Interaction of Radioactive Substances, Salts and Microorganisms in Paddy Soils in Dodoma, Tanzania

博 士 論 文 要 旨

Interaction of Radioactive Substances, Salts and Microorganisms in Paddy Soils in Dodoma, Tanzania

(和題：タンザニア・ドドマ州の水田土壌における放射性物質、塩類、微生物の関係)

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Abstract

Microorganism tolerance to high radioactivity and salinity was discovered in the soils deposited from the weathered granite in Bahi Swamp of Dodoma Region, Tanzania, which has an area of 125 – 140 km^2 and located approximately 50 km to the west of Dodoma. This swamp is underlain by layers of black clayey soils about 110 m thick. Some of the clay layers have been found to be radioactive due to their content of uranium-bearing minerals such as uranyl vanadates; (Ba, Pb)$(UO_2)_2V_2O_8 \cdot 5(H_2O)$. The β (γ)-ray counts and trace elements were analyzed respectively by a scintillation counter and X-ray fluorescence (XRF) spectrometer. Salt and black clayey soils showed their γ -ray radioactivity ranging from 130 to 200 cpm and contained Sr abundant, and Ga, Rb, Y, and Zr slightly less abundant, whereas La, Ce, Th and Ni detected slightly. Besides, their mineralogical characteristics were analyzed by using X-ray powder diffractometer (XRD), which identified clay minerals such as smectite, mica clay minerals and kaolin minerals whereas halite and bischofite in salts. The presence of microorganisms was observed with Scanning Electron Microscopy (SEM) equipped with Energy Dispersive X-ray Analyzer (EDS), and Transmission Electron Microscopy (TEM). Filamentous bacteria were fully covered with flakes of clay minerals, which are recognized as kaolinite from its hexagonal particles. The clay minerals such as kaolinite, smectite, etc. could have functions to alleviate or even absorb the effect of the salinity and the radiation. This is the first report revealing high radioactivity of paddy soils and salt associated with microorganisms in Dodoma, Tanzania.

1 Introduction

High radioactive materials are toxic substance for human health that is widely distributed throughout the hydrosphere, biosphere, and lithosphere of the Earth. Paddy soils and salts in uranium mine environments, associated with weathered granite, support a wide diversity of microbial life in Tanzania. The microbial ecology of environments where uranium mine area discharges radioactive elements and heavy metals to the surroundings is poorly understood. Here, I describe the features of microorganisms in high radioactive-contaminated paddy soils and salt in a uranium mine in Bahi Swamp in Dodoma, Tanzania (Fig. 1.). The Bahi Swamp indicated high β (γ)-ray level at fresh granite, weathered granite, sand, paddy soils and salt. It was understood that under such an environment, microorganism thrive actively by adapting themselves to the local environment.

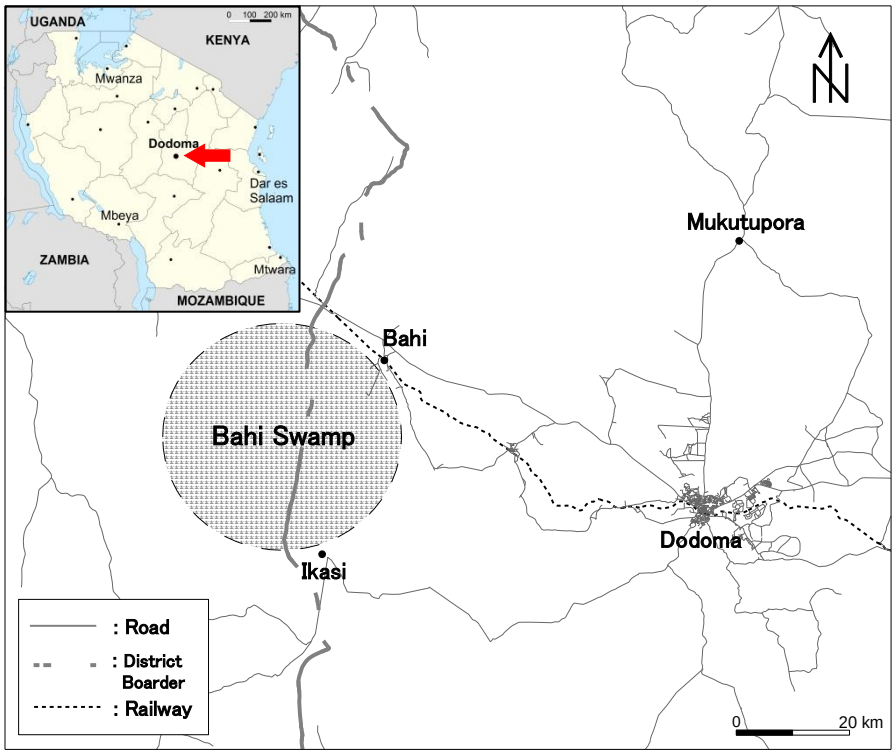


Fig. 1. Location Map of Bahi Swamp, Dodoma, Tanzania

Ministry of Water, Tanzania reports that by June 2014, the access rate to clean and safe water in rural areas of Tanzania has increased to 51% of rural population, which is equivalent to 19.4 million (Tanzania National Bureau of Statistics, 2012). This means that nearly half of rural population depends on the traditional water sources including dug well, river stream, seasonal ponds etc. without any treatment facility. On the other hand, the situation of water supply in the vicinity of Bahi Swamp is worse than that of National level. It was revealed based on the

computation using National Census (2012) data and Water Point Mapping (wpm.maji.go.tz) that approximately 67% of those who live for agriculture, fishing, etc. around Bahi Swamp have no access to protected water sources with a certain water quality allowable to Tanzanian standard.

The occurrence of uranium deposits in and around Bahi Swamp has been reported widely but no effective countermeasure and consideration has not been introduced by authorities responsible for protecting life and health of habitants surrounding Bahi Swamp. Legal and Human Rights Center of Dar es Salaam (2014) reports that a group of farmers expressed their concerns on the impacts from the occurrence of surficial uranium deposit because they got some skin and eye irritations during working in fields around Bahi Swamp. Moreover, Dr. Monburi of Geological Survey of Tanzania indicates high possibility of health impacts from consumption of locally produced agricultural products (rice, fish, salts, etc.) polluted with radioactive substances and heavy metals in and around Bahi Swamp (Monburi et al., 2009).

As summarized in the previous sections, due to the occurrence of uranium deposit and mineral leaching from weathered granites, the radioactivity in the ambient air around Bahi Swamp shows rather higher than other parts of Tanzania and aquifers of groundwater in Bahi Swamp contain high concentration of Na, K, Cl, etc. as major elements of salts. In order to strategize and develop future measures for solve or mitigate negative impacts from radioactive substances and other minerals present around Bahi Swamp, the application of bioremediation could be one of potential approaches, which need to be economically and scientifically relevant. However, there is no such report on the ecology of microorganisms indigenously to Bahi Swamp.

Therefore, the ecology of microorganisms in farming fields (paddy soil) associated with clay minerals under high radioactivity and high salt concentration shall be examined and discussed in this study: Interaction of Radioactive Substances, Salts and Microorganisms in Paddy Soils in Dodoma, Tanzania.

2 Characteristics of radioactive paddy soils, salts and microorganisms in Bahi swamp, Dodoma region, Tanzania

(1) Samples and materials

The fresh and weathered granite, paddy soils, and ground water about 4 - 12 m in depth were collected at nine locations, in December 2010, from Bahi Swamp with an area of 950 km^2 located approximately 35 km to the west of Dodoma, Makutupora well field and Dodoma Town. In this study, salts and paddy soils collected in Bahi Swamp were intensively analyzed.

During the rainy season, surface sediments were wet in Bahi Swamp thereby polluting and accumulating radioactive materials by sediments underneath of the lake bottom, because of the uranium mine under lower layers in this region. The mineral salt occurs on the top of sedimentary deposit in the swamp during the dry season in Bahi. Local farmers made small holes to collect salty water from underground, boiling brown water in the tin, and refine away to be clean white salts. Local farmers have been cultivating this area at the vicinity of uranium mine for growing rice and other cash crops. The salt samples were collected from Bahi Swamp in both 2011 and 2015.

(2) Results and Discussion

A unique ecosystem composed of microorganisms and clayey sediments is able to thrive even in spite of radioactive heavy metal contaminated soils and salt (Tazaki et al., 2015; Tazaki, 2015; Mann et al., 1985; Suzuki et al., 2003).

In this study, it was found that microorganisms could thrive in high salinity and radioactive substances at Bahi Swamp where uranium mine underlays in Dodoma, Tanzania (Table 1, Table 2 and Fig. 2). The SEM and TEM observation, especially, revealed for the first time that radioactive elements were taken up by filamentous microorganisms in paddy soils and salts originated from weathered granite (Fig. 3). Although no radionuclide was so far detected, it was demonstrated by Lovely et al. (1991) that the Fe (III) reducing bacteria *G. metallireducens* and *S. oneidensis* could selectively utilize and absorb radionuclides of U through their metabolism and also suggested the possibility of microbial reduction of radionuclides including ^{230}Th, ^{237}Np, etc. Furthermore, Shimojima (2016) also revealed that microbial mats formed by mainly iron oxidizing bacteria *Leptothrix ochracea* could have absorbed radionuclides such as ^{137}Cs and^{134}Cs originated from a nuclear power plant in Fukushima.

Metal uptake, precipitation and fixation, including absorption by clay minerals (smectite and kaolinite), improves the water quality for surrounding organisms. Microorganisms are as such involved in heavy metal accumulation and converting radioactive elements to clay minerals (Tazaki 2015; Cygan et al., 2014; Mann et al., 1986b).

Table 1. XRF analyses of paddy soils and lake sediment at Bahi in Tanzania

Anzlized on 12 January, 2016

Element	Paddy soils (mass%)	limited	Lake sediments (mass%)	limited
O	50.1	0.22353	44.7	0.21278
Si	27.4	0.00799	8.30	0.00478
Al	7.02	0.00711	4.19	0.0065
Na	3.81	0.01654	17.2	0.02691
Cl	3.20	0.00868	8.74	0.01405
C	2.70	0.07989	3.85	0.09627
K	2.06	0.00531	0.7300	0.00354
Fe	1.32	0.00179	1.17	0.00208
Mg	0.7520	0.00994	2.99	0.01253
Ca	0.6580	0.00258	1.83	0.00326
S	0.5130	0.00157	6.00	0.00347
Ti	0.1840	0.0051	0.1230	0.0046
W	0.1520	0.00297	N.D.	N.D.
Ba	0.0553	0.01991	N.D.	N.D.
Mn	0.0544	0.00195	0.0376	0.0019
Co	0.0221	0.00153	N.D.	N.D.
Br	0.0165	0.00071	0.0317	0.00068
P	0.0154	0.00112	0.0290	0.00123
Sr	0.0141	0.00061	0.0066	0.00061
Rb	0.0125	0.00346	0.0073	0.00338
Zr	0.0123	0.00323	N.D.	N.D.
Cr	0.0078	0.00229	0.0047	0.00214
Pt	0.0040	0.00229	N.D.	N.D.
Ni	0.0027	0.0012	0.0019	0.00112
Cu	0.0020	0.001	0.0014	0.00097
Ga	0.0017	0.00088	0.0010	0.00086
Zn	0.0015	0.00096	0.0020	0.00088
As	0.0011	0.0008	0.0009	0.0008
Sn	N.D.	N.D.	0.0055	0.0022
Total	100.0924		99.95264	

N.D.: Not Detected

Table 2. Trace elements in salt, paddy soils, and weathered granite in Bahi (mg/kg)

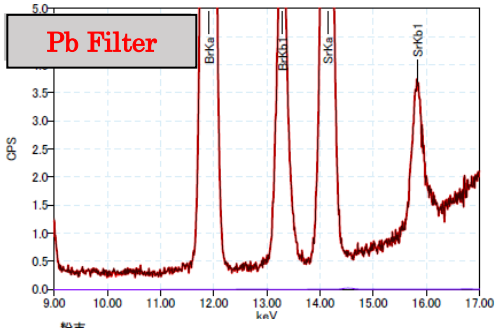
Analyzed on 12 January, 2011

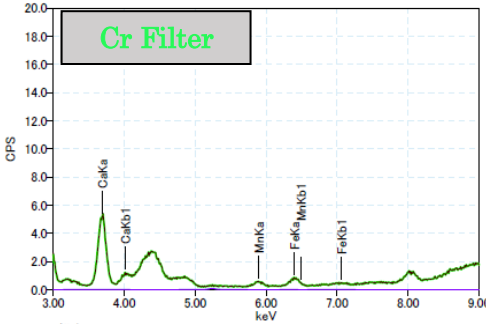
Sample No.	$\beta(\gamma)$ - ray (cpm)	Cd	Pb	Cr	As	Se	Hg	V	Cu	Zn	Ga	Rb	Sr	Y	Zr	Ag	Sn	La	Ce	Th	Ni
Salt	130-150	ND	ND	ND	ND	ND	ND	ND	tr.	ND	ND	**ND**	**abund.**	ND	ND	ND	ND	ND	ND	ND	ND
Paddy soils	160-200	ND	21	120	ND	ND	ND	160	40	94	26	**173**	**118**	29	136	ND	ND	tr.	tr.	tr.	tr.
Paddy wet soils	120-130	ND	31	34	ND	ND	ND	62	22	67	18	**140**	**136**	21	539	ND	16	tr.	tr.	tr.	tr.
Weathered granite	160-180	ND	19	70	ND	ND	ND	92	37	69	23	**131**	**113**	130	373	ND	15	tr.	tr.	tr.	tr.

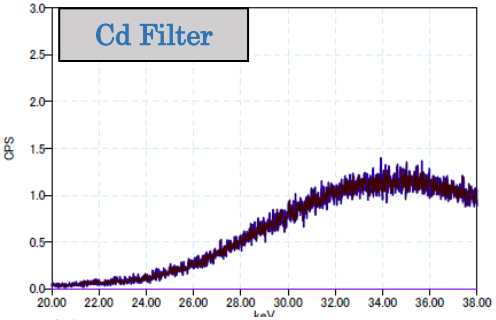
Abund.: Abundant, ND: Not Detected, tr.: Trace

Peak Chart

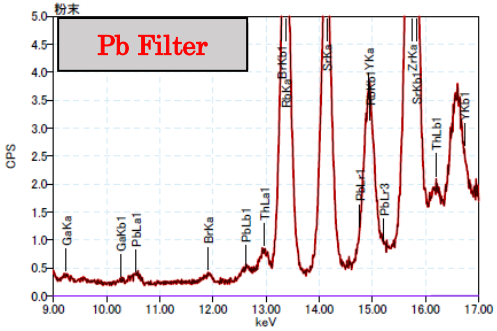
Salt

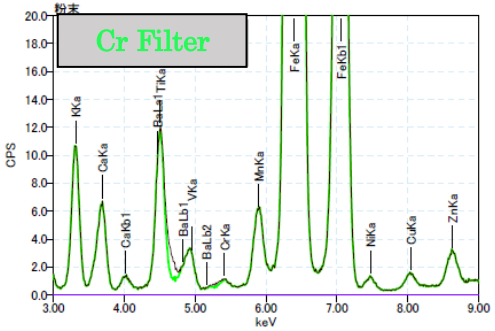


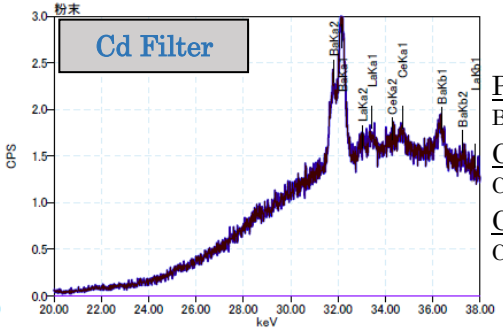




Paddy Soil







Pb filter
Br, Rb, Sr, Y, Zr, Pb, Th

Cr filter
Cd, Ag, Sn, Sb, Ba

Cd filter
Cr, Cu, Zn

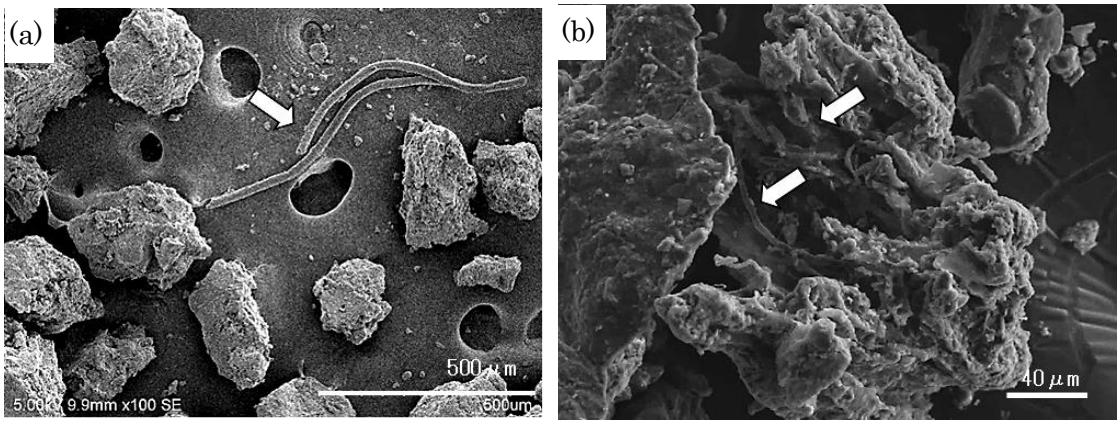


Fig. 2. SEM images (a, b) of paddy soils associated with filamentous bacteria (an arrow) and soil granules aggregating soil particulates and organic substances in Bahi, Dodoma in Tanzania.

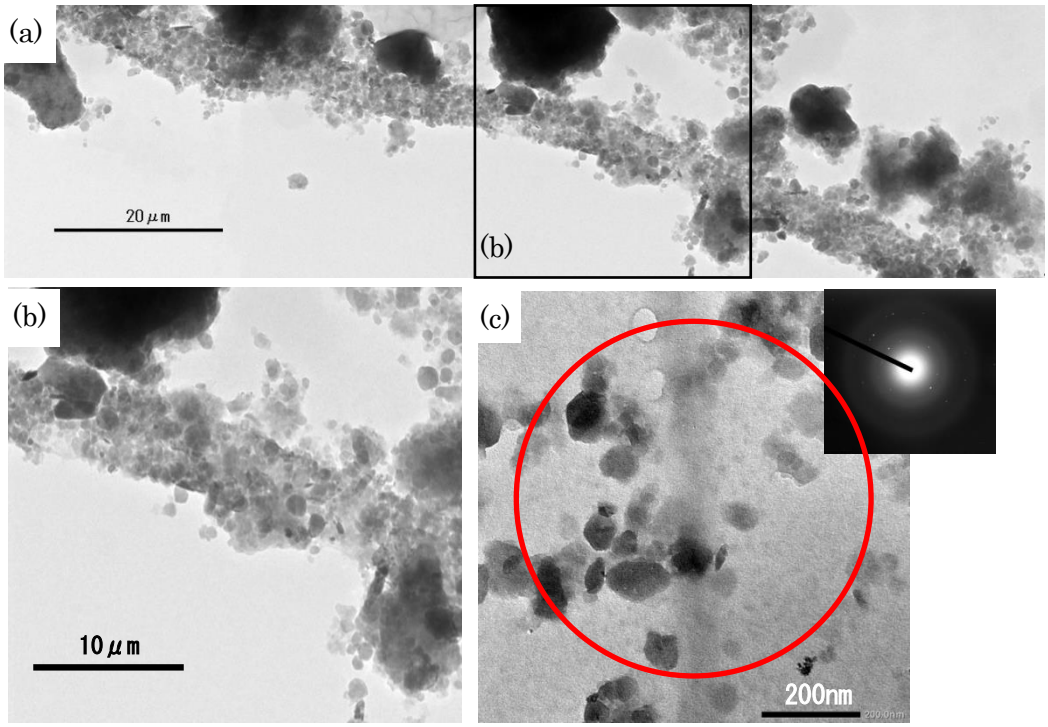


Fig. 3. TEM images of filamentous bacteria covered with minute clay particles (a, b) and results (images) of electron diffraction analysis on filamentous bacteria covered with minute clay particles in paddy soils (c) in Bahi, Dodoma in Tanzania.

Abundant intercellular and cell walls with microcrystalline smectite and kaolinite have been identified in filamentous microorganisms recognized fluently by SEM-EDS and TEM imagery. The microorganisms thrive in uranium mine environments and the groundwater from mines support a wide diversity of microbial life. The microbial ecology of environments, where uranium mine sediments are deposited in paddy soils and salt, are polluted with heavy metals and radioactive substances in Bahi Swamp. Especially, uranium contaminated

paddy soils and salt could cause very serious problems to groundwater for water supply and consequently food chain including rice, maize, fish, and salts which are locally produced in Bahi Swamp. This tendency was detected only in black clayey soils, though Th and U were common elements in Bahi Swamp. Sr was abundant and Rb, Ga, Y, and Zr were slightly less abundant.

Filamentous bacteria were fully covered with flakes of clay minerals, which are recognized as kaolinite from its hexagonal particles, as if they used these clay minerals for their armor in order to protect themselves from high salinity and radiation. Besides, considering absorption of minerals by clay minerals, biosorption of minerals, inclusion of radioactive substances in salts and moreover multilateral interaction within this ecology of Bahi Swamp, it was understood that radioactive substances and heavy metals have been entrapped biologically and physically in the surface soils and salts. This result strongly supports the application of bioremediation using native microorganisms in Bahi Swamp for conserving and improving the life of inhabitants around Bahi Swamp in a sustainable manner. Tazaki et al. (2015) has reported a similar situation where paddy fields in Fukushima, Japan were polluted with salts and radioactive substances caused after the tsunami (March 11, 2011) and Fukushima Daiichi Nuclear Power Plan (FDNPP) accident, respectively. Accordingly, it was found that the clay minerals in paddy soils interacted with radionuclides of I, Cs, Sr, etc., especially Cs and K were adsorbed in the interlayer sites of 7 Å clay minerals (kaolinite), which could have a function to alleviate the radioactivity from radionuclides of Cs and K hazardous to bacteria.

This is the first report revealing the ecology of microorganisms under high radioactivity of paddy soils and salts in Bahi Swamp, Dodoma, Tanzania.

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平成２９年　２月　３日

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。）

Interaction of Radioactive Substances, Salts and Microorganisms in Paddy Soils in Dodoma, Tanzania

（和訳）タンザニア・ドドマ州の水田土壌における放射性物質、塩類、微生物の関係

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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 29 年 2 月 3 日に、第 1 回審査会を開催し審査方針を検討した。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、タンザニア・ドドマ州バヒ湿地帯周辺の空間放射能を測定するとともに、水、塩類、水田土壌を構成する元素及び土壌や塩類に生息する微生物ついて、調査分析を行っている。まず、バヒ湿地帯周辺の水を分析し、その塩分濃度が高いことを定量的に示し、その原因を考察した。また、湿地帯の粘土層や塩類について、微量元素分析及びβ(γ)線測定を行い、Sr 等の微量元素が存在を明らかにし、バヒ湿地帯付近で観測した周囲より高い空間放射線量の原因の可能性を推定した。更に、それらの中に微生物が生息することも確認し、微生物と土壌鉱物及び塩類との関係を、電子顕微鏡観察等によって明らかにした。タンザニアにおいて、このような水田土壌・塩類・微生物の関係を、構成元素やμスケールの観察で解明した研究は例がなく、バヒ湿地帯の環境について、基礎的な情報を与えるものである。得られた情報をもとに更なる考察を加えることにより、バヒ湿地帯の土壌環境についての理解を深めることが期待される。本論文は英語で執筆されていることから国際的コミュニケーション能力があると判断できる。また、本研究の一部は査読付きの学術雑誌に掲載されている。以上のことから、本審査委員会は、本論文はタンザニア地域の環境科学研究の発展に貢献する成果を含んでおり、加藤智弘氏に博士（理学）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）