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 要 旨

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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Abstract

The characteristics of microbial mats formed after “The 2011 off the Pacific coast of Tohoku Earthquake” were clarified by studies of their living environment, chemical compositions and microscopic observation at study sites of Fukushima prefecture.

In Minamisoma city, the microbial mats were formed in paddy fields damaged by tsunami. These microbial mats consist largely of diatom with mineralized halite and gypsum. The radionuclides of Cs by Nuclear Power Plant accident were detected in microbial mats. This suggests that the porous diatoms absorbed and adsorbed the radioactive material. The microbial mats formed by *Nitzschia* sp. was resistant or adapted to the brackish water appeared in the environment change caused by tsunami. This result may become the index on estimating environment before and after the tsunami event in a tsunami deposit distribution area.

In Iwaki Yumoto hot springs area of Iwaki city, hot spring water gushed out at two locations after the aftershock happened on 11th April 2011. In two locations, different microbial mats were formed, and these differences 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. Furthermore, the change of the chemistry of hot spring water was caused by the concentrations of particular elements in microbial mats.

These results show the importance of microbial mats as the index of the environmental change with the disaster.

1 Introduction

"The 2011 off the Pacific coast of Tohoku Earthquake" (henceforth 3.11 Earthquake) occurred on March 11, 2011. Tsunami was occurred by the 3.11 Earthquake. The structures or farmland suffered serious damage by this tsunami. Above all, radioactive materials were scattered on wide area by Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. After one month of the 3.11 Earthquake, the inland aftershock occurred in Hamadori, Fukushima prefecture on April 11, 2011 (henceforth 4.11 aftershock). After the 4.11 aftershock, the 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 also gushed out from the shaft of old coal mine in Izumi-Tamatsuyu, Iwaki city.

The white microbial mats were formed in the paddy field damaged by the tsunami at Krasuzaki, Kashima-ku, Minamisoma city, Fukushima prefecture 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 four places in this area were studied.

The characteristics of the microbial mats were discussed from chemical constitutions by WD-XRF, microbial mats origin minerals by XRD, micro morphological observations by SEM-EDX, water chemistry by ion chromatograph, and radioactive nuclide analysis by Ge semiconductor analyzer. 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 microbial mats formation, and biomineralization, were discussed.

2 Characteristics of Microbial Mats at Krasuzaki, Kashima-ku, Minamisoma City, Fukushima Prefecture

The study site is located in Karasuzaki, Kashima-ku, Minamisoma city in the northeastern part of Fukushima prefecture. This site is a flat land that was used as a paddy field, approximately 29 km to the north from the FDNPP. White microbial mats were formed on the surface of the paddy field, and puddles were also seen in dips in the vicinity. 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.

(1) Formation environment of the microbial mats

In this 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. The water chemistry showed Na+-Cl- type. 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. 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. Foraminifers of species that is characteristic in inner bays were observed in the paddy field in Karasuzaki and they repeated alternation of generations (Nemoto et al., 2014 ; Nemoto et al., 2015). These results may become the index on estimating environment and the paleoenvironment before and after the tsunami event in a tsunami deposit distribution area.

(2) Bomineralization

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 the chemical interactions (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 the element of Ca ibcluded in gypsum is a little in the microbial mats, but the element of 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.

(3) Microorganism and the radionuclide

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

3 Characteristics of Microbial Mats in Azumagaoka Park, Haramachi-ku, Minamisoma City, Fukushima Prefecture.

The study site is located in Azumagaoka Park, Haramachi-ku, Minamisoma city. 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. I studied the elemental concentration action that microbial mats perform in an environment contaminated by radioactivity due to the accident of the FDNPP.

The water chemistry showed Ca^{2+}-HCO_3^- type, that was water chemistry general in springwater. The reddish brown microbial mats in Azumagaoka Park were

formed by iron-oxidizing bacteria *Leptothrix ochracea.* 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 by XRF analyses. The mineralogy of the reddish brown microbial mats, as determined using XRD analysis, contains amorphous materials. On the other hand, the clay underneath the reddish brown microbial mats contains mainly minerals, indicated quartz, feldspars and clay minerals.

The amorphous iron hydroxide was formed by the iron bacteria from SEM observation, XRD and elemental analysis results. The element of Fe is collected in the tubular substance formed by iron oxidation bacteria on elemental content maps, and particular element and trace element were distributed. 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 the element of Sr is confirmed by a XRF analysis, and it is shown that Sr is concentrating on a diatom by the elemental content maps. There are reports on actual cases of efficient collection of radionuclides by iron bacteria in uranium deposits and muck dumping sites (e.g. Waite et al., 1994). 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 spectrum obtained with TEM-EDX (Akai, 2011).

Azumagaoka Park's formation 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.

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

In Iwaki Yumoto hot springs area of Iwaki city, Fukushima prefecture, hot spring water gushed out at two locations. In Uchigo-Takasakamachi, Iwaki City, hot spring water at approximately 25 to 27°C gushed out from the foundation and drainpipes of a house. Reddish brown microbial mats were formed around the foundation and drainpipes of a house. In Izumi-Tamatsuyu, Iwaki City, a large amount of hot spring water at approximately 55°C gushed out from the shaft of abandoned coal mine facilities. White microbial mats were formed on the interface between the hot spring water and the ground. Besides, white microbial mats was observed in the drainage channel to the river. I studied the water chemistry of hot springs, microorganisms and biomineralization, and formation environment by comparing two hot springs.

In Uchigo-Takasakamachi, oxidation-reduction potential was ORP -42 to 78mV (Eh 168 to 284 mV). The hot spring water showed oxidative environment in Uchigo-Takasakamachi. The water chemistry of hot spring water showed the $Na^{+}(Ca^{2+})$-SO_4^{2-} type in Uchigo-Takasakamachi. The reddish brown microbial mats are formed by the iron bacteria *Leptothrix ochracea*, and concentrate iron hydroxides. As for the water chemistry of hot spring water in Uchigo-Takasakamachi, the iron ion concentration rose over 40 times in two years. Iron contained in hot spring water was concentrated by the iron-oxidizing bacteria, so that the water chemistry was changed to one with more iron content. In Uchigo-Takasakamachi, hot spring water gushed out via cracks under the ground. The hot spring water passed through the cracks under the ground, made contact with soil, and mixed with groundwater. This changed the water chemistry and created the oxidative environment. Therefore the iron-oxidizing bacteria formed the reddish brown microbial mats when the hot spring water gushed out.

In Izumi-Tamatsuyu, oxidation-reduction potential was ORP -194 to -331 mV (Eh -3 to -149 mV). The hot spring water showed reductive environment in Izumi-Tamatsuyu. The hot spring water in Izumi-Tamatsuyu gushed out because the 4.11 aftershock destroyed underground pipeline in the Iwaki Yumoto hot spring area. Therefore the water chemistry was the reductive environment. The white microbial mats are formed by the sulfur-oxidizing bacteria, and concentrate sulfur

and gypsum. 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 springs indicated a somewhat different water chemistry since it is the Sulfur-Na^{+}-Cl^{-}・SO_4^{2-} type. This indicates that the white microbial mats formed in the hot spring water concentrated hot spring components SO_4^{2-} and HS^{-} as sulfur by the action of sulfur-oxidizing bacteria. Further, gypsum in the white microbial mats is made of Ca^{2+} and SO_4^{2-} contained as hot spring components. This is another cause of decrease in SO_4^{2-} which is a hot spring component.

The hot spring water in Uchigo-Takasakamachi contained over 10 meq/L of SO_4^{2-}. But the water chemistry was the oxidative environment as compared to Izumi-Tamatsuyu. Therefore, these did not allow sulfur-oxidizing bacteria to live in the hot spring water. On the other hand, the water chemistry was the reductive environment in the hot spring water of Izumi-Tamatsuyu. Therefore, these did not allow iron-oxidizing bacteria to live in the hot spring water. In Iwaki Yumoto hot springs, 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.

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-reduction environment. Different microorganisms concentrate different elements. By analyzing the reration between concentrated element and microorganisms, 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 quality of hot spring water.

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学位論文審査報告書（甲）

１．学位論文題目（外国語の場合は和訳を付けること。）

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

（和訳）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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３．審査結果の要旨（600～650字）

本学位論文について、平成28年1月28日に、第1回審査会を開催し審査方針等について検討した。同日、公聴会を開催した後、第2回審査会を開催し、協議の結果，以下の結論を得た。

本論文は、福島県内のバイオマットの特徴を明らかにし、東北地方太平洋沖地震（以下「東日本大震災」とする）による津波、地盤の変化及び原発事故がバイオマットに与えた影響を調査、考察している。まず、津波の影響について、影響の無かった淡水地域には見られない汽水・海水に適応した珪藻の繁殖及びハライト、石膏などの蒸発鉱物を確認し、珪藻（バイオマット）にはセシウムなどの微量元素が含まれることを見出している。他方、いわき市地域では、住宅地ならびに旧炭田の立坑から地震の影響で噴出した温泉水が、その湧出過程の違いにより泉質が酸化的なものと還元的なものに変化し、その結果、異なるタイプのバイオマットが形成されたことを見出している。以上より、本論文は東日本大震災に起因する津波及び地盤の変化に適応して生成されたバイオマットの特徴を明らかにしており、災害に伴う環境変化の指針としてのバイオマットの重要性を示している。本論文は英文で書かれており、霜島氏には国際的なコミュニケーション能力があると判断できる。更に、本研究の重要な部分は既に学術雑誌等に掲載されている。以上から、本審査委員会は、本論文は環境地球科学分野の発展に寄与する成果を含んでおり、霜島 康浩氏に博士（理学）の学位を授与するに値すると判断した。

４．審査結果　(1) 判　　定（いずれかに○印）　○合　格　・　不合格

(2) 授与学位　　博　士（　理学　）

５．学位論文及び参考論文に不適切な引用や剽窃が無いことの確認

■　確認済み（確認方法：剽窃確認ツール iThenticate を利用して確認した。　　）

□　未 確 認（理　　由：　　）