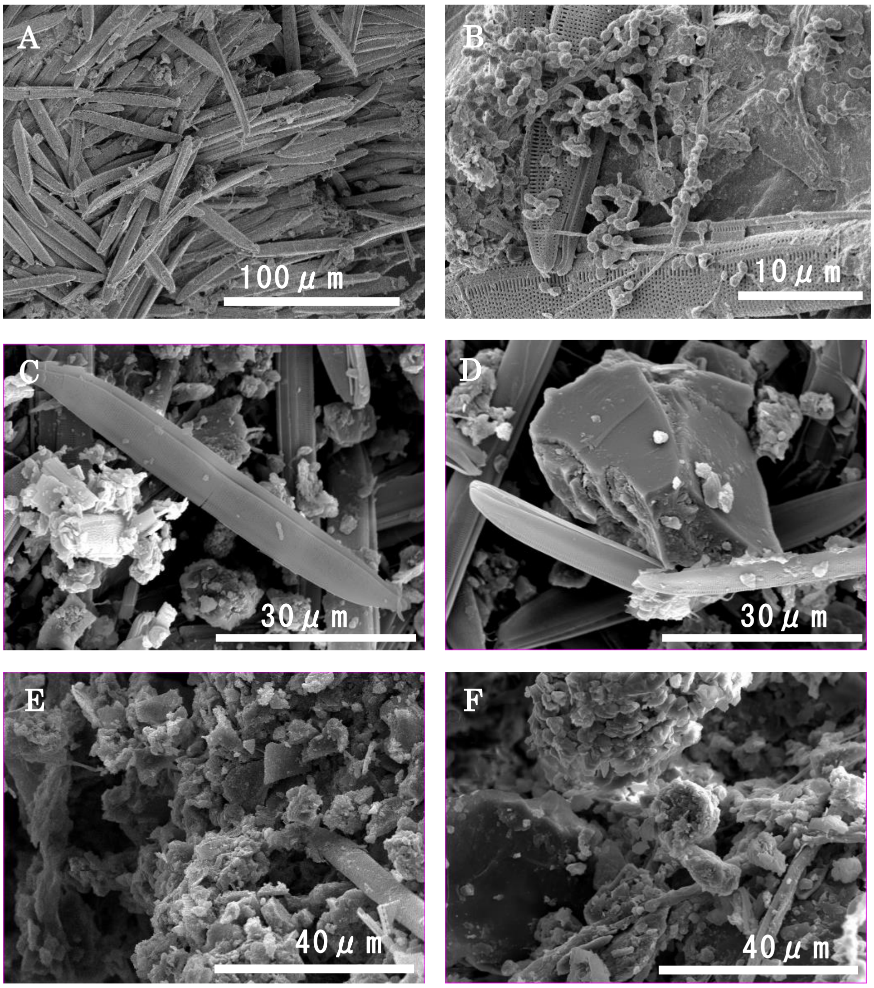

Fig.2-3 Scanning electron micrograph in Karasuzaki

A: The white microbial mats were collections of diatom

B:. Bacteria accepted in the white microbial mats

C: Inorganic materials and Fine mineral particles are scattered between the diatoms.

D: Inorganic materials and Fine mineral particles are scattered between the diatoms.

E: Fine particle mineral, clay mineral and inorganic material are confirmed in the sea sand beneath the microbial mat.

F: Fine particle mineral, clay mineral and inorganic material are confirmed in the sea sand beneath the microbial mat.

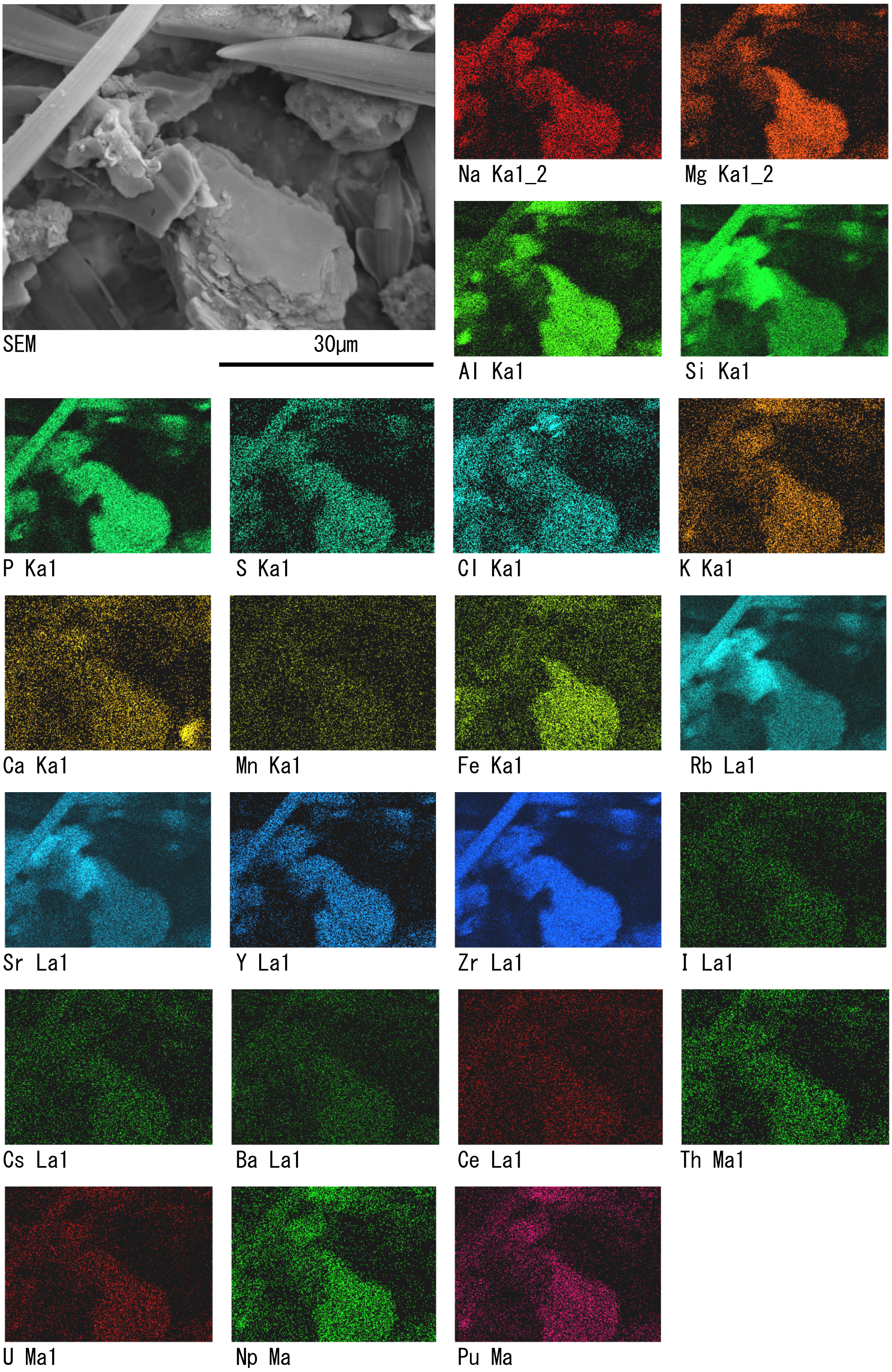

Fig.2-4 Scanning electron micrograph and elemental content maps of diatoms and paddy soils.

This figure was modified from Tazaki et al.(2015c), © Minerals 2015, 5, 849–862, http://www.mdpi.com/journal/minerals.

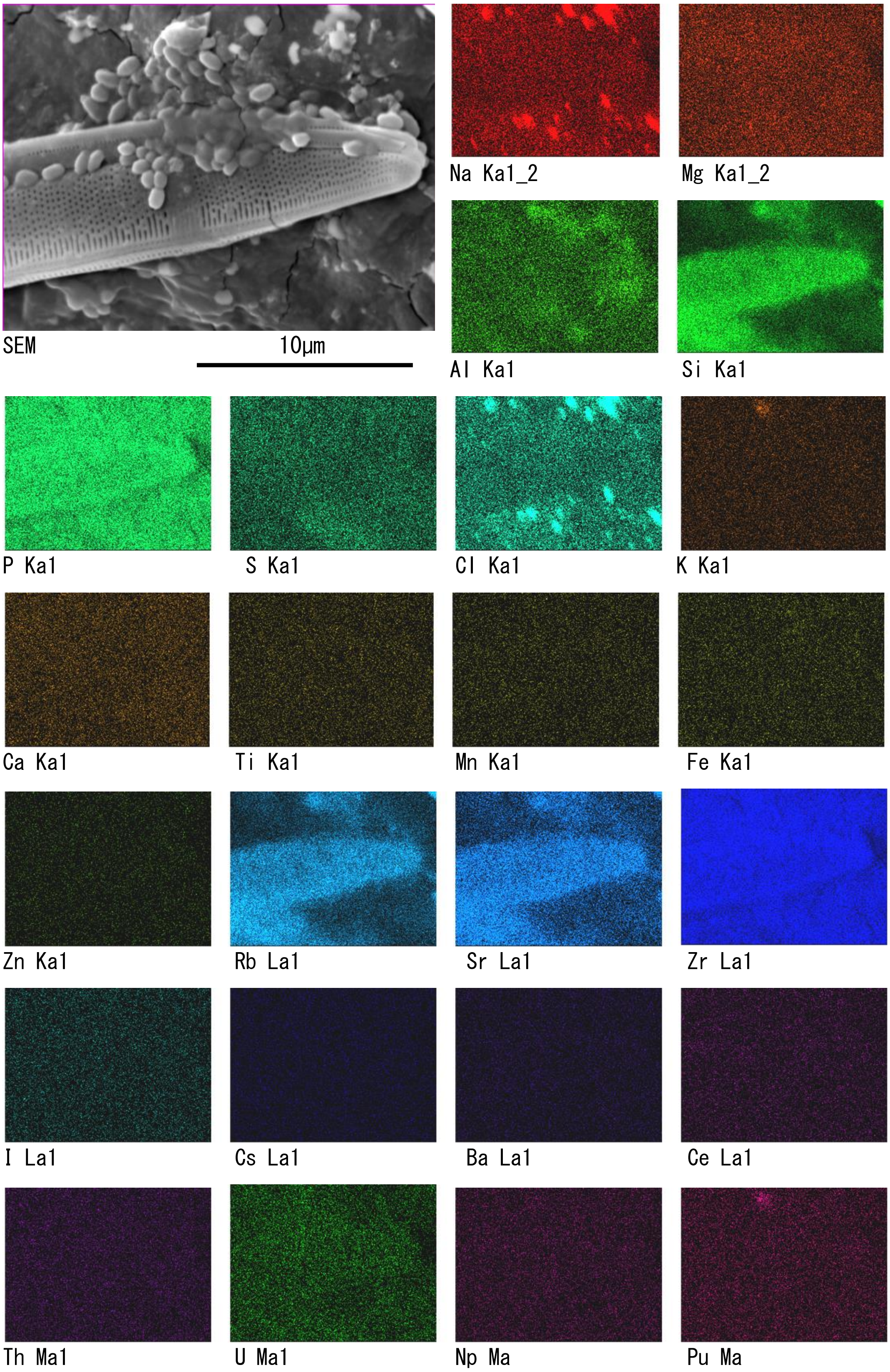

Fig.2-5 Scanning electron micrograph and elemental content maps of diatoms and bacteria.

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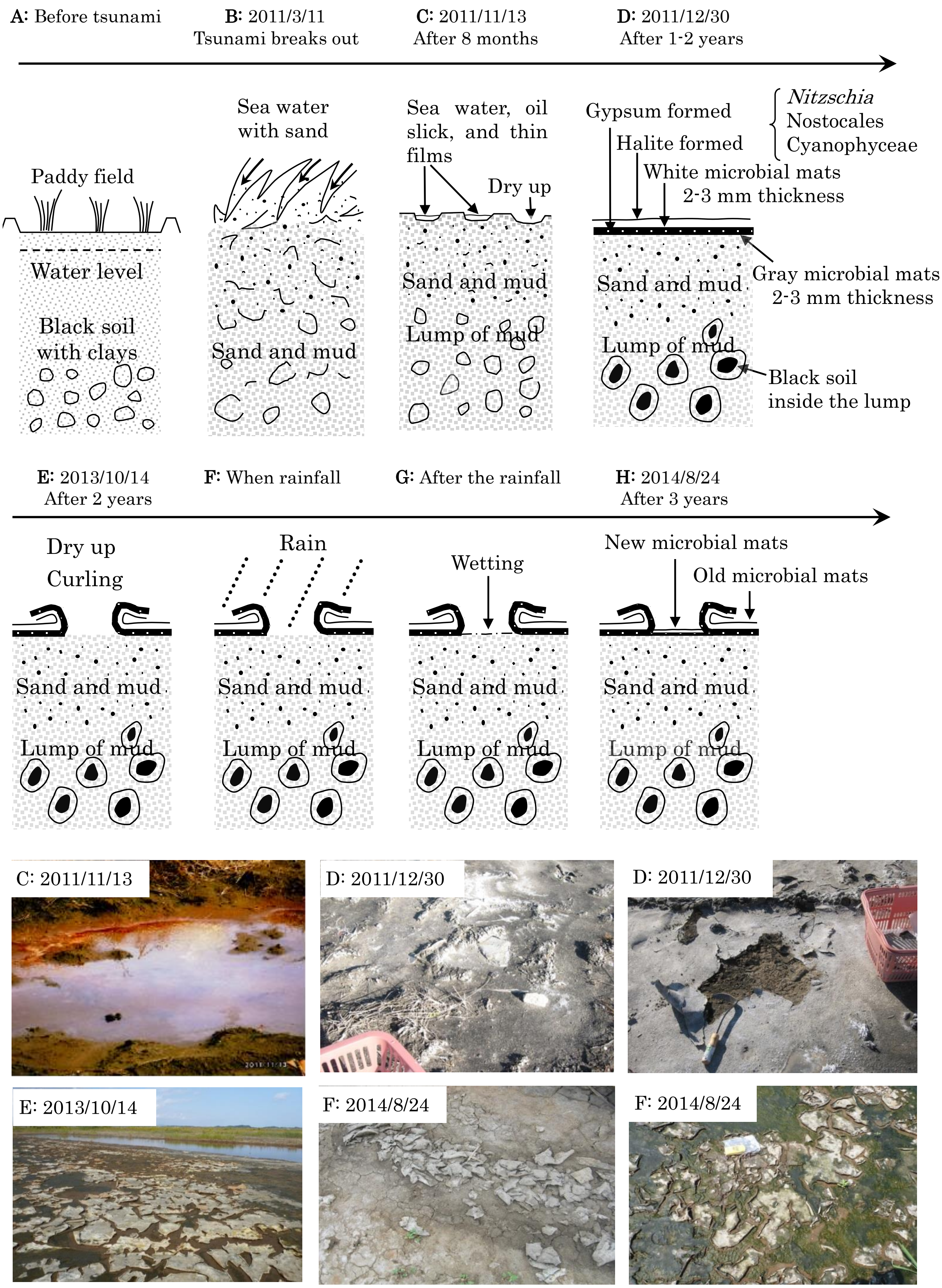

Fig.2-6 Formation processes of microbial mats in Karasuzaki

This figure was modified from Tazaki et al.(2015c),

Chapter 3 Characteristics of microbial mats in Azumagaoka park, Haramachi-ku, Minamisoma city, Fukushima prefecture.

3-1 Background and purpose of this study

The study site is located in Azumagaoka park, Haramachi-ku, Minamisoma city (Fig. 3-1). Azumagaoka park is a nature park located in the hilly area of Haramachi-ku, the center of Minamisoma city, and the park is approximately 25 km away from the FDNPP.

In the park, reddish brown microbial mats were formed in an earth canal where spring water flows (Fig. 3-1A, B). The water does not dry up even if it does not rain. The microbial mats were formed in water in a flocculated state (Fig. 3-1C). It was also possible to observe thin films like oil slicks on the water surface. The canal with the microbial mats is used in the treatment of spring water in the park. Although the canal is partially made of concrete blocks, most of it is an earth canal. When the spring water started to gush and when the microbial mats started to form were unknown. However, they seem to have been there before the earthquake on March 11, 2011. I studied the elemental concentration action that microbial mats perform in an environment contaminated by radioactivity due to the accident of the FDNPP.

3-2 Topography of investigation place and a geological feature

Azumagaoka Park is located in Haramachi-ku, the center of Minamisoma city, and occupies a hilly area as a green space. In the hilly area, mudstones in the Pliocene Epoch of the Neogene Period are distributed as bedrock. Distributed in the highest part of the hilly area are gravel, sand, and mud that are terrace deposit formed in the Pleistocene Epoch of the Quaternary Period (Kubo et al., 1990).

The spring water is seen in the higher part of the hilly area. The microbial mats are formed in the canal for the treatment of surface stream water and spring water, where water always flows slowly. Although the canal is partially made of concrete, distributed on the bottom of the canal is soft clay containing gravel and sand. Since the spring water is seen in the higher part of the hilly area, presumably, water penetrated the terrace deposit and gushed out from the boundary between mudstones that form an aquiclude.

3-3 Materials and methods

3-3-1 Sample collection

Microbial mats samples collected on January 11, 2014. Samples of microbial mats collected by stainless steel net and scoop. Clay in the underside of the microbial mats were also collected in the canal by using a plastic pipe, to compare with microbial mats samples.

About the water, water temperature, pH, electric conductivity (EC), and oxidation-reduction potential (ORP) in field were measured by using HORIBA B-211 Compact pH Meter (pH meter), HORIBA B-173 Compact Conductivity Meter (EC meter), and CUSTOM ORP6041 (ORP meter) on site, respectively. Water samples were collected in polyethylene bottles, and were performed chemical analyses of water in the laboratory. The standard hydrogen electrode potential (Eh) was calculated from the oxidation-reduction potential (ORP) by the following relation.

$$Eh\ (mV) = ORP + 206 - 0.7 \times (t - 25)$$

t : water temperature (°C)

Air radiation dose was measured at height of 1m from a ground level. Radiation dose of microbial mats were measured by sample contact technique on site. Measurements of β (γ)-ray radiation used ALOKA GM survey meter TGS-136 (cpm), and measurements of γ-ray + X-ray radiation used ECOTEST TERRA-P+ (μSv/h) were performed. At GM survey meter, 1000cpm was equivalent to 1.52μSv/h, and there was an error of ± 20 to 30 %. At TERRA-P, there were ± (25+2/10H) of error.

3-3-2 Chemical analyses of water

Inorganic ion ingredient (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-})and iron ion, manganese ion of water samples were analyzed. Chemical analyses of water were performed using an ion chromatograph (Metrom, 883Basic IC Plus). Chemical analyses of water samples were performed by an official analysis based on JIS K 0102 or a water supply test method. Ion chromatography was developed for the analysis of the inorganic ion and it was used in many environment-related fields, (Goto, 2011). Chemical analyses of water were performed by NSS Co., Ltd. (Tsubame city, Niigata prefecture).

3-3-3 X-ray powder diffraction measurement(XRD)

XRD measurements for samples were performed using RIGAKU RINT 2200 with Cu Kα radiation. The θ/2θ scanning technique was adopted over the 2θ = 2 - 60°

with a scan step of 0.02°. The applied acceleration voltage and current were 40 kV and 30 mA, respectively.

3-3-4 Wavelength dispersive X-ray flourescence analysis (WD-XRF)

Chemical compositions of the samples were determined by wavelength dispersive X-ray fluorescence analyzer (WD-XRF ; RIGAKU ZSX100e) using Rh-Kα ray, which operated at an accelerating voltage of 30kV, under vacuum condition, and FP bulk qualitative and sem-quantitative analysis technique were used. WD-XRF analysis were performed by Yamato Environmental Analysis Co., Ltd. (Kawakita town, Ishikawa prefecture).

3-3-5 Scanning electron microscopy and energy dispersive X-ray analysis (SEM-EDX)

The scanning electron microscopy equipped with energy dispersive X-ray analyzer (SEM-EDX ; HITACHI S-3400N and HORIBA, EMAX X-act) was used for micro morphological observations, semi-quantitative analyses, and elemental content maps of the samples under 15kV accelerating voltage and current 70-80μA, with an analytical time of 1,000 seconds, and an area of 10mm x 10mm on the carbon double tape with Pt coating. The energy dispersive X-ray analyzer (HORIBA, EMAX X-act) used in this study could be analyzed even small amount of element and displayed on the elemental content maps. Even in the cases that samples include unexpected elements and microparticles, the apparatus can distinguish them (HORIBA, 2010; Burgess et al, 2013; Shozugawa, 2014). The semi-quantitative analyses were performed. The peak separation software was used for analyses of coexist elements with similar fluorescence energies, and a value of zero is indicated in the case that lists of fluorescence energy are overlapped.

3-4 Results

3-4-1 Water chemistry and radiation measurement

The water chemistry showed Ca^{2+}-HCO_3^- type, that was water chemistry general in springwater. The dissolved amount of iron ion shows 24.0 mg/L by the chemical analyses of water. There was measurement in the field of the water temperature 7.5°C, pH 6.9, EC 36 mS/m, ORP 22 mV (in the field), Eh 240 mV (Table3-1).

Radiation dose of samples was measured in Azumagaoka park, radiation value detected on microbial mats was (200-300cpm, 0.33 μSv/h), the moss around canals (200cpm, 0.38 μSv/h), and air dose (110-160cpm, 0.14-0.16 μSv/h) on January 11,

2014. Radiation dose of samples was measured in Kanazawa, detected radiation microbial mats (80cpm) and air dose (70-80cpm) on January 14, 2014(Table3-2).

3-4-2 XRD analysis

The mineralogy of the reddish brown microbial mats, as determined using XRD analysis, contains amorphous materials. The reddish brown microbial mats indicated that the broad amorphous background peak is located at d = 3Å and 2 Å (Fig.3-3A). On the other hand, the clay underneath the reddish brown microbial mats contains mainly minerals, indicated quartz (d = 3.34 Å), feldspars (d = 3.25 Å) and clay minerals (d = 7 Å and 14 Å) (Fig.2-3B).

3-4-3 WD-XRF analysis

The chemical composition of microbial mats and clay collected in Azumagaoka park were analyzed by WD-XRF analyses (Table3-3).

The Fe of 50.00wt% was detected in the microbial mats sample, the other hand the Fe of 4.20wt% was detected in the clay sample. The Fe contents on the microbial mats were higher than those of the clay. The contents of Si (3.00wt%) and Al (0.20wt%) on the microbial mats sample were lower than that on the clay sample including quartz and feldspar minerals (Si(24.00wt%) and Al(12.00wt%)). Major chemical composition was different in the reddish brown microbial mats and clay, and it became clear that iron was gathered in the microbial mats.

3-4-4 SEM-EDX analysis

In the SEM observation of the reddish brown microbial mat in the Azumagaoka park, long tubular substances are densely aggregated (Fig.3-3A, B), and a long tubular structure with thickness of 1-2 μm and length: 10-50 μm and the large number of deposited substances around it are observed (Fig.3-3C,D). The long tubular substance shows a linear form without a branch. The hollow long tubular substance is found to be an aggregation of iron oxidizing bacteria, *Leptothrix ochracea*, from its morphology and elemental analysis results. *Leptothrix ochracea* are distributed in fresh water containing iron, and often detected as a dark brown to reddish brown cotton-like colony in the underground water and subsoil water or from a swamp and a groove even in Japan (Kojima et al., 1995). They are also often detected at places such as the sump water portion of cliff edge and rice paddies where the water appears red-brown, including very frequently the iron-oxidizing bacteria to form a filament (Kojima et al., 1995). The cells with diameter of about 2 μm are lined with bacillus having in a row in a substantially straight sheath with the same width, and since the stalks have unbranched flagella and are motile, the cells are often undetected, leaving only the sheath with deposited iron (Japan Water Works Association, 1999). These mentions and observation results are consistent.

The semi-quantitative chemical analyses of microbial mats in Azumagaoka park were performed by SEM-EDX analyses (Table3-3).

Fe (49.01-61.93 wt%) were detected most in the microbial mats sample, and Si,Cl,Ca,Zn were also detected in the sample. Furthermore, there is a possibility that radionuclides of Th and U were detected in the microbial mats. SEM observation, XRD and elemental analysis results show the amorphous iron hydroxide was formed by the iron bacteria.

Fe is collected in the tubular substance formed by iron oxidation bacteria are found on elemental content maps (Fig3-4, 3-5). In XRF chemical composition of microbial mats, Sr of trace element were detected, Sr is detected on tubular substance at elemental content maps (Fig3-5). It is suggested that the microbial mats were collected trace elements.

3-5 Discussion

Microorganisms can uptake and concentrate some elements into their cells and then mineralizing them. The iron hydroxide was concentrated in the reddish brown microbial mats in Azumagaoka park by iron-oxidizing bacteria, *Leptothrix ochracea.* Since the water in the canal had the quality of spring water, it seems that the microbial mats were formed by iron and iron-oxidizing bacteria taken into the spring water from the strata while the spring water flowed through there.

The iron-oxidizing bacteria that form the reddish brown microbial mats oxidize divalent iron into trivalent iron to obtain energy to metabolize iron precipitate. Further, there are reports on actual cases of efficient collection of radionuclides by iron bacteria in uranium deposits and muck dumping sites (Waite et al., 1994; Lioyd and Macaskie, 2000; Yong and Mulligan, 2004; Mulligan et al., 2007; Krejci et al., 2011; Cygan and Tazaki, 2014). There are also an electron micrograph of microorganisms adsorbing uranium (Sakaguchi, 1996; Tazaki, 2013), an electron micrograph of native microorganisms in Fukushima prefecture adsorbing cesium by an experiment, and a secium spectrum was obtained with SEM-EDX (Akai, 2011). Microorganisms' uptake of uranium is not performed through substance metabolism. It is mostly physicochemical adsorption into cell substances (Sakaguchi, 1996). Considering the fact that results of radiation dose measurement at the site indicated the higher dose of the reddish brown microbial mats than air dose, and considering elemental analysis results and the elemental content maps obtained with WD-XRD and SEM-EDX, it seems that the reddish brown microbial mats in Azumagaoka park took into not only iron but also trace elements and radioactive materials. In addition Sr is confirmed by a XRF analysis, and it is also shown that Sr is concentrating on a diatom from the obtained elemental content maps. Iron hydroxide formed through mineralization by the microbial mats is unlikely to produce a compound with radionuclides. Therefore, radionuclides are considered to be adsorbed into microorganisms, organic substances, and a trace of clay minerals. This suggests that maintaining an environment where the microbial mats are formed also maintains the formation of the microbial mats to allow adsorption and immobilization of newly-supplied specific substances.

3-6 Conclusion

The reddish brown microbial mats collected in Azumagaoka park were formed by iron-oxidizing bacteria *Leptothrix ochracea.* It seems that the microbial mats started to be formed before the earthquake. Since the water in the canal had the quality of spring water, it seems that the microbial mats were formed by iron-oxidizing bacteria and iron ion taken into the spring water from the stratum while the spring water flowed through there. Results of XRF chemical composition and elemental content maps of microbial mats, were suggested that the microbial mats were collected trace elements.

Azumagaoka park's environment did not change significantly before and after the 3.11 Earthquake. Therefore, newly-supplied specific substances will be adsorbed and immobilized through the microbial mat formation process. This suggests that maintaining an environment where the microbial mats are formed also maintains the formation of the microbial mats to allow adsorption and immobilization of newly-supplied specific substances.

Table 3-1 Water chemistry in Azumagaoka park.

Sampling 2014/1/11

Point name		Azumagaoka Park
Microbial mats		Reddish brown microbial mats
Characteristics of the water		Ca^{2+}-HCO_3^- type Spring water
Ion equivalent (meq/L)	Na^+	0.30
	K^+	0.03
	Ca^{2+}	2.30
	Mg^{2+}	0.40
	Cl^-	0.15
	HCO_3^-	2.79
	SO_4^{2-}	0.10
	NO_3^-	0.00
Iron (mg/L)		24.00
Manganese (mg/L)		3.30
pH		6.9
EC(mS/m)		36
ORP (mV)	E (field)	+ 22
	Eh (Reduced value)	+ 240
Oxidation-reduction state		Oxidative
Water temperature (℃)		7.5

Table.3-2 Radiation dose in Azumagaoka park

Date	Sample	Radiation dose	
		cpm	μSv/h
2014/1/11	Air dose (Haranomachi St.)	110-160	0.14-0.16
	Microbial mat	200-300	0.33
	Moss	200	0.38
2014/1/14 (Kanazawa)	Air dose	70-80	
	Microbial mat	80	

Table.3-3 WD-XRF analyses of reddish brown microbial mats

Unit : wt(%)

element	Reddish brown microbial mats	Soil present underneath the microbial mats
C	9.10	5.90
N	ND	ND
O	36.00	50.00
Na	ND	0.49
Mg	0.06	0.86
Al	0.20	12.00
Si	3.30	24.00
P	0.02	0.06
S	0.04	0.25
Cl	0.03	ND
K	0.04	1.30
Ca	0.69	1.00
Ti	ND	0.47
Cr	ND	0.02
Mn	0.28	0.06
Fe	50.00	4.20
Ni	ND	0.01
Zn	ND	0.01
Rb	ND	0.01
Sr	0.01	0.01
Zr	ND	0.01

ND : not detected

Table.3-4 SEM-EDX analyses of reddish brown microbial mats.

Unit : wt(%)

element		Azumagaoka park	
		Reddish brown microbial mats	Reddish brown microbial mats
		Fig..3-4	Fig.3-5
Mg	K	0.58	1.35
Al	K	0.00	0.00
Si	K	2.97	2.04
P	K	0.00	0.00
S	K	0.34	1.41
Cl	K	1.63	3.45
K	K	0.44	1.47
Ca	K	3.71	2.59
Mn	K	0.28	0.36
Fe	K	61.93	49.01
Zn	L	2.48	9.20
As	L	0.00	0.00
Sr	L	0.00	0.00
Y	L	0.00	0.00
Zr	L	0.00	0.00
Ag	L	0.00	0.00
I	L	0.20	0.22
Cs	L	0.10	0.00
Ba	L	0.30	0.11
Ce	L	0.00	0.26
Nd	L	0.00	0.35
Th	M	0.59	3.27
U	M	0.01	0.00
Np	M	0.00	1.11
Pu	M	0.00	0.00
O		24.41	18.27

0.00 : Analysis value shows zero

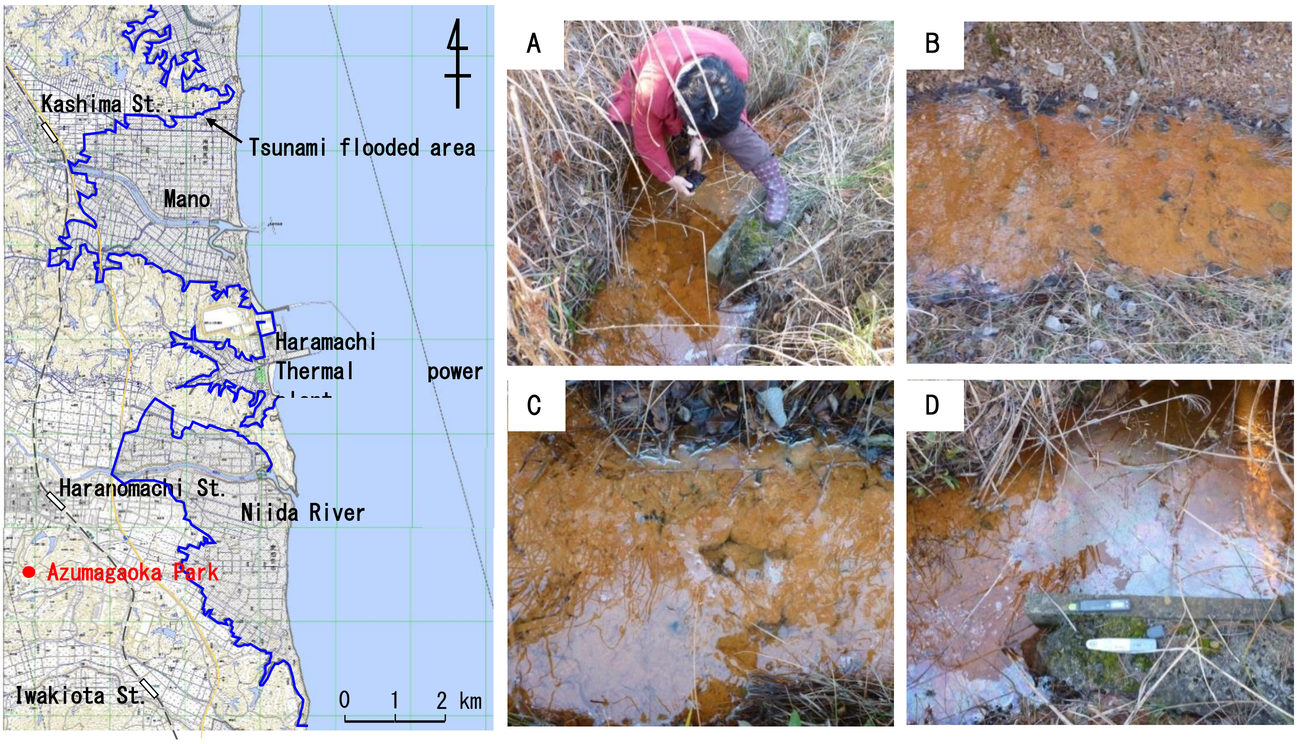

Fig.3-1. Location map of the reddish brown microbial mats were formd in canal in Azumagaoka park, Haramachi-ku, Minamisoma, Fukushima, Japan

A: Formation situation of the reddish brown microbial mat in canal.

B: The reddish brown microbial mats were formd on the entire canal .

C: The reddish brown microbial mats were formd like flock.

D: Biofilm were recognized like oil slick.

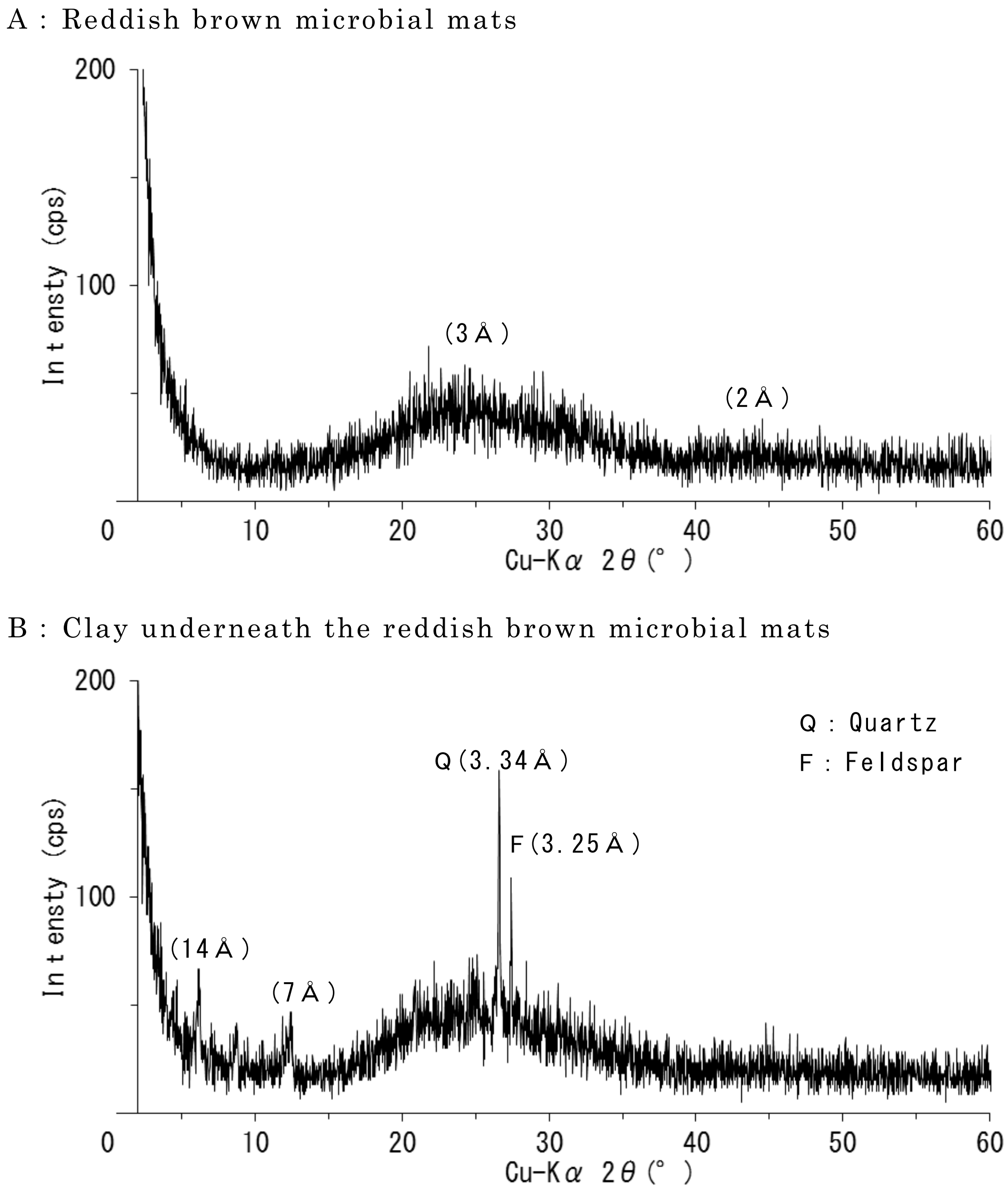

Fig. 3-2. X-ray diffraction pattern of the reddish brown microbial mats and clay.
A: Reddish brown microbial mats
B: Clay underneath the reddish brown microbial mats.

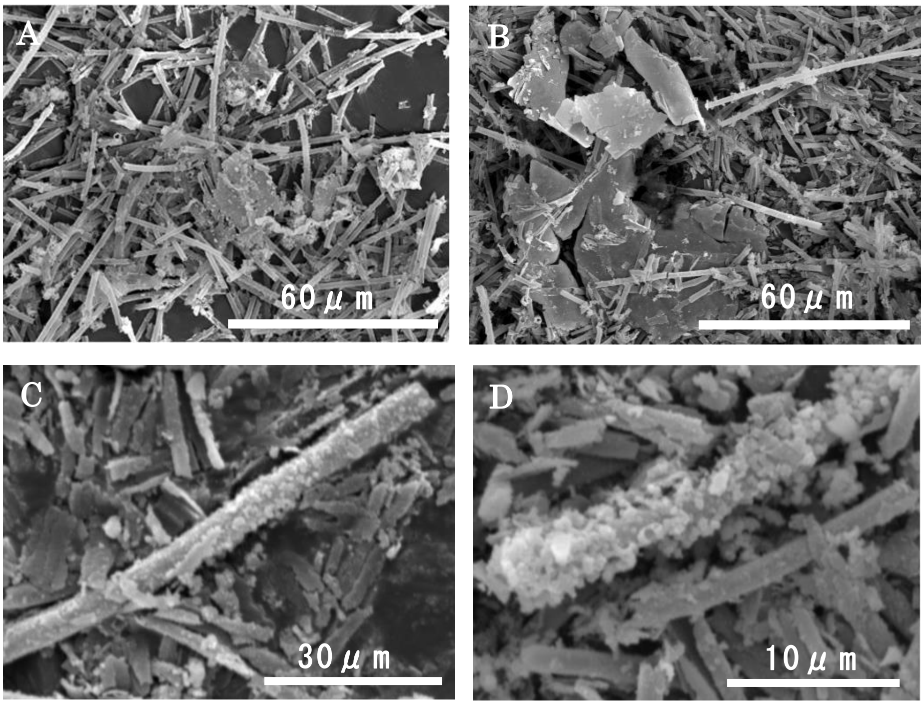

Fig.3-3 Scanning electron micrograph in Azumagaoka park

A : Long tubular substances are densely aggregated in the reddish brown microbial mat

B : Long tubular substances and Inorganic materials

C : Long tubular structure with thickness: 1-2 μm and length: 10-50 μm

D : The deposited substances around it are observed in a large number

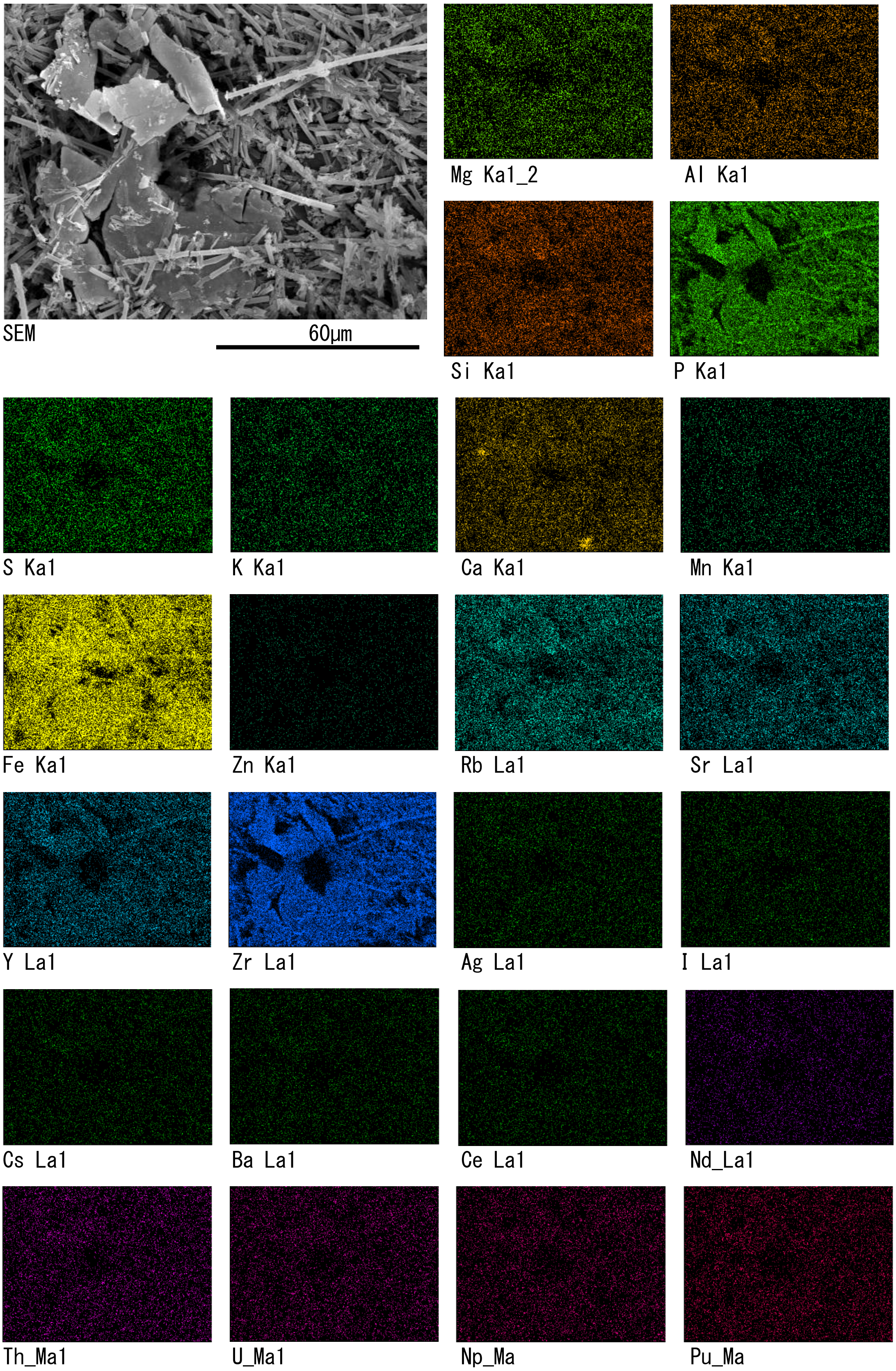

Fig.3-4 Scanning electron micrograph and elemental content maps of reddish brown microbial mats.

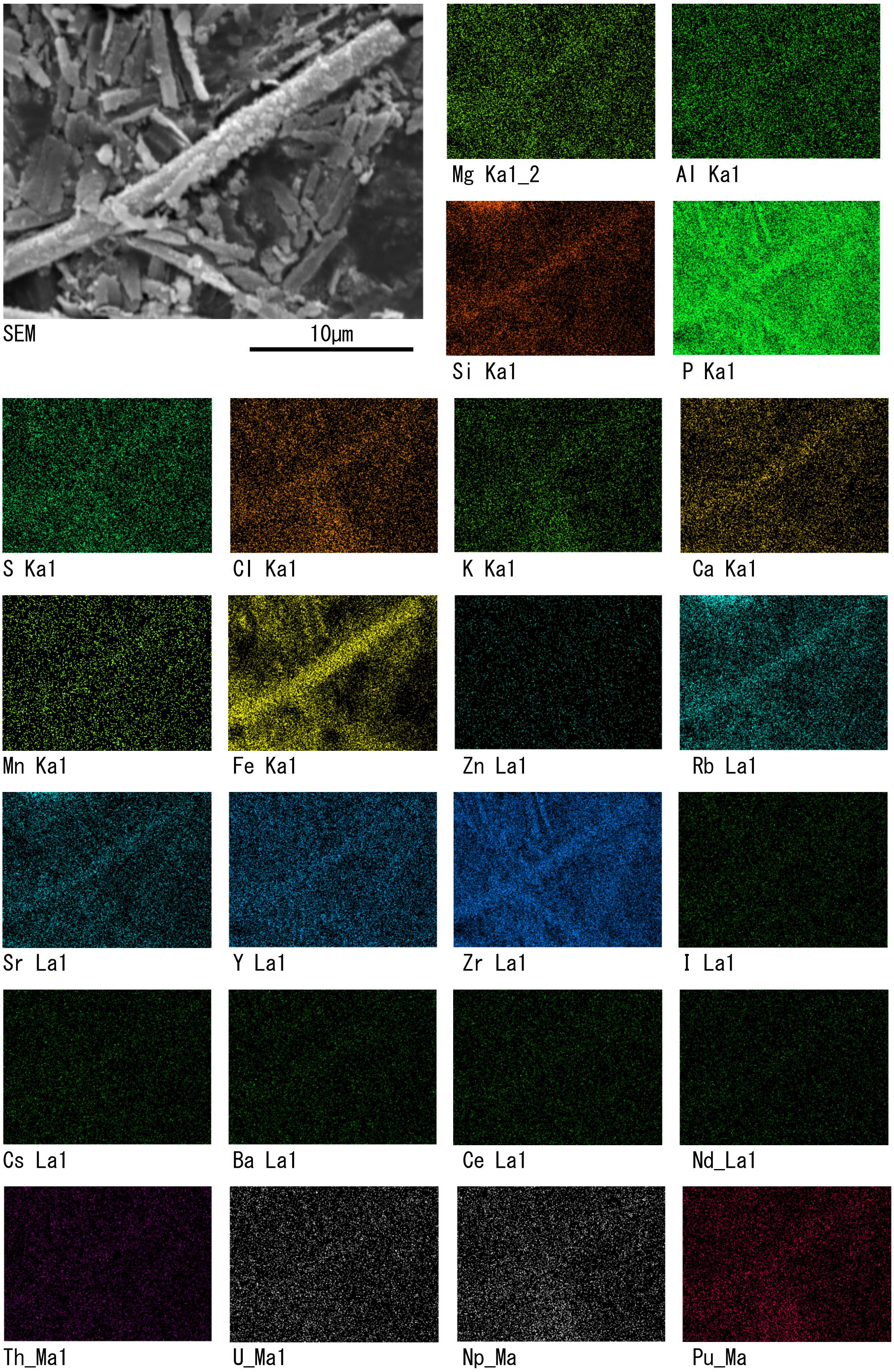

Fig.3-5 Scanning electron micrograph and elemental content maps of reddish brown microbial mats and iron oxidizing bacteria cell.

Chapter 4　Characteristics of microbial mats and hot spring water of two locations in Iwaki city, Fukushima prefecture, that gushed out after the Earthquake.

4-1 Background and purpose of this study

In Iwaki Yumoto hot spring area of Iwaki city, Fukushima prefecture, hot spring water gushed out from housing foundations and drainpipes in Uchigo-Takasakamachi, Iwaki city due to the 3.11 Earthquake. Besides, a large amount of hot spring water gushed out from the vertical shaft (exhaust port) of abandoned coal mine facilities in Izumi-Tamatsuyu, Iwaki city due to the 4.11 aftershock. As of January 2014, hot spring water still gushes out from these hot springs. Since no countermeasure to stop the gush has been found, and it is discharged into side ditches and rivers. This raises concern about an environmental change due to a water chemistry change caused by the hot spring water. One month after the 3.11 Earthquake, a 7.0-magnitude epicentral aftershock occurred in a shallow hypocenter at a depth of approximately 5 km in the Hamadori district, Fukushima prefecture around 17:16 on April 11, 2011. This aftershock is the biggest inland earthquake that occurred along an active fault immediately after the 3.11 Earthquake. It suggests a relation with huge earthquakes that occur at an oceanic trench (Ishiyama et al., 2011; Sugito et al., 2011; Japan Meteorological Agency, 2012).

There are many reports on the mechanisms of the active fault and hot spring gush related to these earthquakes. However, no report is available on scientific data about the hot spring gush due to the earthquakes and microbial mat formation. I studied the water chemistry of hot springs, microorganisms and biomineralization, and environment by comparing two hot springs.

4-2 Topography of investigation place and a geological feature

Two investigation sites in the present study are Uchigo-Takasakamachi and Izumi-Tamatsuyu in Iwaki city in the southeast area of Fukushima prefecture. The Joban coal field is widely distributed around these sites. The Joban coal field is known as a coal field with hot spring water, and Iwaki Yumoto hot spring is located at the approximate midpoint between the two investigation sites (Fig. 4-1). Yumoto and Uchigo regions within the Joban coal field had problems on discharged hot water, and it was drained into the Yumotogawa River, Shinkawa River, etc. (The association of COAL FIELD, 2008).

The Iwaki Formation and Shirasaka Formation that are classified in the Shiramizu group in the Pliocene Epoch of the Neogene Period are distributed around these investigation sites. The Iwaki Formation is made up of fine to medium grained sandstones and gravel and has a coal bed. The Shirasaka Formation is a mudrock formation. Under these formations in the Neogene Period, granites and diorites in the Cretaceous system are distributed (Fukushima Prefecture, 1994, 1995). These areas are close to the Idosawa fault and Yunodake fault, and these faults are considered to have acted due to the earthquake on April 11, 2011. Due to this earthquake, a surface deformation that was considered to be a surface earthquake fault emerged for 10 km in length along the Idosawa fault and Yunodake fault (Geography and Crustal Dynamics Research Center, 2011).

Uchigo-Takasakamachi, Iwaki city was a slag heap of the Joban coal mine. The slag heap was leveled off so that a residential area was developed. In the housing premises in Uchigo-Takasakamachi, hot spring water has been gushing out from about 20 spots such as drainpipes and drainpipes due to the 3.11 Earthquake. Although the hot spring water gushed out from two spots at first, they moved to different positions. Once hot spring water stops gushing out from one spot, after it gushes out from another spot. This sank the surrounding ground level. In the premises, the ground surface became soft for a while enough to sink the feet in the soil and the ground level dropped by approximately 20 cm (Fukushima Minpo, 2011). The amount of discharge between 2011 and 2013 was 2 to 6 L/sec and its temperature was approximately 27°C (Geological Survey of Japan, AIST, 2013). When the amount of discharge was measured on January 12, 2014, the total amount of several spots was 2.8 L/sec, and no significant change was observed in amount.

Uchigo region was the center of Iwaki northern area of the Joban coal field. After coal mining was closed in Uchigo region in 1957, the slag heap was leveled off in 1967 so that the Takasaka housing development was developed this area. That is, houses in Uchigo-Takasakamachi were built in a place where the slag heap had been leveled down and then the land had been reclaimed, which resulted in loose ground. Therefore, it seems that hot spring water gushed out due to the earthquake on March 11, 2011.

Izumi-Tamatsuyu, Iwaki city is located at the western end of the Joban coal field and has an abandoned vertical shaft (exhaust port) extending from a mineshaft at approximately 600 m below ground. Due to the earthquake on April 11, 2011, a large amount of hot spring water gushed out from the vertical shaft. When its western coal pit was closed in 1977, the Joban coal mine was entirely closed (Ishiyama, 2002). After the closing, Jobanyumoto onsen Co., Ltd. was founded at Jobanyumotomachi, Iwaki City in 1979 to pipeline the mineshaft at approximately 600 m below ground where hot spring water gushes out to supply hot spring water to the hot spring district in Iwaki Yumoto area. This pipeline is extended to the vertical shaft in Izumi-Tamatsuyu.

It is considered that since the hot spring water gush mechanism is related to crustal movement due to earthquakes, the earthquake on April 11 squeezed the earth crust, so that hot spring water present in cracks was pressured to gush out on the ground surface (Geological Survey of Japan, AIST, 2013).

4-3 Materials and methods

4-3-1 Sample collection

Microbial mats sample collected on January 11, 2014 and January 12, 2014. Reddish brown microbial mats collected by stainless steel net and scoop in Uchigo-Takasakamachi. White microbial mats collected by stainless steel net at scupper of hot spring water to Kamado-gawa river in Izumi-Tamatsuyu.

About hot spring waters of two locations, water temperature, pH, electric conductivity (EC), and oxidation-reduction potential (ORP) in field were measured by using HORIBA B-211 Compact pH Meter (pH meter), HORIBA B-173 Compact Conductivity Meter (EC meter), and CUSTOM ORP6041 (ORP meter) on sites, respectively. Water samples collected in polyethylene bottles, and were performed chemical analyses of water in laboratory. The standard hydrogen electrode potential (Eh) was calculated from the oxidation-reduction potential (ORP) by the following relation.

$$Eh\ (mV) = ORP + 206 - 0.7 \times (t - 25)$$

t : water temperature (°C)

Air radiation dose was measured at height of 1m from a ground level. Radiation dose of microbial mats were measured by sample contact technique on site. Measurements of β (γ)-ray radiation used ALOKA GM survey meter TGS-136 (cpm), and measurements of γ-ray + X-ray radiation used ECOTEST TERRA-P+ (μSv/h) were performed. At GM survey meter, 1000cpm was equivalent to 1.52μSv/h, and there was an error of ± 20 to 30 %. At TERRA-P, there were ± (25+2/10H) of error.

4-3-2 Chemical analyses of water

Water chemistry analyzed inorganic ion ingredient (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}) and iron ion, manganese ion. Chemical analyses of water were performed using Ion Chromatography (Metrom, 883Basic IC Plus). Water chemistry were performed an official analysis by JIS K 0102 or a water supply test method. Ion chromatography was developed for the analysis of the inorganic ion and it was used in environment-related many fields, and methods were determined (Goto, 2011). Chemical analyses of water depended on NSS Co., Ltd. (Tsubame city, Niigata prefecture).

4-3-3 X-ray powder diffraction measurement(XRD)

XRD measurements for samples were performed using RIGAKU RINT 2200 with Cu Kα radiation. The $\theta/2\theta$ scanning technique was adopted over the $2\theta = 2 - 60°$ with a scan step of 0.02°. The applied acceleration voltage and current were 40 kV and 30 mA, respectively.

2-3-4 Wavelength dispersive X-ray flourescence analysis (WD-XRF)

Chemical compositions of the samples were determined by wavelength dispersive X-ray fluorescence analyzer (WD-XRF ; RIGAKU ZSX100e) using Rh-Kα ray, which operated at an accelerating voltage of 30kV, under vacuum condition, and FP bulk qualitative and sem-quantitative analysis technique were used. WD-XRF analysis were performed by Yamato Environmental Analysis Co., Ltd. (Kawakita town, Ishikawa prefecture).

4-3-4 Optical microscope observation

The optical microscope (OLYMPUS ; BX-60) was used in order to observe the form of reddish brown microbial mats and black microbial mats which was sampling at Uchigo-Takasakamahi.

4-3-5 Scanning electron microscopy and energy dispersive X-ray analysis (SEM-EDX)

The scanning electron microscopy equipped with energy dispersive X-ray analyzer (SEM-EDX ; HITACHI S-3400N and HORIBA, EMAX X-act) was used for micro morphological observations, semi-quantitative analyses, and elemental content maps of the samples under 15kV accelerating voltage and current 70-80μA, with an analytical time of 1,000 seconds, and an area of 10mm x 10mm on the carbon double tape with Pt coating. The energy dispersive X-ray analyzer (HORIBA, EMAX X-act) used in this study could be analyzed even small amount of element and displayed on the elemental content maps. Even in the cases that samples include unexpected elements and microparticles, the apparatus can distinguish them (HORIBA, 2010; Burgess et al, 2013; Shozugawa, 2014). The semi-quantitative analyses were performed. The peak separation software was used for analyses of coexist elements with similar fluorescence energies, and a value of zero is indicated in the case that lists of fluorescence energy are overlapped.

4-4 Results

4-4-1 Characteristics of sampling site in Uchigo-Takasakamachi

When the first investigation was conducted on November 14, 2011, hot spring water of approximately 25 to 27°C gushed out from a drainpipes on the north side of a housing foundation (Fig. 4-2A) and another drainpipes on the west side (Fig. 4-2B, C, D), and the formation of reddish brown microbial mats and black ones was observed (Fig. 4-2). At that point, hot spring water had stopped gushing out from several ones at about 20 drainpipes, and the traces of dried reddish brown microbial mats were observed there. As of January 2014, microbial mats are widely formed around spots from which a large amount of gushing hot spring water. However, due to decrease in amount, there was a drainpipes whose wet part was its central part only (Fig. 4-2C). Although the innermost microbial mats on the west side were dried, new gushing hot spring water was observed from the lower layer, and black microbial mats were also observed in a wet state around the hot spring water (Fig. 4-2C).

According to the information from local residents, this area was used as a water tank for storing groundwater pumped up by the Joban coal mine. Even after the water tank was filled with soil, residents saw rusty water always flowing out from the ground in this area. After that, land development was carried out so that houses were built there. When the hot spring water started to gush out, a large amount of hot spring water gushed out on the west side. However, after it stopped, hot spring water started to gush out from another drainpipes. Fig. 4-2B and C show microbial mats that were dried due to the halt of gush of hot spring water. Thus, the information and the photographs correspond to each other.

4-4-2 Characteristics of sampling site in Izumi-Tamatsuyu

The hot spring water that gushed out in Izumi-Tamatsuyu, Iwaki city was led with sandbags and gutters to the Kamadogawa River flowing on the south side (Fig. 4-3). The formation of white microbial mats and green algae on the interface between the hot spring water and the ground was observed. Besides, a large amount of white string-like substance called sulfur turf was observed in the drainage channel to the Kamadogawa River (Fig. 4-3B, C). Sulfur turf is named by Miyoshi, M. (1897) (Sugimori, 1994). Sulfur turf is microbial mats, which develop in hot spring flowing water including hydrogen sulfide at a high temperature and neutrality, and is a colony of sulfur-oxidizing bacteria. Sulfur turf is appearing to be white to yellowish-white to the naked eye (Maki et al., 2004).

In the Iwaki Yumoto hot spring district, hot spring water that gushed out in the mineshaft at 600 m below ground is led via a pipe to a hot water tank at 40 to 50 m below ground and then pumped up to the ground. This fact may be consistent with the fact that the pipe installed in the mineshaft was destroyed due to the earthquake and therefore hot spring water gushed out from the vertical shaft.

4-4-3 Water chemistry and radiation measurement

The result of the radiation measurement is shown in Table4-1, the result of water chemistry measurement is shown in Table4-2 in Uchigo-Takasakamachi and Izumitamatsuyu, Iwaki city.

Water chemistry analysis was performed about 3 samples in the reddish brown microbial mats formation spot, the black microbial mats formation spot in Uchigo-Takasakamachi and Izumitamatsuyu where water. The samplings were done on January 3, 2012 and 2 hot spring water samples extracted at the reddish brown microbial mats formation spot in Uchigotakasakamachi and Izumitamatsuyu on January 12, 2014. For reference, I show a value in terms of an ion equal amount from an ingredient list for the Iwaki Yumoto hot spring (Table4-1).

From the results of radiation dose measurement at November, 2011 in Uchigo-Takasakamachi, the air dose was 100-140cpm, on the other hand the dose of microbial mats were 430cpm, and the dose of a road and the drainage were 400-430cpm. The dose of radioactivity of the flowing out hot spring water was 110-140cpm (Table4-2).

In Uchigo-Takasakamachi, the following water chemistries were measured, water temperature 25 ℃, pH 7-8, EC 129-162 mS/m, ORP -42 to 78mV (Eh 168 to 284) (Table4-1). These results of hot spring water showed the $Na^{+}(Ca^{2+})$-SO_4^{2-} type in Uchigo-Takasakamachi(Table4-1). The density of iron became more than 40 times than an analysis level of that for 2012 with the hot spring water which formed the reddish brown byte mat which was sampled in 2014 (Table4-1).

From the measured radiation dose of November, 2011 in Izumi-Tamatsuyu, the air dose was 150-180cpm, on the other hand the dose of microbial mats were 430cpm, the gutters were 330cpm (Table4-2).

In Izumi-Tamatsuyu, the following water chemistries were measured, water temperature 55 ℃, pH 8, EC 323-408 mS/m, ORP -194 to -331 mV (Eh -3 to -149) (Table4-1). This hot spring water showed remarkably reductive environment in Izumi-Tamatsuyu. The water chemistry of hot spring water showed the Na^{+}-Cl^{-} type, and the content of SO_4^{2-} is 1.9-2.9 meq/L in Izumi-Tamatsuyu(Table4-1). On the other hand the water chemistry of hot spring water in Iwaki Yumoto hot springs showed the Sulfur-Na^{+}-Cl^{-} ・ SO_4^{2-} type, and the content of SO_4^{2-} is 7.1meq/L.

4-4-4 XRD analysis

The mineralogy of the reddish brown microbial mats in Uchigo-Takasakamachi was determined using XRD analysis. The XRD results shows this sample indicated the amorphous background is located at 3Å and 2Å(Fig.4-4A). The concentration of iron was in the hot sparing water was larger than that of the biomats, and iron was strongly detected by the chemical compositions analysis by WD -XRF and SEM-EDX. From these results that the sample includes amorphous iron hydroxide.

The XRD results show that white microbial mats in Izumi-Tamatsuyu included native sulfur(d = 3.85Å, 3.44Å and 3.21Å) and gypsum (d = 7.63Å, 4.28Å, 3.07Å, 2.87Å and 2.68Å) (Fig.4-4B). Gypsum is considered to have been formed from Ca^{2+} and SO_4^{2-} in the hot spring water. It shows that SO_4^{2-} included in ingredients of hot spring water was also used for formation of gypsum as well as native sulfur by a white microbial mat.

4-4-5 WD-XRF analysis

The chemical composition of microbial mats collected at two location in Iwaki city, were analyzed by WD-XRF analyses (Table4-3).

O(44.00wt%) and Fe(30.00wt%) detected in the microbial mats sample in

Uchigo-Takasakamachi. And C, Al, Si and Ca are also detected.

O(38.00wt%), C(24.00wt%), S(13.00wt%) and N(10.00wt%) detected in the microbial mats sample in Izumi-Tamatsuyu. And Al, Si Ca and Fe are also detected. The trace elements of Sr detected slightly in the microbial mats, but the other trace elements not detected.

4-4-6 Observation by optical microscope and SEM

From the optical microscope observation of the reddish brown microbial mat in Uchigo-Takasakamachi, many long tubular structure with thickness of 1-2 μm and fine brown deposits around it are observed (Fig.4-5A). The long tubular substance shows a linear form without a branch. In the SEM observation, a wide variety of long tubular microorganisms with thicknesses of 1-2 μm and 7μm and length of 10-50 μm and spherical substances deposited on them are remarkable (Fig.4-5B, C). From its morphology and elemental analysis results, the hollow long tubular substance is found to be an aggregation of iron-oxidizing bacteria, *Leptothrix ochracea*. Clay particles are deposited on the surface of tubular substance. *Leptothrix ochracea* are distributed in fresh water containing iron, and also often detected as a dark brown to reddish brown cotton-like colony in the underground water and subsoil water or a swamp and a groove even in Japan (Kojima et al., 1995). They are also often detected in the places such as the sump water portion of cliff edge and rice paddies where the water appears red-brown, giving the highest detection frequency as the iron-oxidizing bacteria to form a filament (Kojima et al., 1995). The cells with diameter of about 2 μm are lined with bacillus having in a row in a substantially straight sheath with the same width, and since the stalks have unbranched flagella and are motile, the cells are often undetected, leaving only the sheath with deposited iron (Japan Water Works Association, 1999). These mentions that observation results are congruent. In addition, black microbial mat was found to be an aggregation of algae by the optical microscope observation (Fig.4-5D).

In the SEM observation of the white microbial mat in Izumi-Tamatsuyu, a filamentous substance whose surface became smoothed with an adhesive substance was observed (Fig.4-5E). Microorganisms form sulfur-turf which allegedly produce cellulose (Maki et al., 2004), and the adhesive material is considered to be an organic material composed mainly of cellulose. Cylindrical bacilli with a size of 40-50 μm are also observed in the filamentous substance (Fig.4-5F). Sulfur turf is microbial mats, which develop in hot spring water including hydrogen sulfide at a high temperature and neutrality, and is a colony of sulfur-oxidizing bacteria. Hydrogen sulfide contained in the hot spring water is first oxidized by sulfur-oxidizing bacteria to become elemental sulfur particles, depositing on the mats and appearing to be white to yellowish-white to the naked eye (Maki et al., 2004). The bacteria constituting the sulfur turf are classified into three types - A: *Thiovibrio miyoshii*, B: *thiobacillus* sp. and C: *Thiothrix miyoshii* (Sugimori, 1994). From the morphological features and elemental analysis results, the white microbial mat is found to be an aggregation of sulfur-turfs that sulfur-oxidizing bacteria capture the sulfur components (such as SO_4^{2-} and HS^-) of hot spring water to grow.

4-4-7 SEM-EDX analysis

The chemical composition by semi-quantitative analyses of microbial mats in Uchigo-Takasakamachi and Izumi-Tamatsuyu were analyzed by SEM-EDX analyses (Table4-3).

In Uchigo-Takasakamachi, the elements of Fe(12.15wt%), Si(11.13wt%), C(13.34wt%) detected in the microbial mats. The elements of Al, S, Cl detected in the range of 1.0-2.5w%.

From the element map data, the elements of Fe, Ca, S and Cl were concentrated in the tubular substance formed by iron oxidation bacteria, and the elements of C and O were detected in the wide area. This result is consistent with the water chemistry and the chemical composition. On the other hand, Al and Si are detected as localized fine particles, and it is considered clay minerals or feldspars (Fig.4-6).

In Izumi-Tamatsuyu, the quadrangular diatom is found somewhere in the white microbial mats by SEM observation. The S (16.80wt%) was detected in the microbial mats, and abundant C and O also were detected. The Ca of 4.23 wt% was detevted which is a constituent element of gypsum (Table4-3).

The elemental content map showed S, C and O concentrates in the wide area, and Ca, Cl and K were also detected. C and O are considered to be the elements which constituted cellulose. S and Ca are considered to be elements which constitute native sulfur and gypsum concentrated in microbial mats in hot spring water. S concentrated on a shell of the diatom in the nicrobial mats. But the concentration of sulfur is not recognized in the diatom, and it show the elements concentrated with the microorganisms are different (are not sulfur). In addition, platinum is detected in the sample of Izumitamatsuyu by SEM-EDX analysis, but this is influence by the vapor deposition of the sample.

The trace element including the radionuclide was undetected by the SEM-EDX analysis as well as WD-XRF analysis for samples of Uchigo-Takasakamachi and Izumi-Tamatsuyu

4-5 Discussion

Leptothrix ochracea are precipitating as? iron hydroxides in an amount five times or more of the inorganic precipitates (Sato et al., 2004), it show that iron bacteria concentrated iron.

Reddish brown and black microbial mats were formed in the hot spring water that gushed out in Uchigo-Takasakamachi after the earthquake on March 11, 2011. The reddish brown microbial mats are formed in such a manner that the iron bacteria *Leptothrix ochracea* form a population and concentrate iron. As for the water chemistry of hot spring water in Uchigo-Takasakamachi, the iron ion concentration increased over 40 times in two years. In contrast, no significant increase of iron content was observed in Izumi-Tamatsuyu region. The iron concentration by *Leptothrix ochracea* is efficient over five times of the inorganic deposition (Sato et al., 2004), and this indicates that the iron bacteria concentrated iron. Besides, it is known that iron bacteria also concentrate manganese (Yagi, 2001). WD-XRF analysis of the reddish brown microbial mats in Uchigo-Takasakamachi indicated a manganese concentration approximately 27 times higher than the white microbial mats in Izumi-Tamatsuyu. Iron contained in hot spring water was concentrated and deposited(?) by the iron-oxidizing bacteria, so that the water chemistry was changed to one with more iron content.

Since hot spring water passes through stratal cracks in Uchigo-Takasakamachi, it seems that the hot spring water mixed with groundwater in a shallow stratum, so that Na^{+} and Cl^{-} were diluted. Further, since spring water called rusty water containing iron was observed around Uchigo-Takasakamachi, it seems that stratal iron was taken in the hot spring water and therefore the iron-oxidizing bacteria formed the reddish brown microbial mats.

The hot spring water in Izumi-Tamatsuyu gushed out after the aftershock on April 11, 2011 because which destroyed a water pipe in the Iwaki Yumoto hot spring district. While the water chemistry of the hot spring water in Izumi-Tamatsuyu indicated Na^{+}-Cl^{-} type, the hot spring water in the Iwaki Yumoto hot spring district indicated a somewhat different water chemistry since it is a sulfur-containing salt sulfate hot spring. This indicates that the white microbial mats formed in the hot spring water concentrated SO_4^{2-} and HS^- contents as sulfur by the sulfur-oxidizing bacteria. Further, gypsum in the white microbial mats is made of Ca^{2+} and SO_4^{2-} which contained as hot spring components. This is another cause of decrease in SO_4^{2-} of a hot spring.

Comparing the hot spring water of Uchigo-Takasakamachi with that of Izumi-Tamatsuyu, presumably, that of Uchigo-Takasakamachi passed through filling soil that had formed the slag heap before to take in stratal components and mix with groundwater to change its water chemistry to be oxidized. In contrast, the hot spring water in Izumi-Tamatsuyu has the water chemistry close to that of Iwaki Yumoto hot spring and is in the reduced state because the hot spring water gushed out in the mineshaft of the coal mine and blew out via the vertical shaft extended to the ground level. Although the hot spring water in Uchigo-Takasakamachi contained over 10 meq/L of SO_4^{2-}, it was in the oxidative environment as compared to Izumi-Tamatsuyu. Therefore, presumably, lack of multiplication of sulfur-oxidizing bacteria led to lack of formation of sulfur and gypsum. In Izumi-Tamatsuyu region, iron was not taken into the hot spring water because the hot spring water did not make contact with any stratum or mixed with groundwater. In addition, it was in the reductive environment after gushing out. Therefore, presumably, these did not allow iron-oxidizing bacteria to live in the hot spring water. In Iwaki Yumoto hot spring, the hot spring water collected through the pipeline is stored in the underground tank and then pumped up through a well extended down to the underground tank. It seems that the hot spring water thus did not make contact with any stratum or soil, and therefore, the hot spring water was in an environment where microbial mats are unlikely to form (Fig. 4-8).

The difference between the microbial mats of these two regions was brought about by the difference in water chemistry depending on the hot spring water gush processes and microbe habitat segregation depending on whether microorganisms are in the oxidation or reduction environment. Further, in the elemental content maps for Izumi-Tamatsuyu, elemental concentration distribution varied between sulfur turf and diatoms. This indicates a fact that microorganisms concentrate specific elements. Different microorganisms concentrate different elements. By analyzing the reration between concentrated element and microoоganism, we may determine what mineral is formed by the biomineralization. Thus, it indicated that microorganisms concentrate specific minerals to form microbial mats and this changes the water chemistry of hot spring water.

4-6 Conclusion

In Uchigo-Takasakamachi, hot spring water gushed out via cracks under the ground. This created a somewhat oxidative environment, and accordingly, iron-oxidizing bacteria multiplied proliferously there. The hot spring water passed through the cracks under the ground, contacted with soil, and mixed with groundwater. This changed the water chemistry and created the oxidative environment.

In Izumi-Tamatsuyu, hot spring water gushed out from the vertical shaft to make contact with air and soil to form the microbial mats, and this changed the water chemistry. The hot spring water rapidly gushed out through the pipeline and the vertical shaft. This created a reductive environment, and accordingly, sulfur-oxidizing bacteria multiplied proliferously to form sulfur turf.

In the microbial mats of Izumi-Tamatsuyu, sulfur turf and diatoms concentrated different elements. The elemental content maps for Izumi-Tamatsuyu demonstrated that microorganisms concentrate specific elements such as S, Ca.

The difference between the microbial mats of these two regions was brought about by the difference in water chemistry depending on the hot spring water gush processes and microbe habitat segregation and also depending on whether microorganisms are in the oxidation or reduction environment.

Thus, it was demonstrated that formed microbial mats concentrate specific minerals, and the water chemistry of hot spring water accordingly changes over the years.

Table 4-1 Water chemistry in Iwaki city.

Point name		Uchigo-Takasakamachi			Izumi-Tamatsuyu		Iwaki Yumoto hot springs
Sampling Date		2012/1/3	2012/1/3	2014/1/12	2012/1/3	2014/1/12	Hot springs notice data 2015/7/28
Microbial mats		Reddish brown microbial mats	brack microbial mats	Reddish brown microbial mats	Whaite microbial mats	Whaite microbial mats	—
Characteristics of the water		$Na^+(Ca^{2+})$-SO_4^{2-}	$Na^+(Ca^{2+})$-SO_4^{2-}	$Na^+(Ca^{2+})$-SO_4^{2-}	Na^+-Cl^-	Na^+-Cl^-	Sulfur-Na^+-Cl^-・SO_4^{2-}
		Hot spring water	Hot spring water	Hot spring water	Hot spring water	Hot spring water	Hot spring water
Ion equivalent (meq/L)	Na^+	8.26	8.26	8.70	31.32	32.19	23.51
	K^+	0.08	0.07	0.06	0.28	0.33	0.18
	Ca^{2+}	8.48	7.98	8.48	4.99	5.99	3.00
	Mg^{2+}	1.15	1.15	1.07	0.43	0.25	0.10
	Cl^-	2.20	2.37	2.60	31.03	33.85	17.42
	HCO_3^-	4.75	4.59	4.75	3.28	2.13	0.00
	SO_4^{2-}	10.62	10.62	10.41	2.91	1.89	7.13
	NO_3^-	0.00	0.00	0.01	0.00	0.01	0.00
Iron(mg/L)		0.07	<0.03	3.00	0.07	0.13	<0.10
Manganese(mg/L)		1.10	0.44	1.10	0.07	0.05	—
pH		8.1	8.0	7.2	7.8	8.0	8.0
EC(mS/m)		129	124	162	323	408	300
ORP(mV)	E (field)	78	-82	-42	-194	-331	—
	Eh (Reduced value)	+ 284	+ 126	+ 164	- 3	- 149	—
Oxidation-reduction state		Oxidative	Oxidative	Oxidative	Reductive	Reductive	—
Water Temperature (℃)		25.5	22	24.3	46.9	59	58

Table.4-2 Radiation dose in Iwaki city

(unit:cpm)

Date	Uchigo-Takasakamachi		Izumi-Tamatsuyu	
2011/11/14	Air dose	100-140	Air dose	150-180
	Road	300	Gutter	330
	Gutter	430		
	Hot spring water	110-140		
	Microbial mat	430		
	Concrete	400		
	Road	140-165		
	Gap of stairs	140		
2014/01/12	Air dose	100-140		
	Microbial mat	60-90	Sulfur turf	60-70
	Crack of road	150	After drying	80
	Dried grass	100		
	Lichen	230		

Table.4-3 WD-XRF analysis of reddish brown microbial mats and white microbial mats

Unit : wt(%)

element	Uchigo-Takasakamachi Reddish brown microbial mats	Izumi-Tamatsuyu White sulfur turf
C	7.70	24.00
N	ND	10.00
O	44.00	38.00
Na	0.40	0.58
Mg	0.35	0.47
Al	3.20	2.30
Si	8.00	4.90
P	0.07	0.92
S	0.37	13.00
Cl	0.15	0.48
K	0.43	0.48
Ca	2.90	3.00
Ti	0.19	0.14
Cr	ND	ND
Mn	1.10	0.04
Fe	30.00	1.50
Cu	ND	ND
Zn	0.03	0.01
As	ND	ND
Rb	ND	0.01 <
Sr	0.03	0.01
Y	ND	ND
Zr	0.01	0.01 <
Ag	ND	ND
I	ND	ND
Cs	ND	ND
Ba	ND	ND
Ce	ND	ND
Nd	ND	ND
Eu	ND	ND
Tb	ND	ND
Pt	ND	ND
Hg	ND	ND
Ra	ND	ND
Th	ND	ND
U	ND	ND
Np	ND	ND
Pu	ND	ND

ND : not detected

Table.4-4 SEM-EDX analysis of reddish brown microbial mats and white microbial mats.

Unit : wt(%)

Element		Uchigo-Takasakamachi Reddish brown microbial mats	Izumi-Tamatsuyu White sulfur turf
Mg	K	0.10	ND
Al	K	2.55	0.09
Si	K	11.13	2.20
P	K	0.00	ND
S	K	1.56	16.80
Cl	K	0.20	0.10
K	K	0.30	0.49
Ca	K	1.03	4.23
Cr	K	*	ND
Mn	K	0.20	ND
Fe	K	12.15	0.31
Cu	L	*	ND
Zn	L	0.00	ND
As	L	ND	*
Rb	L	0.00	ND
Sr	L	0.00	ND
Y	L	0.00	*
Zr	L	25.22	*
Nb	L	*	*
Pd	L	*	*
Ag	L	ND	0.85
I	L	ND	ND
Cs	L	ND	0.00
Ba	L	ND	ND
Ce	L	0.00	*
Nd	L	ND	*
Eu	L	0.00	*
Tb	L	*	*
Pt	M	*	42.93
Hg	M	0.00	1.89
Ra	M	*	0.00
Th	M	0.00	0.00
U	M	0.00	0.00
Np	M	0.00	0.00
Pu	M	0.00	0.00
O	K	30.02	18.27

* : Element that is not set
0.00 : Analysis value shows zero
ND : not detected
—: Not analyzed

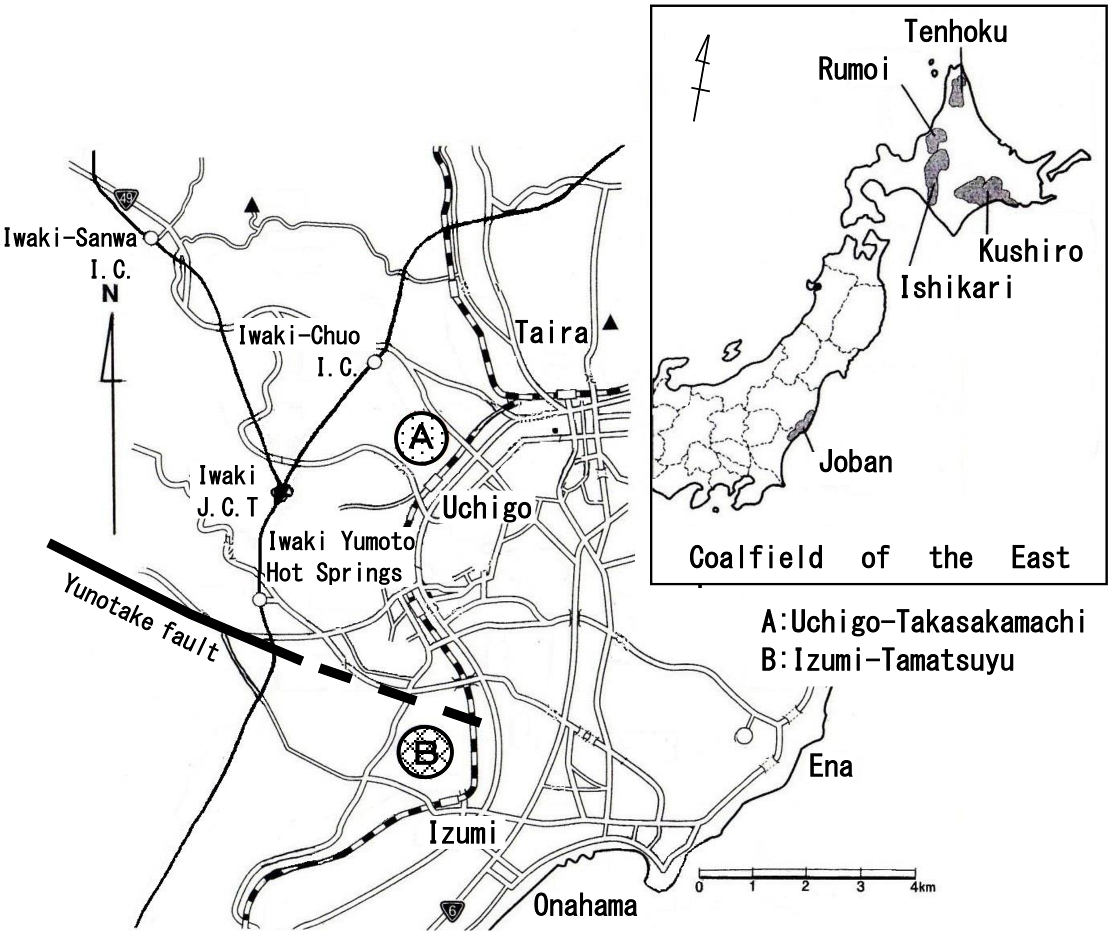

Fig.4-1. Location map at Iwaki, Fukushima, Japan

A: Uchigo-Takasakamachi .

B; Izumi-Tamatsuyu

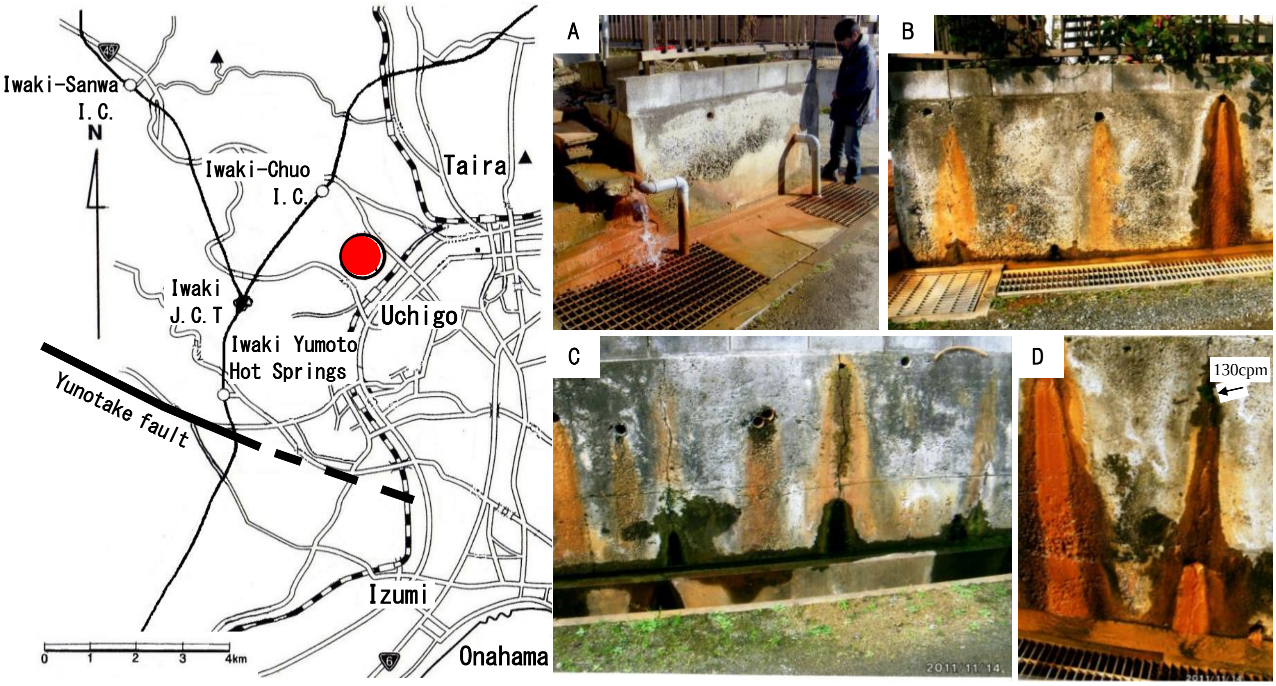

Fig.4-2. Location map of hot spring water gushed out from the foundation and drainpipes of a house in Uchigo-Takasakamachi, Iwaki, Fukushima, Japan

A: Hot spring water gushed out from the foundation of a house at the north side

B; Hot spring water gushed out from drainpipes, showing reddish brown microbial mats at the west side

C; Showing black microbial mats at the west side

D;. Radiation dose of reddish brown microbial mats (130cpm).

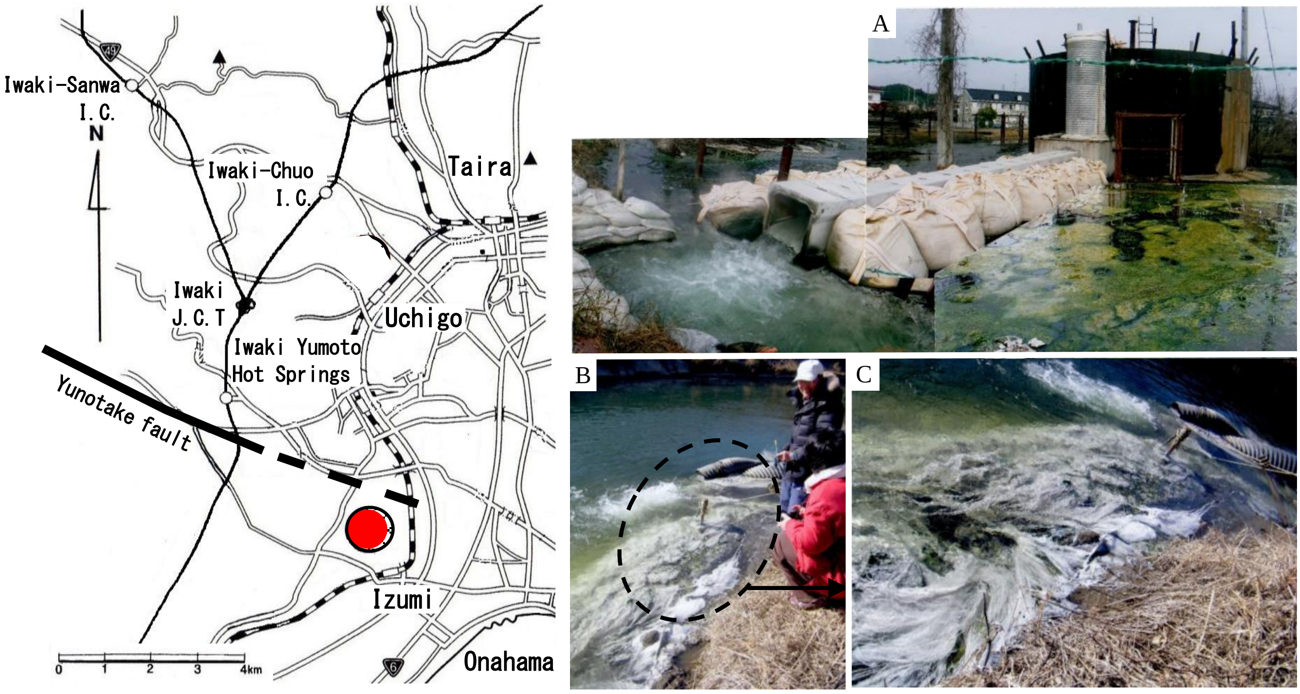

Fig.4-3. Location map of hot spring water gushed out from the shaft in Izumi-Tamatsuyu, Iwaki, Fukushima, Japan

A: Green algae and the white microbial mats were formed in the periphery of the shaft.

B; The white microbial mats were formed near drainage pipe.

C; The white microbial mats were formed of filamentous.

Fig.4-4 X-ray diffraction pattern of the reddish brown microbial mats and the white microbial mats.

A: Reddish brown microbial mats in Uchigo-Takasakamachi.

B; White sulfur turf in Izumi-Tamatsuyu.

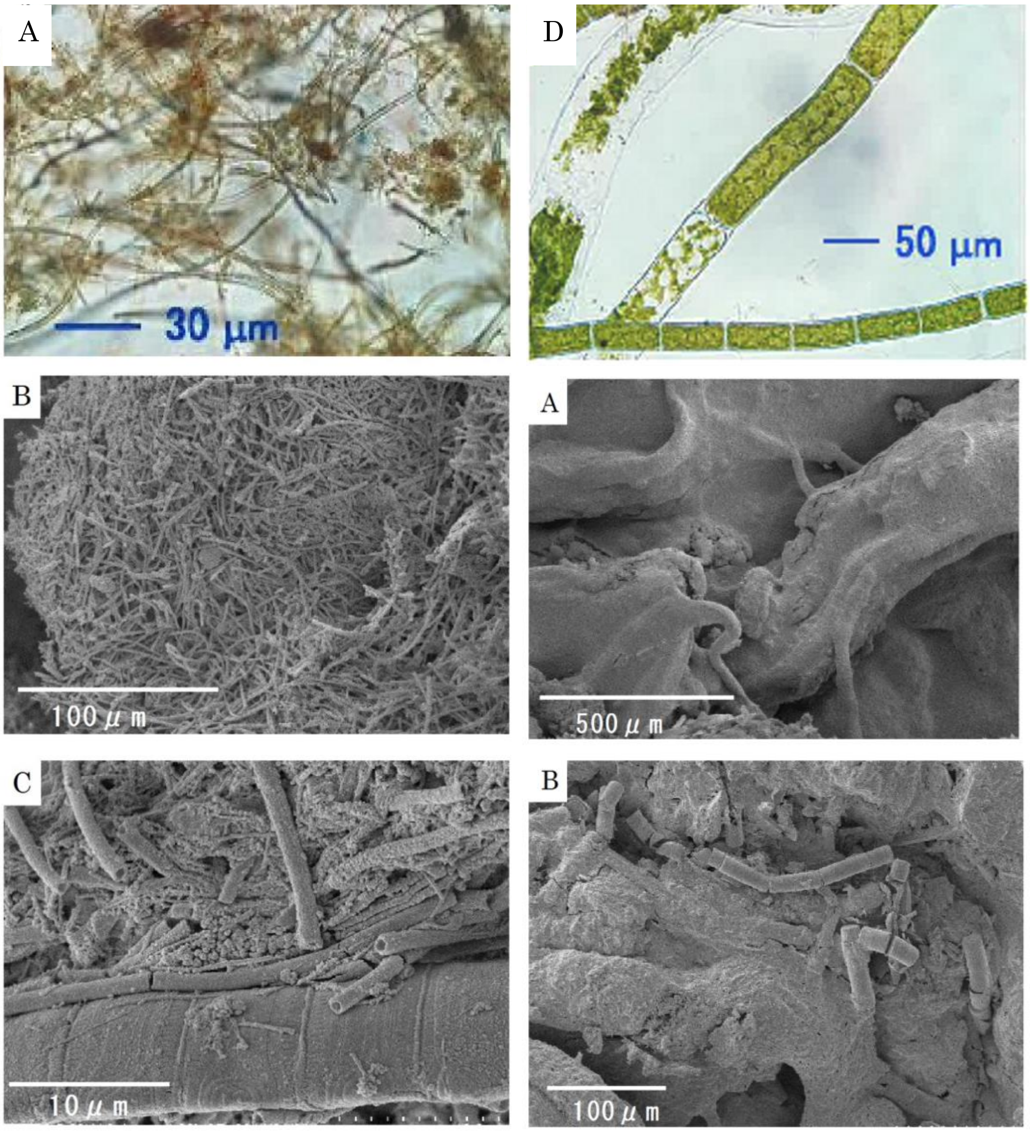

Fig.4-5 Microbial mats observation of Iwaki Yumoto Hot Springs area.

A: Optical microscope observations of reddish brown microbial mats in Uchigo-Takasakamachi.

B: Scanning electron microscopic observation of the reddish brown microbial mats in Uchigo-Takasakamachi.

C: Length 20-30μm, thickness 2-10μm in Uchigo-Takasakamachi.

D: Optical microscope observations of the black microbial mats in Uchigo-Takasakamachi.

E: Scanning electron microscopic observation of the white microbial mats in Izumi-Tamatsuyu

F: Scanning electron microscopic observation of the white microbial mats in Izumi-Tamatsuyu

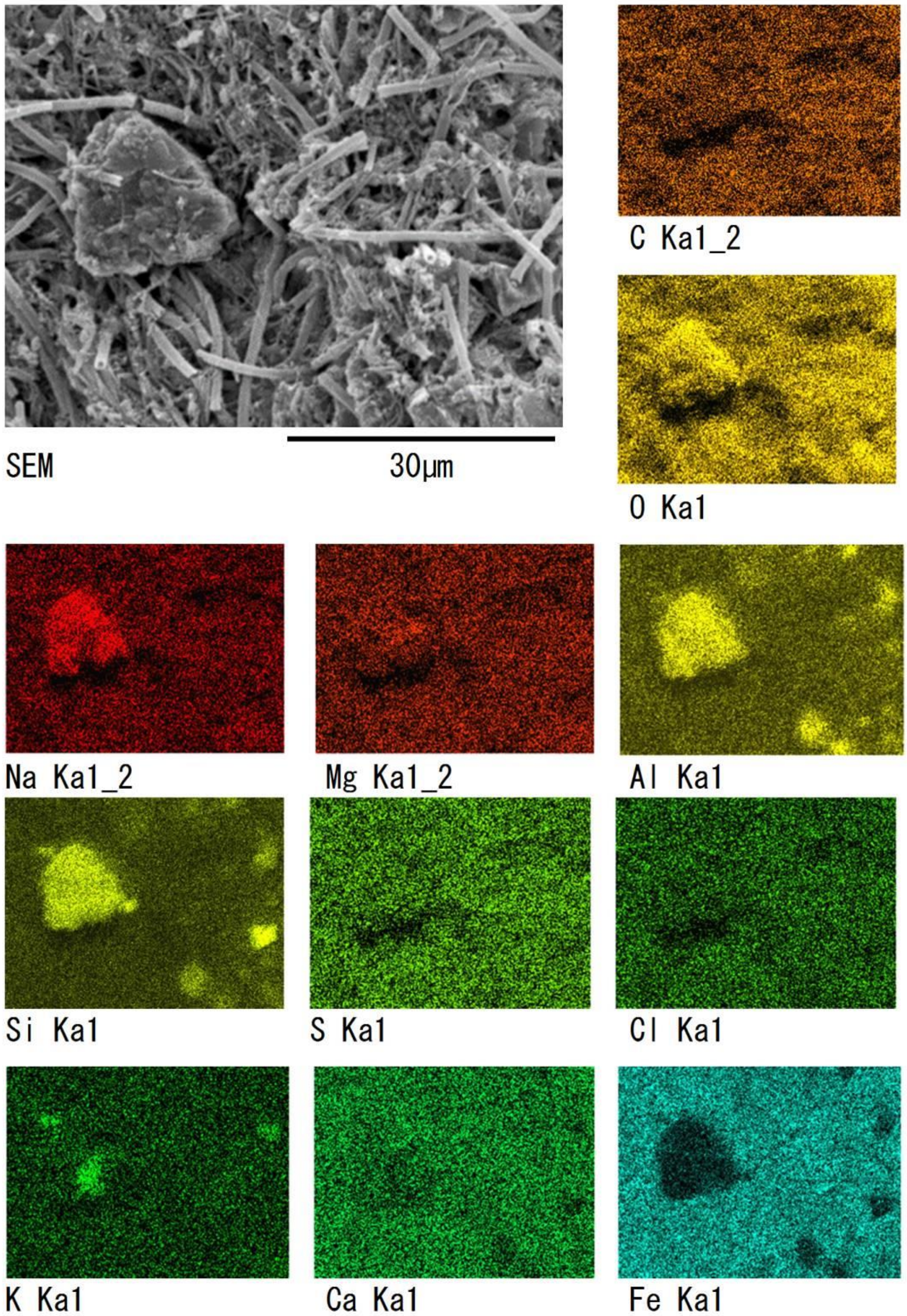

Fig.4-6 Scanning electron micrograph and elemental content maps of the reddish brown microbial mats.

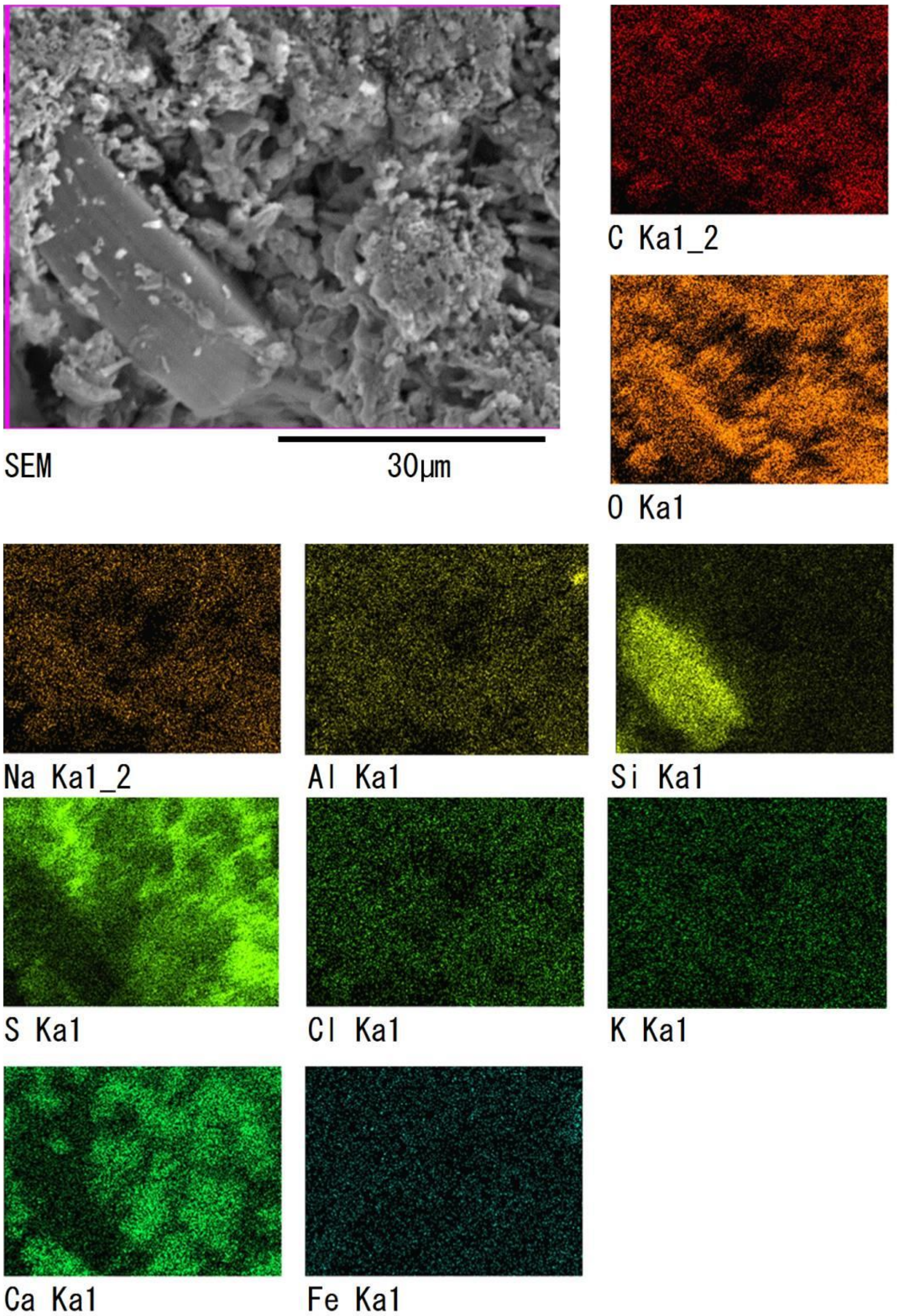

Fig.4-7 Scanning electron micrograph and elemental content maps of the white microbial mats and diatom.

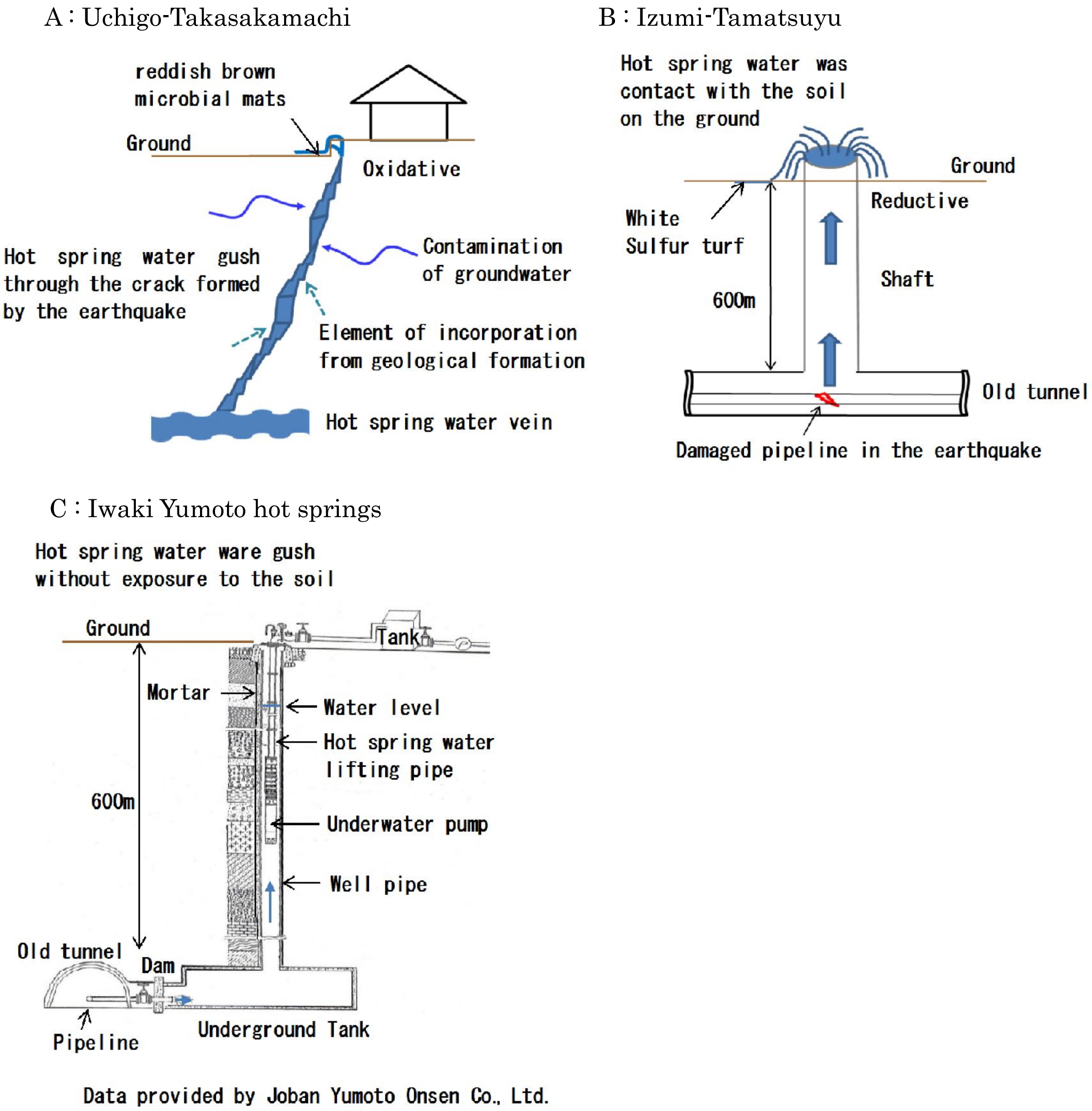

Fig.4-8 Gushed out process of hot spring water

A: The hot spring which was managed by pipeline and well at Iwaki Yumoto hot springs.

B; Hot spring water which climbs into the crack by 4.11 aftershock in Uchigo-Takasakamachi.

C; Hot spring water gushed out from shaft in Izumi-Tamatsuyu

Chapter 5 General discussion

Chapter 1 described the significance and purpose of the present study. Chapter 2 presented a case of formation of the white microbial mats in the paddy field damaged by tsunami in Karasuzaki, Kashima-ku, Minamisoma city, Fukushima prefecture, along with considerations on the microbial mat formation by limited microorganisms due to the environmental change and on the sustainable microbial mat formation environment. Chapter 3 presented a case of the reddish brown microbial mats in the canal for treating spring water in Azumagaoka Park in Haramachi-ku, Minamisoma city, Fukushima prefecture, along with a consideration on concentration of specific trace elements due to the environmental change. Chapter 4 presented a case of microbial mats formed in the hot spring water that gushed out after the earthquake in two regions in Iwaki city, Fukushima prefecture, along with considerations on concentration of specific elements by the microbial mats formed in different water chemistry environments and on a water chemistry change due to the microbial mat formation.

Microorganisms that form microbial mats vary between the two regions. Water chemistry and oxidation-reduction environment also vary between the regions (Table 5-1). This chapter organizes these discussions and discusses microbial mat formation environment, concentration of radionuclides and specific elements, and the possibility of environmental restoration by microorganisms that form microbial mats.

5-1 The formation of the microbial mats

Microorganisms that form microbial mats were observed in different environments such as brackish, spring water, and hot spring water environments. Microorganisms varied between the studied sites, and formed microbial mats also varied therebetween. The paddy field damaged by the tsunami shifted from fresh water environment to brackish environment. Because of this, among a wide variety of microorganisms that had lived there before the tsunami, those who could not adapt to this environmental change could not fully multiply proliferously while limited species who could adapt to the brackish environment drastically multiplied proliferously to form microbial mats. Besides, due to water salination, diatom-eating planktons died, and aquatic insects and fresh water fish were also prevented from propagating. Further, since the submersion was a temporary one due to the tsunami, invasion of marine creatures was also limited. These contributed to the creation of an environment where *Nitzschia acicularis* can drastically multiply proliferously. This seems to be one factor of the microbial mat formation. Likewise, among foraminifers that carried by seawater, new species resistant or adapted to the brackish environment multiplied proliferously to repeat alternation of generations. Thus, it was discovered that the creation of new environment by tsunami allows microorganisms adapted to the new environment to form microbial mats. These results may become the index on estimating environment and the paleoenvironment before and after the tsunami in a tsunami deposit distribution area.

Besides, it was discovered that these microorganisms concentrate elements that brought about by tsunami to form halite and gypsum. Radionuclides were also observed in the microbial mats, and they are adsorbed and immobilized by porous diatoms that make up the microbial mats. The fact that the adsorbed radionuclides decreased in dose by a fifth in a short period suggests that diatoms shielded radiation. In the former paddy field repeating drying and re-moisturization, the microbial mats peeled up, and the formation of new microbial mats on the exposed ground surface was observed. Thus, even though microbial mats are removed, re-moisturization allows sustainable formation of microbial mats.

It is expected that the concentration of halite and gypsum in microbial mats promotes salt removal from paddy fields to contribute to environmental restoration. Besides, radionuclides were observed in the white microbial mats. This suggests that porous diatoms absorbed them. Removal of microbial mats that concentrated specific elements in themselves will help environmental restoration. In particular, microbial mats adsorb halite and radionuclides. Therefore, although it takes a certain period, they may be usable in salt removal or radiation decontamination. Even after four years from the earthquake, some places are left abandoned. The recovery from the earthquake disaster is thus taking time. In such places, environmental restoration using microbial mats will be effective until full-scale recovery is started. Agricultural production is limited in some radiation-contaminated farm lands. There is a possibility that adsorption and immobilization of radionuclides by microbial mats prevent food from taking radionuclides in to prevent secondary contamination to realize earlier recovery of agricultural lands.

Azumagaoka Park's environment did not change significantly before and after the earthquake. Therefore, newly-supplied specific substances will be adsorbed and immobilized through the microbial mat formation process. This suggests that maintaining an environment where the microbial mats are formed also maintains the formation of the microbial mats to allow adsorption and immobilization of newly-supplied specific substances.

In the Joban hot spring district, Iwaki city, different microbial mats were formed since microbe habitat segregation occurred due to the difference in water chemistry depending on the hot spring water gush processes and the difference in oxidation-reduction environment. These microbial mats concentrated specific elements. Microbial mats are formed in watery environments in addition to places where microorganisms can make contact with a stratum, soil, or air. In contrast, microbial mats are unlikely to form in environments where microorganisms can hardly make contact with a stratum or soil, such as managed hot spring facilities.

5-2 Relationship between formation of microbial mats and the water chemistry

The microbial mats were observed in different water environments such as brackish, spring water, and hot spring water environments, and this demonstrated that microorganisms resistant or adapted to local environments lived there. In Karasuzaki, freshwater diatoms were observed although the former freshwater environment had shifted to the brackish environment due to the influence of tsunami. This suggests that such microorganisms are resistant and adapted to a change in water environment.

In the Iwaki Yumoto hot spring district, different microbial mats were observed between two places with gushing hot spring water. This difference is the result of microbe habitat segregation depending on the hot spring water gush processes and on oxidation-reduction environments. In Uchigo-Takasakamachi, hot spring water was oxidized since it slowly rose along cracks under the ground. Meanwhile, the hot spring water took in stratal components and mixed with groundwater to have different water chemistry. This water chemistry change is considered to have created an environment where iron-oxidizing bacteria can easily multiply proliferously. In Uchigo-Takasakamachi, spring water containing iron had been observed even before the hot spring water gushed out. Therefore, presumably, the strata and groundwater in Uchigo-Takasakamachi were suitable for the multiplication of iron-oxidizing bacteria. This suggests that the hot spring water was also affected by this during rising in the strata. Since the total iron concentration was low in Izumi-Tamatsuyu, the hot spring water in Uchigo-Takasakamachi seems to have taken in stratal iron. Besides, it appears that its oxidative environment prevented sulfur-oxidizing bacteria from multiply proliferously unlike Izumi-Tamatsuyu's case, and accordingly, SO_4^{2-} did not decrease. In Izumi-Tamatsuyu region, in contrast, its reductive environment was maintained since the hot spring water gushed out from the vertical shaft at a burst. In addition, the hot spring water did not make contact with any stratum until it reaches the ground surface, and accordingly, hardly underwent a water chemistry change. Because of this, sulfur turf was formed by action of sulfur-oxidizing bacteria.

Microorganisms have adaptability to a wide water temperature range. Although water piled up in the former paddy field in Karasuzaki had wide temperature changes, microbial mats were observed there. In both Azumagaoka Park and Uchigo-Takasakamachi too, microbial mats were formed by iron-oxidizing bacteria even in spring water at a temperature of few degrees and in hot spring water at a temperature around 25°C. In Izumi-Tamatsuyu, sulfur turf was formed even at an ejection hole for hot spring water at approximately 60°C and in a place in a river where discharged hot spring water decreases water temperature. Thus, it was demonstrated that microorganisms that form microbial mats had adaptability to a wide water temperature range.

5-3 Environmental change incident to forming of microbial mats

The microbial mats in Karasuzaki contained radionuclides in addition to tsunami-derived halite and gypsum. Therefore, the formation of microbial mats will promote salt removal from paddy field soil and radionuclide reduction. Thus, microbial mat formation is expected to contribute to environmental restoration of soil contaminated by the tsunami and nuclear accident. The hot springs gushing out in Iwaki city are located in the Joban coal mine area, and Iwaki Yumoto hot spring is located at the approximate midpoint between the two regions. Therefore, presumably, the water chemistry of hot spring water stored under the ground in these regions is not different significantly from that of Iwaki Yumoto hot spring. However, hot spring water that gushed out in these regions after the earthquake had somewhat different water chemistry from that of Iwaki Yumoto hot spring. In addition, specific elements in the hot spring water increase or decrease due to water chemistry changes over the years. Microbial mats thus concentrate specific elements to change water chemistry. In Uchigo-Takasakamachi, iron-oxidizing bacteria multiplied in the hot spring water to raise the total iron concentration and even change the water chemistry. It appears that in Izumi-Tamatsuyu region, its reductive environment was maintained since the hot spring water gushed out from the vertical shaft at a burst, and accordingly, the formation of sulfur turf and the action of sulfur-oxidizing bacteria concentrated sulfur to decrease SO_4^{2-} in the hot spring water to change water chemistry. Gypsum was also formed there, which contributes to reduction of SO_4^{2-}. Thus, water chemistry was changed by action of microorganisms that form microbial mats.

Hot spring water in the Iwaki Yumoto hot springs area contains SO_4^{2-} and sulfur. Therefore, there is a concern that the sulfur component increases environmental load on rivers and canals. With reductive environment formation, the formation of sulfur turf and the action of sulfur-oxidizing bacteria will allow a reduction in SO_4^{2-} to decrease environmental load.

5-4 Elemental concentration by microbial mats

Microorganisms that form microbial mats have an action of adsorbing and immobilizing specific elements. Using this action, removal of microbial mats that concentrated specific elements in themselves may allow environmental restoration. Microorganisms that form microbial mats undergo habitat segregation depending on water chemistry and oxidation-reduction environment, and concentrate specific elements. Different microorganisms concentrate different minerals and give a different influence to water chemistry.

The presence of radionuclides was confirmed by radionuclide analysis in Karasuzaki. Presumably, radionuclides were adsorbed and absorbed by diatoms because they were porous.

The chemical composition analysis and elemental content maps using WD-XRD and SEM-EDX on the microbial mats in Karasuzaki and Azumagaoka in the north of the FDNPP appear that trace elements and elements that may contain radionuclides were concentrated by microorganisms that formed the microbial mats. In contrast, trace elements containing radionuclides were hardly observed from microbial mats in two spots in Iwaki city which is 40 km to the south of the nuclear plant. In the FDNPP accident, radioactive materials diffused mainly to the northwest. These facts suggest that the microbial mats took in radionuclides.

Microorganisms also adsorb mineral particles around their cells. Therefore, when microorganisms with adsorbed radionuclides further adsorb other mineral particles and clay minerals, the radionuclides are covered with them. This may allow shielding of radiation and prevention of diffusion of radionuclides. The fact that covering radiation-contaminated soil with diatom earth reduces its radiation dose also suggests that covering radionuclides adsorbed by microorganisms with clay minerals makes it possible to shield radiation. In particular, the fact that the microbial mats in Karasuzaki are made up of diatoms suggests that they have a high radiation shielding effect. There observed microorganismspecies with an action of concentrating radionuclides. Therefore, it is expected that when radionuclides taken into microorganisms are immobilized together with clay minerals etc., the diffusion of radionuclides and radiation can be suppressed so that secondary exposure due to intake by large animals and plants can be reduced.

The iron-oxidizing bacteria that formed the reddish brown microbial mats observed in Azumagaoka Park and Uchigo-Takasakamachi convert bivalent iron into trivalent iron to obtain energy to metabolize iron precipitate. Iron-oxidizing bacteria also have an action of concentrating manganese in addition to iron. There also reported their action of adsorbing radionuclides. This suggests that when mineral particles and clay minerals other than radionuclides are adsorbed by such microorganisms, radionuclides can be shielded.

It is expected that removal of microbial mats that concentrated specific elements in themselves allows removal of elements that can add environmental load, and this allows environmental restoration and prevention of environmental change.

Table.5-1 Characteristics of microbial mats and water chemistry in Fukushima prefecture

Location	Karasuzaki	Azumagaoka Park	Uchigo-Takasakamachi	Izumi-Tamatsuyu
Date	2013/10/14	2014/1/11	2014/1/12	2014/1/12
Sample	White microbial mats	Reddish brown microbial mats	Reddish brown microbial mats	White sulfur turf
Water chemistry	Na^{+}-Cl^{-} type Brackish water (Tsunami)	Ca^{2+}-HCO_3^{-} type Spring water	Na^{+}(Ca^{2+})-SO_4^{2-} type Hot spring water	Na^{+}-Cl^{-} type Hot spring water
Water temp. (℃)	21.0, 22.0	7.5	24.3	59.0
pH	7.5, 7.7	6.9	7.2	8.0
EC (mS/m)	3480, 4520	36	162	408
ORP (mV)	78, 88 287, 296	22 240	-42 164	-331 -149
Oxidation-reduction state	Oxidative	Oxidative	Oxidative	Reductive
Bacteria	Diatom	Iron oxidizing bacteria	Iron oxidizing bacteria	Reducing bacteria
Mineral	Halite, Gypsum	iron hydroxide	iron hydroxide	Sulfur, Gypsum

Chapter 6　General conclusion

The present study presented actual cases of formation of microbial mats in Fukushima prefecture, along with discussions on their formation situations, formation conditions, formation mechanisms, microorganisms' adaptability to environmental changes, and environmental cleanup by the action of concentrating specific minerals on the basis of water chemistry analysis, chemical composition, form observation, etc. The summary of the research results is as follows.

Since the study site in Karasuzaki was a paddy field, freshwater diatoms were living there. Due to the environmental change caused by the tsunami, the study site was salinated, and seemingly, *Nitzschia* acicularis adapted to the environmental change limitedly multiplied proliferously to form the white microbial mats.

Since the water in the paddy field was salinated, diatom-eating planktons died and aquatic insects and freshwater fish were also prevented from propagating. Further, since the submersion was a temporary one due to the tsunami, invasion of marine creatures was also limited. These seemingly contributed to the creation of an environment where *Nitzschia* acicularis can drastically multiply proliferously. The fact that foraminifers of a characteristic species in inner bays were observed in the paddy field in Karasuzaki and they repeated alternation of generations demonstrated that microorganisms adapted to the environment were living there.

XRD and chemical composition analysis revealed that halite and gypsum were adsorbed and immobilized to the white microbial mats.

Radionuclide analysis revealed that a radioactive material cesium was concentrated in the white microbial mats. The diatoms that make up the microbial mats are porous diatoms. This suggests that the diatoms absorbed and adsorbed the radioactive material.

The water chemistry of water flowing in the canal in Azumagaoka Park indicated figures equivalent to those of common spring water and shallow groundwater. Iron bacteria multiplied proliferously and took in stratal iron to form the microbial mats.

Azumagaoka Park's environment did not change significantly before and after the earthquake. Therefore, newly-supplied specific substances will be adsorbed and immobilized through the microbial mat formation process. This suggests that maintaining an environment where the microbial mats are formed also maintains the formation of the microbial mats to allow adsorption and immobilization of newly-supplied specific substances.

In the Iwaki Yumoto hot spring district, the water chemistry of Iwaki Yumoto hot spring and that of hot spring water that gushed out after the earthquake were compared to infer microbial mat formation environment that varies depending on water chemistry and a water chemistry change due to the microbial mat formation.

In Uchigo-Takasakamachi, the hot spring water rose along cracks formed by the earthquake to gush out on the ground surface. The water chemistry seemingly changed because the hot spring water took in stratal elements, or the hot spring water components seeped into a stratum, and the hot spring water mixed with groundwater. It seems that when the hot spring water gushed out on the ground surface, iron-oxidizing bacteria that concentrate iron existed in strata multiplied proliferously to form the microbial mats. The reason why Na+ and Cl- concentrations were low in Uchigo-Takasakamachi is seemingly due to mixture of the hot spring water with groundwater.

Even though drying and re-moisturization were repeated as in the case of Karasuzaki, microbial mats were formed sustainably. In Uchigo-Takasakamachi, besides, microbial mats cannot be formed in a place dried due to the halt of gush of hot spring water. Any microbial mat formation environment has water, soil, and air. The presence of them allows sustainable formation of microbial mats. With the formation of such environment, a new microbial mat is sustainably formed even if an existing microbial mat is removed, and specific elements are concentrated, so that environmental restoration can be promoted. A drastic action cannot be expected from environmental restoration by microbial mats. In areas damaged by the earthquake disaster, some places have not been recovered even after 4 years from the earthquake. Microbial mats may be usable in a place requiring a long recovery period or as a supplementary means until full-scale recovery is started. There observed microorganismspecies with an action of concentrating radionuclides. Therefore, when radionuclides are adsorbed and immobilized to microbial mats, the diffusion of radionuclides can be suppressed and radiation can also be shielded. Accordingly, radionuclides cannot be taken into food. Such prevention of secondary contamination may allow earlier recovery of agricultural lands.

As for the water chemistry of the hot spring water in Uchigo-Takasakamachi, iron was concentrated by the formation of the reddish brown microbial mats, so that the iron content in the hot spring water rose over 40 times in two years. In Izumi-Tamatsuyu, the formation of sulfur turf decreased SO42- in the hot spring water by approximately two thirds in two years. SO42- is approximately one fourth of that of hot spring water of Iwaki Yumoto hot spring. Not only sulfur but also gypsum was formed in the white sulfur turf. This is a cause of the decrease in $SO4^{2-}$. Thus, microorganisms concentrate specific elements by biomineralization to change even water chemistry environment. The difference between the microbial mats formed in the two regions was brought about by a water chemistry difference caused in the hot spring water gush processes and by microorganisms' habitat segregation due to an oxidation-reduction environment change. Nonetheless, it was demonstrated that the formation of microbial mats changes water chemistry.

The hot spring water that gushed out in the Iwaki Yumoto hot springs area is discharged into side ditches and rivers without being treated, which raises concern about a water environment change. Such a water chemistry change by the formation of microbial mats will help the recovery of water environments.

Microorganisms that form microbial mats have an action of concentrating specific elements such as iron and sulfur contained in hot spring water. Even in a place that underwent a significant environmental change, microorganisms resistant or adapted to the environment multiply proliferously to form microbial mats to concentrate specific minerals. Even if drying and re-moisturization are repeated, these microorganisms sustainably form microbial mats.

The elemental concentration action by microbial mats and sustainable microbial mat formation can be expected to contribute to environmental restoration of contaminated water and watersides. Besides, the radioactive element adsorption action can be expected to contribute to decontamination of radioactive materials raising concern about their long-term influence after the nuclear plant accident. The environmental restoration by native microorganisms observed in the present study does not require large-scale machines or plants. Therefore, it can be expected as an inexpensive method. In addition, a sustainable effect can be expected. The use of native microorganisms will allow prevention of a significant ecosystem change and prevention of a change in original local environment.

Fukushima prefecture is facing damages caused by the tsunami due to the earthquake as well as radiation contamination caused by the FDNPP. Particularly, the radiation contamination has a long-term influence. Therefore, decontamination is important. Decontamination work is carried out in urban areas and residential areas. However, wastes generated in the decontamination work are packed into large sandbags and piled outdoors. Further, there has been a delay in construction of interim storage facilities. When decontamination using microbial mats becomes available, a sustainable effect can be expected and microbial mats used in decontamination can be significantly downsized through water removal or incineration. Therefore, it can be expected as an effective decontamination means. Radionuclides can be immobilized by action of microorganisms so that the diffusion of radionuclides can be prevented. Besides, secondary exposure due to intake by other animals and plants can also be prevented.

Since decontamination requires a long period, it must be carried out continuously, and it is also important to maintain and manage machines and facilities. Decontamination using microbial mats made up of native microorganisms can be carried out at simple facilities, and the maintenance and management are also easy. Therefore, it does not require advanced scientific knowledge or complicated machine operation. Thus, it is a method that local residents can also carry out. Environmental restoration using microbial mats takes time. Therefore, it will be effective to supplementally carry out environmental restoration using microbial mats until full-scale recovery is started in abandoned places falling behind in recovery.

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