What we learned from the large-scale soil sampling for radioactive nuclide released from the Fukushima Dai-ichi Nuclear Power Plant accident? [1]

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Abstract

After the accident of the Fukushima Dai-ichi Nuclear Power Plant (NPP) of Tokyo Electric Power Company (TEPCO) in March 2011, radioactive nuclei were spread out over the Fukushima area. We started to monitor the radioactive nuclei discharged by the accident at the Fukushima Dai-ichi NPP. We compiled results of measured ambient dose rates at a 1-m height above the ground surface, and the "soil deposition density map" for deposition densities of radioactive nuclide deposited on soil. The area within 80 km from the Fukushima Dai-ichi NPP was divided into 2 km×2 km grids, whereas the areas between 80 km and 100 km and areas of Fukushima prefecture out of that scope were divided into 10 km×10 km grids. Ambient dose rates were measured at one location in divided grid (nearly 2,200 locations in total), and soil samples were collected at five points at each location. Measurement of ambient dose rates and soil sampling was performed by 440 persons from 107 different organizations, including researchers from Osaka University, Kyoto University, University of Tsukuba, the University of Tokyo, the Japan Atomic Energy Agency, and the Japan Chemical Analysis Center, and members of the local support team of the Federation of Electric Power Companies of Japan,. Nuclide analysis of soil samples was performed by 291 persons from 21 organizations. After making a brief report on the large-scale soil sampling for radioactive nuclide emitted from the TEPCO Fukushima Dai-ichi NPP accident, the following subjects are discussed in the present article: 1) what happened in the Japanese academic community after the TEPCO Fukushima Dai-ichi NPP accident? 2) what were the main concerns about the radioactive fallouts? 3) what were the difficulties in organizing an emergency team consisting of many independent research institutes? 4) what do we learn from the NPP accident for future? and 5) how should we prepare for urgent accidents?

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1 Fukushima Dai-ichi NPP Accidnet

Japan had 54 nuclear power plants (NPP's). 17 NPP 's were in operation and supplied 29% of the total electric power requirement in Japan in March 2011. An earthquake with a magnitude of M9 struck the east coast in the afternoon on March 11, 2011. Soon after the strong vibration due the M9 earthquake, a huge tsunami struck many seashore towns along the eastern coasts of Iwate, Miyagi, Fukushima, and Ibaraki prefectures. Television news showed, online, the real situations of these tsunami disasters attacking many seashore towns in the whole afternoon on March 11.

Multiple nuclear reactors at Fukushima Dai-ichi, Fukushima Dai-ni and Tokai were also attacked by huge tidal waves. The cooling functions of reactor cores were lost due to seawater flood in the NPP buildings where electric generators were installed for emergency use. Especially, in case of the Fukushima Dai-ichi NPP's, the prolonged loss due to unrecoverable electric power and associated cooling systems caused extraordinary heat of nuclear fuel rods in the core, and brought about the serious NPP accident of level 7. A series of explosions occurred at the reactor buildings. Radioactive materials were released into atmosphere. Meltdown of the reactor cores resulted in leaking of radioactivity through broken parts into the ocean. Almost all Japanese people believed that his kind of serious NPP accidents could not occur in high technology country like Japan. It turned out that this illusive belief was not true any more.

Later, we learned of more unbelievable facts: Serious accidents had proceeded at the site of the Fukushima Dai-ichi nuclear power plants. The time sequences of the level-7 NPP accident concerning radiation dose rate observed at the main gate located at south-west of Fukushima Dai-ichi site were as follows:

1. Small scale venting from Unit-1 NPP was carried out during 10.00-13.00 on 12 March. Explosion of Unit-1 happened at 15.36 on 12 March. Radiation dose rates were still not high and were less than 0.5 mSv/h.

2. Small scale venting from Unit-3 were carried out three times at 9:20 and around 13:00 on 13 March, and 6:20 on 14 March. Explosion of Unit-3 happened at 11:01 on 14 March. Radiation dose rates were less than 1 mSv/h.

3. Venting of Unit-2 was carried out at 20:25 on 14 March. Radiation dose rates increased to 3.5 mSv/h.

4. On 15 March, explosions of Unit-4 and Unit-2 happened at 6:00 and 6:10, sequentially, and fire accidents happened during 6:00-11:00. Radiation dose rates increased up to 12 mSv/h.

5. Radiation dose rates increased to 8 mSv/h at midnight on 15 March. The reason was not well understood.

6. Smoking accidents happened at Unit-1 during 10:00-12:00 on 16 March. Inside air of the Unit-3 building was evacuated during 10:45-11:30 on 16 March.

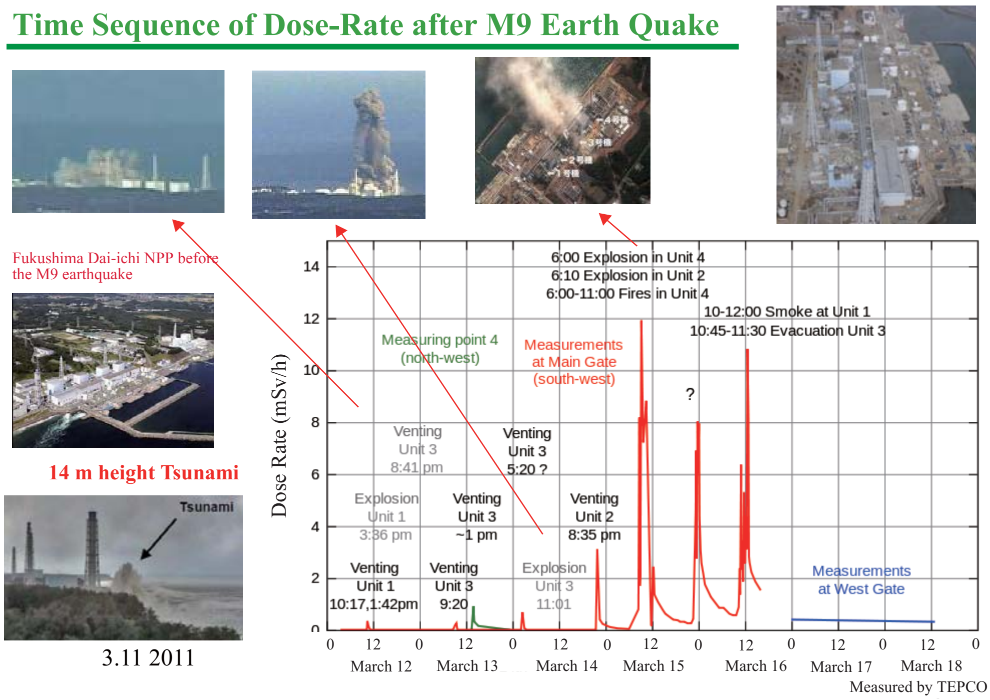

Figure 1: Time sequence of radiation dose date after the M9 Great East Japan Earthquake. Dose rates were measured at the main gate located at south-west of Fukushima Dai-ichi site. Some epoch-making photos about the Fukushima Dai-ichi NPP accident after the earthquake are inserted.

Figure 1 summarizes the aforementioned events. Information on these radiation dose rates was obtained freely through the internet from the website of Tokyo Electric Power Company (TEPCO). Japanese scientists working on nuclear physics frequently shared opinion and information on the Fukushima Dai-ichi NPP accidents through the nuclear physics network in Japan. When they realized that the NPP accident was an event of an unbelievable scale in Japan, all felt what they should contribute something as nuclear scientists whose work is directly related to nuclear energy. Especially, this feeling was much strengthened after 15 March when the radiation dose level exceeded 12 mSv/h.

2 Discussions of Nuclear Scientists

There were heated discussions on the possibility of meltdown of the Fukishima Dai-ichi NPP. TEPCO reported that ^{38}Cl activity was observed outside the NPP site. There was a news from the media that seawater including NaCl was used to prevent meltdown of the nuclear fuels. If the re-criticality happened in the core with seawater, the ^{37}Cl$(n,\gamma)^{38}$Cl reaction would be induced and the ^{38}Cl activity was produced. There was a contradiction, however. TEPCO did not report anything about the ^{24}Na radioactivities which should have been produced because of the much higher (n,γ) cross sections. This was a serious inconsistency. There were many TEPCO reports full of mistakes due to chaotic situations of the Fukushima NPP accident. We concluded that there was no experienced nuclear scientist at the TEPCO NPP site. We thought that nuclear scientists should somehow help the people suffering from the Fukushima Dai-ichi NPP accident.

There were passionate communications to organize an emergency meeting for discussing what nuclear scientists could contribute to the Fukushima Dai-ichi NPP accident. The traffic systems and buildings in Tokyo and Sendai were seriously damaged by earthquake so that there was no possibility to have such a meeting in Tokyo and Sendai. There was no earthquake damage in Osaka, located about 700 km south-west from the epicenter of the M9 earthquake. We decided to organize an emergency meeting in Osaka in order to discuss how to measure the radioactivities released from the NPP site. The announcement of the meeting was sent at 2:00 am on 15 March. The meeting was held at the Research Center for Nuclear Physics, Osaka University, in the afternoon on 16 March.

3 Emergency Meeting at RCNP

More than fifty participants attended the meeting in the afternoon on 16 March. People came from Kobe, Osaka, Kyoto, Nara, Tokyo, Sendai, Niigata, and Kyushu. Tanihata joined after taking a morning flight from Beijing. This was beyond our expectations. The subjects to be discussed in the meeting were 1) how to make the map of the radioactivities released to the wide area in Fukushima Prefecture from the NPP accident, and 2) how to prevent the associated accidents. We knew that detailed measurements were not performed for making the radioactivity map in the surrounding area of the Chernobyl NPP accident site in 1986. The soil radioactivity measurements were performed only three years later. Before the dissolution of the Soviet Union in 1991, serious confusion in the politics of Ukraine had already started around 1986 on the pathway towards being free from the Soviet Union. Thus, Ukraine and Russian people could not afford the soil radioactivity measurements around Chernobyl until 1989. This lack of information on the radioactivity dose rates for 3 years from 1986 to 1989 was well known to have led to serious health hazard for the residents living in the polluted areas near the Chernobyl NPP site. Japan should not repeat the health hazard due to the fallout radioactivities, and not make the mistake of not preparing a radiation dose-rate map in the Fukushima area. This was what the participants felt in the meeting. Especially, urgent action was needed to prepare the soil radioactivity measurements of short-life radioactive iodine-131, ^{131}I ($T_{1/2}$=8 d), which is notoriously dangerous for increasing thyroid cancer risk, in particular, among children.

We asked Hiroyasu Ejiri to communicate with Mr. Kiyosi Shimuzu, Vice-Minster of the Ministry of Education, Culture, Sports, Science and Technology (MEXT), on 17 March in order to discuss the direct way for the professional contribution of the nuclear physics group in Japan to the disaster caused by the Fukusima Dai-ichi NPP accident. Mr. Shimizu talked to Mr. Kanji Fujiki, one of the bureau chiefs of MEXT. Mr. Fujiki consulted Mr. Takashi Gouda who was responsible for taking measures against the Fukusima Dai-ichi NPP accident. After the Fukushima Dai-ichi accident, MEXT had established a support team for residents affected by radioactivities under the Nuclear Emergency Response Headquarters. Mr. Gouda asked Mr. Daichi Saito to take work with us on the project to make the fallout radioactivity map.

4 Resident Screening

MEXT had not yet taken necessary budgetary steps for the map project. It was still necessary to wait for budget allocation. This was a direct reply sent to Fujiwara and Tanihata by mail from Mr. Ichiro Ikeda of MEXT. It was our big surprise to know that Japan did not have any imminent budgets for the NPP accidents although the Japanese government always sent urgent support teams to help the residents in case of earthquakes, river water floods. and tsunamis etc. Mr. Ikeda asked us to support the radioactive screening task since the nuclear scientists were thought to have professional skills to measure the radiation dose rates. This screening task had already started on 17 March in order to check the dose rates of residents who were forced to be evacuated from their homes in neighbouring villages of the Fukusima Dai-ichi NPP site.

The staff people from Kyoto University Research Reactor Institute (RRI), National Institute of Radiological Sciences (NIRS), Research Institute for radiation Biology, Medicine (RBM), the Federation of Electric Power Companies of Japan (FEPC), and MEXT had already been chosen to be sent on radioactivity screening mission to Fukushima according to the emergency request from the medical team of Fukushima prefecture to MEXT. This kind of emergency call system had been established after the Tokaimura nuclear accident (JCO accident) at the factory of Japan Nuclear Fuel Conversion Co. in 1999 [1].

5 Movement of Osaka University

Mr. Gouda of MEXT was very careful to contact the Japanese Universities; Only the decision of RCNP was not enough to let nuclear scientists join the screening program in Fukushima, but also the permission of the head quarter of Osaka University was necessary to get the full support for the program. We consulted Kenshi Doi, a member of the board of trustees, and the head of Department for the Administration of Safety and Hygiene, Osaka University. The decision making was very quick. T. Kishimoto (director of RCNP), Tanihata, Fujiwara, Doi, and Yamamoto came together at the head-quarter building of Osaka University in the late afternoon on 17 March, and discussed the strategy of how we could proceed with the screening program by talking with Mr. Gouda in Tokyo.

The medical school of Osaka University sent a helicopter to Tohoku area soon after the M9 earthquake for disaster victim assistance. This helicopter passed above the Fukushma Dai-ichi NPP site. When the helicopter returned at the heliport on the building roof of Osaka University hospital, it was found that radioactivities were deposited on the helicopter surface. Although the deposited radioactivities were washed out soon, Osaka University sent a formal message "The staff of Osaka University should not willfully participate in the disaster victim assistance". Mr. Gouda knew this fact. He wished to have a full agreement of Osaka University to proceed with the supporting program for the Fukushima Dai-ich NPP disaster through RCNP.

Osaka University decided to fully support the activities for earthquake disaster reconstruction. This decision was formally agreed to by the board of trustees of Osaka University on 18 March. This was the emergency support statement for the M9 earthquake disaster, under the excellent leadership of Kiyokazu Washida, the president of Osaka University. Osaka University also decided to prepare the necessary budget for supporting the activities. RCNP started to call scientists to participate in the screening program in Fukushima. This call was not only sent to nuclear scientists, but also to many scientists working in various fields such as radiochemistry, medical physics, environmental sciences, radiological sciences, etc.

The formal call for the professional participation in the MEXT screening project was issued from RCNP on 19 March. Subsequently, in the afternoon on 20 March, we held a meeting to discuss the strategy on the method of the participants' transportation to Fukushima. On 21 March, the planned project actually started. We first traveled to Tokyo by train, and came together at RIKEN. The RIKEN staff joined the project team. Dr. En'yo provided official cars of RIKEN for road transportation from Tokyo to Fukushima. Since railways from Tokoyo to Fukushima were partly damaged at various locations, there was no way for traveling by train for Fukushima. Dr. Shimbara of Niigata University arrived at Fukushima by car. Dr. Tamii played the role of the leader of the screening team of the nuclear physics group.

6 Participation in Screening Project

In the morning on 21 March, the support of the screening program by the nuclear physics group started. At 7:30 in every morning, support members came together at the head-quarter office of the residents' association building nearby Fukushima prefecture government. After a short briefing, the screening teams went to the villages where the radioactive dose rates were as high as 10 μSv/h. About 20 nuclear physicists from Osaka University, University of Tokyo, RIKEN, and other universities, and 80 staff people from RRI, NIRS, RBM, FEPC, MEXT had joined the screening project in March and April 2011. The staff members of MEXT and the Ministry of Health, Labour and Welfare had managed to let these 100 people work smoothly every day in a close contact with the emergency measure head office in Tokyo for the Fukushima NPP disaster.

The Fukushima Dai-ichi NPP accident forced many residents to leave their homes in the villages near the NPP site. First, the government ordered to evacuate the residents living in the 3 km radius around the Fukushima Dai-ichi NPP site on 11 March, 2011 immediately after the accident. When the accident became serious, the areas of evacuation were expanded. On March 12, after the first explosion at the nuclear reactor Unit-1, the government expanded the areas of evacuation within 20 km radius from the Fukushima Daiichi NPP site (20 km zone). On March 15, after the explosion happened at the reactors Unit-2 and Unit-4, the government instructed the residents living in the zone between 20 km and 30 km radii from the NPP site to stay inside their houses. The predictions of Environmental Emergency Dose Information (SPPEDI) were not properly reflected for evacuation of residents on 15 March. The residents did not know the fact that wind and rain had resulted in spreading radioactive nuclides to the north west direction from the NPP site.

During screening work for the people who escaped from towns and villages within the 20 km zone, and from the Iitate village, I could hear several claims fragmentarily about the serious situation of the refugees. The staff who attended the screening radioactivity measurement felt a big fear in the panic situation with the wide and rapid spread of radioactivity. Even in case of experienced experimental nuclear physicists, it was very rare to stay one hour in the radioactivity environment with several μSv/h. Young nuclear physicists without such experience really felt a strong fear of high radiation exposure. The exposed radiation dose in the screening work was monitored using a pocket radiation monitor. The daily radiation dose was at most 30 μSv.

The evacuated residents relied on the results of radioactivity measurements by nuclear physicists in screening work. We heard a rumour that young persons of the Federation of Electric Power Companies in refugee screening work on March 15 and 16 honestly said to evacuated residents "We also feel fear of a high exposure of radioactivities". This fact tells us that

Japan needs to prepare well-trained emergency teams responsible for rescue work for NPP accidents.

For example, France has established Institute de radioprotection et de sûreté nucl'eaire (IRSN) in 2001 as the support organization of the Nuclear and Industrial Safety Agency and the radiation protection. The annual budget of this institute is about 5 Million US dollars, Varoius activities of IRSN are:

1. to directly check all the NPP operation parameters in France;
2. to have a training function for emergency due to the NPP accidents;
3. to work on the radiation safety issues including the international collaboration for the Fukushima Dai-ichi as well as Chernobyl NPP accidents;
4. to study the environmental effects by radioactivities; and,
5. to develop new radiation monitors and to develop new instruments for radiation safety, etc.

The United States has also a similar team "Nuclear Regulatory Commission (NRC)" for answering to emergencies. The Japan Atomic Energy Agency in Japan corresponds to these organizations. However, no organization corresponding to NPP emergencies had been discussed under the name of "mythical atomic safety". All the budgets for NPP emergencies were cut because of false belief in this "mythical atomic safety". The Japanese NPP system is somewhat different from those of the other countries. Fujiwara heard a very strange fact from the head of IRSN when Fujiwara visited the IRSN headquarter in Paris in December 2012. He told Fujiwara, "To my knowledge, you are the second Japanese visitor to IRSN". This means that any staff working on the nuclear safety in Japanese government has no international relationship, at least, with France. On the other hand, we know that some government people contacts with NRC in USA. A cynical person told me that the emergency cooling water system was placed in the underground room because the NPP of USA was designed under the safety consideration for tornado attack, not for tsunami attack. Apparently, we need to re-examine the Japanese NPP safety system by studying the ISRN and NRC organizations in view of the global standard and by studying what happened at the Fukushima Dai-ichi NPP accident. Otherwise, we should call Japan a great economic nation with a faint awareness as a cultured nation.

It is inferred that the cumulative damage by the Dai-ichi Fukushima NPP accident will be the equal to the annual government budget of Japan for more than 30 years. A big criticism stemmed from the fact that an emergency organization of Japan didn't function in case of this unprecedented accident. The vertically-segmented organization system would also have to be re-examined by taking into account a new, completely different look. There is a sentence:

"The business should be run securely, adjusted in accordance with changing circumstances and financial state, and should never be run carelessly by chasing easy money." This is the famous motto of the Sumitomo group. It's necessary for us to remember that a small mistake with small amount of money often leads to a great loss of an individual person, a company, as well as a country.

7 Thyroid Measurement for Children and Radioactivity Screening

During the screening work, there was a request for nuclear scientists to measure ^{131}I concentrations in the children's throats since the thyroid in children was a cause for people's concern. Although it was expected that precise measurements to check the ^{131}I radioactivities absorbed by children would be impossible under the high radiation background situation, MEXT wished to wipe out this concerns. The SPEEDY indicated that there was a possibility that the ^{131}I activities flowed to northeastern Iitate village and to southern Iwaki-city. Measurements were performed by touching a NaI survey meter at children's throat. However, it was almost impossible to obtain reliable results at public hall in Yamakiya area of Kawamata-town because the background dose was high as 2-3 μSv/h even inside a wooden house. The radiation shield by walls of wooden houses was not effective. The observed results fluctuated in the range of 0.3 μSv/h.

To improve such situations, we decided to repeat the same thyroid measurement by changing the measurement place to a cave room with no window in concrete building where γ-rays from the outside are well shielded by the concrete walls, and the dose rates were less than 0.1 μSv/h We could not find any children who accumulated ^{131}I activities in the throat with radiation exposure higher than 0.1 μSv/h. The detection limit of the radiation exposure was around 0.1 μSv/h. It would have been possible for us to decrease the detection limit to lower than 0.1 μSv/h if we could find a low-background cave equipped with a good ventilation system. However, it was not possible to find such well-shielded rooms in Fukushima. The children's thyroid exposure measurements for ^{131}I were continued until 30 March 2011. Since two half-lives of ^{131} ($T_{1/2}$=8 d) passed and there was no significant exposure detection of ^{131}I, we stopped this work at the end of March. An accumulated number of thyroid checks for children amounted to 890 on 30 March.

At first, the screening activity was carried out by about 100 people, consisting of about 10 teams every day from March 21 to April 9. An accumulated number of the screened people from 21 March to 9 April amounted to 6,100. Around the middle of April, the screening activity gradually tapered off because the number of the residents escaping from the high dose area decreased. The support by RCNP, Osaka University, to send nuclear scientists was not necessary any more. Therefore, this mission ended around August

20, 2011.

8 Fallout Radioactivity Nuclide Map

8.1 Preparation of Proposal

During the participation in the screening work in Fukushima, Masaharu Hoshi of Hiroshima University and Mamoru Fujiwara discussed, on 25 March 2011, how we could make a rough draft on the project to make ”Fallout radioactivity nuclide map after the Fukushima Dai-ichi NPP accident and its post-processing for resident’s healthcare”. Hoshi is a professor famous from the study of the radioactivity health effects after the atomic bomb was dropped on Hiroshima in 1947 and after the Chernobyl NPP accident in 1986. Hoshi had a special connection with Dr. Valeriy F. Stepanenko, who was a director of Medical Radiological Research Center (MRRC) in Obninsk, Russia. Stepanenk had rich medical experience from his long and direct association with the Chernobyl NPP accident. Hoshi asked Stepanenk by an e-mail to list the key things that Japan should do for residents after the Fukushima Dai-ichi NPP accident. The answer came just quickly in 2-3 days later. The key things written in English was rewritten into the Japanese document by taking into account the Fukushima situation. This Japanese document was completed on March 29 after several e-mail exchanges between Fujiwara and Hoshi.

Fujiwara returned to Osaka after the screening job in Fukushima around the end of March. The document prepared by Hoshi and Fujiwara was polished up with the help of Tanihata. Shojiro Nishio, a trustee of Osaka University, had already contacted Tanihata concerning the measurement of radioactive nuclide map. Since Nishio wanted to discuss the importance of the map project in the Science Council of Japan (SCJ) and in MEXT, he wished for us to send the prepared document to him. Our document was sent to him on 31 March. A further succinct document was prepared by Nishio for his explanation at SCJ and MEXT on 4 April. The document title was ”Proposal for the radioactive nuclide measurement in Fukushima”. Tokushi Shibata, a commissioner of SCJ, asked us to send the prepared document on 2 April. Our proposal was discussed in SCJ on 4 April. Finally, a SCJ 2nd urgent proposal for the East Japan Earthquake disaster was declared with the title ”Importance of the radioactive nuclide investigation after the Fukushima Dai-ichi NPP accident” with the date stamp of 4 April. In the final SCJ proposal, the map points were substantially increased compared with those in our first Osaka proposal, and the measurement area was expanded to cover the whole area of Fukushima prefecture. It was inferred that this drastic increase of map points and the areal expansion had been suggested by Shibata and other committee members of SCJ.

8.2 Procurement of soil sampling devices

There was an informal notice from Mr. Gouda of MEXT to Osaka University at the end of March. This informal notice concerned the project for making the radioactive nuclide map in Fukushima. Mr. Gouda wished to start this project as soon as possible. This notice from MEXT was received by Osaka University as a signal for the actual preparation to execute the radioactive nuclide measurements. According to the notice from MEXT, Administration of Safety and Hygiene (ASH) and Research Center for Nuclear Physics (RCNP) of Osaka University had begun to procure huge numbers of soil sampling kits and consumable goods in order to start the projects officially during the holiday season in early May 2011. We decided to use a plastic container with a diameter of 5 cm and a height of 5 cm (U-8 type). However, it was just about impossible for us to procure 12,000 U-8 containers in the market. We asked a company to produce this huge numbers of U-8 containers within two months. Later, part of produced U-8 containers were delivered to us during the soil sampling project in accordance with our request, and a large numbers of U-8 containers and soil sampling devices were stocked in the storehouse of SANSHIN METAL WORKING company in Fukushima prefecture. Since many parts of highways in the Tohoku-district and the Tokyo area were seriously damaged by the earthquake, we felt great anxiety to send large numbers of soil sampling kits, as many as 12,000 from Osaka to Fukushima through the Tokyo area. However, one truck company in Osaka could manage to transport the soil sampling kits through special highway routes to Fukushima although the car driver might feel fear of radioactivity.

Osaka University decided to have community support projects for the Great East Japan Earthquake within the limited budget as a national university. RCNP and Osaka University decided to have the administrative work to support the travelling expenses to screening activity to scientists from public institutes in Japan. For this purpose, we had a kind of implicit Memorandum of Understanding (MOU) between MEXT and Osaka University, although the budget was not yet officially decided completely. The board of trustees of Osaka University decided on a 10 million yen budget with which ASH could manage to buy the soil sampling kits through the university finance department. These decisions allowed us to buy many iron pipes for ground radioactivity permeation distribution measurement, many U-8 containers for fallout radioactivity measurement, GPS receivers for positioning the sampling locations, gloves, napkins, plastic bags, and many other required goods. The project procurement was accelerated towards the actual strategic start of the radioactivity nuclide map project during the holiday season in May 2011.

The activities for the procurement of the soil sampling devices at Osaka University are summarized as follows:

1. Procurement activity of soil sampling devices had started from March 28, 2011. 10,000 iron pipes for sampling soil were prepared in order

to measure the permeation distribution of fallout radioactivities from the ground surface. Procurement had been completed after the holiday season in May.

2. Osaka University provided JPY 10,000,000 for preparing devices necessary for the radioactivity nuclide map measurements without any detailed budget requirement document.

3. About 70 High-Purity Germanium (HPGe) detectors were estimated to be needed for γ-ray measurements to identify the fallout radioactivities such as ^{131}I, ^{134}Cs, and ^{137}Cs and to extract the radioactivity intensity in unit of Bq/m^2. We confirmed that the radiochemistry society could provide 30 HPGe detectors, and the nuclear physics society could provide 40 HPGe detectors. We called the collaborators from various research institutes to measure the γ-ray intensities from the collected soil samples using the HPGe detectors.

4. On 15 April, MEXT suggested Osaka University to request the special coordination fund for the promotion of the radioactivity nuclide map measurement and the travel expenses to dispatch scientists for sample extraction and radiation dose measurement to Fukushima.

5. We estimated that collected soil samples could amounts to about 10 tons in total. Therefore, we needed to prepare a library collecting a vast number of soil samples with a web retrieval form. There were candidate buildings such as the old cyclotron facility in Toyonaka campus and the free electron laser (FEL) facility in Hirakata campus. These facilities ware not used for the scientific researches. However, we could not get the agreement in Osaka University to use these facilities for the new construction of the radioactive soil sample library, because the relevant scientists just wished to keep the building facilities without any concrete research projects.

8.3 Towards the pilot program

At the beginning of April, the University of Tokyo requested MEXT to find the special research fund budget, independent of the movement of Osaka University. This request was rejected. The budget requested by The University of Tokyo was judged not to be executable with only one university. Koji Otsuka, director of the Center for Nuclear Study (CNS), the University of Tokyo called various research groups of nuclear physics, radiochemistry, atmosphere and ocean, agricultural environment, living resources, and global environment in order to discuss the strategy for the radioactive nuclide map measurement. The meeting was held on 14 April 20111 at the building of Department of Physics, Faculty of Science, The University of Tokyo. Yuichi Onda (University of Tsukuba), who had contact with the Ministry of Land, Infrastructure, Transport and Tourism (MLIT), received informal consent by which the soil sampling was possible around rivers even if we did not get full agreement with each town and village of Fukushima prefecture. We agreed to execute a pilot program starting before consecutive holidays in May.

It was decided that the CNS should play an important role to accumulate soil samples from Fukushima, and to distribute the soil samples to various institutes for independent radioactivity measurements. This independent radioactivity measurement was very important to assure the reliability of the measured results, and to finalize the protocol manuscript for soil sampling and radioactivity measurements. We could reach a consensus that RCNP, Osaka University, would play the role to manage the travel expenses and lodgings for the soil sampling. All of Japanese researchers cooperated in the soil radioactivity nuclide map program, resulting in the de facto recogniton of MEXT. The pilot program of the soil sampling was put onto the schedule to be performed during the consecutive holidays season in May 2011. It was also decided that Hoshi and Onda would prepare the detailed protocol for the program execution.

After the meeting on 14 April, an urgent nuclear physics committee meeting was held at the University of Tokyo on April 20. The subject to be discussed was how a nuclear physics group should cooperate in this unprecedented big project and led to heated, but very positive argument. On 25 April, a meeting to request the cooperative work of Fukushima Prefecture was held at the Fukushima government building. Eiji Nakashima (Atmosphere and Ocean Research Institute, University of Tokyo), Haruo Tsuruta (Atmosphere and Ocean Research Institute, University of Tokyo), Naohiro Yoshida (Interdisciplinary Graduate School of Science and Engineering, Tokyo Institute of Technology), Koji Otsuka (CNS, University of Tokyo), Masaharu Hoshi (Research Institute for Radiation Biology and Medicine, Hiroshima University), Yuichi Onda (University of Tsukuba), Mamoru Fujiwara (RCNP), Isao Tanihata (RCNP), and Hitoshi Yamamoto (ASH) attended this meeting asking for cooperation in the radioactivity nuclide map measurements.

The Chernobyl accident happened in 1986 when the Soviet Union was involved in the breakdown crisis. The radioactivity nuclide map measurement was actually performed by a leadership of IAEA after the Russian Revolution in 1989. Three years passed after the accident. Thus, important data on ^{131}I radioactivity was lost completely in case of the Chernobyl accident. Japan could not afford to follow the same path as Russia. The spirit of the participants in the Fukushima meeting was uplifted toward the immediate radioactive nuclide map measurements.

In MEXT, a committee entitled ”Investigation and Study of the Secondary Distribution of Radioactive Substances due to the Accident at the Fukushima Dai-ichi Nuclear Power Plant” was started around April 22. This large-scale project was planned to be carried out with the responsibility of ”Japan Atomic Energy Agency” (JAEA). JAEA took this project for budget execution. A question still remains about the reason why a member of Osaka University was not included as a member of this committee. Independently, the pilot program was carried out during the consecutive holiday season in May. On the basis of the γ-ray measurements from soils sampled in Fukushima, the original protocol draft written by Hoshi and Onda was further polished up after long discussions among many scientists. The final protocol was submitted to the MEXT committee on 5 May. This protocol greatly contributed to obtain highly reliable results because of a standardized process for soil samplings and γ-ray measurements with HPGe detectors, which were performed by more than 500 inexperienced people.

8.4 Logistics and Storage of soil sampling kits

It was hard to find a transport company to send a huge number of soil sampling kits to Fukushima just after the Great East Japan Earthquake. A major trouble came from difficulties to look for a person who could drive a truck from Osaka to Fukushima. As mentioned above, first, the truck drivers felt a serious fear to go to the unknown areas with a high radioactivity dose in Fukushima. Second, many sections of highways connecting from Tokyo to Fukushima were closed. Another problem was that we could not find a warehouse to keep a huge number of sampling kits in Fukushima. Fujiwara asked Mr. Koyama, an officer of the Fukushima prefecture government, to look for an appropriate warehouse in the neighborhood of Fukushima city. But it was almost impossible for him to find such a convenient warehouse because many companies and people seriously damaged by the big earthquake and the subsequent Fukushima Dai-ichi NPP accident could not find it in time of confusion.

Finally, on 6 May 2011, Hitoshi Yamamoto, the director of ASH, Osaka Univesrity, made a telephone call to Hiromasa Arai, the president of Sanshin Metal Working Co. Ltd. to ask the possibility whether Osaka University could borrow the warehouse of the factory in the Hirata village near the Fukushima airport. Mr. Arai immediately gave us positive answer. We felt with great appreciation that the private connection between Yamamoto and Arai played an important role at most urgent situation.

Two trucks with a huge number of iron pipes started from the Horiguchi Iron Work, Co. Ltd of Hyogo to the Fukushima factory of the Sanshin Metal Working Co. Ltd in Hirata village on 10 May. They arrived at Fukushima on 11 May. Soon afterward, Sanshin Metal Working Co. Ltd. allowed us to store a huge number of soil sampling kits including U-8 containers and other various materials like gloves and napkins. It should be noteworthy that it would have been just about impossible for us to start the soil sampling project in Fukushima without the courtesy of Sanshin Metal Working Co. Ltd. We held a small town meeting to explain the details about the radioactivity and its health effect to employees of Sanshin Metal Working company and residents living in Hirata-village on 8 June during the soil sampling work. I.

Tanihata, H. Yamamoto, M. Fujwiara, and H. Nakajima of Osaka University attended this meeting. The speakers in this meeting were Fujiwara and Nakajima.

8.5 Toward soil sampling

The government officers belonging to MEXT and the Ministry of Finance needed to obtain the formal allowance of the budget enforcement for the radioactivity nuclide map project from the Ministry of Finance. This was a technical problem of the budget usage of the government. The Ministry of Finance had to check the outline of the project since the proposed project was quite new in Japan. Government administrative officers had to understand the details of the project. All the technical problems were solved on the government side before the end of May. It was decided that the soil sampling work would formally start from June 4. The delay of this starting date due to the official process resulted in losing important information on the fallout radioactivity of ^{131}I with a short half life of 8 days.

We arranged to set up the headquarters at the "Fukushima Adatara Fureai Center" in Nihonmatsu city. Soil sampling work started in cooperation with scientists who lodged at the neighboring hotels. First, a hotel with the accommodation up to 100 people was not found even in Fukushima-city, and the neighboring big towns such as Kohriyama-city because of damages due to the M9 earthquake. We encountered a serious difficulty to start the soil sampling work. This difficulty was solved soon. Fortunately, we could find "Azuma ryokan", a Japanese-style hotel in the Adatara hot-spring resort area near the headquarter. The "Azuma ryokan" was temporarily closed down because of the earthquake and the following radioactive fallout due to the Fukushima Dai-ichi NPP accident. We negotiated with the owner of the "Azuma ryokan" to offer 100 people the lodging rooms and the daily meals for the soil sampling work.

8.6 Soil Sampling Work

More than 400 volunteers from more than 90 organizations took part in the soil sampling from June 4 to July 8, 2011. To ensure the quality of the soil sampling and spectrometry, the soil sampling procedures underwent with careful pre-examination beforehand. The protocol describing the standard methods for soil sampling and γ-ray spectrometry was determined after extensive discussions with specialists, and was approved by the Committee on the Construction of Maps for Radiation Dose Distribution [2]. All soil samplings were carried out according to this protocol. Sometimes, planned sampling locations could not be reached by car because the access roads had been severely damaged by the earthquake or tsunami. Overall soil sampling was performed at 2,138 locations.

After the pilot soil sampling work in May, it was concluded that the permeation depth of deposited radioactive nuclide was less than 5 cm from the ground surface. Thus, the surface soil was dug up to 5 cm depth using a special soil sampling device. Collected soil was sufficiently mixed to ensure reliable measurements since the detection systems were calibrated assuming that radioactivities were uniformly distributed in the container. The sampled soil was put into a plastic container with a diameter of 5 cm and a height of 5 cm (U-8 type). Five soil samples were collected at one location in order to compensate for a large deviation of deposited radioactivity with the sampling point. When sampling was not easy, soil samples less than five were collected. In total, 10,915 soil samples were collected.

Figure 2: Photo of the meeting at "Adatara Fureai Center" in Nihonmastu city on 4 June. About 100 people came together and heard the explanation on the protocol describing the details for the soil sampling work.

MEXT took the role of obtaining the agreement with each mayor of cities, towns and villages of Fukushima, Mityagi, and Yamagata prefectures for the radioactive nuclide map project and its associated soil sampling work. Almost everyday, MEXT officers made a telephone call to mayors to obtain the full allowance for the soil sampling work on each day at individual places at parks, empty spaces, and rice fields. Day by day, MEXT officers got each agreement with relevant mayors for soil sampling. It was greatly to the MEXT's credit that they completed this mission successfully. Everyday, the JAEA group led by Kimiaki Saito decided the soil sampling points, the staff numbers necessary for the soil sampling work, and arranged the taxis to carry them to the soil sampling points. Each team collected soil samples at several locations per day. Every day, up to 30 teams worked on soil sampling. In the 20-km zone where entry was restricted, the Federation of Electric Power Company of Japan, which was responsible for routine monitoring in the zone, supported the soil sampling work. JAEA sent the soil samples collected everyday to Tokyo University CNS for the next process of the measurement of γ-rays from soils. Figure 2 shows a photo of the morning meeting at the "Adatara Fureai Center".

RCNP and ASH of Osaka University were in charge of supplying the soil sampling devices, and the lodging of people. Decision of the measurement points often was made at midnight every day because the agreement with mayors was always delayed in the complicated situation after the Fukushima Dai-ich NPP accident. The staff assignments could only be made thereafter. Takashi Saito of ASH, Osaka University, played this important role to make the human arrangement with only a short or rest every night and day. It should be noted that the soil sampling work from 4th June to the end of June would not have been completed well without the great work and devotion of Takeshi Saito.

Figure 3 shows the photo after the sampling work. Many people worked to check the correspondence between collected soils in the U-8 container and their working record describing the detailed properties of sampled soil locations.

Everyday, collected soil samples were sent to the CNS, Tokyo University, by using a express home delivery service. Susumu Shimoura served to improve the methods to obtain reliable results of the γ-ray measurements from soils. Radioactive fallouts had percolated through soil. The density of the radioactivities was dense at the surface of the ground, and decreased as increasing the depth. In order to avoid large variation of γ-ray measurements, the soil taken by using a U-8 soil sampler was once moved into a plastic bag, and was stirred to homogenize the contained radioactivities. Even after these processes, there still were sometimes large deviations among five independent soil samples taken at one location. Shimoura examined all the qualities of the γ-ray measurements.

The radioactive nuclide map program was an unprecedented large-scale measurement with many people at many locations. Each measured result with a HPGe γ-ray detector was stored in the data file with a corresponding log note. We wish to mention my appreciation to the work by MEXT which played a leading role for this purpose, and the work by the team led by Kimiaki Saito at JAEA.

However, the one-month delay to start the project, which had first been scheduled during the holiday season of May, left the problem that we missed the precision data for the radioactivity map of ^{131}I with a half-life of 8 days. This regrettable thought remains with us. If we could procure a number of Compton suppressed HPGe γ-ray detectors, we could extract the ^{131}I intensities from the peak at E_γ=365 keV on top of a huge background originated

Figure 3: Photo after the soil sampling work. All soil samples and data sheets were collected for the subsequent γ-ray measurements.

from ^{137}Cs, ^{134}Cs, and other radioactive nuclei, because of suppression of the background at the low energy part of the γ-ray spectrum. Several months later, ^{137}Cs, ^{134}Cs, and other soil radioactivity maps were completed. These achievements were the result of a great cooperative work of Japanese scientists.

8.7 γ-ray Spectrometry and data analysis

Twenty-one organizations helped to measure the γ-ray intensities from each soil sample capsuled in a U-8 container with HPGe detectors. People in some institutes had no expertize in measuring environmental samples. It was very important to have preliminary tests using mock-up samples for checking fundamental reliability of measured results. The measured results were compared with those independently obtained at different institutes. We found that rather good agreement could be obtained to ensure the reliability of the measurements.

The obtained results were calibrated by comparing with those for the standard sources provided by the International Atomic Energy Agency (IAEA) and the Japan Chemical Analysis Center (JCAC). Peak counts in the spectrometry were converted to the radioactivity intensities according to the calibration data. The radioactivity intensities were averaged over soil samples collected at one location, where we took usually five samples. The averaged radioactivity intensity was converted to the values per unit ground area (Bq/m^2).

Radioactive ^{134}Cs and ^{137}Cs were detected at every location. In case of other radioactive nuclides, statistically significant data were obtained from soil samples at a limited number of locations because of short half-lives and deposited radioactivity levels. When statistically significant data were obtained at least for one soil sample at one location, deposition was judged to have occurred, and the average deposition density was estimated.

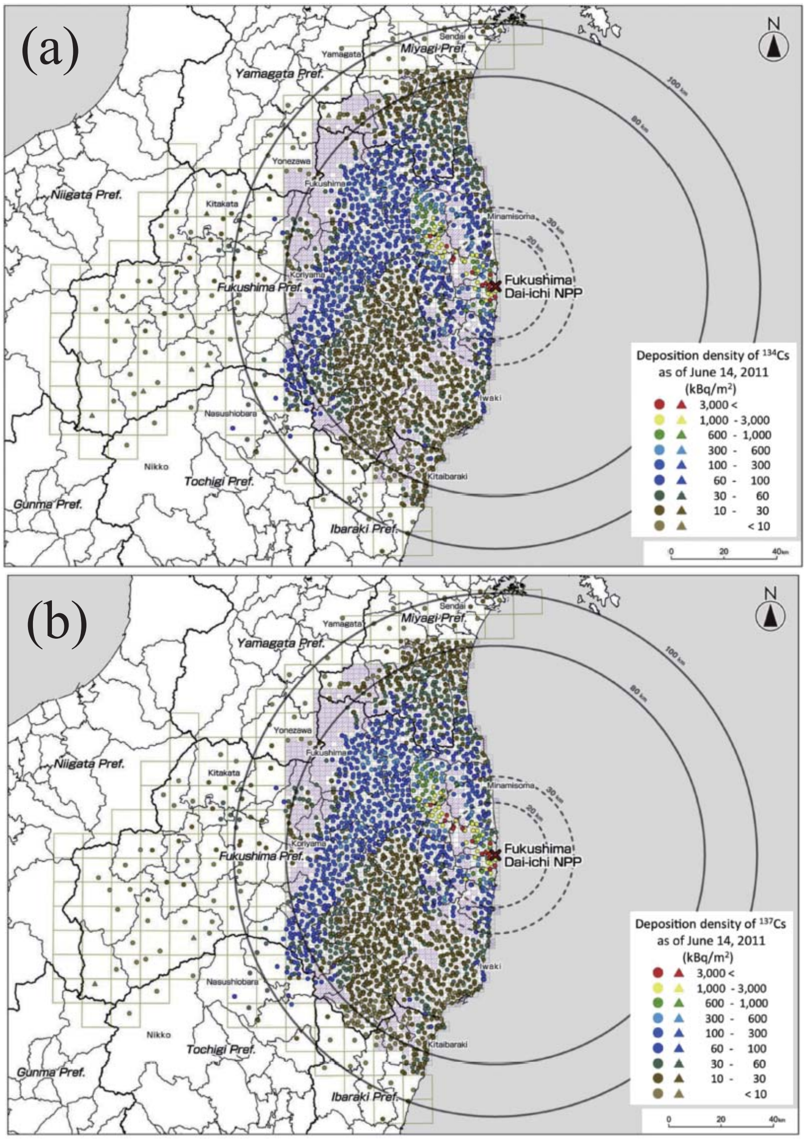

Figure 4: Deposition density maps for (a) ^{134}Cs and (b) ^{137}Cs. The radioactivity per unit ground area is shown by the colored mark at the soil sampling location. For the details, see Ref. [3].

9 Results

The details of the obtained results has been reported in Ref. [3] for ^{134}Cs ($T_{1/2}$=2.065 y), ^{137}Cs ($T_{1/2}$=30.16 y), ^{131}I ($T_{1/2}$=8.02 d), ^{110m}Ag ($T_{1/2}$=249.9 d), ^{129m}Te ($T_{1/2}$=33.6 d). In Ref [3], the distribution maps of the activity ratios of ^{131}I, $^{129\mathrm{m}}$Te and $^{110\mathrm{m}}$Ag, respectively, to ^{137}Cs are given. The regional distributions of the ratios indicated similar tendency for ^{131}I and $^{129\mathrm{m}}$Te. The ratios were relatively high at the coastal regions south of the Fukushima Dai-ichi NPP site when compared with those in the other regions. Various results for the survey of Fukushima soil radioactivities are displayed in the RCNP website [4].

9.1 Deposition density maps for ^{134}Cs and ^{137}Cs

Two results of the deposition density maps for ^{134}Cs and ^{137}Cs are displayed in Fig. 4. The ^{134}Cs and ^{137}Cs radioactivity doses were corrected on June 14, 2011 by taking into account the dose decrease due to the decay of these nuclei. As seen in Fig. 4, the basic pattern of the ^{134}Cs map was similar to those of ^{137}Cs. The radioactivity ratio of ^{134}Cs/^{137}Cs is almost constant to be 0.91 at many locations in the map. This result suggests that the cesium radioactivities were released with the same isotope ratio of ^{134}Cs/^{137}Cs from three reactors in the Fukushima NPP accident.

Fission fragment yields of cesium isotopes from the ^{235}U(n, f) reaction are ^{133}Cs (6.09%), ^{134}Cs (7.6×10^{-6}%), ^{135}Cs (4.9×10^{-4}%), and ^{137}Cs (6.3%) [5]. The ^{134}Cs isotopes with a half-life of 2.065 y are not directly produced as fission fragments from the ^{235}U(n, f) reaction. The slow-neutron capture process with 29 barns converts the stable ^{133}Cs to ^{134}Cs. The ratio of ^{134}Cs/^{137}Cs is expected to be zero at the initial operation time of a nuclear reactor if only the ^{235}U fuel is assumed to contribute the heat generation. During the operation of a nuclear reactor, the ^{134}Cs/^{137}Cs ratio increases gradually. However, when the operation time is longer than the half-life of ^{134}Cs, this ratio tends to decrease because the production and decay speed of ^{134}Cs reaches an equilibrium while the relative yield of ^{137}Cs with a longer half-life of $T_{1/2}$=30.16 y increases. Thus, the observed ratio of ^{134}Cs/^{137}Cs, 0.91, is a good indicator to estimate the operation time of a nuclear reactor. In case of the Fukushima Dai-ichi NPP, it is reported that the NPP operation stopped for about 3 months every year [6]. Some of nuclear fuel rods were expected to be exchanged during the maintenance time.

Figure 5 shows a simple calculation by Oyamatsu without taking into account such maintenance times [8]. The comparison indicates that the ratio of 0.91 corresponds to the NPP operation time of 600 days. Taking into account the long maintenance time of about 3 months every year, the increase of the ^{134}Cs/^{137}Cs ratio is expected to be prolonged in time because of the faster decay of ^{134}Cs ($T_{1/2}$=2.065 y) compared with those of ^{137}Cs ($T_{1/2}$=30.16 y). A detailed analysis to reproduce the obtained ^{134}Cs/^{137}Cs ratio of 0.91 would

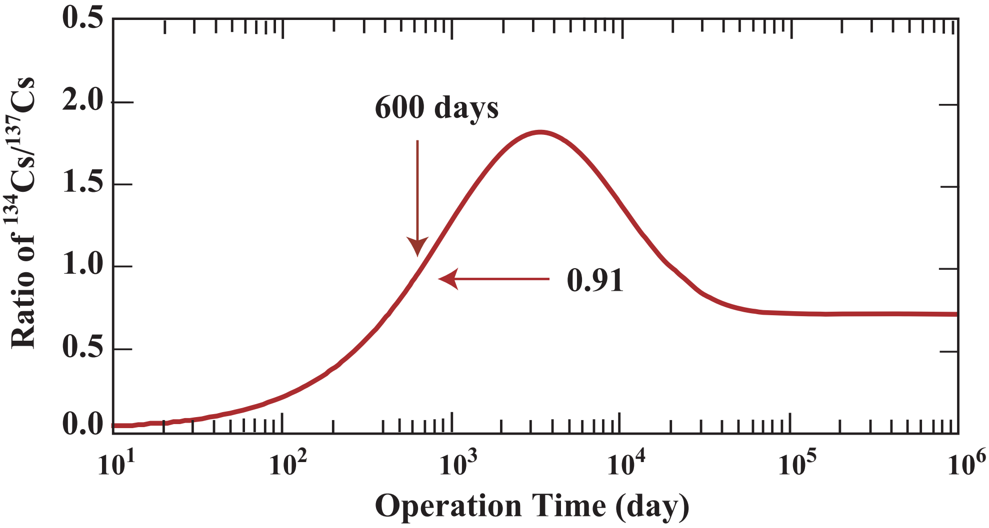

Figure 5: The deposition isotope ratio of ^{134}Cs/^{137}Cs as a function of the operation time calculated by Oyamatsu [7, 8]. In this simple calculation, the maintenance time of the NPP, and the regular change of nuclear fuel change are not taken into account. From the measured ratio of 0.91, it is approximately inferred that the Fukushima Dai-ichi NPP operation time has been around two years before the accident.

be beneficial to understand what happened in the Fukushima Dai-ichi NPP before the accident.

9.2 Radioactivity ratio of ^{131}I

Figure 6 shows the map for the deposition densities of ^{131}I. The radioactivity densities per unit ground surface, (Bq/m^2), on June 14, 2011 are indicated by colored marks at soil sampling locations. The major soil sampling work was completed on this date. Thus, the ^{131}I radioactivity doses were corrected for to this date by taking into account the dacay effect of ^{131}I ($T_{1/2}$=8 d). It was regrettable that the first campaign of the soil sampling work started on 4 June and ended on 14 June when about 90 days had passed after the NPP accident so that we could not obtain a good radioactivity density map for ^{131}I because the ^{131}I radioactivities had decayed out significantly. However, some features of the ^{131}I fallout activities could be recognized.

The area situated in the northwest of the Fukushima Dai-ichi NPP site was most contaminated. Since the wind direction rotated clockwise according to the passage of elapsed time from March 11 to March 20 in Fukushima prefecture, there appeared a densely contaminated area along the southern coastal regions. The deposition densities for ^{131}I shows good correlation with those for ^{137}Cs in the southern coastal regions. Those in the other regions showed different correlations. This suggests that at least two dominant

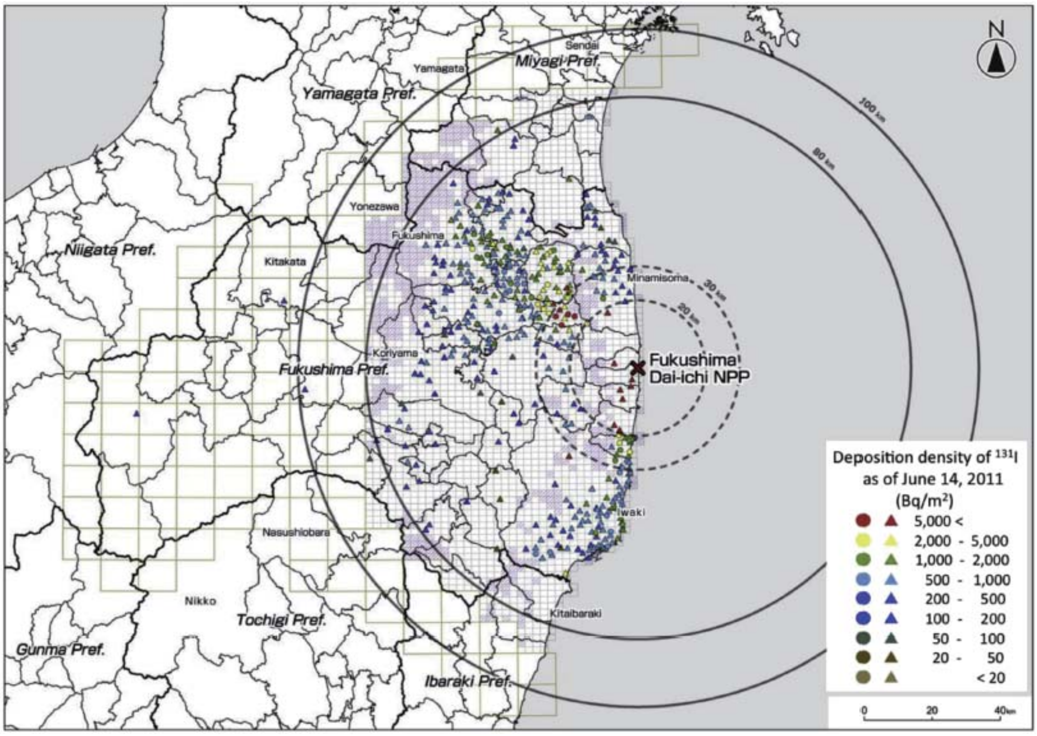

Figure 6: Deposition density map for ^{131}I. The radioactivity per unit ground area is shown by the colored mark at the soil sampling location. For the detailed discussion, see Ref. [3].

pathways for contamination existed due to different radioactive plumes or deposition processes.

According to the review of atmospheric diffusion simulations carried out after the Fukushima Dai-ichi NPP accident by Yamazawa *et. al.* [9], the radioactive nuclide deposition in the southern coastal regions was due to the plumes released on March 14, 15 and after March 20. The monitoring data in Ibaraki prefecture indicated that relatively high fractions of ^{131}I appeared in the radioactive plumes from the Fukushima Dai-ichi NPP site to the southern direction during these periods [10]. The observed data were consistent with the results from the atmospheric diffusion simulations.

10 Radioactivities on plant leaves

Many towns in Fukushima prefecture are surrounded with beautiful landscapes consisting of small cultivated land, rivers, and forests in a mountainous regions. About three-quarters of Fukushima's land area is mountainous and two-thirds is covered with forest. The mountain percentage is much high in rural villages.

We can calculate the total amount of the ^{134}C and ^{137}C radioactivities by using the measured soil radioactivity mapata. The total amount of spread-out radioactive isotopes is estimated to be less than a few gram.

Radioactive Cs isotopes released from the Fukushima Dai-ichi NPP were water soluble, and were transported by wet plumes and clouds to the northwestern direction. Then, snowy weather happened in cold days in March 2011. Snow and rain dropped at plain areas and at mountain areas in the northwestern part of Fukushima prefecture like Iiitate village. Mountain areas in Fukushima prefecture are mostly covered with the evergreen trees such as Ceders and Japanese Cypresses. Fallout snow or rain first drops on tree or grass leaves. Radioactive Cs isotopes on these leaves soon combined with organic substances since Cesium is chemically very reactive. Therefore, the fallout radioactivities are expected to leave an evidence on a leaf as an finger print of deposition.

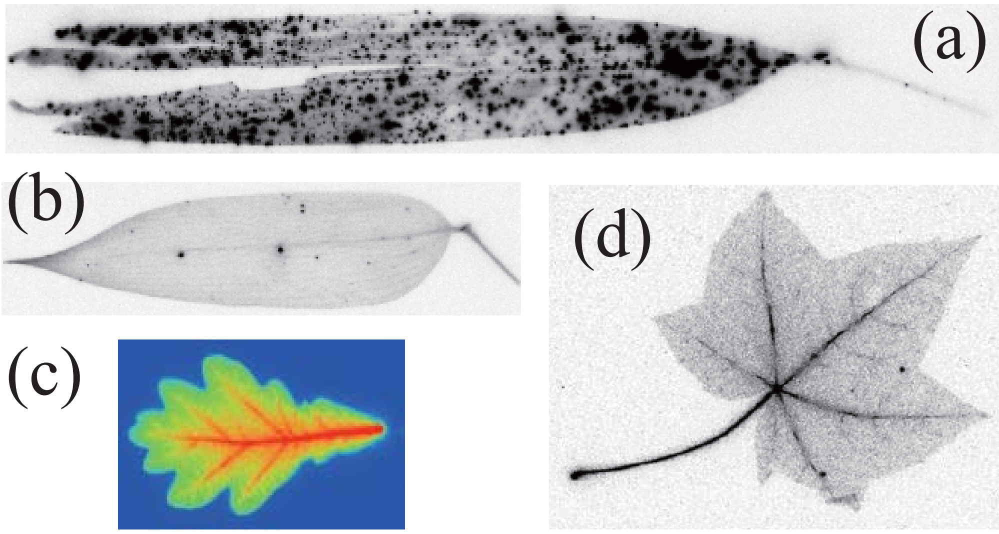

Figure 7: Semi-quantification pictures of radioactive distributions taken using the imaging plate. (a) Fallout radioactivity distribution on an old bamboo leaf. (b) Fallout radioactivity distribution on a young leaf. These leaves were sampled at the mountain site in Iitate village along the route 115 car road, located 42 km from Fukushima Dai-ichi NPP when a 109 day period had passed after the NPP accident on 12 March 2011 (see Ref. [12] for details). (c) Radioactivity distribution absorbed into an oak leaf sampled on an high radioactivity are in Masani village, Belarus in 1997, 11 year after the Chernobyl NPP accident. (d) Radioactivity distribution absorbed into a Japanese maple leaf, which was sampled in September 2014 at a Cedar forest in Iiitate village.

Figures 7(a) and (b) show the radioactivity images for an old bamboo leaf grown before the NPP accident, and for an young bamboo leaf grown after the accident, respectively [12]. A bamboo leaf grows in springtime and lives as a green leaf for a few years. After a few years, the dead leaf falls to the ground. Therefore, snow or rain with radioactivities in March 2011 were expected to fall on the old bamboo leaf sampled in Iiitate village. On the other hand, as seen in Fig. 7(b), radioactivities did not fall on the young bamboo leaf grown after April 2011. In Fig. 7(a), radioactivites are observed as dot images with various sizes. Generally speaking, this is a typical evidence that snow or rain including radioactive cesium isotopes have fallen on tree leaves, and are trapped with organic substances in the surface of the leaves.

Figure 7(c) shows the radioactivity image of an oak leaf sampled at a highly contaminated area in Masani village, Belarus in 1977, 11 years after the Chernobyl NPP [12]. Oak is a deciduous tree. In every autumn, oak leaves fall to the ground as death-leaves. Thus, the image shown in Fig. 7(c) indicates that radioactivities are absorbed from the polluted ground soil through the oak tree root, and arrives at the leaf veins. Radioactivities are continuously spread out from the leaf veins to the leaf apex. The smooth image pattern of radioactivity distribution for oak leaf is totally different from those for the old bamboo leaf which consists of many dot images due to fallout radioactivities.

Figure 7(d) shows the image of radioactivity distribution absorbed into a Japanese maple leaf sampled in September 2014 in a Cedar forest in Iitate village, 4.5 years after the Fukushima Dai-ichi NPP accident. In every autumn, dead maple leaves drop down to the ground. It is evident that radioactivities are absorbed from the ground soil through the maple tree root, and arrive at the maple leaf through veins.

It is inferred that the radioactivity circulation in the natural environment has started. The leaf cycle of evergreen trees is 2-4 years. Radioactivities attaching on withered leaves of evergreen trees fell down on the ground around 2014, and made litter layers with dead-leaves and radioactivities deposited when the Fukushima Dai-ichi NPP accident happened. It is expected that all radioactivities due to the Fukushima Dai-ichi NPP accident had fallen to the ground in 2014. All radioactive materials have once stopped at a ground surface as a litter layer in a forest without permeating the ground soil. Dead leaves proceed to be gradually decomposed by bacteria, and to assimilate into forest soils in future. These radioactively polluted dead-leaves will be partly blown away from mountains to plains by the wind. Radioactivities are now in the slow process toward the equilibrium state. In a very long period, all radioactivities will be diluted in the natural environment, and reach at the equilibrium state. After a long time passage, all radioactivity densities will be equally distributed based on the law of entropy increase in physics (the second law of thermodynamics). No body cannot escape from this basic law in physics. The problem to be answered is to look for the speed of this equilibration process in nature. No body knows the answer at present. It is important for us to study the transient slow process toward the radioactive equilibrium as a global environmental problem.

11 Summary and Final Comments

In thie article, we have reported on what happened following the Fukussima Dai-ichi NPP accident triggered by the Great East Japan Earthquake on 11 March 2011. Especially, we have reported the activities of the nuclear scientists in Japan. Since many important view-points seem to be still lacking, there may be some unfair descriptions from the standpoints of other persons.

The deposition densities of γ-emitting radioactive nuclides have been obtained by collecting a large number of soil samples around the Fukushima prefecture. All collected soil samples have been carefully analyzed using Ge detector spectrometry. The deposition density maps have been constructed for the critical γ-ray emitting nuclides of ^{134}Cs, ^{137}Cs, ^{131}I, ^{129m}Te and ^{110m}Ag. All details of the soil sampling, the γ-ray measurement with HPGe detectors, and the obtained results have been reported in Refs. [3, 11].

In order to summarize the present report, we wish to discuss what we came to realize during the soil sampling work and later.

1. It was really regrettable that we could not start the soil sampling in May 2011. Administration work took an inordinately long time. Because of this delay, the deposited ^{131}I radioactivity map was not obtained in any useful way. In order to avoid these kinds of delay for the nuclear power plant accidents, it is important for Japan to organize an implicit emergency team as a national agency for nuclear safety & victim's aid before any future nuclear power plant accident. There are such organizations for nuclear emergency in the United State.and France. We should not artificially create a nuclear-safety mythology to avoid critical discussions about NPP operations.

2. In case of radioactive fallout mapping, there was no protocol prepared before the Fukushima Dai-ichi NPP accident in Japan. To prepare the protocol for sampling radioactive soils and the γ-ray measurements, it was essential to first examine the validity of the applied methods. It took a long time for the protocol to be implemented for the actual measurements. Certainly, we need to polish up the present protocol prepared in 2011.

3. In-situ Ge detectors are a powerful alternative in comparison with the soil sampling method and radioactivity measurements. We can use in-situ Ge detectors for urgent γ-ray measurements for identifying the amounts of radioactive ^{131}I isotopes with a half-life of 8 days. However, the number of such Ge detectors are not enough to perform the urgent measurements needed at the next time when the NPP accident will happen in Japan. One of the ways is to strategically prepare in-situ Ge detectors at the radiation safety devisions in many universities and national research institutes. These in-situ Ge detectors are very useful for the daily radiation safety work as well as the urgent work for the accidents in relation to radioactivities.

4. Obviously, it is very difficult for Japan to prepare many in-situ Ge detectors in one country. Nuclear power plants have been already constructed in many countries in Asia, Europe, and America. When the NPP accident happens in other countries in future, it is inevitable for Japan to have an international collaboration for making the deposited radioactivity maps. This kind of international collaboration is commonly performed in case of the disasters due to big earthquakes, tsunamis, and catastrophic floods. Japan always joins such international collaborations to contribute the peace making efforts in the world.

5. We need to understand that the radioactive pollution is totally different from the normal soil pollution with chemical substances including lead, arsenic, fluorine and cadmium. In case of chemical pollution, chemical elements are mixed with the soil as an impurities at the level of 10^{-6} (ppm order), which is a lower limit of the normal chemical purification technique. However, the radioactive pollution level is much lower. For example, when we have a ^{137}Cs deposition density with 3000 Bq/m^2, the corresponding chemical impurities is at 10^{-14} level, which is significantly lower than the feasibility of chemical separation. Therefore, the removal of the radioactive Cs strongly bound with organic materials in soils is practically impossible.

6. The 662 keV γ-rays of ^{137}Cs can pass 100 meters in air with 40% intensity decrease. They can go 200 meters in the air with intensity decreased to 16%. This aspect is totally different from the pollution due to chemical materials which are almost fixed in soil. In addition, there is Compton scattering due to oxygen and nitrogen in the air. This causes the so-called sky shine effect. Around 14% of the 662 keV γ-rays comes to the ground from the air by the sky shine effect even if we put a heavy wall surrounding the house. The researchers of French IRSN have performed a test simulation to estimate how much radioactivity came from the outside when we decontaminate a 10 m radius area within a flat plain with a 1-km radioactively polluted radius. The radiation dose rate decreases to only around 35% by such a decontamination procedure. The decontamination effect is expected to decrease at the mountainous areas in Fukushima.

7. Many news media reported the leak of radioactivities due to the Fukushima Dai-ichi NPP accident because of an overwhelming feeling of fear and anxiety against radioactivities. On the other hand, it is an undeniable fact that human beings have continuously receive radiations from environmental radon gas and from the food containing ^{13}C and ^{40}K radioactivities. From the view point of nuclear scientists, correct understanding of radioactivities and their biological behavior seems to be still lacking even in the university education level. Unfortunately,

most of university teachers and students do not have correct knowledge of the phenomenon of radioactivity. At present, most of education at higher education organizations seem to teach the techniques for students to earn money because of the economically severe situations for young people. The education for radioactivities may be of secondary importance. However, it is of utmost importance that Japan should offer good education opportunity at the university level for correctly understanding radioactivity and its biological effects. Otherwise, we cannot hope to improve the economy situation in the Fukushima area as well as in Japan.

As final remarks of the present report, it should be noted that the Fukushima story is not yet over; After passage of four year after the Fukushima Dai-ichi NPP accident, health fears have decreased, but economical fear for future has increased. Depopulation in the radioactive contamination area in Fukushima has already accelerated. We have to contrive drastic strategy to improve the present situation by taking into account people's feelings in Fukushima. There are many things to be done for Fukushima at present, and in future.

12 Acknowledgments

The radioactivity deposition map project was financially supported by the Ministry of Education, Culture, Sports and Science (MEXT). We thank all people who directly and indirectly supported the project. This large-scale project could not have been completed within a short time after the Fukushima Dai-ichi NPP accident without such support. We also thank very much the staff members of the Nuclear Emergency Response Headquarters in MEXT, Tokyo, for the hard work to realize and complete the project.

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