Report on “Radiation Disaster Recovery Studies”

Course’ Environmental Radioactivity Protection
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○Regarding “Radiation Disaster Recovery Studies”

（Describe your thoughts, the process you engaged in and your research progress regarding Recovery from Radiation Disaster. ）

Fukushima Daiichi Nuclear Power Plant accident occurred in March, 2011. We noted with great concern that the Fukushima nuclear disaster and radiation protection measures introduced during recovery phases brought significant consequences for the daily life of the afflicted people. For example, the people had to be reallocated with strict measures and restrictions on foods which made their lives difficult. The media was also flooded with lots of information which ought to be scrutinized properly as some added anxiety to the people. Affected people also needed medical attention and examination for radiation doses.

Therefore, through the Hiroshima University Phoenix Leader Education Program (Hiroshima Initiative) for the “Renaissance from Radiation Disaster”, which was initially funded by the Ministry of Education, Culture, Sports, Science and Technology came up with a worthwhile program fostering social recovery, environmental radioactivity protection and radiation disaster medicine for not only the people of Fukushima but also in preparation for the future unforeseeable nuclear disasters.

Throughout my studies at Hiroshima university, I have been exposed to all these three disciplines with much emphasis on my major, Environmental radioactivity protection which was also affiliated to graduate school of science-chemistry department.

This is the course that has been preparing the scientists how they can deal with radiation catastrophe emanating from natural disasters, even nuclear terrorist attacks. Frankly speaking, I did enjoy this program in that we have had opportunities of hands-on instrumentation in our research-work and also, we had a lot of education tours during leadership seminars, conferences and field-work visits where we shared knowledge with experts in the field. We had social recovery gatherings with the people of Fukushima in which we discussed a number of issues as regards radiation levels in Fukushima and also measured the radioactivity of their foods publicly using portable gamma spectrometer. This reduced panic among the people as they saw for themselves how the radioactivity

levels in foods had dropped with time. Above all in this program I accumulated a combination of global skills, management and interdisciplinary skills by encountering with experts in various fields. We realized lessons that were learned from the Chernobyl disaster had also to be applied to our program for Fukushima nuclear disaster residents. Environmental radioactivity protection program dealt much more mitigation measures to deal with anthropogenic radionuclides aftermath of nuclear disaster in particular ^{90}Sr, $^{134+137}Cs$ and ^{131}I. The half lives of ^{90}Sr, ^{134}Cs and ^{137}Cs are 28.8, 2.1, and 30.1 years, respectively. Iodine-131 has a shorter half-life of 8 days and soon after the disaster much emphasis had been put on it because it accumulates in thyroid gland. Thus, in order to ensure that people were safe in Fukushima we took initiative of venturing into field sampling in Kawauchi village and Fukushima city where we collected soil and rice samples from disturbed and undisturbed forestry and paddies. Further, to ensure that people were consuming rice free of radioactivity we had to determine the transfer factor, i.e. the ratio of amount of radiocesium transferred to the rice plant to that in soil. We had also studied critically why the transfer factors between Kawauchi and Fukushima City paddies were different by looking into other physical and chemical properties of soils in the two areas. Next, we did really collaborate with the University of Tokyo as long-term internship to analyze the soil core and bulk samples in Kawauchi village and Fukushima city for distinction in migration between ^{137}Cs and ^{90}Sr, more particularly in undisturbed forestry, lake sediment and disturbed paddies.

The results were indeed compelling, Kawauchi soil despite having lower concentration of ^{137}Cs in its soil, it showed 5-fold transfer factor than that of Fukushima. And there was a difference in migration between ^{137}Cs and ^{90}Sr.

We believe this information can easily be disseminated to the people through social recovery sensitization programs now that the levels of radioactivity are on their decline although forests are still containing high levels of radioactivity. The results of this research together with other findings concerning the lowering of levels of radioactivity in the arable land of Fukushima has given hope to the displaced people of Fukushima who have so far sought to return to their abandoned areas.

Above all the course on environmental radioactivity protection prepared me to become a specialist in environmental radiation protection emergency preparation and response.

○Title of Doctoral Thesis;

Difference in Migration of Radioactive Element Originating from Fukushima Daiichi Nuclear Power Plant Accident: Factors affecting transfer factor of ^{137}Cs from soil to rice and Difference in migration between ^{137}Cs and ^{90}Sr in the Environment

○Summary of Doctoral Thesis

(Describe so as to be easily understood, by relating it to "Radiation Disaster Recovery Studies".)

In my doctoral research the focus has been on ^{137}Cs and ^{90}Sr in Kawauchi village and Fukushima city. This is because both these radionuclides are produced in high yield during nuclear fission reactions inside core of nuclear reactor. Thus, they are common in radioactive wastes and are present in the environment following Chernobyl and Fukushima nuclear disasters. ^{90}Sr concentration was about 1/1000 that of ^{137}Cs in the soil after FDNPP disaster. The gamma and beta radiation emitted by the radioactive decay of ^{137}Cs and ^{90}Sr have to be taken into consideration as they may damage the cells of living organisms leading to cancers.

Strontium-90 will be accumulated in the bone and its biological half-life is about 50 years, thus it is of concern just like ^{137}Cs which affects the whole body although it has a shorter biological half-life of 70-100 days compared to ^{90}Sr. The study of distribution and migration of both nuclides is important from the point of protection against external and internal exposure. The present Doctor of Science (DSc) thesis includes two topics. The first I considered the factors to control transfer factor (TF) of ^{137}Cs from soil to rice plant (First paper). The second I investigated the difference in distribution and migration of ^{137}Cs and ^{90}Sr in the soils of Kawauchi and Fukushima (Second paper).

In the first paper the relation among soil characteristics (soil particle size distribution, exchangeable cation, ^{137}Cs, and ^{40}K, mineral composition, Fe oxidation state and ^{137}Cs transfer factor) was investigated by analyzing the soil and rice samples from Fukushima City and Kawauchi Village. This study ensured the comparison of two study areas unlike other studies which have only focused on one particular area.

The samples were prepared accordingly and analyzed by High Purity Germanium detector. In order to investigate other physico-chemical properties of the soils following variation in TF between the two areas. The major exchangeable cations were analyzed by using Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) after extraction using ammonium acetate, crystal nature of the soils was studied by X-ray diffractometry, while the effect of Fe(II) and Fe(III) on the uptake of ^{137}Cs was done by Mössbauer spectroscopy. We also investigated the particle size distribution of the soils in relation to ^{137}Cs sorption by plants.

Cesium-137 transfer factor between Kawauchi and Fukushima samples was compared. The TF of ^{137}Cs was higher in Kawauchi paddy field compared to that of Fukushima despite the soils of Fukushima being more radioactive than that of Kawauchi.

Cesium-137 was also compared in Fukushima and Kawauchi soils before and after treatment with 1M ammonium acetate at pH 7. From this, exchangeable ^{137}Cs was obtained for both soils. The

amount of exchangeable ^{137}Cs was larger in Fukushima paddy than in Kawauchi paddy. The percentage (%) of exchangeable ^{137}Cs to total ^{137}Cs was higher in Kawauchi paddy than Fukushima paddy. On the other hand, % of exchangeable K^+ was higher in Fukushima paddy than in Kawauchi paddy. The results revealed that TF becomes higher when % of ^{137}Cs is higher, while it is restricted when % of exchangeable K^+ is high.

Furthermore, it was shown that Kawauchi soil contains more silt and clay and less coarse and gravel than Fukushima soil. However, Kawauchi soil that contains more silt and clay has higher percentage of exchangeable ^{137}Cs than Fukushima soil that contains more fine and medium sands. The finding in this study showed that the ^{137}Cs exchangeable rate cannot be easily explained only based on the soil particle size distribution.

The analysis of powder X-ray diffraction pattern (PXRD) using Match software showed that Kawauchi soil contains more quartz constituents and slightly less mica minerals than Fukushima soil and Fukushima soil contains more vermiculite, zeolite and phlogopite. Fukushima soil ^{137}Cs is adsorbed to the clay particles more strongly compared to Kawauchi probably because Fukushima soil is rich in vermiculite, mica and zeolite which possibly retard ^{137}Cs transfer from the soil to rice plant.

Mössbauer spectra did not show significant magnetic component in Kawauchi soil compared to that in Fukushima. The ratio of Fe(II)/(Fe(III) + Fe(II)) of Kawauchi soil happened to be larger than that of other Fukushima soils. The differences between Fe (II) and Fe (III) signify the influence of the redox potential of the soil. Other studies have shown that microbially mediated reduction of Fe(III) in chlorite and biotite by *Shewannella oneidensis* MR-1 leads a significant reduction in sorption of both Cs and Sr compared to the abiotic system

In the second paper, the difference in distribution and migration between ^{90}Sr and ^{137}Cs in the soil of disturbed area, Kawauchi (KP) and Fukushima paddy rice fields (FP), and undisturbed area, Lake Ogi valley sediments (LS) and its catchment forestry area (CA), was investigated. The sediment to soil ratio for ^{90}Sr and ^{137}Cs was used to analyze the horizontal migration difference between ^{90}Sr and ^{137}Cs.

^{90}Sr and ^{137}Cs distributions were investigated by analyzing their concentration in the soil of Kawauchi (KP) and Fukushima paddy rice fields (FP), and the depth distribution of ^{90}Sr and ^{137}Cs in Lake Ogi valley sediments (LS) and its forestry catchment area soil (CA) and the sediment to soil ratio were also investigated. ^{90}Sr is a pure ß-emitter and its daughter nuclide (^{90}Y) is also radioactive. ^{90}Sr was extracted from the soil and after the radioactive equilibrium was attained, the radioactivity was measured using liquid scintillation counter. The recovery ratio was measured promptly by Inductively Coupled Plasma-Mass Spectrometry-Triple Quadrupole (ICP-MS-QQQ).

The rough correlation between ^{90}Sr and ^{137}Cs concentrations was observed. The relation deviates from original point (0, 0), which suggests the global fallout of ^{90}Sr. The coefficient of variation for ^{90}Sr was larger than that of ^{137}Cs for Fukushima and Kawauchi paddy. Furthermore, the coefficient of variation for ^{90}Sr was also larger than that of ^{137}Cs for Ogi Lake Sediment (LS) and its catchment forestry area (CA). The results suggest that the migration of ^{90}Sr is more affected by external factor.

The sediment to soil ratio (LS/CA ratio) of ^{90}Sr was greater than that of ^{137}Cs, which might relate to ^{90}Sr faster horizontal migration than ^{137}Cs.

The depth dependence of ^{90}Sr and ^{137}Cs at Catchment Forestry area was investigated. Cesium-137 and ^{90}Sr activity concentrations decrease exponentially with depth. However, the vertical profile between the two is different, which showed faster ^{90}Sr vertical migration than ^{137}Cs. Depth dependence of ^{137}Cs and ^{90}Sr at Lake Ogi sediments (LS) revealed that the exponential change for both ^{137}Cs and ^{90}Sr was largely interfered near the surface (0 to 4 cm) for the sediment. One of the possibilities is that surface sediments become relatively uniform by the disturbance near surface (0~4 cm). ^{137}Cs adsorbed strongly to sediment becomes uniform, while ^{90}Sr weakly adsorbed to sediment re-dissolves to the lake water. The re-dissolved ^{90}Sr is absorbed to the sediment again (0~1 cm). Therefore, the surface sediment (0~1 cm) has significantly higher ^{90}Sr than that of deeper sediment layer.

Strontium-90 penetrated more than ^{137}Cs in the soils of catchment forest and LS/CA ratio of ^{90}Sr is greater than the LS/CA ratio of ^{137}Cs, which might relate to ^{90}Sr faster horizontal migration than ^{137}Cs. The $^{90}Sr/^{137}Cs$ ratio for Fukushima soil and sediment samples ranged from 0.0001 to 0.0019 with an average value of 0.0007±0.0005, which is in good agreement with some previous results.

In conclusion there was a difference in migration between ^{137}Cs and ^{90}Sr. However, in future research there is need to take core soil samples from the paddies also in order to compare well with the core samples from undisturbed forestry zones. Further, considering 5 folds values TF of Kawauchi compared to that of Fukushima, it is vital in future research to consider also how ^{137}Cs transfer factor might be influenced by the adsorption kinetics.

○Other theses published in academic research journals

・Title of academic research journals （Impact Factor, Peer Review）

1. W.C. Bekelesi, T. Basuki, S. Nakashima,^{137}Cs Soil to Rice Transfer Factor and Soil Properties: Fukushima and Kawauchi Case Study. *Radiation Safety Management* Vol. 21 (1–12) (2022). [doi:10.12950/rsm.220131].
2. W.C. Bekelesi, T. Basuki, S. Higaki, S. Nakashima, Distinction of strontium-90 and cesium-137 migration of Fukushima soil and sediment following Fukushima accident. *Radiation Safety Management* Vol. 21 (26–35) (2022).[doi:10.12950/rsm.220527].

・Title of thesis

1. W.C. Bekelesi, E.O. Darko, A.B. Andam, Activity concentrations and dose assessment of 226Ra, 228Ra, 232Th, 40K, 222Rn and 220Rn in soil samples from Newmont-Akyem gold mine using gamma-ray spectrometry. *African Journal of Environmental Science and Technology*. 2017 May 31;11(5):237-47. https://academicjournals.org/journal/AJEST/article-abstract/759E8E064123
2. W.C. Bekelesi. Thesis, Master of Philosophy Degree (2015). http://ugspace.ug.edu.gh/handle/123456789/8571.

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1. T. Basuki, W. C. Bekelesi, M. Tsujimoto, S. Nakashima, Air dose rate to ^{137}Cs activity per unit area ratio for different land use 7 years after the nuclear accident -Case of the slope catchment, Ogi reservoir, Fukushima-, *Radiation Managements,* 137 (2020). https://doi.org/10.1016/j.radmeas.2020.106424.
2. T. Basuki, W. C. Bekelesi, M. Tsujimoto, S. Nakashima, Investigation of radiocesium migration from land to waterbody using radiocesium distribution and soil to sediment ratio: A case of the steep slope catchment area of Ogi reservoir, Kawauchi Village, Fukushima, *Radiation Safety Management*, 19, 23-34 (2020). DOI 10.12950/rsm.190924.
3. M. A. Habib, T. Basuki, S. Miyashita, W. Bekelesi, S. Nakashima, K. Techato, R. Khan, A. B.K. Majlis, K. Phoungthong, Assessment of natural radioactivity in coals and coal combustion residues …… *Environmental Monitoring and Assessment*, 191: 27 (2019). DOI10.1007/s10661-018-7160-y.
4. M. A. Habib, T. Basuki, S. Miyashita, W. Bekelesi, S. Nakashima, K. Phoungthong, R. Khan,M. B. Rashid, A. R. M. T. Islam, K. Techato, Distribution of naturally occurring radionuclides in soil around a coal-based power plant and their potential radiological risk assessment, *Radiochimica Acta* (2018).DOI 10.1515/ract-2018-3044.