As Japan’s sole comprehensive R&D institute in the field of nuclear energy, we are conducting R&D to promote the decommissioning of the TEPCO’s Fukushima Daiichi NPS (1F), as well as environmental restoration after the 1F accident (Fig. 1-1). Our research results are described below.

Towards the decommissioning of 1F, the Collaborative Laboratories for Advanced Decommissioning Science (CLADS), as an international research hub based on the Mid- and Long-term Roadmap formulated by the Inter-Ministerial Council for Contaminated Water and Decommissioning Issues, is promoting research and development on: 1) clarification of 1F-accident-progression scenarios (Topics 1–1 and 1–2); 2) retrieval of fuel debris from nuclear reactors (Topics 1–3 and 1–4); 3) treatment and disposal of radioactive wastes generated by the decommissioning work (Topics 1–5 and 1–6); and 4) remote-controlled instrumental technologies (Topics 1–7–1–9).

The CLADS main building began operation in Tomioka Town in FY2017 (Fig. 1-1, right), and has integrally promoted R&D and human-resource development by industry, academia and government. Also, we have held many Fukushima research conferences attended by domestic and international researchers in Fukushima prefecture (Fig. 1-1, bottom right). The Research Fund for Promoting Projects on Decommissioning Research was founded by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) in FY2018. The purpose of this fund is to promote fundamental R&D and human-resources development (HRD), mainly at CLADS. With this fund, we will reinforce cooperation with universities and research institutes so that we can continuously achieve a more stable environment for R&D and HRD.

The Naraha Center for Remote Control Technology Development, which began operation in FY2016 in Naraha Town, is available for external users who are interested in the development of the remote-control devices to be used in decommissioning work and nuclear disasters (Fig. 1-1, left). The number of facility users from industry, academia, and government exceeded one hundred (38 in FY2016 and 64 in FY2017) (Fig. 1-1, bottom left). We are improving the test equipment by developing robot simulators and robot test methods to encourage facility use.

The Okuma Analysis and Research Center is intended to analyze radioactive wastes and fuel debris for development of long-term waste management. The Center consists of three buildings: an Administration Building, Laboratory-1, and Laboratory-2 (Fig. 1-1, top right). The Administration Building, which consists of office rooms, meeting spaces, workshop, etc., was opened in March 2018 in Okuma Town. Laboratory-1, which is now under construction, will be used for analyzing low-to-medium-radiation-level rubbles and secondary waste. Laboratory-2, which is at the design phase, will be used to analyze high-radiation-level material, such as fuel debris.

These three centers described above are regarded as decommissioning-related facilities that will play a part in the Fukushima Innovation Coast Framework, and will therefore contribute ever more to 1F decommissioning.

For environmental restoration, the Fukushima Environmental Safety Center is carrying out R&D in accordance with the Medium- and Long-Term Activities of the Centre for Environmental Creation (CEC), in collaboration with Fukushima Prefecture and the National Institute for Environmental Studies (NIES) (Fig. 1-1, top left). We have cooperated with Fukushima Prefecture and the NIES of the CEC to investigate and analyze the environmental impact due to the forest fire that occurred inside the difficult-to-return zone of Namie Town in April 2017. The Fukushima Environmental Safety Center has been developing technologies for environmental monitoring and mapping (Topics 1–10 and 1–11) in order to establish methods for monitoring contaminated forests, river areas, inshore areas and so on. Moreover, the Center conducts research on environmental dynamics (Topics 1–12–1–15) to predict and resolve the migration of radioactive materials in the environmental, as well as research on decontamination and volume reduction (Topics 1–16–1–18). The Center is disseminating R&D results with a web-based system that will meet a wide variety of needs for researchers, municipalities, local residents, and so on (Topic 1–19).

We will continue to offer technical expertise to promote environmental restoration and decommissioning of 1F by concentrating expertise. We are contributing to Fukushima’s revitalization through development of regional industries and HRD by network buildings and cooperation with regional industries, research institutes, and educational institutes.
During severe accidents (SAs), fuel debris (a mixture of fuel and the other materials) is generated by melting of reactor cores and is relocated to the lower part of reactors. Information concerning fuel debris, including its distribution, composition, and re-criticality, is required for its removal. Therefore, obtaining this information is important for decommissioning of the TEPCO’s Fukushima Daiichi NPS (1F). However, melting and relocation of fuel debris are complicated phenomena, making this information quite difficult to obtain. Numerical simulations are effective tools, but conventional SA-analysis codes are of little use because they include various assumptions about phenomena and simplify the shapes of reactor-core structures to shorten the computational time.

Therefore, the fuel-debris-melting and relocation process must be simulated precisely. To this end, we have developed a numerical-simulation code for melting and relocation behavior called JUPITER, based upon a mechanistic methodology. JUPITER can simulate melting and relocation without assumptions or simplification using basic equations governing the thermal-hydraulic behavior of the fluids.

We numerically simulated melt-relocation behavior to the lower part of the primary containment vessel (pedestal) using JUPITER. In this simulation, we decided the largest-density material, molten UO$_2$, should be poured first, followed by the lower-density materials SUS, Zry, and B$_4$C. As shown in Fig.1-2(a), UO$_2$ spreads over the pedestal and is accumulated in two cavities (sump pits) with solidification. Then, fuels and other materials mix in a complicated fashion (Fig.1-2(b)–(d)).

Fig.1-3 shows a cross-sectional view of the pedestal along its center to illustrate the accumulation state inside the sump pits. As shown in Fig.1-3, the melt near wall of the pedestal is solidified by heat transfer to the wall. The heavier material, UO$_2$, is located at the bottom and the lighter material, Zry, is located at the upper layer. This distribution is given by mechanistic estimation for melt relocation and solidification based upon the physical governing equations.

In addition, we try to evaluate the re-criticality of fuel debris in the pedestal using the continuous-energy-neutron-transport Monte Carlo code MVP. The required fuel-debris distribution for re-criticality analysis is evaluated using the above-mentioned fuel-debris-relocation simulation of JUPITER. In a normal re-criticality analysis, it is assumed that the fuel debris is uniformly mixed. Here, with the distribution given by JUPITER, re-criticality can be calculated using a more realistic condition. If water exists in fuel debris, re-criticality occurs readily in theory. In this simulation, we calculated re-criticality with and without water in the fuel debris. It is found that re-criticality does not occur except where an unrealistic amount of water exists in the fuel debris.

In the near future, we will validate the applicability of JUPITER to the relocation and accumulation of fuel debris by comparing simulation results with experimental ones and perform debris-accumulation simulation under several parameters. In addition, we will contribute to the decommissioning of 1F by evaluating the characteristics and re-criticality of fuel debris.

Reference
Emission of insoluble cesium (Cs) particles was detected in environmental samples collected during the early stage of the TEPCO’s Fukushima Daiichi NPS (1F) accident. Particles with diameters of 1 μm to 1 mm were particular ejecta of the accident. There were at least two types of particles of which the main element is silicate (Fig.1-4). This silicate makes the particles insoluble in water. Type A is a few μm in diameter and contains a small amount of radioactive Cs within it. However, the concentration of Cs is so large that characteristic X-rays can be detected using energy-dispersive X-ray spectrometry (EDS), which has a much higher detection limit than other methods. By contrast, Type B particles are hundreds of microns in size and can be seen with the naked eye, but the concentration of Cs is lower than in Type A. The radioactivity concentration per unit volume is called the specific radioactivity, but in order to clarify the above relationship, when two kinds of particles are illustrated by the particle volume and the Cs concentration contained, the inclination of Type B is smaller than that of Type A. The results of previous studies (● and ● in Fig.1-5) of particles considered to have been released at the same time (midnight on March 14) as Type A also agreed with specific radioactivity relationships.

Type-B particles with low specific radioactivity were confirmed to have been released by the 1F Unit 1 reactor hydrogen explosion on March 12. Moreover, Type-B particles were deposited over a limited region north of the polluted nuclear plant immediately after the explosion took place. Observing Type B, fibrous silicate compounds were confirmed to adhere surface (Fig.1-6(a)). Based on this discovery, we investigated the silicate compounds used around the reactor building. The elemental composition of the heat-insulating material used inside the building and the constituent elements of Type B nearly agreed (Fig.1-6(b)). This result shows that cesium filled in the reactor building, adsorbed onto fibrous insulation made of silicate compounds, shrunk, spread due to the hydrogen-explosion heat and the blast, and scattered as Type-B particles into the environment. On the other hand, Type-A particles, whose generation process has many unknown features due to their high specific activity, are a subject for further study.

Reference
Chemical Reaction between Stainless Steel and Cesium
— Estimation of the Cesium Distribution within the TEPCO’s Fukushima Daiichi NPS —

At the TEPCO’s Fukushima Daiichi NPS, information on cesium (Cs) distributions within the nuclear reactors is necessary for managing the radiation exposures of workers, because Cs is a major source of $\gamma$ ray. In particular, evaluation of Cs-chemisorption behavior, which is a reaction between Cs vapor and the stainless steel (SS) used in structural materials, is essential. Thus, such Cs-chemisorption amounts are estimated using severe-accident-analysis codes with Cs-chemisorption models. However, there is a large discrepancy among the analytical codes. One reason for this is that the chemical forms treated in their models differ.

Since Cs has various compounds with differing vapor pressures and water solubilities, the revaporization and transport behaviors of Cs from structural materials to gaseous and aqueous phases significantly depend upon which Cs compounds are formed. Therefore, clarification of the chemical forms of Cs adsorbed onto SS, which is widely used inside reactors, or clarification of reactions between SS and Cs vapor, is important for improving the estimation of Cs distributions within nuclear reactors. However, although Cs has been found to react with silicon (Si) impurities in SS, various Cs silicates have been reported and the major chemical form remains unclear, possibly due to the micron sizes of the reaction products.

Thus, we aim to develop an analytical technique to identify the chemical forms of micro-sized compounds by combining micro-hard-X-ray photoelectron spectroscopy (HAXPES) with scanning electron microscopy (SEM). Furthermore, since the chemical forms may depend upon the Si content in SS, Cs chemisorption tests were performed using different Si-content samples. As a result, the compounds formed by chemical reaction between SS and Cs vapor were successfully identified, and their distributions were clarified (Fig.1-7). In the future, we will try to improve estimates of the Cs distribution within nuclear reactors by establishing a model that considers the chemical properties of the identified Cs compounds.

Reference
Zirconium Hinders the Oxidative Dissolution of Nuclear Fuel
— Effect of Zirconium Incorporation in Uranium Oxide upon Inhibiting Oxidative Dissolution —

Fig.1-8 Reaction scheme of U dissolution by H2O2
Oxidative dissolution by H2O2 is a well-known degradation process for UO2, but little is known about the process for fuel debris. Thus, the oxidative U dissolution was examined through experiments with simulated fuel debris.

Fig.1-9 Scanning-electron-microscope (SEM) images of the U oxide samples and a schematic illustration of the setup for the H2O2 reaction experiment
The reactions of H2O2 with UO2 and with the simulated fuel debris in aqueous solution were examined.

Fig.1-10 Comparison of the H2O2 reaction kinetics between UO2 and the simulated fuel debris
The reaction of H2O2 with UO2 results in the dissolution of U. However, the reaction with the simulated fuel debris scarcely caused the U dissolution and resulted in catalytic decomposition of H2O2, producing oxygen and water molecules.

We have shown that uranium dioxide (UO2) becomes chemically stable by forming solid solution containing zirconium (Zr). The stabilization by Zr indicates that the molten fuel debris in the TEPCO’s Fukushima Dai-ichi NPS (1F) would be stable against dissolution by chemical reactions and would remain in the reactors until it is retrieved.

Our research on the chemical reaction of the molten fuel debris is based on an understanding of the chemical degradation of spent nuclear fuel under the conditions of deep geological disposal. The spent fuel emits intense ionizing radiation and produces hydrogen peroxide (H2O2) by decomposing water molecules. In the case of direct contact of the fuel with water, H2O2 consequently reacts with the fuel, inducing dissolution of uranium (U). The U dissolution is induced by oxidation of U at the surface of the fuel (Fig.1-8). In the UO2 matrix of the fuel, U is mainly in tetravalent state that is scarcely soluble in water. However, if tetravalent U is oxidized into hexavalent state, its solubility drastically increases. This sequence of chemical reactions is expected to occur for the fuel debris in 1F. However, little is known about the chemical behavior of the fuel debris in water, because the composition of fuel debris is far different from that of the usual UO2 fuel due to melting with the materials in the reactor core.

We have performed experiments regarding the dissolution of U by H2O2, and shown that it is significantly inhibited by the incorporation of Zr (Fig.1-9). U oxide containing Zr was used as simulated fuel debris, because the fuel debris is expected to contain Zr by melting with the fuel-cladding material. When the simulated debris was exposed to H2O2, the reaction of H2O2 proceeded at a rate comparable with that of UO2. However the dissolution of U was significantly inhibited (Fig.1-10). For example, the U dissolution from the simulated debris containing 50% Zr in atomic ratio was only 4% of that from UO2. This inhibited dissolution resulted from the effect of Zr promoting H2O2 decomposition on the surface. This reaction mechanism was confirmed by analysis of gaseous products of the reaction. The reaction of H2O2 with the simulated debris produced nearly the stoichiometric amount of oxygen (O2).

We are going to further investigate the basic chemistry of molten fuel in order to support technological development for the safe retrieval and management of the fuel debris in 1F.

Reference
Characterization of Waste Generated by Contaminated Water Treatment — Analysis of Carbonate Slurry —

The Multiple Radio-nuclides Removal System (MRRS) is a contaminated water treatment facility that decontaminates the radioactive water at the TEPCO’s Fukushima Daiichi NPS (1F). The treatment generates secondary wastes, e.g., slurry and used adsorbent. Since such secondary waste is not generated during the operation of a normal nuclear power plant, a method of safely storing, processing, and disposing this waste is imperative. For this purpose, the properties of this waste must be investigated.

The carbonate slurry must be characterized preferentially because it contains highly concentrated radionuclides and is generated abundantly by the MRRS. We collected and analyzed four samples of the waste generated at different times from different sampling positions in the High Integrity Container (HIC), which stores the carbonate slurry generated by the MRRS of 1F.

We found high dose rates (surface dose rates of the vial) in the sampled carbonate slurry (10–100 mSv/h from a 10-mL sample in the polyethylene terephthalate vial (Fig.1-11)).

Fig.1-12 is a micrograph of the carbonate slurry. As a result of measuring the particle size distribution in this slurry, there were many particles of several μm.

Radiochemical analysis results of the carbonate slurry sample. In the slurry, $^{90}$Sr was present at a concentration higher than those of other radionuclides (white bar denotes the lower detection limit, which varies among the nuclides).

This work includes part of the results of the “Development of technology for treatment and disposal of accident waste”, funded by the Ministry of Economy, Trade and Industry (METI), Japan.

Reference
Safe Long-Term Storage of Secondary Wastes after Decontamination of Radioactive Water
— Evolution of Radiolysis Studies to Support Practical Issues in the Decommissioning of 1F —

Following the accident at the TEPCO’s Fukushima Daiichi NPS (1F), we have focused upon water decontamination and secondary-waste storage, and performed R&D on cesium adsorption, hydrogen (H₂) generation, corrosion of structural materials, and gas and heat flows. We then provided the data obtained as information indispensable for safety measures to TEPCO and 1F-related companies. Moreover, according to the medium- and long-term roadmap for decommissioning 1F, we have quickly complied with requests for studying and solving urgent problems and events in cooperation with TEPCO and 1F-related companies. Moreover, according to the medium- and long-term roadmap for decommissioning 1F, we have quickly complied with requests for studying and solving urgent problems and events in cooperation with TEPCO and the companies.

R&D upon H₂ generation is related to the explosion of this gas just after the accident; it is considered to be the most dangerous phenomenon in the management of radioactive materials. Since water radiolysis in H₂-generation sources is brought about by ionizing radiations emitted from radioactive materials, the temperature need not be higher than several hundred degrees centigrade; this process takes place continuously anywhere the materials for it exist. Thus, studies on water radiolysis have been performed at every step of the roadmap.

Unlike that derived from other sources, H₂ formed from water decomposition by radiation is emitted into the air through numerous processes and reactions (Fig.1-14). H₂ generation is complexly affected by many conditions and factors. Using practical materials under real conditions, several effects upon the generation have been investigated in detail and determined to be important for 1F decommissioning (Table 1-1).

Fig.1-15 shows representative experimental results. Seawater supplied at the accident works as a coolant as well as pure water (PW), while more H₂ was generated in seawater than in pure water (PW). In submerged zeolites (b), pores and spaces affect H₂ generation, leading to an enhancement that cannot be explained only by the water content within the zeolites.

Table 1-1 H₂-generation factors relevant to the decommissioning of 1F

<table>
<thead>
<tr>
<th>Effect factor</th>
<th>Main phenomenon (this work)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Seawater salt (halide ions, X⁻)</td>
<td>X⁻ scavenging of oxidizing -OH radical in its H₂ oxidation</td>
</tr>
<tr>
<td>Zeolites; solid oxides</td>
<td>Surface interaction with (binding to) oxidizing products including -OH</td>
</tr>
<tr>
<td>Liquid depth; viscosity</td>
<td>H₂ escaping from reactions in liquid phase (water) dependent on time for reaching gas phase</td>
</tr>
<tr>
<td>Liquid flow</td>
<td></td>
</tr>
<tr>
<td>Dose rate</td>
<td>Reaction between H₂ and -OH present in different isolated spurs</td>
</tr>
</tbody>
</table>

Reference
Seeing invisible radiation is important not only for protecting workers from radiation exposure, but also for understanding the contamination situation inside the TEPCO’s Fukushima Daiichi NPS (1F). Inside its buildings, radioactive substances are adhering to various objects such as floors, walls, equipment, and scattered rubble, and the contamination due to radioactive substances exists in three dimensions (3D). There is also an area with such a high dose rate that workers cannot enter or remain there. Drawing a radiation-distribution map indicating the distribution condition of radioactive substances inside 1F is extremely important for predicting the risk to workers and decreasing the amount of radiation exposure for performing decommissioning tasks. Such a map would help workers to easily recognize the locations of radioactive substances in the work environment. In addition, the information would be effective for establishing detailed decontamination plans. We have newly developed a method for visualizing the radioactive substances scattered inside the 1F building in 3D.

To this end, we fabricated a compact Compton camera that could be installed in remote equipment such as a drone or crawler robot, as shown in Fig.1-16. This camera weighs about 680 g and was fabricated based on handheld-Compton-camera technology jointly developed by Waseda University and Hamamatsu Photonics. In high-dose-rate environments, it was necessary to equip the γ-ray sensor with a large and heavy shield for radiation measurements using a conventional Compton camera. On the other hand, since our Compton camera is compact, it is possible to perform measurements with a compact shield while maintaining portability within the 1F buildings. Using this Compton camera, we conducted a measurement of the distribution of radioactive substances inside 1F3’s turbine building. We succeeded in detecting a high-dose-rate region (up to 3.5 mSv/h) inside the turbine building with an air dose rate of 0.4 to 0.5 mSv/h. Fig.1-17 shows the result of the radiation measurement, which visualizes the radioactive substances (mainly 137Cs) by superimposing their image onto a photograph of the measurement environment. The measurement time required for visualization was under 1 minute.

In addition, we also succeeded in drawing a 3D-radiation-distribution map by superimposing the image of the radioactive substances obtained by the Compton camera onto a 3D-structural model of the measurement environment. Two contaminated regions with a higher dose rate than the surroundings are displayed at the wall and floor surfaces of the measurement environment.

Fig.1-16 Photograph of a compact Compton camera
A compact Compton camera consists of a γ-ray sensor, signal-processing unit, and an optical camera.

Fig.1-17 Visualization result of radioactive substances inside 1F3’s turbine building
Strong contamination was detected on the hose located on the floor surface.

Fig.1-18 3D-radiation-distribution map
A radiation-distribution map was created by superimposing visualized images of radioactive substances onto a 3D-structural model of the measurement environment.
Remote Laser-Sampling Technologies
— Development of Concrete-Sampling Technologies Using High-Power Lasers —

Safe and secure decommissioning of the TEPCO’s Fukushima Daiichi NPS is a national issue. To evaluate the soundness of the reactor-pressure vessel and the primary containment vessel over the long period of decommissioning, we should develop suitable inspection technologies. On the other hand, maintaining safe and reliable conditions for aging public infrastructure such as roads and tunnels built during rapid economic growth is also a national issue. Thus, securing worker safety and enabling efficient inspection by workers are common shared issues. Therefore, we should develop remote technologies such as robots that incorporate advanced inspection technologies to overcome a high-dose environment or perform high-elevation works.

High-power fiber lasers are an attractive technology for material processing due to their advantages over mechanical-removal technologies, such as the capability to work over long distances with noncontact processing and ease of coupling with remote control. In this study, we have developed remote-sampling technology using a quasi-continuous wave (QCW) fiber laser. The advantages of the QCW fiber laser with a high peak power include a smaller heat-affected zone (HAZ) and a higher controllability with a smaller and lighter processing head than a continuous-wave laser. The results of melt ejection and the temperature profile during the QCW laser sampling are shown in Fig.1-19. When a laser pulse interacts with concrete, the surface is quickly heated to high temperature via the absorption of laser energy, causing the concrete to be melted and vaporized. A vapor plume was created immediately at the beginning of the laser-sampling process. As the surface temperature reached the vaporization point, part of the molten concrete was further vaporized by the pressure created by the beam upon the molten concrete’s surface. Figs.1-19(d) and (e) show the melt-ejection process, which continued even after the laser. Fig.1-19(f) shows the concrete surface kept at a sufficiently high temperature to control the molten phase until the next laser pulse.

Fig.1-20 shows that increasing the peak power to 6 kW enables us to sample a piece of concrete up to 50 mm with a depth of 10 mm. The total sampling time was 32 s. (b) Circularly shaped sampling with a diameter of 10 mm and depth of 50 mm was demonstrated with a 6-kW fiber laser.

Reference
After the accident at the TEPCO’s Fukushima Daiichi NPS (1F), areas within 1F have high radiation-dose rates, so remotely operated robots need to be deployed there for physical and radiation surveys. It is difficult to remotely maneuver the robots inside reactor buildings due to natural obstacles that include staircases and obstacles created by debris from the accident. Remote robot operation is complicated by frequent difficulties with signal transmission between the operator site and the robots. Therefore, it is necessary to develop robots for specific tasks and to train skilled robot operators. To meet these needs, we are developing a robot-simulator system to support performance evaluation of remotely operated robots and to improve robot-operator skills (Fig.1-21 and Fig.1-22(a)).

As the basis of the robot simulator, we used a Choreonoid that was developed by the National Institute of Advanced Industrial Science and Technology (AIST). Additional functions can be designed and implemented on the Choreonoid as plug-ins. Therefore, we developed required functions and installed a 3D digital model inside the 1F reactor building to realistically simulate remotely operated robot-operational cases in the decommissioning process.

We developed a plug-in function for simulating the submerged behavior of underwater remotely operated vehicles (ROVs). The plug-in calculates the effects of buoyancy, hydrodynamic-lifting force, and fluid resistance upon the ROV’s body in response to motion commands.

A plug-in function for camera-view-disturbance effects was also developed. We designed these functions to add the effects of magnification, distortion, noise, and view-blocking obstacles. Distortion effects occur in the underwater environment due to refraction. Fish-eye distortion is a typical example of image distortion. Noise on the image is simulated to present an unclear-visual-recognition situation to the operators (Fig.1-22(b)).

Another function that we developed simulates the effects of signal- and data-transmission trouble that occurred during past missions of remotely operated robots at 1F. This function is implemented by installing a computer called the communication-traffic controller, between the computer that runs the robot simulator and the computer used to operate the simulated robot. The communication-traffic controller manages the communication states and can vary bandwidth limitations, communication delay, packet loss, and jitter effect both upstream and downstream. The developed simulation functions are implemented in the robot simulator and a 3D digital model is installed inside the 1F reactor building. The user can experience realistic remote operations by the robot (Fig.1-21).

We will continue to develop and implement the required functions to the robot simulator to promote robot development and operator training to contribute to the decommissioning of 1F.

Reference
Detection of Radionuclide Depth in Soil by Aerial Radiation Monitoring

Technology for Estimating the Vertical Distribution of Radionuclides in Soil Based on the γ-ray Spectrum

Seven years have passed since the accident at the TEPCO’s Fukushima Daiichi NPS (1F). It has been reported that radiocesium deposited by the accident was transferred into deeper soil with rainfall, inversion tillage (a type of decontamination work) and disturbance by wild animals. It is necessary to readily detect radiocesium depth in the soil from the ground surface. This method would be helpful for determining the depth of decontamination. We have developed an aerial-radiation-monitoring method using an unmanned helicopter (R-Max G1, YAMAHA Co., Ltd.), which is used to rapidly and easily investigate the spread status of radiocesium on a wide scale.

In the present study, we succeeded in developing a method for acquiring radiocesium soil depth based on the characteristics of the γ-ray spectrum obtained by aerial radiation monitoring. To validate the theory in Fig.1-23, the contribution ratio of scattered γ-rays to direct γ-rays is compared to the vertical distribution of radiocesium in core soil at the same point in the farmland (Fig.1-25). Inversion tillage was performed in the farmland, except in the southern area. The value of radiocesium in soil was expressed as a parameter, effective relaxation mass depth \( \beta_{\text{eff}} \). As \( \beta_{\text{eff}} \) increased, radiocesium distributed more deeply into the soil.

As shown in Fig.1-24, good agreement was observed between the RPC and \( \beta_{\text{eff}} \). The map of estimated \( \beta_{\text{eff}} \) in the entire field in Fig.1-25 was created based on the above-mentioned equation. The result of core soil sampling is shown as a black circle in Fig.1-25 was created based on the above-mentioned equation. The estimated \( \beta_{\text{eff}} \) value calculated by the equation in Fig.1-24 is well fitted to the actual result. This figure implies that the estimated \( \beta_{\text{eff}} \) value is the average vertical distribution of radiocesium in soil in the selected area.

The estimated \( \beta_{\text{eff}} \) value calculated by the equation in Fig.1-24 is well fitted to the actual result. This figure implies that the estimated \( \beta_{\text{eff}} \) value is the average vertical distribution of radiocesium in soil in the selected area.

\[ \beta_{\text{eff}}(\text{g cm}^{-2}) = 0.0281x + 9.5843 \]

\[ r = 0.616, n = 29 \]

\[ \gamma \text{-rays (50–450 keV) to that of direct } \gamma \text{-rays (450–760 keV) for } \gamma \text{-rays obtained by aerial radiation monitoring. We used the LaBr}_3(Ce) \text{ detector that showed good energy resolution for } \text{Cs}. \]

The vertical distributions of radiocesium in the core samples (0–60 cm) were obtained by random soil sampling using a core sampler in farmland. The vertical distribution of radiocesium in soil was expressed as a parameter, effective relaxation mass depth \( \beta_{\text{eff}} \). As \( \beta_{\text{eff}} \) increased, radiocesium distributed more deeply into the soil.

As shown in Fig.1-24, good agreement was observed between the RPC and \( \beta_{\text{eff}} \). The map of estimated \( \beta_{\text{eff}} \) in the entire field in Fig.1-25 was created based on the above-mentioned equation. The result of core soil sampling is shown as a black circle in the result for aerial radiation monitoring over all farmland. The vertical distribution of radiocesium from farmland on a wide scale was investigated by aerial radiation monitoring using an unmanned helicopter. This method would be helpful for performing decontamination in the difficult-to-return zone over a wide scale.

This research was supported by grants from the Project of the NARO Bio-oriented Technology Research Advancement Institution “the special scheme project on regional developing strategy”.

Reference

Highly Accurate Measurement of Radiation-Dose Distributions
— Realization of Highly Accurate Radiological Airborne Monitoring on Rugged Terrain —

Radiological airborne monitoring is one radiation-monitoring tool that can measure air dose rates and radionuclide deposition rapidly over a wide area by measuring radiation at 300 m above the ground (Fig.1-26(a)). In radiological airborne monitoring, not only manned helicopters, but also unmanned helicopters, drones, and other flying objects are used. Measurement results collected by radiological airborne monitoring are employed for a number of purposes including assessment of the contamination situation. Recently, the measurement result has also been used to estimate inhabitants’ exposure dose. Therefore, improvement of data accuracy is regarded as one of the most important issues.

In radiological airborne monitoring employed to date in Japan, the terrain of a target area has been regarded as flat in the process of data analysis. However, due to topographical relief, the air dose rates may potentially be overestimated or underestimated, because the measured counting rates change depending on the terrain effect in spite of having the same contamination level (Fig.1-26(b)). Unlike Europe and the United States, most of Japan’s land area is covered by mountains. Therefore, we evaluate the effects of terrain upon data accuracy and develop a new method to decrease the error.

In the analytical process, an air dose rate (μSv/h), D, can be obtained by dividing a counting rate (cpm), C, by a conversion factor (cpm/(μSv/h)), CD, according to the following equation: D = C/CD

CD changes depending on the terrain. We performed data analysis considering the terrain effect. CD values for arbitrary terrain were obtained based on the calculated results of γ-ray flux at 300-m height. Gamma-ray flux was calculated as shown in Fig.1-27(a). First, a triangle-surface map was created from a digital elevation map. Then, small triangles in this map were regarded as small radiation sources and generated γ-rays. In the end, by summation of the number of γ-rays that reach to the detector position at a height of 300 m, the counting rate was obtained.

For 217 points in Fukushima prefecture, we compared D obtained with the traditional method and D obtained with the new method including the terrain effect to D obtained by ground measurement and evaluated the conversion accuracy. In the evaluation, the frequency distributions of the ratio between the radiological-airborne-monitoring and ground-measurement D values were used. If the two D values were equivalent, the distribution shows as a green bar in Fig.1-27(b). From the frequency distribution, the average was improved from 1.75 to 1.12 and the standard deviation was improved from 0.53 to 0.33 using a new analysis method considering terrain effect. By considering the terrain effect in data analysis, we succeeded in improving conversion accuracy.

We are going to apply this method to the regular operation of radiological airborne monitoring in Japan.

Reference
How is Radiocesium Distributed in a Town? — Investigation of the Radiocesium Distribution in Residential Areas —

The distribution and migration of radiocesium in diverse terrestrial environments has been well-studied; however, there has been little analysis of its behavior in residential areas. Since residential areas comprise diverse components such as paved ground, roofs, walls, and permeable ground, the radiocesium dynamics should be different from other terrestrial environments. To evaluate the dynamics of radiocesium in a residential area, $^{137}$Cs inventories (activity per unit area) were measured on major components (i.e., roofs, rooftops, walls, planar permeable fields, and paved ground) for 11 building lots in the evacuation zone in January 2015. The $^{137}$Cs inventories were converted to relative $^{137}$Cs inventories, which are defined as the relative values of the $^{137}$Cs inventory on each component to the initial $^{137}$Cs inventory on a nearby planar permeable field, enabling comparison of $^{137}$Cs inventories among sites with different initial deposition amounts. The initial $^{137}$Cs inventory on planar permeable fields on March 23, 2011 was estimated by only physical-decay correction, since the $^{137}$Cs inventory on planar permeable fields has been reported to decrease almost as predicted by its physical-decay constant, without any detectable wash-off effects.

The average relative $^{137}$Cs inventories on the components are shown in Fig.1-28. The value on paved ground accounted for about 20% of that on the planar permeable field. Other components, such as roofs, rooftops, and walls also showed values less than 10% of that on the planar permeable field. This study was carried out in the evacuation zone in which decontamination had not been conducted. Therefore, these results indicate that large amounts of radiocesium deposited in the residential area were removed by initial run-off and the following wash-off effects due to rainfall over the four years following the accident, even without decontamination.

The average relative $^{137}$Cs inventories on roofs and rooftops showed large coefficients of variation over 100%. Studies in Europe reported that porous materials such as unglazed tiles have larger absorption capacities for $^{137}$Cs than other materials. Variations in the relative $^{137}$Cs inventories depending on roof and rooftop materials were also observed in this study (Fig.1-29); smooth-surface materials such as glazed tile, metallic slate, and resin showed low relative $^{137}$Cs inventories, while unglazed mortar showed large values. Since the roof and rooftop are major components in residential areas, these results indicate that the migration of radiocesium deposited in the area largely depended upon the materials of roofs and rooftops.

The results in this study are expected to contribute to decontamination planning and exposure-dose simulations in residential areas.

Reference
Prediction of Radiocesium Behavior in Upstream Catchment
— Sediment and Radiocesium-Transport Simulation during Approximately Five Years Following the 1F Accident —

Due to the accident at the TEPCO’s Fukushima Daiichi NPS, significant amounts of radiocesium (RCs) remain in the top surface soil because RCs have the characteristic of adsorbing strongly to soil particles including clay minerals. Surface-soil erosion by heavy rainfalls such as typhoons is important for predicting RCs behavior in the watershed; however, we need to describe both surface and subsurface water flow to simulate this behavior with and without precipitation periods. In this study, we applied the watershed model GETFLOWS, which can describe sediment and RCs transport to an Oginosawa river catchment (Fig.1-30) in Fukushima.

Fig.1-31 shows the amount of sediment erosion/deposition and the fraction of the residual $^{137}$Cs inventory from May 2011 to December 2015. Sediment erosion was found to have occurred significantly and the $^{137}$Cs inventory decreased in the vicinity of the river channel and forest gullies. On the other hand, sediment erosion did not occur much in the forested area far from the rivers, with the result that the $^{137}$Cs inventory remained. Therefore, the physical decay of $^{137}$Cs is the most important factor in forested areas far from rivers. Calculation of $^{137}$Cs sources supplied to the river at this catchment showed that contributions from the vicinity of the river channel were an order of magnitude higher than that from the forested area far from rivers. This is because surface-water flow during heavy-rainfall events can cause high sediment-erosion rates.

In the future, $^{137}$Cs discharge to the river is expected to decrease due to the decreasing $^{137}$Cs inventory in the vicinity of the river and the migration of $^{137}$Cs from top-surface soil to deeper soil. We need to watch the downstream influence of $^{137}$Cs discharge by decontamination or starting cultivation in the future.

Verification and improvement of this study, especially dissolved RCs which is high bioavailability, can be used to understand mechanisms of dissolved RCs behaviors in environment in the future.

Reference
Research on the Distribution of $^{137}$Cs in Seabed Sediments
— Relationship between the $^{137}$Cs Inventory and Seafloor Topography —

The accident at the TEPCO’s Fukushima Daiichi NPS that occurred following the Great East Japan Earthquake and the resulting tsunami in March 2011 resulted in extensive release of radioactive cesium into the Pacific Ocean. Understanding the features of $^{137}$Cs transport from contaminated mountain forests to coastal sinks is key to the revitalization of marine industries. The $^{137}$Cs distribution in shallow seas, which were major settlement areas for particle-sorbed $^{137}$Cs, is very important when studying $^{137}$Cs transport from contaminated mountain forests to coastal sinks. Therefore, we examine the heterogeneity of the $^{137}$Cs distribution in seabed sediments at sea depths <30 m. A bathymetric survey and sonic prospecting were conducted to map the occurrences of seabed sediments, and core sampling of such sediments was performed to describe the vertical profile of the $^{137}$Cs distribution therein.

The bathymetric map shown in Fig.1-32 indicates that terrace-like seafloors consisting of gently descending slopes opposite to the coastal sides (dip side; around point A, Fig.1-32) and steeper ascending slopes on their coastal sides (up-dip side) extend in the investigation area. In particular, point B is located in a semicircular depression surrounded by steep sides. According to the results of sonic prospecting, the surface ratio of the seabed sediments is limited to a quite narrow zone of semicircular depression (point B).

The $^{137}$Cs-inventory values collected at point B are greater than those at point A, and the grain sizes (D50 values) at point B are homogeneously silt (Fig.1-33). The current velocities of the bottom layer at point B, which were observed using current profilers towed by ships, were lower than those at point A. These results suggest that the steep slope around the semicircular depression area appears to play a major role as a topographic barrier in reducing the current velocity, prompting the settlement of fine-grained particles. In other words, the features of the seafloor topography are determined to be significant factors controlling the horizontal and vertical distributions of $^{137}$Cs in the seabed sediments. Areas with similar seafloor topographies of the semicircular depression were not identified in the investigation area, suggesting that distributions with remarkably large $^{137}$Cs inventories (more than several thousands of kBq m$^{-2}$) are strictly limited.

Our research also revealed the continuous $^{137}$Cs distribution at depths greater than 81 cm (Fig.1-33(b)). Vertical $^{137}$Cs profiles in seabed sediments obtained in previous studies have been insufficient to evaluate $^{137}$Cs concentrations because information about the distribution of $^{137}$Cs at the lowermost depths of these sediments is lacking, owing to short core-sampler lengths (almost less than 20 cm).

Therefore, these results indicate that estimation of the $^{137}$Cs inventory in shallow seas should be performed carefully, considering the extent of the $^{137}$Cs distribution along the vertical depth and the surface ratio of the seabed sediments.

Reference
Prediction of the Dispersion of Radionuclides Released into the Ocean — Dispersion Simulation of Radionuclides from Coastal Regions to the Open Sea —

Cesium-137 ($^{137}$Cs) was released to the ocean by the accident at the TEPCO’s Fukushima Daiichi NPS (1F). Many observational and simulation studies have been carried out to clarify the oceanic dispersion. Numerical simulation is a powerful methodology for understanding the spatiotemporal characteristics of oceanic dispersion of accident-derived radionuclides. Unfortunately, the quality of oceanic-dispersion simulations is degraded by uncertainties in the source term and simulated ocean currents. In this study, oceanic-dispersion simulations were carried out by an oceanic-dispersion model using output data from five oceanic general-circulation models. By comparing these simulation results, we objectively analyzed the effect of different oceanic data upon the $^{137}$Cs-dispersion simulation. Then, we analyzed the $^{137}$Cs dispersion from the coastal region of Fukushima to the North Pacific Ocean.

In this study, we applied the SEA-GEARN oceanic-dispersion model developed at JAEA. Input oceanic data were calculated by oceanic general-circulation models of the Japan Marine Science Foundation (JMSF), the Japan Agency for Marine-Earth Science and Technology (JAMSTEC), the Meteorological Research Institute (MRI) of the Japan Meteorological Agency (JMA), and the National Oceanic and Atmospheric Administration (NOAA). The data-assimilation method was applied to these oceanic general-circulation models to assimilate observed data into numerical simulations. The direct-release rate of $^{137}$Cs into the ocean was estimated by means of the observed sea surface $^{137}$Cs concentrations near the northern and southern discharge channels of 1F. To provide the $^{137}$Cs-deposition amounts at the sea surface, we conducted an atmospheric-dispersion simulation using the latest-estimated $^{137}$Cs release rate into the atmosphere.

Compared to the simulation with a lower horizontal resolution, the higher-resolution simulation reproduced well the $^{137}$Cs concentration observed in the coastline and offshore of Fukushima prefecture. The $^{137}$Cs was suggested to have been spread along the coast in the north–south direction during the first few months after the 1F accident. The simulations for the western part of the North Pacific and the whole North Pacific reproduced the main ocean current favorably using the data-assimilation method, despite the relatively low resolution. This suggests that the Kuroshio Extension plays a large role in the transport process of $^{137}$Cs from the coast to the outer ocean (Fig.1-34).

Analysis of the depth distribution of $^{137}$Cs by the dispersion simulation showed that most of that transferred to the ocean by direct release or deposition from the atmosphere existed in the surface layer (0–211 m from the sea surface) shortly after the accident. However, as time passed, it was transported from the surface layer to deeper layers (Fig.1-35). The $^{137}$Cs amounts in the surface, intermediate (211–510 m), deep (510–1050 m), and bottom layers (deeper than 1050 m) one year after the 1F accident were 71, 19, 4, and 0.8% of the total release amount, respectively.

This study suggests that the accident-derived $^{137}$Cs was dispersed widely into the North Pacific and gradually dispersed from the surface to the deeper layers by simulations, regardless of the different ocean-current data. A future task is to carry out oceanic-dispersion simulations over the whole North Pacific for several decades to quantify the time-series change of $^{137}$Cs abundance.

Reference
Determining the Conditions for Effective Forest Decontamination
— Simulation of the Effect of Reducing Air-Dose Rate by Decontamination —

In the accident at the TEPCO’s Fukushima Daiichi NPS, forests were polluted by radioactive-cesium (Cs) release. Radioactive Cs remains in forest soil. The decontamination of such Cs is expected to reduce the air-dose rate. However, if decontamination is applied to the forest as a whole, a huge amount of soil will have to be removed. Therefore, there will be a tremendous burden on its management and expenses. In addition, such decontamination increases the possibility that forest soil will leak out, damaging the multifaceted functions of forests such as water-source protection and disaster prevention. Thus, as a method of forest decontamination, the Forestry Agency has recommended gradually removing the surface layer of fallen leaves and forest soil.

In this research, the A0 and A1 layers were assumed to be radiation sources. The A0 layer is the surface of the forest soil including sedimentary organic materials such as fallen leaves. The A1 layer contains the soil. The air-dose rate in a residential area was analyzed using the three-dimensional transport calculation code MCNP on the condition that only the A0 layer was decontaminated. The shapes of radiation sources were given as one or three forest slopes. Fig.1-36 shows the shape of the radiation source for a single forest slope. The three forest slopes were assumed for the case of a house surrounded by three forests. For each radiation-source shape, cases where an inclination angle of the forest soil and the amount of radioactive Cs in the soil were changed—and also a case where the contamination of the forest soil was planarly non-homogeneous—were analyzed. For all of those cases, we analyzed the extent to which the air-dose rate at that point in the residential area was reduced when the range of the A0 layer was removed.

It was found that, even if the A0 layer was removed, the air-dose rate in the residential area (especially the second floor of the residence) was hard to lower compared with the forest edge, regardless of the number of slopes or the inclination angle.

Also, if there is more radioactive Cs in the A0 layer and the contamination is planarly homogeneous, it is effective to remove the A0 layer up to 20 m from the forest edge. But, for instance, as shown in Fig.1-37, in the case where the concentration of radioactive Cs included in the forest soil at over 20 m distant from the forest edge was tripled compared with its concentration included in the forest soil from the forest edge up to 20 m, the decontamination of the A0 layer from the forest edge up to 40 m was revealed to be needed to effectively reduce the air-dose rate in the residential area.

This simulation has made it possible to effectively determine the forest-decontamination method according to the distribution of contamination.

Reference
It is known that radioactive cesium ($^{137}$Cs) is present at the soil surface and strongly adsorbed to clay minerals following the accident at the TEPCO’s Fukushima Daiichi NPS. A basic understanding of Cs adsorption and removal is required to develop a decontamination method with a low environmental load. However, it is not easy to study the adsorption behavior of Cs because the clay contains an extremely small amount of Cs with an irregular size of less than several microns. Synchrotron-radiation-photoemission electron microscopy (SR-PEEM) is an analytical technique that can visualize the chemical state of individual elements with nanoscale spatial resolutions (Fig.1-38). Despite its superiority to other methods, a significant problem called charging arises for an insulator sample, e.g., clay. In this study, we developed an effective way to avoid this problem with a thin-carbon-film deposition to the particle surface. Here, the results of nanoscale chemical analysis of weathered-biotite clay particles containing Cs are shown.

In Fig.1-39(a), a clear Cs-distribution image was obtained for a clay mineral about several microns in size. We found that Cs was not localized at the particle surface. In addition to the information on particle identification and element distribution, the chemical state of Cs at a specific position indicated by a red full-circle could be visualized in Fig.1-39(b). We found that the chemical state of Cs in clay is similar to CsNO$_3$ and that Fe with a trivalent-oxidation state (Fe$^{3+}$) is present. The presence and distribution of Fe$^{3+}$ were also revealed. The real-space information on the chemical states would deepen our understanding of Cs-adsorption mechanism.

Nanoscale visualization of chemical states can be applied to nuclear-research fields such as analysis of decontamination soil and mock samples of fuel debris contributing to decommission of the reactor. Our new approach to analyzing insulating materials using SR-PEEM is expected to be used for the development of many advanced materials in nanotechnology, environmental, and new-energy studies.

Reference
Radioactive cesium was released into the environment due to the accident at the TEPCO’s Fukushima Daiichi NPS in March 2011. Part of it was strongly adsorbed onto the topsoil and became the main cause of the evacuation of residents. Decontamination was carried out on a large scale, and the radioactivity and air-dose rate were successfully reduced. However, a huge amount of waste soil was produced by the decontamination due to the use of a method that removes contaminated topsoil. Therefore, processing and management of the waste soil are emerging as problems.

For the development of volume reduction technology and risk assessment for long-term management, it will be helpful to know how cesium is adsorbed onto the soil. It is known that radioactive cesium strongly adsorbs to clay minerals in soil. However, its adsorption mechanism is unknown. One reason for which it is difficult to investigate this mechanism is that adsorption occurs over a small area that cannot be observed, even with the latest experimental equipment. As shown in Fig.1-40, clay minerals have several surface structures, each of which has been thought to exhibit different adsorption strengths. However, it is very difficult to observe the details.

Computer simulation is a powerful tool for research on such a small area. We collaborated with research institutes in the United States and the National Institutes for Quantum and Radiological Science and Technology to model the atomic-scale surface structures shown in Fig.1-40. The adsorption energies of cesium on the surfaces were evaluated using the supercomputer by changing the distance between cesium and the model surfaces at the atomic scale (Table 1-2). Based on the results of this systematic numerical simulation, it was shown that a wedge-shaped structure called “frayed edge” with a size of about a nanometer gouged by weathering adsorbs cesium most strongly (Table 1-2). A hypothesis concerning the strongest adsorption by the frayed edges was proposed about 50 years ago, but experimental proof was difficult to obtain due to the difficulty of directly observing atomic-scale structures. We succeeded in validating this hypothesis by investigating the adsorption on each surface through numerical simulations.

Based on the above results and the findings obtained from previous experimental studies, we also evaluated promising volume reduction technologies and risks during long-term management. In the future, based on the adsorption mechanism revealed in this research, we will continue research and development to contribute to reducing the volume of waste soil.

Reference
Even now, radionuclides dispersed in the Fukushima environment by the accident at the TEPCO’s Fukushima Daiichi NPS largely remain in forests. Over time, these radionuclides can pass out to the ocean and living environments under the action of wind and water flows. People are concerned with the potential risks that this poses to health and daily life.

In order to understand this issue in its entirety, evaluate its effects, and decide on countermeasures, JAEA and other research institutes have been conducting environmental monitoring since the accident. However, the findings of their studies are dispersed between various web pages, reports, research papers, pamphlets, and so forth. Therefore, it takes a great deal of time and effort for residents and local municipalities to gather all necessary information on the subject. We have gathered scientific findings on the migration of radioactive cesium and the recovery of the environment in Fukushima from various institutes. We have combined these with our own findings, and created a system to allow users to search easily for all relevant information. We are now enhancing this system so that evaluation of the environmental dynamics of radioactive cesium is possible. The system is called a comprehensive evaluation system (Fig.1-41).

The research results are arranged in a common format in the environmental-monitoring database. Visualization of the results is possible in this system. The system allows users to find various information on environmental-radioactive cesium dynamics with the description level matched to their needs (the knowledge base on environmental recovery), without referring to other media. For users who wish to know more details, the simulation unit gives access to simulation results on the transport of radioactivity in the environment, with land-use, topography, soil type, vegetation type, and initial radioactivity-deposition amounts taken into account. In addition, the various results have been summarized in an easy-to-understand Q&A format in the knowledge base on environmental recovery. Thus, the system provides information according to the needs of individual users.

It is recognized that understanding the re-circulation of radioactive cesium in forest environments and the sources of dissolved cesium discharged into rivers are important issues. We are therefore advancing research in these areas in response to concerns amongst local inhabitants. We will continue to update the system in accordance with the release of new research findings.

References