Thesis Summary

Properties of the Soil in Rice Fields and Transfer of Cesium to Rice Plants (水田土壌の性質とセシウムの米への移行)

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On 11th March 2011, The Great East Japan Earthquake of magnitude 9.0 and the following 15 m tsunami hit the Tohoku area. This led to the cause of Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, where ¹³⁴Cs and ¹³⁷Cs were released to the air and made land and rice fields contaminated. As a student from The Phoenix Leader Education Program (Hiroshima Initiative) for Renaissance from Radiation Disaster, I learnt the radiation disaster and restoration from the disaster in Fukushima Prefecture. After the accident, the food safety has emerged as a very important problem. Contaminated rice from paddy fields was observed in Fukushima City. How the contaminated rice is reduced was the basis for my doctoral dissertation. Although there are many studies concerning the contamination of radioactive cesium and the transfer of radioactive cesium from soil to ears of rice in the paddy fields of the Fukushima after the FDNPP accident has been carried out, the mechanism of the up-taking of cesium into rice, the migration of cesium and the factors that affect are still required. An investigation in 2013 showed the concentration of radioactivity of ¹³⁷Cs and ¹³⁴Cs in rice which was collected from one of neighbor rice fields was relatively high. This study aims to investigate the contamination of radiocesium in rice and paddy field's soil, and the factors that affect the contamination and the transfer of ¹³⁷Cs and ¹³⁴Cs into rice. In order to understand the factors, the soil samples were collected, and the radioactive concentrations, the depth distribution, the soil size distribution were investigated and the oxidative/reductive atmosphere in the paddy field was investigated using ⁵⁷Fe Mössbauer spectroscopy. Background and objective of the research are given in Chapter 1.

The chapter 2 shows the method. Our study performs in 4 paddy fields (which are named as A, B, C, and D) in Fukushima City, which are 60 km Northwest far from Fukushima Dai-ichi Nuclear Power Plant. We monitored the fields from 2014 to 2018. The paddy fields are irrigated by a nearby pond and partial water from Abukuma River. Four fields were cultivated in 2013 and 2014, although the upper two fields were not cultivated from 2015. The samples were taken from 4 paddy fields with 5 sample sites for each field. For radioactivity measurement, the samples are contained in U-8 vessels and measured for the radioactivity by Ge semiconductor detector (GEM 30-70, ORTEC). The radioactivity was calibrated using MX033U8PP source set (Japan Radioisotope Association). ⁵⁷Fe Mössbauer spectroscopic measurement was performed at room temperature with a ⁵⁷Co (Rh) radiation source moving in a constant acceleration mode on Wissel MB-500. The Mössbauer parameters were obtained by least-squares fitting to Lorentzian peaks. The spectra were calibrated by the six lines of α -Fe, the center of which was taken as zero isomer shift.

Chapter 3 is the study about depth distribution of radioactive cesium in soil after cultivating and the difference by the year of the uptake of radioactive cesium in rice in these fields. The average

value of five points in each field was compared to each other. The ¹³⁷Cs concentration decreased depending on the depth. The same trend was also observed in the depth dependence of ¹³⁴Cs concentration. The ratio of radioactivity concentration of ¹³⁴Cs to ¹³⁷Cs was 0.373. This value corresponds well with the expected ratio (0.376) on the sampling day declined according to reduction of ¹³⁴Cs by shorter half-life than that of ¹³⁷Cs, given that the same amount of radioactivity of ¹³⁷Cs and ¹³⁴Cs was released from the FDNPP reactor. These fields were cultivated, ploughed and irrigated before transplanting rice seedlings in 2013. Nevertheless, the concentrations of ¹³⁷Cs and ¹³⁴Cs in soils were not uniform and there was depth dependency in 2014. It is expected that small particles such as clay include a large amount of radioactive cesium. One of the possibilities is that bigger particles fell down quickly but the small particles such as clay fell down slowly in the sedimentation process after ploughing and irrigating, although the soils were mixed. There is a difference between Fields A, B and Fields C, D. The radioactivity of ¹³⁷Cs and ¹³⁴Cs for 0-5 cm depth was higher in Fields C and D than in Fields A and B, while the radioactivity for ⁵⁻¹⁰ cm was found at greater depth in Fields A and B than in Fields C and D.

In Chapter 4 the ⁵⁷Fe Mössbauer spectroscopy was used to determine the oxidative/reductive characteristic of soil samples. The iron concentration variation was also investigated based on soil size distribution. It was suggested that the oxidative/reductive condition of iron state may reflect the features of the soils and then affect the solubility of potassium and radioactive cesium in the field. We checked the change of ⁵⁷Fe Mössbauer absorption depending on the soil size. Consequently, the iron in the larger size soil is much abundant in Fields A and B compared with that in Fields C and D. It is known that the small sized soil includes clay much. And the radioactive cesium is adsorbed strongly to the clay. It is also thought that the cesium adsorbed to large sized soil is relatively easily desorbed and absorbed to rice body. The amount of iron may affect desorption of radioactive cesium from large sized soil. One of the possibilities is that the iron works as catalyst to dissolve radioactive cesium from soil. In conclusion, oxidative atmosphere and in particular more abundant iron in larger size soil may affect the transfer of radioactive cesium from soil to rice grain.

Chapter 5 shows the change of radioactive Cesium in soil by year and uptake of radioactive Cesium in rice. The distribution of grain size of soil was investigated to know the changing depending on time. There is a difference between radioactive cesium which was accumulated depending on particle size of soil in the cultivated fields and fallowed fields. Fields A and B had more abundance of medium sand than Fields C and D. On the other hand, Fields A and B had less abundance of clay and silt than Fields C and D. It is expected that the radioactive cesium penetrated more deeply in the soils having more medium sand. If we compare the average concentration from 0 to 30 cm, the value becomes 647, 674, 708, and 602 Bq/ kg for A, B, C, and D, respectively. This might suggest that the radioactive cesium migrated from Fields A and B to Field C. But, the radioactive cesium did not migrate much from Field C to D. In 2015, C-4 and D-4 had much higher radioactivity than other sites. There is a possibility that the lost clay and silt in C-4 and D-4 were not compensated from Fields A and B because of the fallowing of Fields A and B since 2015.

Chapter 6 shows general conclusion.