Time series of atmospheric radiocesium just after the accident at two SPM monitoring sites near the Fukushima Daiichi Nuclear Power Plant

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The first retrieval of hourly atmospheric radiocesium concentrations during March 12-23, 2011 just after the Fukushima Daiichi Nuclear Power Plant (FD1NPP) accident was already published by measuring radionuclides in Suspended Particulate Matter (SPM) on the filter-tapes installed in SPM monitors with beta-ray attenuation method at 40 sites operated by local governments in the air pollution monitoring network of eastern Japan (Tsuruta et al., Sci. Rep., 2014). Since then, hourly atmospheric radionuclides in SPM have been measured at around 100 SPM monitoring stations, and the dataset has been also published on a website (Oura et al., JNRS., 2015). After that, the used SPM filter-tapes at two SPM monitoring stations were offerred, which were operated even after the Great East Japan Earthquake and tsunami on March 11, 2011. One (site F) was located in the Futaba town, about 4 km northwest of the FD1NPP, and the other (site N) was in the Naraha town, 16 km south of the FD1NPP. At first, we checked if these SPM monitors were normally operated even after the Great Earthquake and tsunami, or not. Considering from various points of view, we judged that the SPMs at these sites were properly collected on the filter-tapes as usual. Then, Cs-134 and Cs-137 in the SPMs were measured by Ge detectors. The radiation dose rates at the monitoring stations located outside the FD1NPP and FD2NPP by the Fukushima prefecture, and inside by TEPCO, and the meteorological data by the Japan Meteorological Agency were also used for further analysis. In this paper, we will briefly introduce new findings from the hourly Cs-137 concentrations during March 12-23, 2011, which had not been recognized by the previous analyses. At site F, six peaks of high Cs-137 concentrations (Cs-137>100 Bq m^{-3}) were found on March 12-13, 15-16, and 18-20, 2011. Most of these high radionuclides were transported as plumes/polluted air masses to the northwest or north, which were already recognized in the previous paper. In addition, at site N, six peaks of high Cs-137 concentrations were also found on March 15-16, 20-21, 2011. Most of them were also transported to southern Fukushima and the Tokyo Metropolitan Area. Some of them at sites F and N, however, were identified as new plumes/polluted air masses. Furthermore, the peak times of radiation dose rates measured at the monitoring stations located near two SPM sites, were well correlated with those of Cs-137 concentrations. These findings greatly contribute to understand how radionuclides were released and transported from the FD1NPP just after the accident. We greatly thank to the Fukushima prefecture for offering the used filter-tapes at two SPM monitoring stations.

Keywords: Fukushima Daiichi Nuclear Power Plant, Radiocesium, Suspended particulate matter, Air pollution monitoring network, Time series

Investigation of chemical and physical properties of radioactive aerosols sampled from SPM tape filters using multiple synchrotron radiation X-ray analyses

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Large amounts of radioactive materials were released into the environment after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident caused by the Tohoku Earthquake and Tsunami on March 11th 2011. As one form of radioactive material emitted into the atmosphere, Cs-bearing radioactive spherical microparticle, known as Cs ball, has gathered attention in recent years. The Cs-ball was first found in aerosols collected at Tsukuba during March 14th and 15th 2011[1)] and identified as a water-insoluble glass material containing various heavy metals derived from fission products (FPs) of the nuclear fuel[2)]. While recent investigations suggest the widespread distribution of similar particulate radioactive materials in the environment such as soils sampled from the Fukushima prefecture[3)], the distribution of such radioactive particles in the Kanto region including the capital Tokyo just after the accident is still unclear. The present study thus focuses on suspended particulate matter (SPM) filter samples collected by operational air pollution monitoring stations[4)]. Radioactive aerosols were sampled from the filters collected at several stations during March 15th 2011 to investigate their chemical and physical properties in the nondestructive manner. The synchrotron radiation (SR)-X-ray microbeam of SPring-8 was used as an analytical probe for chemical composition analysis by X-ray fluorescence analysis (SR-μ-XRF), chemical state analysis by X-ray absorption near edge structure analysis (SR-μ-XANES) and crystal structure analysis by X-ray diffraction (SR-μ-XRD).

All particles sampled from the SPM filters in the present study have similar physical properties: a spherical shape and ~1 μm of diameter. Based on $^{134}Cs/^{137}Cs$ ratios (~1.0) of individual particles, it is expected these particles were emitted from the reactor No.2 or 3 of the FDNPP. These properties are similar to those of Cs-balls reported in previous studies[1,2)] except for their sizes (Cs-ball: ~2 μm in diameter). Various heavy elements (Rb, Mo, Sn, Sb, Te, Cs, Ba etc.) possibly derived from FPs were commonly detected from all these particles by the SR-μ-XRF. In addition, it was revealed that some particles contain trace amount of U. While we examined chemical states of four metal elements (Fe, Zn, Mo and Sn) contained in these particles by the SR-μ-XANES, all analytical results indicated that these elements exist as a glass state with high oxidation states in these particle. Results of the SR-μ-XRD also verified that these particles are amorphous materials. Because of these obvious similarities of chemical and physical properties between radioactive particles sampled from the SPM filters collected at the Kanto region and the Cs-balls, we thus concluded that particulate radioactive materials equated with the Cs-ball passed through the Kanto region on March 15th 2011. At the same time, our analytical results demonstrated the U possibly originated from the fuel certainly arrived to the Kanto region just after the accident.

Acknowledgments: We thank to all local governments for allowing us to investigate the SPM filter samples.

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Keywords: Fukushima Daiichi Nuclear Power Plant accident, radioactive material, Synchrotron radiation X-ray analysis, aerosol

Unsteady source term estimation of the Fukushima dai-ichi release using contributed radiological measurements

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A new methodology is presented for reconstructing the unsteady release rate of an atmospheric nuclear release using a Lagrangian atmospheric transport and dispersion (T&D) model, contributed concentration measurements, and stochastic search techniques. The methodology was applied to the reconstruction of the 2011 Fukushima nuclear release using an atmospheric T&D model and contributed radiation data from the Safecast citizen science project.
First, the Safecast contributed measurements were compared to official aerial surveys completed by the Department of Energy (DOE) and the National Nuclear Security Administration (NNSA) in 2011. It is shown that the Safecast data provides a reliable spatial estimation of radiation concentration when compared to official data.
Then a T&D model is run to simulate the nuclear release using high resolution terrain and meteorological data. The model simulates multiple sequential releases with a constant rate. The model values are decayed to the dates for which Safecast data are present, and an error is then computed between the observed and simulated concentrations. A statistical process is performed to find scalar multipliers that minimize the error between observed and simulated values.
The outcome of the optimization is the non-steady release rate concentration for the Fukushima nuclear release over land. This is because the release rate is estimated for primarily land-based data so the estimation does not include consideration for the plume of radiation that spread over the ocean. Environmental concerns and human impacts can be better addressed with long-term monitoring that confirms the prediction of models representing dynamic behavior of radionuclides dispersed in the environment.

Keywords: dispersion, radiation, fukushima, spatio-temporal analysis

Mechanism of resuspension of radiocesium in summer and autumn

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.Radionuclides emitted in the Fukushima dai-ichi nuclear power plant (FNDPP) accident in March 2011 have been deposited on the soil, ocean and vegetation. Re-suspension of radioactive cesium from the soil and vegetation to the atmosphere may be one of significant path in the diffusion of radionuclides after the accident.
We have measured the concentration of atmospheric Cs-134/137 radioactivity at Namie, Fukushima, where deposition amount of Cs-134/137 is relatively high. Atmospheric suspended particles have been collected on a sheet of quartz fiber filter with high-volume air samplers mounted at a playground site and forest site, and gamma-ray emission from them were measured with Ge detector to obtain the atmospheric activity concentration of Cs-134/137. A small part of each filter was used to measure chemical composition and microscope particle observation.
The atmospheric Cs radioactivity concentration increased from late May to September, summer and autumn. It was higher in the forest than that at the playground in these season. The measured concentration of atmospheric Cs-134/137 was positively correlated with amount of carbonatious particles in these seasons. Bioaerosol sampling and analyses showed that major coarse particles in these seasons were spores of fungi and stain. We counted the spores collected on the sample filters to evaluate their number density, and found the number density was positively correlated with the atmospheric Cs radioactivity concentration. We collected fungi at Namie to sample its spores. About half of Cs-137 in the spores was removed by pure water, being consistent with similar experiment for the atmospheric particle samples. These results indicated that spore emission from fungi significantly contributes to the resuspension of radioactive Cs to the atmosphere in summer and autumn. However, Cs-137 radioactivity in the collected spores may be too small in comparison with atmospheric Cs activity concentration by considering atmospheric spore particle concentration. Other process may contribute to the re-suspension.

Keywords: radioactive cesium, atmospheric resuspension, bioaerosol, fungi spore

Model inter-comparison of atmospheric Cs-137 from the Fukushima Daiichi Nuclear Power Plant accident

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After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011, atmospheric simulation models played important roles in understanding the atmospheric behaviors of radionuclides. For the evaluation of the validity and variability of model results, model inter-comparison provides valuable and useful information. In this study, we compared results of seven atmospheric transport models to simulate atmospheric ^{137}Cs released from the FDNPP. All of model results used in this analysis were submitted for the model inter-comparison project of Science Council of Japan (2014). Model reproducibility was assessed from comparison with the observed hourly atmospheric concentrations of ^{137}Cs at 90 sites in Tohoku and Kanto regions (Tsuruta et al., 2014).

Tsuruta et al. (2014) identified 9 plumes (P1 –P9) which passed over Tohoku and/or Kanto regions. P1, P5 and P6 passed through the northern coastal area of Fukushima prefecture to the Pacific Ocean from FDAPP in the night of March 13, morning of 18th and afternoon of 19th, respectively. P2, P4, P7 and P9 reached Kanto region in the morning of 15th, morning of 16th, afternoon of 20th and morning of 21st, respectively. P3 and P8 widely spread over Fukushima prefecture in the afternoon of 15th and night of 20th, respectively.

On average, performance of the models was the best for P2 with FA2 (fraction of simulated data that reproduced the observations within a factor of 2) of 10% ~ 40%. Model performance for P1, P3 and P8 was moderate with FA2 of 0%~10%. The models generally reproduced the observed ^{137}Cs concentrations in plumes which widely spread inland of Tohoku or Kanto regions (P2, P3, and P8). By contrast, the models largely underestimated the observed ^{137}Cs concentrations for P4, P5, and P6, which passed coastal areas of Japan.

Ensemble average of seven models showed reasonable performance for most of plumes, and no individual models reproduced better that the ensemble average for all the plumes. These results suggest that ensemble average is effective for reliable and stable simulation of radioactive plumes.

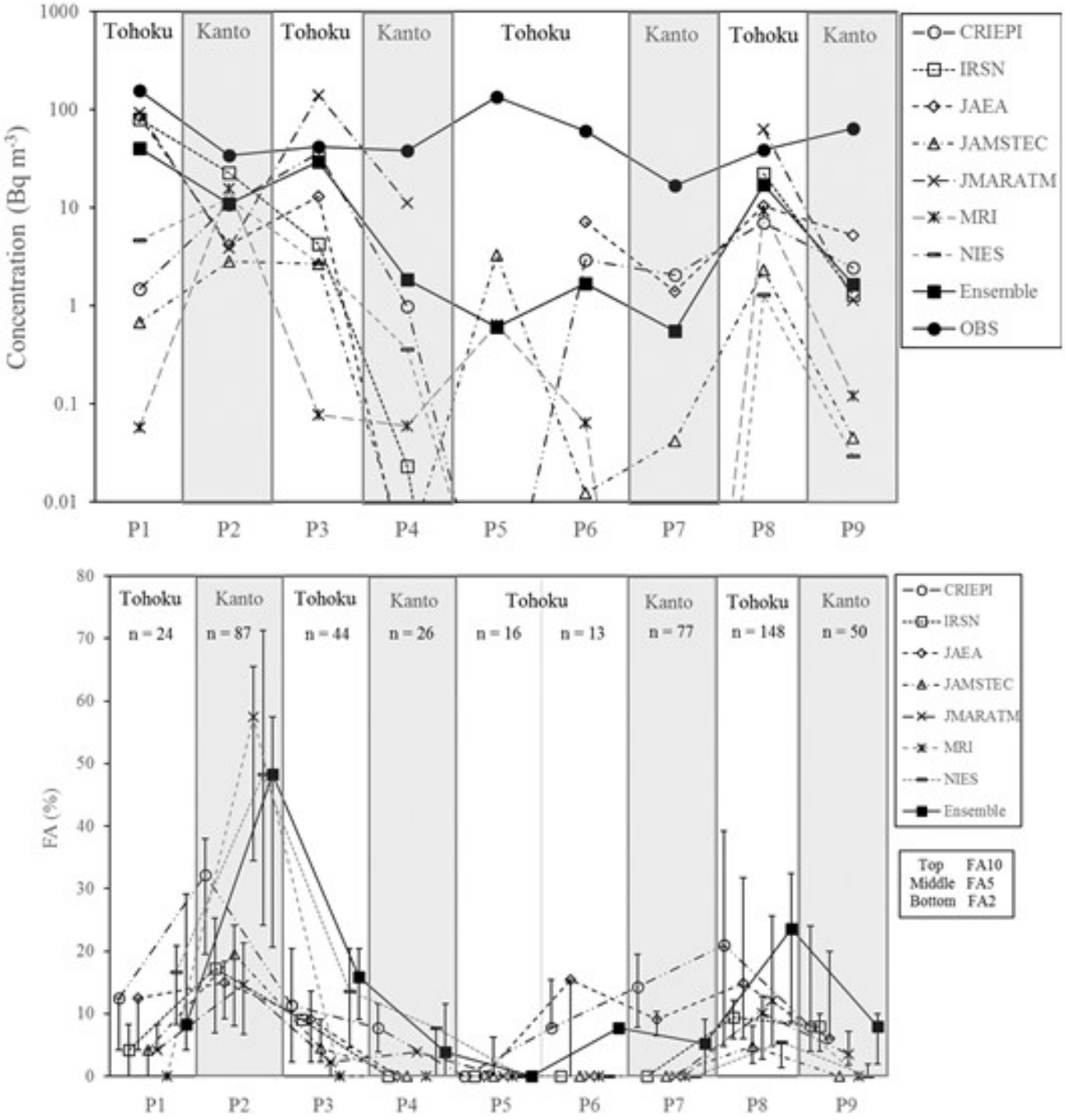
References

Science Council of Japan, September 2, 2014.

Tsuruta et al., Scientific Reports, 4:6717, 2014.

Fig. (top) Observed and simulated average concentrations of ^{137}Cs in nine radioactive plumes ([Observed-^{137}Cs] 10 Bq m^{-3}). (bottom) Fraction of simulated data that reproduced the observations within a factor of 2, 5, and 10 (FA2, FA5, FA10, respectively).

Keywords: Model inter-comparison, Cs-137, Fukushima Daiichi Nuclear Power Plant accident



Development of meta-database system for radiation monitoring related to the Fukushima Dai-ichi reactor accident.

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Since March 2011, there have been various efforts made by the government, local communities, private companies, researchers and local residents in order to measure radiation exposed to the environment from the Fukushima Dai-ichi reactor. A part of the measured data has been compiled and open for the public. But most of the data have not been yet compiled, and even their locations are not well known to others. From this point of view, we have developed meta-database system which compiles all the meta-data information for the existing radiation monitoring data. With this system one can find data location sorted by meta-data keys such as observables, location and date of measurement and so on. We will report status of this meta-database system and discuss future prospects.

Keywords: meta-database, Fukushima Dai-ichi reactor accident, Radiation monitoring data

Investigation of soil radionuclude near the Fukushima Dai-ichi Nuclear Power Plant 5-year after the accident by JpGU-JNRS team

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[Introduction] A large-scale soil sampling project for radionuclides from the Fukushima Dai-ichi Nuclear Power Plant accident was conducted in June 2011 by a research group including the Japan Geoscience Union (JpGU) and the Japan Society of Nuclear and Radiochemical Sciences (JNRS). New research project following the 2011 project was also implemented by JpGU-JNRS team with the objective to know the transition process of radioactive cesium in soil and the current contamination in 2016 when five years passed since the accident.
In this report, the outline of the project will be presented. We will also present some preliminary results of the inventory of 134Cs and 137Cs in the soil, comparing 2011 and 2016. More detailed results wll be presented in another report.

[Sampling and Measurement] Between June and September 2016, 176 researcher of total had worked to measure the air dose rate and collect soil samples at 105 locations near the Fukushima Dai-ichi nuclear power plant, mainly on the difficult-to-return zone, for 9 days in total. Surface soil was sampled at 5 points per one location as well as air dose rate at 5 cm and 1 m from the ground surface. The sample from depths of 5 cm was divided into two parts of 0 - 2.5 cm depth and 2.5 - 5 cm depth, filled in a U-8 container after drying. Radiocesium was quantified using gamma ray spectrometry with Ge detectors. To understand the transion of radiocesium in soil, 30-cm depth soil core samples were obtained at about 1/4 of all locations.

Keywords: Fukushima Dai-ichi Nuclear Power Plant, Radiocesium, soil deposition density

Radioactivity in soil from near the Fukushima Dai-ichi Nuclear Power Plant at five years after the accident

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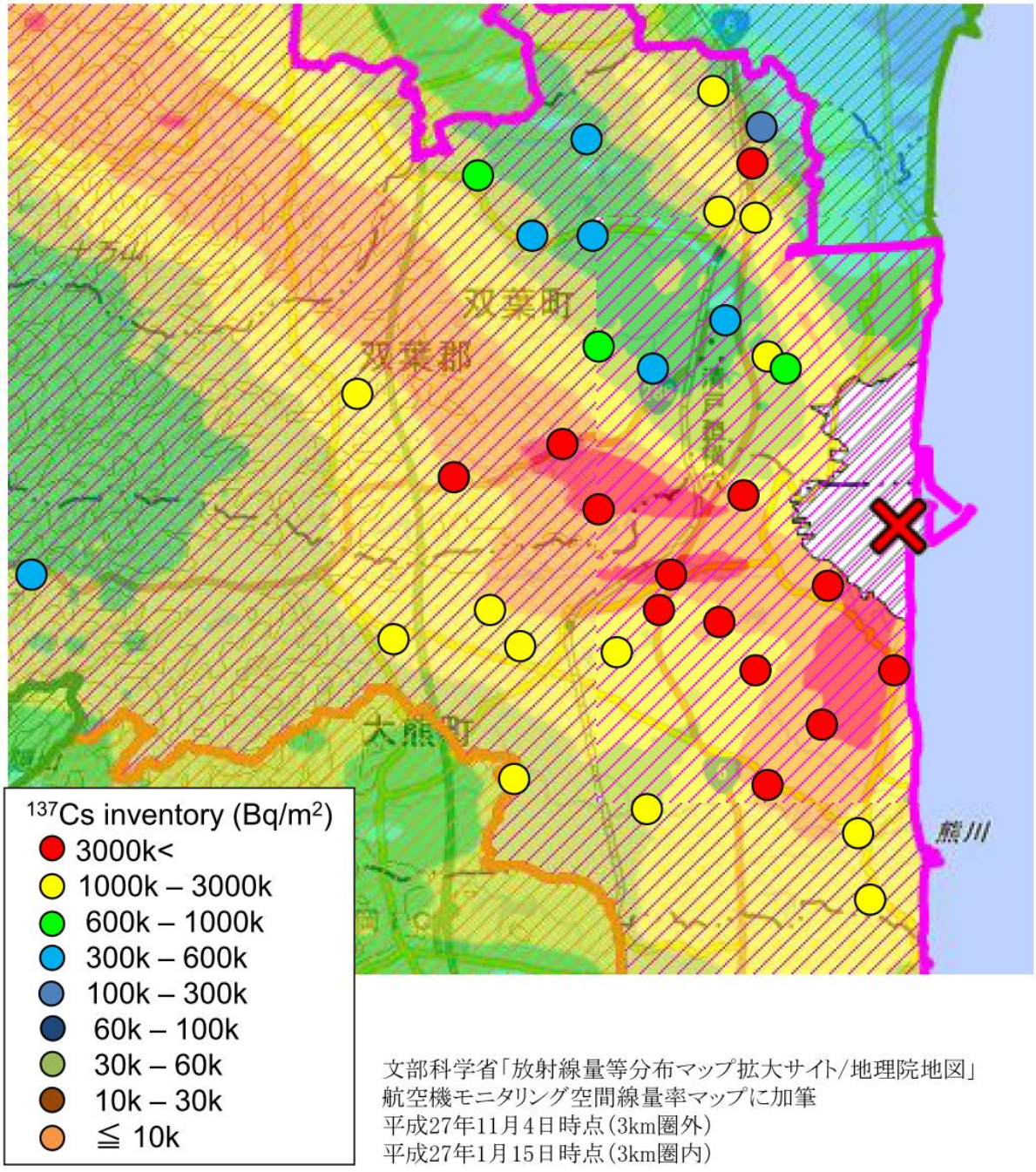
[Introduction] A large-scale soil sampling project for radionuclides from the Fukushima Daiichi Nuclear Power Plant accident was conducted in June 2011 by a research group on the Japan Geoscience Union and the Japan Society of Nuclear and Radiochemical Sciences. New research project followed by was implemented with the objective to know the transition process of radioactivity and the current contamination in 2016 when five years passed since the accident.
Between June and September 2016, 176 researcher of total were work to measure the air dose rate and collect soil samples at 105 locations near the Fukushima Dai-ichi nuclear power plant, mainly on the difficult-to-return zone, for 9 days in total. The outline of the project will be reported in another presentation. In this report, we will present the results of the inventory of ^{134}Cs and ^{137}Cs in the soil, the degree of destribution, the ratio of ^{134}Cs/^{137}Cs concentration, and the correlation with the air dose rate.
[Sample and Measurement] Surface soil of 5 samples per one location was collected from 36 public facilities in the difficult-to-return area of Futaba-machi or Okuma-machi in Fukushima Prefecture in June and July 2016. The sample from depths of 5 cm was divided into two parts of 0 - 2.5 cm depth and 2.5 - 5 cm depth, filled in a U-8 container after drying. Radiocesium was quantified using gamma ray spectrometry with Ge detectors.
[Results and Discussion] The results are shown in Fig.1 The maximum ^{137}Cs concentration was 68400 kBq/m^2 in inventory and 1180 kBq/kg(dry) in specific activity. The inventory clearly correlated with the air dose rate. There was large difference in soil distribution degree (the ratio of radioactivity of 2.5 - 5.0 cm and 0 - 2.5 cm in depth) for each sample, and the values were almost less than 1. Moreover, it was found that there are points where the concentration ratio of ^{134}Cs /^{137}Cs clearly has a low value of 0.87 - 0.93 in the north-north-west direction from the nuclear power plant.

Keywords: Fukushima Dai-ichi Nuclear Power Plant, radiocesium, Cs-134, Cs-137, soil deposition density



文部科学省「放射線量等分布マップ拡大サイト/地理院地図」
航空機モニタリング空間線量率マップに加筆
平成27年11月4日時点(3km圏外)
平成27年1月15日時点(3km圏内)

図1. 2016年の土壌調査による^{137}Csインベントリ
(2016年7月1日時点) 36地点(双葉町18地点、大熊町18地点)

Numerical study on sorption kinetics affecting vertical profile of radiocesium in soil and air dose rates

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Despite its affinity for binding to soil, radiocesium gradually migrates deeper into the ground over time. This results in a faster rate of reduction of air dose rates than would be expected by radioactive decay. Field measurements of the activity profile of radiocesium with depth in soil generally show exponential-shapes, which are often followed by a long tail at large depths. The vertical migration of radiocesium in soil has been successfully reproduced with the modified Diffusion-Sorption-Fixation (mDSF) model, which is based on an advection-dispersion equation coupled with kinetic models of reversible/irreversible sorption. Using radiation transport methods, this study calculated the trajectory of air dose rates over time given the evolution in the depth profile predicted by the model. The results indicate a faster reduction in dose rates than the rate of radioactive decay in the first ten years following fallout, as there is a gradual migration of radiocesium in soil over this period. The rate of reduction of dose rates over the following years is then set by the rate of radioactive decay alone as the majority of the radiocesium has become fixed to the soil matrix.

Keywords: Fukushima NPP accident, radiocesium vertical distribution, exponential-shape distribution, sorption kinetics

Temporal changes of radiocesium transfer and ambient dose rate in forest of Fukushima Prefecture

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We investigated the transfer of canopy-intercepted radiocesium to the forest floor during 6 years following the Fukushima Daiichi Nuclear Power Plant accident. The cesium-137 (Cs-137) contents in throughfall, stemflow, and litterfall were monitored in two coniferous stands (plantation of Japanese cedar) and a mixed deciduous broad-leaved forest stand (Japanese oak with red pine). Temporal changes of radiocesium deposition flux onto forest floor have been determined based on the analysis of radiocesium concentration in collected samples. We also measured the ambient dose rate (ADR) at different heights in the forest using a survey meter and a portable Ge gamma-ray detector. The ambient dose rate in forest exhibited height dependency and its vertical distribution varied with forest type and stand age. The ambient dose rate showed an exponential decrease with time for all the forest sites, however the decreasing trend differed depending on the height of dose measurement and forest type. The ambient dose rate at the canopy (approx. 10 m-height) decreased faster than that expected from physical decay of the two radiocesium isotopes, whereas those at the forest floor varied between the three forest stands. Our monitoring results suggested that the ambient dose rate in forest environment varied both spatially and temporally reflecting the transfer of radiocesium from canopy to forest floor. In addition to that, the measured ambient dose rate within forest showed different decreasing trend between initial 4 years (2011-2014) and the following 2 years (2014-2016). The results of our monitoring experiment suggested that understanding the mechanism of radiocesium transfer and spatio-temporal evolution of radiocesium distribution in forest must be essential for predicting long-term trend of ambient dose rate in forest environment.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident, Forest Environment, Radiocesium, Ambient dose rate

Migration of sediment and ^{137}Cs on decontaminated arable lands

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To mitigate radiological risks to residents in Fukushima, decontamination, such as removal of contaminated surface soil and covering contaminated soils with clean soil (i.e., soil dressing), has been conducted in areas of heavy radiological contamination. Although removing and concealing radioactive materials are expected to reduce ambient dose rates, such anthropogenic alteration of soil may result in changes to the hydrological responses and sediment dynamics of the area under the decontamination. Few studies have investigated the dynamics of sediment and ^{137}Cs associated with decontamination efforts. Our research studies the release of sediments and ^{137}Cs from decontaminated lands to elucidate the influence of decontamination. Soil erosion plots were installed on four contaminated land use types (uncultivated farmland, cultivated farmland, grassland, forest) and two decontaminated land use types (uncultivated farmland, cultivated farmland) to observe sediment discharge and ^{137}Cs wash-off. The ^{137}Cs activity concentration of eroded sediments from decontaminated lands were approximately 10 times lower than those from contaminated lands. The amount of sediment eroded from the decontaminated farmland was comparable to those from contaminated farmlands, but higher than those from contaminated grassland and forest. ^{137}Cs wash-off rates on decontaminated lands were lower than those on contaminated farmlands and higher than those on grassland and forest. These results suggest that decontamination results in a decrease of ^{137}Cs wash-off on erodible agricultural lands and increase relative to less erodible lands (grasslands and forests). Surface runoff coefficients and contribution of fine sediment to discharges of both sediment and ^{137}Cs were lower on decontaminated farmlands than those on contaminated farmlands. One possible explanation is that high permeability of decontaminated land resulted in inactive discharge of fine sediment. Elucidation of soil erosion processes on decontaminated lands based on investigation of its soil physical properties, such as permeability and particle size distribution, are required to better understand the influence of the decontamination.

Keywords: Decontamination , Soil erosion, Slope, 137Cs

The rapid deline of Cs-137 concentration in Fukushima rivers

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The Fukushima Daiichi Nuclear Power Plant accident has released massive amount of radiocesium into the terrestrial environment, and the radiocecium have been transferred through terrestrial hydrological pathways in the last 5 years. The ^{137}Cs attached fine particle of soil has been by washing off by erosion, then transported by river in which particulate (suspended) materials, play the 90-99% of the transported materials in Japanese rivers.
In the Chernobyl affected area, land use has been found to be affected the cleanup of the contaminated terrestrial environment , but precipitation and the land use is quite different affected by the radionuclides by FDNPP accident. Here we show the results by intensive field monitoring campaign, started in June, 2011, 3 months after the accident, monitoring 30 river stations for detailed monitoring of activity concentration of radiocesium in suspended sediment and their flux flown through the river.
Total 13 TBq of Cs have been transported to ocean from Abukuma river, this corresponds to 2.5 % of the total fallout from July 2011 to August 2015. Analysis of the riverine transport by upstream landuse reveals that higher percentage of Paddy field having largest rate of decline and also the highest flux, but the forest area showing lowest decline and smallest flux flowing downstream. The entrainment coefficient of dissolved water is quite low; due to high initial decrease by active landuses, combining with high flux, typically 1 order lower than the lower end by the report of Chernobyl affected area, suggesting the rapid recovery of Fukushima contamination. We also analyze the effect of decontamination works on the activity and flux of the suspended sediment in the rivers.

Keywords: Cs-137, Fukushima NPP accident, runoff from rivers

Effect of Cesium-bearing Microparticles to the Solid-Liquid Distribution of Cesium in Rivers

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Introduction: The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred on 11th March 2011 and a lot of radionuclides were emitted into the environment from the reactors (Yoshida and Takahashi, 2012). About 1.3×10^{16} Bq of Cesium-137 (Cs-137) was deposited to the ground by dry and wet depositions (Chino et al., 2011). According to Konoplev et al. (2016), solid-liquid distribution coefficient K_d values in Fukushima rivers are by 1-2 orders of magnitude higher than those in Chernobyl and it is possibly because the presence of Cs-bearing microparticles and higher Radiocecium Interception Potential (RIP) values of soils in Fukushima than those in Chernobyl. Cesium-bearing microparticle is a glassy, water-insoluble particle (Adachi et al., 2013). Cesium and many other elements from the reactor were found in the microparticles (Abe et al., 2014). The calculation of isotope ratio of Cs shows that the particles are from unit 2 or 3 (Nishihara et al., 2012). In this paper, the microparticles in suspended particles on the filters in river waters will be shown, focusing on the effect of the microparticles to the K_d values in the rivers. If there are the microparticles on the filter, apparent K_d value will be higher than intrinsic K_d value related to the adsorption-desorption reaction to the clay minerals.

Method: Suspended particles (3-63 μm) were collected from some rivers in Fukushima Prefecture by filtering water (60-90 L) in 2014 - 2016. After the filters were dried, total radioactivity of Cs-137 on the filters was measured by high purity Germanium semiconductor detector (HPGe). Planar distribution of radiation of filter was measured by autoradiography with Imaging Plates (IPs). After microparticle with high radioactivity was found on the filter, it was separated from other particles by water and NaI scintillation counter. Our method can separate microparticles more quickly and efficiently than previous methods (Adachi et al., 2013). Also, the microparticles can be more readily found by Scanning Electron Microscope (SEM) without using glue in the separation processes. The microparticles were measured by SEM coupled with Energy Dispersive x-ray Spectroscopy (EDS) to determine that the particles were similar to the particles reported in Adachi et al. (2013) etc. In the end, radioactivity of Cs-137 in the microparticle was measured by HPGe.

Result: Some hot spots were found on filters of Kuchibuto River collected in 2014 and 2015 by autoradiography. No hot spots were found on the filters from other rivers. The results of SEM-EDS showed that these hot particles and reported Cs-bearing microparticles were almost similar because they consist of elements such as Si, Cs, Fe, and Zn. Radioactivities of Cs-137 in the microparticles were about 0.8-3.0 Bq. Their isotope ratios of Cs (Cs-134/Cs-137 around 1.1) showed that the microparticles were from unit 2 or 3 of FDNPP. On filters collected on 3rd May 2014, 30% of Cs-137 on the filters was from the microparticles, while 46% of Cs-137 on filters was from the microparticles for the samples of 11th November 2015. If K_d value is calculated using these filters, the value will be overestimated compared with intrinsic K_d value that assumes adsorption-desorption reactions on clay minerals etc. The filters after 1-3 years from the accident and from different places should be measured successively. These results may explain the phenomenon that K_d values in rivers in Fukushima are higher than those in Chernobyl. The

effect of the Cs-bearing microparticles must be considered when K_d values of Cs in Fukushima are discussed.

Keywords: Fukushima, Cesium, Cesium-bearing particle, nuclear power plant accident, radioactivity, Solid-liquid distribution

Radiocaesium in the North Pacific Ocean derived from atmospheric weapons tests and Fukushima accident: A review of past and present

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1, Two major sources terms of radiocaesium to the Ocean from the Fukushima accident and fallout from atmospheric weapons tests before the accident

The ^{137}Cs derived from atmospheric weapons test conducted late 1950s and early 1960s and the inventory in the North Pacific Ocean in 1970 was 290 +- 30 PBq (Aoyama et al., 2006). Some portion of the ^{137}Cs in the North Pacific Ocean were transported to South Pacific Ocean and Indian Ocean and also radioactive decay occurred with a half-life of 30.7 years, the ^{137}Cs inventory in the North Pacific Ocean before the FNPP1 accident decreased to 69 +- 7 PBq as of 2011 (Aoyama et al., 2016).

There are two major sources of radionuclides to the environment derived by the TEPCO Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident in 2011. The largest and earliest source of artificial radionuclide was atmospheric release from FNPP1, which led to atmospheric deposition on both land and in the ocean. Atmospheric release peaked mid of March 2011 and total amount of atmospheric release of ^{137}Cs was estimated to be 15.2-20.4 PBq and same amount of ^{134}Cs was also released because activity ratio of ^{134}Cs vs. ^{137}Cs was almost 1(Aoyama et al., 2016). About 20 % of released radiocaesium fell on land and 80% of released radiocaesium fell on the ocean. Therefore 11.7-14.8 PBq of ^{137}Cs was injected in the North Pacific Ocean as atmospheric deposition.

Second largest source was the direct discharge of contaminated waters to the ocean since 26 March 2011 and peaked on 6 April 2011 based on analysis of 131I vs. ^{137}Cs activity ratio (Tsumune et al., 2012). Total amount of released ^{137}Cs was estimated to be 3.5 +- 0.7 PBq. A combined input to the North Pacific Ocean of ^{137}Cs from both atmospheric deposition and direct discharge was therefore estimated to be 15.2 –18.3 PBq.

2, Three major pathways of FNPP1 derived radiocaesium in the North Pacific Ocean

The fastest pathway of FNPP1 derived radiocaesium might be surface pathway. FNPP1-derived radiocaesium injected at north of Kuroshio front by atmospheric deposition and direct discharge spread eastward in surface water up to 200 meters by the North Pacific Current across the mid-latitude North Pacific (Aoyama et al., 2016). In 2013 main body of FNPP1 radiocaesium in surface layer was already in the eastern Pacific. A model simulation (Tsubono et al., 2016) also shows good agreement with the observed radiocaesium activities in the Pacific Ocean reported by several studies.

The second pathway is subduction of central mode water (CMW). A maximum of radiocaesium activity in June/July 2012 was observed at potential densities of 26.1–26.3 at 34 deg. N–39 deg. N, 165 deg. E, which correspond to 400 meters depth. The density is in a range of density of CMW and radiocaesium activity was higher than those in the surrounding waters, including STMW. In June-July 2015 and June 2016 at 36˚N–44˚N along 165˚E, there are only very week signal of subduction of FNPP1 radiocaesium. This means that subducted radiocaesium might move eastward from this region. Before the Fukushima accident, ^{137}Cs maximum corresponding CMW region was observed, however, it located at 20˚N, 165˚E because it was 40 years after subduction (Aoyama et al., 2008).

The third pathway is subduction of subtropical mode water (STMW). FNPP1-derived radiocaesium injected at south of Kuroshio front by atmospheric deposition transported to southward rapidly due to subduction of STMW at potential densities of 25.1–25.3. In 2015 along 165 deg. E, FNPP1 radiocaesium corresponding STMW spread entire subtropical gyre and a part of them reached 2 deg. N and recirculated in the subtropical gyre and reached Japanese coast.

3, Mass balance of FNPP1 radiocaesium in the North Pacific

134Cs inventory was estimated to be 8 PBq in surface layer in summer 2012 (Inomata unpublished). Kaeriyama et al. (2016) estimated that 134Cs inventory in STWM in 2012 was about 4 PBq. We believe that FNPP1 derived 134Cs injected in the North Pacific was 15.2 - 18.3 PBq. Therefore 134Cs inventory can be estimated 3-6 PBq in CMW at this moment based on a mass balance of FNPP1 radiocaesium. The ^{137}Cs inventory in the North Pacific Ocean before the FNPP1 accident, which was derived mainly from nuclear weapons testing, was 69 PBq at the end of 2010. Thus, the FNPP1 accident increased the ^{137}Cs inventory of 15.2 - 18.3 PBq by as much as 22 - 27%.

Keywords: Fukushima accident, global fallout, North Pacific Ocean

Oceanic dispersion of radioactive cesium derived from Fukushima Dai-ichi Nuclear Power Plant accident in the North Pacific during six years

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After the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, many data on radioactive cesium (^{134}Cs and ^{137}Cs) in seawater had been collected and published. Present study summarizes details the radioactive cesium dispersion pattern in the North Pacific based on observational data obtained by FRA. Briefly, the Fukushima-derived radioactive cesium dispersed eastward as surface water, and was also observed via a southward intrusion to subsurface waters as Subtropical Mode Water and Central Mode Water. The radioactive cesium movement related to mode water is important in terms of the circulation of cesium into the ocean interior. The concentration of Fukushima-derived radiocesium in Subtropical Mode Water and water column inventory of ^{137}Cs were gradually decreased between 2012 and 2015.
The most remarkable temporal changes of Fukushima-derived radioactive cesium off the coast near the FDNPP site were observed during the first six months of 2011. After that, continuous decreasing trend has been observed until 2016. Although higher concentrations of radioactive cesium than the background levels measured before the accident are still detected. Continued monitoring is still necessary.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident, Radioactive cesium, North Pacific, Mode water

Transport of Fukushima-derived radiocesium into the coastal ocean via submarine groundwater discharge: an approach from geochemical character of pore water

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Submarine groundwater discharge (SGD) has been recognized as a route of Fukushima-derived radiocesium (Cs) from land to the ocean tougher with rivers. However, it is difficult to take large volume SGD samples compared with river water, which can be collected more easily. Therefore, quantitative evaluation of its impact as a source of Cs has not been carried out. In this study, we analyzed Cs activity and chemical component in pore water in Matsukawa-ura Lagoon to explore the transport and flux from land to ocean including SGD.
The measured ^{137}Cs activity in pore water samples and the overlying water were 1,398 mBq/L and 117.7 mBq/L, respectively. This indicates that a significant amount of Cs in sediment has been desorbed into pore water. The ^{137}Cs flux between the sediment and overlying water was calculated to be 11.3 mBq/cm^2 /h, using Fick’ s First Law of diffusion. Moreover, the ^{137}Cs flux from the bottom to overlying water of the lagoon was estimated to be 0.08 GBq/day based on the weighted average of ^{137}Cs activity in bottom sediment of the lagoon and the Kd value between the sediment and the pore water. This value accounts of the majority of ^{137}Cs which supplies the lagoon. Thus, it can be expected that large quantities of Cs have been supplying the coastal area by pore water exchange, that is to say, recycled SGD (RSGD). In the coastal area and the open ocean, it is suggested that SGD is vitally important as a source of Cs to assess it.

Keywords: Fukushima-derived radiocesium, Submarine Groundwater discharge, Pore water, Matsukawa-ura Lagoon

Regional-scale oceanic simulations of ^{137}Cs, ^{90}Sr, ^{3}H radioactivity directly released by the Fukushima Dai-ichi Nuclear Power Plant accident

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A series of accidents at the Fukushima Dai-ichi Nuclear Power Plant (1F NPP) following the earthquake and tsunami of 11 March 2011 resulted in the release of radioactive materials to the ocean by two major pathways, direct release from the accident site and atmospheric deposition. Additional release pathways by river input and runoff from 1F NPP site with precipitation and were also effective for coastal zone in the specific periods before starting direct release on March 26 2011. The activities attributable to the direct release were observed adjacent to the 1F NPP site. The sea side impermeable wall was closed at 26 October 2015. We estimated the direct release rate of ^{137}Cs, ^{90}Sr and ^{3}H for more than four-and-a-half years after the accident by the Regional Ocean Model System (ROMS).

Direct release rate of ^{137}Cs were estimated by comparing simulated results and measured activities adjacent to the 1F NPP site (adjacent to 5,6 discharge and south discharge). Direct release rate of ^{137}Cs was estimated to be 2.2×10^{14} Bq/day and decreased exponentially with time to be 3.9×10^{9} Bq/day by 26 October 2015. Estimated direct release rate have exponentially decreased with constant rate since 4 November 2011. Apparent half-life of direct release rate was estimated to be 346 days. The estimated total amounts of directly released ^{137}Cs was 3.6±0.7 PBq from 26 March 2011 to 26 October 2015. Simulated ^{137}Cs activities attributable to direct release were in good agreement with observed activities, a result that implies the estimated direct release rate was reasonable. Simulated ^{137}Cs activity affected off coast in the Fukushima prefecture.

$^{90}Sr/^{137}Cs$ activity ratio of stagnant water was 0.05 in the basement of the 1F NPP reactor 2 turbine building on 27 March 2011. Direct release rate of ^{90}Sr was estimated to be 1.1×10^{13} Bq/day from 26 March to 6 April 2011 using the activity ratio in stagnant water because the stagnant water released to the ocean in this period (Tsumune et al., 2012). And the temporal change of direct release rate was estimated by the measured ^{90}Sr activity adjacent to 1F NPP. Directly release rate decreased exponentially to 3.9×10^{10} Bq/day by 30 April 2011. The direct release rate was constant and decreased exponentially from 27 June to 16 December 2013. And the direct release rate was 2.9×10^{9} Bq/day by 26 October 2015. The estimated total amounts of directly released ^{90}Sr was 208 ±42 TBq.

$^{3}H/^{137}Cs$ activity ratio of stagnant water was 8.7×10^{-3} in the basement of the 1F NPP reactor 2 turbine building on 27 March 2011. Directly release rate of ^{3}H was estimated to be 1.9×10^{12} Bq/day from 26 March to 6 April 2011 and decreased exponentially by 16 April 2011. The rate was decreased exponentially with constant rate by 26 October 2015. The direct release rate was estimated to be 7.7×10^{9} Bq/day at 26 October 2015. The estimated total amounts of directly released ^{3}H was 131 ±26 TBq.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident , regional ocean model, direct release, radiocaesium, radiostrontium, tritium

Radiocaesium activities in the North Pacific Ocean Water from 1945 to 2020 Calculated by eddy-resolving ROMS

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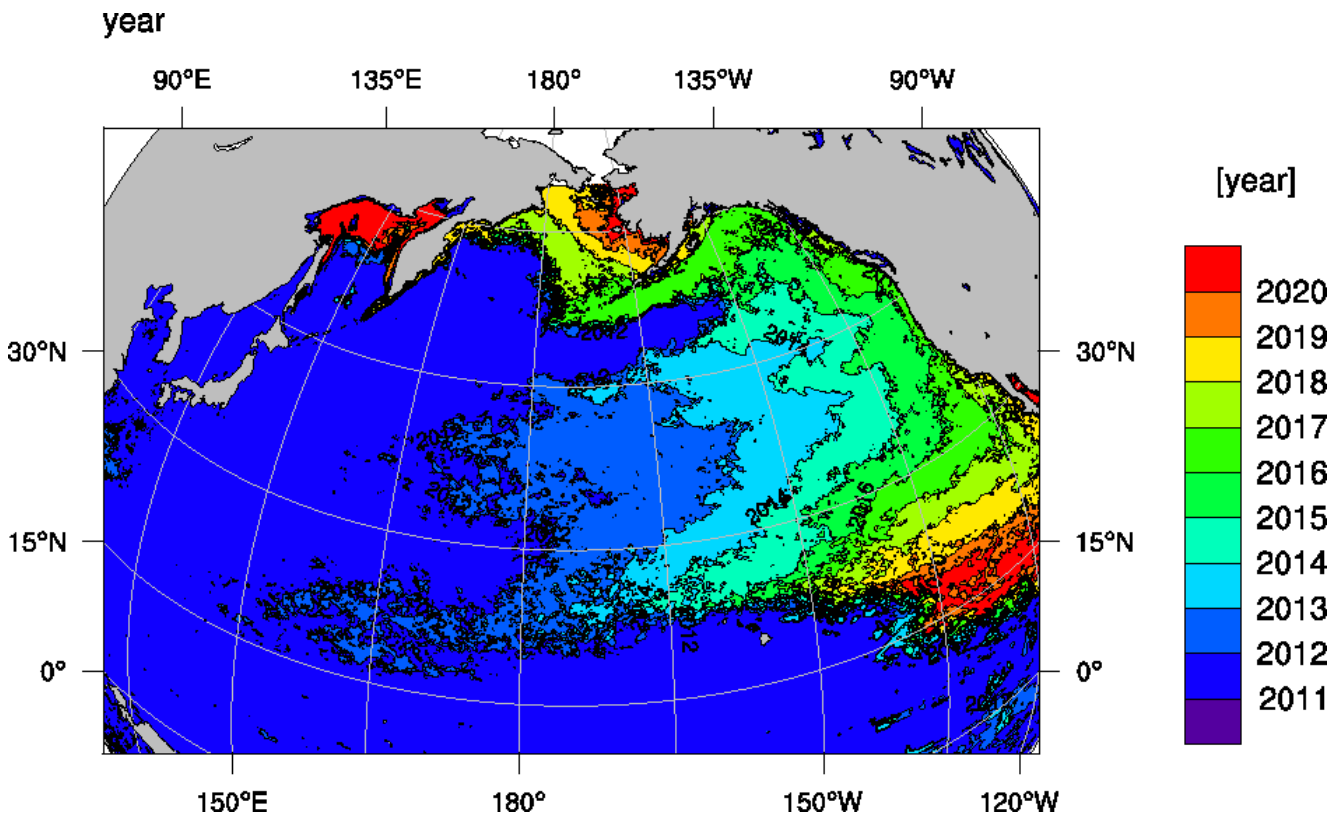
We conducted the ensemble simulation of ^{137}Cs activity in the North Pacific Ocean (NPO) water from 1945 to 2020, before and after the Fukushima Dai-ichi Nuclear Power Plant (1F NPP) accident, because we used the estimations of ^{137}Cs activity flux, but climatology as physical forcing . Using the Regional Ocean Model System (ROMS) with high resolution (1/12º-1/4º in horizontal, 45 levels in vertical), of which domain was the NPO, we preliminarily estimated a factor multiplying the total of ^{134}Cs fluxes, which have been estimated for the atmospheric deposition and the direct discharge from the accident. The direct comparison of the observed and calculated ^{134}Cs showed that the total ^{134}Cs Flux was 1.6 times greater than the previous estimate. We re-calculated the ^{134}Cs activities in the NPO water using the flux multiplied by 1.6 and confirmed the improvement of the simulation by the multiplied flux, which suggested that each the ^{134}Cs and ^{137}Cs inventories in the NPO increase by about 16PBq due to the accident. For the hindcast and forecast of the ^{137}Cs activity in the NPO water, we calculated the ^{137}Cs activity in the NPO water from 1945 to 2020 by using the global fallout flux due to atmospheric nuclear weapons' tests and the Chernobyl accident and the estimated fluxes of the 1F NPP accident. For the calculation, five ensemble calculations of ^{137}Cs activity were conducted by moving the start period of the input flux for one year. The ^{137}Cs activity in the surface water showed that the plume due to the 1F NPP accident with relatively higher activity than 5 Bq m^{-3}was transported to the western area of 135˚W in 2015, while the activity of the plume was rather lower than that in 1985. The peak year of the ^{137}Cs activity can be estimated from the hindcast and forecast. The ^{137}Cs activity in the surface water north of 30˚N shows that the peak activity of ^{137}Cs reached 180˚ in 2011, but in 2012 it moved near 180˚ and in 2017 moved around 90˚W. The total inventory of ^{137}Cs in the NPO increased up to 77 PBq in 2011 and gradually decreased to 61PBq in 2018 by transport outside of the domain, which is almost the same as that in Dec. 2010. The whole amount of ^{137}Cs in the subsurface layer (200-600m depth) is larger than that in the surface layer (0-200m depth) since the 1F NPP accident except 2011.

Keywords: 137Cs, North Pacific ocean, eddy-resolving model



Radioactive cesium-bearing particles in various environmental samples

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Radioactive materials released by the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident caused extensive radioactive contamination. Adachi et al. (2013) discovered radioactive Cs-bearing particles (Cs-bearing particles) from air filter in Tsukuba (170 km from FDNPP). This finding is an important, since the particle which is likely to be emitted directly from FDNPP may contain various information on the phenomena occurring in FDNPP during the accident. However, because of the difficulty of separating Cs-bearing particle from environmental sample, comprehensive information on physical and chemical properties of the particles as well as distribution of particles in Fukushima contaminated areas is limited. In this study, the distribution and physical and chemical properties of Cs-containing particles were investigated using various types of environmental samples, such as suspended particles in river and surface seawater, tree leaves, and road dust (noted as black substances).
Radioactive cesium-bearing particle in the sample was separated from other particles by the wet-separation method using a NaI scintillation counter. The separated particle was identified by a scanning electron microscope (SEM) equipped with an energy dispersed X-ray spectrometer (EDS). The activities of ^{134}Cs and ^{137}Cs in the identified particle were measured by non-destructive gamma-ray spectrometry.
Spherical particles with diameters of approximately $< 5\ \mu$m were found in any samples. Particles of this type are similar in terms of chemical composition to those reported so far, and were estimated to be derived from Units 2 or 3 of FDNPP estimated by the ^{134}Cs/^{137}Cs activity ratio. In the dust samples of the northwest direction within 20 km from FDNPP, particles with diameters of several tens to several hundreds of microns were found. This area has been reported to be contaminated with radioactive materials from Unit 1 (Satou et al., 2015). Most of the particles in this region were not uniform in shape. The main components of the particles in this region were Si, Ca, K, and Al derived from Unit 1 from the ^{134}Cs/^{137}Cs activity ratio. In addition, although it is reported that the shape of the particles in this region is not uniform, some spherical particles with a diameter larger than $> 20\ \mu$m were also found.
From the facts above, it was found that three kinds of radioactive particles are widely present in various environmental samples.

Keywords: Fukushima Daiiti Nuclear Power Plant, Cs-bearing particle, Environmental sample, Radioactivity

Atmospheric radioCs in case of the local dust event

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Atmospheric radioCs seems suspended along with surface soil dust when the strong wind creates local dust event. Local surface contamination may have resulted in the enhancement of the radioCs concentration when the local dust event occurred. Although Ishizuka et al. (2017) made the radioCs resuspension computation scheme due to the dust uplift in the air by wind blow, the effect of the local dust event has not been evidenced nor confirmed in detail so far. Furthermore, though Kajino et al. (2016) has simulated yearly resuspension in 2013 due to the dust suspension and those from the forest, event-based case study has not been carried out. In this presentation, some of such events are analyzed in terms of the radioCs resuspension. The dust event occurred in March 2013 was analyzed for meteorology and temporal change in radioCs concentration observed at Meteorological Research Institute in Tsukuba. The simulated results of radioCs concentration in Tsukuba is compared with the observed results. These analysis would suggest qualitatively minor effect of the local dust events on the enhancement of the radioCs concentration.

Keywords: radioCs, local dust event, concentration

Long-term Changes in the Distribution of Atmospherically Deposited Radioactive Cs in a Small Forest in Fukushima Prefecture

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Radioactive Cs concentration in fresh leaves/needles, litter, surface soil, stream sand in a deciduous broadleaf forest and cedar forest in Minamitsushima, Namie-town was continuously investigated from June 2012 except for snow-cover period. A car-borne survey from Fukushima city to Minamitsushima showed that air dose rate declined faster than the values estimated by physical attenuation, due to the radioactive decontamination, other than the forest area. Radio-Cs concentrations (^{137}Cs+^{134}Cs) in litter and surface soil in deciduous broadleaf forest were constant at 52.0, 102 Bq kg-dry^{-1} respectively from 2014. However, those in fresh needle and litter in cedar forest continued to decline, probably because of washing and leaching by throughfall, from 2012 to 2016 and accumulated in surface soil (106 Bq kg-dry^{-1} in 2016). Radio-Cs infiltrated into soil deeper in deciduous broadleaf forest (buffer depth: 1.26 cm) than in cedar forest (buffer depth: 1.14 cm) in spring in 2013, but the relationship between the two reversed in winter in 2015 (buffer depth: 1.5 cm in broadleaf forest and 2.6 cm in cedar forest). Radio-Cs were concentrated in smaller bottom sand in stream water (over 2 cm: 3.04, 0.2-2 cm: 10.2, under 0.2 cm: 54.5 Bq kg-dry^{-1} in downstream near the broadleaf forest and over 2 cm: 2.67, 0.2-2 cm: 7.95, under 0.2 cm: 41.3 Bq kg-dry^{-1} in upstream near the cedar forest) and the outflow of a part of that as suspended sand were concerned. Though the relative radio-Cs concentrations in smaller bottom sand to those in surface soil have declined (2013:0.54, 2016: 0.29 in downstream and 2013: 1.4, 2016: 0.31 in upstream), floating male flowers of cedar containing high radio-Cs (23.8 Bq kg-dry^{-1}) could be another transport media in spring.

Keywords: Forest, Surface soil, Fresh leaves/needles, Litter, Stream sand, Edge effect

Radioactive Cs distribution of litter and soil in forests in each kind tree for 5years after Fukushima Dai-ichi Nuclear Power Plant accident

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The Fukushima Dai-ichi Nuclear Power Plant fall-out contaminated the surrounded areas with high levels of radioactive materials in march 2011. We have been investigated based on soil samples as part of a cooperative pilot survey involving both government and academia since May 2011. The surveys detected widespread radioactive contamination throughout the forested areas in Fukushima prefectures, where approximately 71% of the land area is forested. And, there is an artificial forest more than 35% in Fukushima forest. Usually, artificial forest is made with a specific kind tree. They are processed to plant mushrooms and/or to build houses. Mushrooms are cultured on *Quercus serrata*. It's a deciduous tree. It means that Quercus serrata had not put leaves in the accident. Another hand, houses are built of *Cryptomeria japonica*. It is an evergreen tree. It means that fall-out was contaminated canopy of *Cryptomeria japonica*. Radionuclides deposited in forested areas by either wet or dry processes encounter the canopy. Most radioactive Cs (>90%) deposited onto the canopy is intercepted and retained by tree leaves and branches and subsequently transferred to the forest floor as a result of weathering by rainwater and wind.
The fall-outed radioactive Cs contaminated any kind tree forest. It is important to know the distribution of radioactive Cs in each kind tree forest for decontamination of forests.
Therefor we studied to know radioactive Cs distribution of litter and soil in the forest floor in each kind tree from 2011 to 2016.
Forests were separated for *Pinus densiflora* forest (PDF), *Pinus densiflora* Mix forest (PD Mix), decicuous tree forest (DTF), *Cryptomeria japonica* forest (CJF) and other coniferous tree forest (OCF). Air dose rate measured 3 times in 2011, 2014 and 2016. The number of samples were each 26, 56, 153, 71 and 52 in 2011, each 7, 28, 88, 52and 33 in 2014, and each 7, 13, 24, 33 and 9 in 2016. The litter and soil radioactive Cs measured in 2011 and 2016.
Each kind tree forests air dose rate was positive correlated an initial deposition radioactive Cs. And, each forests air dose rate decreased with time. 2011-2014 air dose rate decrease was faster than 2014-2016 decrease in PDF, PD Mix, DTF and OCF. Another hand, 2014-2016 air dose rate decrease was faster than 2011-2014 decrease in CJF. It means that the timing of air dose rate decrease was delay in CJF. Each forests air dose rate decrease was faster than the physical decay of radioactive Cs. Another hand, each radioactive Cs inventory of litter and soil decrease was slower than the physical decay of radioactive Cs. CJF's radioactive Cs inventory of litter and soil increased significantly faster than PDF's and DTF's. It means that each forest air dose rate was negative correlated radioactivity of the litter and soil from 2011 to 2016. The litter's radioactive Cs inventory decreased faster than radioactive Cs physical decay in PDF, PD Mix, DTF and CJF. DTF's rate of decrease was significantly faster than PD Mix's and CJF's. The soil's radioactive Cs inventory decrease slower than radioactive Cs physical decay in PDF's and OCF's. And, the radioactive Cs of soil of PD Mix, DTF and CJF in 2016 were more than the radioactive Cs inventory of soil in 2016. And, CJF's rate of radioactive Cs inventory of soil increased significantly higher than PDF's. It means that radioactive Cs distributed from litter to soil in each forest.
Therefore, air dose rate of forest was decreased faster than radioactive Cs physical decay from 2011 to 2016. And, litter's radioactive Cs inventory decreased from 2011 to 2016. Soil's radioactive Cs inventory increased from 2011 to 2016. It was thought that radioactive Cs distributed from litter to soil in every forest from 2011 to 2016.

Temporal changes of Cs-137 concentration and its flux (dissolved fraction, suspended sediment, and coarse organic matter) at small Headwater Catchment in Fukushima after Fukushima Dai-ichi Nuclear Power Plant Accident

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Radiocesium migration from headwater forested catchment is important perception as output from the forest which is also input to the subsequent various land use and downstream rivers. In this study, dissolved Cs-137 concentration of stream water, subsurface water, suspended sediment (SS) and coarse organic matter (Org) were measured. Observations were conducted from 2011 at four headwater catchments in Yamakiya district, located 35 km northwest of Fukushima Dai-ichi Nuclear Power Plant (FDNPP).
Stream water discharge was monitored by combination of parshallflume and water level recorder. Stream water was sampled manually at steady state condition in 2-4 month interval. Also intense few hours interval sampling of stream water was conducted during rainfall events using automated water sampler. Suction lysimeters were installed for sampling the soil water. Boreholes were installed for the groundwater sampling. Stream water and groundwater samples were collected for 40 L each. All the water samples were filtered through 0.45 μm pore-size membrane. Water samples with less than few L were concentrated by evaporative concentration. Water samples with more than few L were concentrated using the ammonium molybdophosphate (AMP)/Cs compound method. The SS was sampled using time-integrated SS sampler. Turbidity sensor was set in the streambed. The output of the turbidity (mV) was converted to SS concentration (mg/L). A 15 mm mesh net was placed in the stream channel to trap Org sample carried by the stream flow. The SS and Org samples were dried at 105 degree in an oven for 24 h and then finely crushed. The radioactivity of the samples was measured using gamma-ray spectroscopy.
The fast decline of first phase and secondary comparatively gradual decline were observed in time series of dissolved Cs-137 concentration in stream water and therefore fitted to two-component exponential line. During the rainfall events, the concentration of dissolved Cs-137 in stream water increased temporarily. Also the declining trend of Cs-137 concentration in SS and Org were fitted to exponential line however, those components were started to collect from August 2012 and the fast declining phase were not observed therefor fitted to single-exponential line. After 5 years passed from the accident, the difference of declining trend between catchments are shown.
In conjunction with Cs-137 concentration results, Cs-137 discharge flux were calculated for three components (dissolved fraction, SS, and Org). As a result, it is shown that more than 96% of the Cs-137 discharge was due to the discharge by SS. The total discharge ratio of Cs-137 to its deposition amount at each catchment was ranged 0.002–0.3% per year.

Keywords: Cs-137, FDNPP, headwater catchment

Dynamic association of radioactive cesium between sediment and suspended sediment in river

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Abstract

After earthquake and tsunami of 11 March 2011,a lot of radionuclides ^{134}Cs and ^{137}Cs were released into environment due to Fukushima Dai-ichi Nuclear Power Plant(FDNPP) accident. For the safety of people and the support of decontamination, the Japanese Ministry of Education,Culture,Sports,Science and Technology(MEXT) has taken a long term monitoring system,in which about river sediment,surrounding environment and water quality,in areas such as Iwate,Miyagi and Fukushima Prefectures since August 2011.Meanwhile,7 sites were newly opened in Nakadori and Hamadori since July 2011,24 sites were opened since October 2012,to monitor the dynamics of water quality and suspended sediment.However due to previous researches,we can see the obviously tendency of declining radiocesium concentration in suspended sediment,which slowed down after one year from the accident.But we cannot conclude the same ideal by analyzing the sediment samples which was taken by MEXT during 2011-2012,owing to the low correlation and widely variation between time and concentration caused by the influences of particle size.So during this survey we reanalyzed the particle size of samples and correction the particle size effect on the concentration of ^{137}Cs.As a result,80% sites' declining rate and the correlationship between time and concentration were improved.On the other hand,we compared the decline rate between suspended sediment and river sediment in the same watershed,we find there are same tendencies and numerical correlations of radiocesium between suspend sediment and river sediment.

Keywords: Radiocesium, Sediment, Suspended Sediment, Persistent

Distributions of radiocaesium activity in sediment and overlying water off the Fukushima

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After the accident at the Fukushima Dai-ichi Nuclear Power Station (FDNPS) happened in March 2011, large amounts of radionuclides including radiocaesium also released from the FDNPS into the terrestrial and marine environments. In marine environment, parts of particulate radiocaesium have transported in seawater and accumulated to seafloor. Then, radiocaesium in sediment have partly re-suspended as particulate form and re-eluted as dissolved form due to several factors such as bottom current and deformation. The characters of seafloor topography are more different in the area off the coast of northern and southern part of Fukushima Prefecture, dividing areas at the Onahama port (Mogi and Iwabuchi, 1961). Because the wave bases in fine and stormy weather are about 20 and 80 m, respectively (Saito et al., 1989), it seems that the area of shallower than 100 m is also affected by erosion and re-sedimentation near seafloor with ocean wave degree. Thus, it is necessary to elucidate interaction for radiocaesium between sediment and seawater close to seafloor with more stations in order to guess radiocaesium activity variation at long times. For example, in the case of collected bottom-layer water with the Conductivity-Temperature-Depth (CTD) system, it is very difficult to collect seawater close to sediment because it is careful not to touch CTD system seafloor. This study was aimed at elucidating the relationship for radioacesium activity concentration between sediment and trapped water on sediment collected using Multiple Corer, which is considered as overlying water.
Sediment samples were collected using a Multiple Corer during UM14-04 cruise in May 2014 at three stations: I01 (37˚14’ N, 141˚07’ E, water depth:60 m), I02 (37˚14’ N, 141˚13’ E, water depth:120 m) and C (36˚55’ N, 141˚20’ E, water depth:190 m).Overlying waters were collected using tube for 2 hours later from collected sediment. In laboratory, collected sediment sample are dried and overlying water samples were filtered through a 0.2-μm pore size filter and was concentrated by the ammonium phosphomolybdate (AMP) method (Aoyama and Hirose, 2008). The radiocaesium activity concentrations in each sediment and overlying water samples were measured by gamma-ray spectrometry using a high-purity Ge-detector and corrected to sampling date.
In overlying water, the dissolved ^{137}Cs activity concentrations (mBq/l) were 3.1-16 and the activity at I01, I02 and C in order from the higher. In the surface-layer sediments (core depth 0-3cm), the activity concentrations (Bq/kg-dry) were 8.4-286 and the high activities at I01 and I02 have characters of relatively high percentage for silt to clay particle compared to those at C. At I02 and C, the activity in overlying water were same value compared those in bottom-layer of seawater, which collected above water depth 10 m from seafloor. On the other hand, the activity in overlying water at I01 was five time higher than those in bottom water. The calculated K_d' (L/kg) of apparent distribution coefficient using ^{137}Cs activity concentrations in surface-layer sediment and overlying water were 8.8×10^2-1.5×10^4 and within rages of recommended K_d value of 2.0×10^3 for caesium by IAEA TRS422.

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Keywords: radiocaesium, overlying water, sediment, Fukushima Dai-ichi Nuclear Power Station accident

Fukushima-derived radiocesium in the western subtropical gyre of the North Pacific Ocean in 2015/2016

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Accident of Fukushima-Dai-ichi Nuclear Power Plant (FNPP1) on 11 March 2011 resulted in a large amount release of radiocesium (^{134}Cs and ^{137}Cs) into the North Pacific Ocean. Oceanographic observations have revealed that the Fukushima-derived radiocesium deposited on and discharged directly into coastal area of Japan is transported eastward in surface layer along the North Pacific Current (Kumamoto et al., 2016). In addition, radiocesium deposited on an area south of the Kuroshio/Kuroshio Extension Current had been transported southward to about 15 north degree through subsurface layer due to subduction of the subtropical mode water (STMW) by the end of 2014 (Kumamoto et al., 2014). On the other hand, in coastal areas of nuclear power plants in Hokkaido, Niigata, Ishikawa, Fukui, Shimane, Saga, Kagoshima, Ehime, and Shizuoka prefectures, activity concentration of radiocesium in surface seawater was increasing between 2011 and 2015 (NSR, 2016). Aoyama et al. (2017) also reported increase in radiocesium activity in surface seawater in the coastal area. Contribution of the Kuroshio Current water is relatively large in these coastal area where the activity concentration of radiocesium increased. These results suggest that the Fukushima-derived radiocesium spread into the western subtropical area has returned to the coastal area of Japan along a clockwise subtropical gyre current. However, temporal and spatial variation in the Fukushima-derived radiocesium in the western subtropical gyre is not clear. In 2015/2016 we measured vertical profile of radiocesium in seawater from surface to 800 m depth in the western subtropical area south of the Kuroshio/Kuroshio Extension Current. Seawater samples (10-20 liter) were collected using a bucket or Niskin Sampler during research cruises of KS15-14 (October 2015), KH16-03 (June 206), and KM16-08 (September 2016). In a laboratory, the seawater sample was acidified using nitric acid and then radiocesium in the seawater was concentrated onto ammonium phosphomolybdate (AMP). Radiocesium in the AMP was measured using gamma-ray detectors. Uncertainty of the radiocesium measurement was estimated to be about 8 %. A vertical profile of activity concentration of ^{134}Cs, which corrected to the FNPP1 accident date for radioactive decay, in 30-32 north degree/144-147 east degree in 2015 and 2016 was compared to that observed in 2014 (Kumamoto et al., 2017). In surface mixing layer between surface and about 100m depth, ^{134}Cs increased from about 1 Bq/m^3 in 2014 to about 1.5-2.5 Bq/m^3 in 2015/2016. In subsurface maximum layer (300-400 m depth), whose water density agrees with that of STMW, 3-4 Bq/m^3 of ^{134}Cs activity concentration did not change between 2014 and 2016. According to Kumamoto et al. (2017), in 34 north degree/147-150 east degree just south of the Kuroshio/Kuroshio Extension Current, decay-corrected activity concentration of ^{134}Cs in the surface mixed layer increased from below the detection level (about 0.1 Bq/m^3) in 2012 to about 1 Bq/m^3 in 2014 while the concentration in the subsurface layer decreased from about 16 Bq/m^3 in 2012 to 3-4 Bq/m^3 in 2014. These observational results clearly suggest that the Fukushima-derived radiocesium transported southward due to subduction of STMW has come back to the northern subtropical area south of Japan along the clockwise subtropical gyre current. If the other sources (e.g. riverine water input) are negligible, the ^{134}Cs increase in surface seawater from 2012 to 2016 (from less than 0.1 Bq/m^3 to about 1.5-2.5 Bq/m^3) was probably derived from entrainment of the subsurface maximum into the surface mixed layer. We will show additional data of dissolved radiocesium in coastal area of Japan in 2015/2016 in the coming presentation. This work was partially supported by Grant-in-Aid for Scientific Research on Innovative Areas, the Ministry of Education,

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Keywords: Fukushima Dai-ichi nuclear power plant accident, radiocesium, western subtropical gyre, subtropical mode water

Estimate of spatial and temporal variation of radiocaesium amount derived FNPP1 accident in the North Pacific Ocean

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^{134}Cs and ^{137}Cs (radiocaesium) were released to the North Pacific Ocean by direct discharge and atmospheric deposition released from the TEPCO Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident in 2011. It is recognized that estimation of the total amount of released ^{134}Cs and ^{137}Cs is necessary to assess the radioecological impacts of their release on the environment. It was reported that the inventory of ^{134}Cs or ^{137}Cs on the North Pacific Ocean after the FNPP1 accident was 15.2-18.3 PBq based on the observations (Aoyama et al., 2016a), 15.3±1.6 PBq by OI analysis (Inomata et al., 2016), 16.1±1.64 PBq by global ocean model (Tsubono et al., 2016). These suggest that more than 75 % of the atmospheric-released radiocaesium (15.2-20.4 PBq; Aoyama et al., 2016a) were deposited on the North Pacific Ocean. It was revealed that these radiocaesium existed in the Subtropical Mode Water (STMW, Aoyama et al., 2016b; Kaeriyama et al., 2016) and Central Mode Water (CMW, Aoyama et al., 2016b), suggesting that mode water formation and subduction are efficient pathway for the transport of FNPP1 derived radiocaesium into the ocean interior within 1-year timescale. Kaeriyama et al. (2016) estimated the total amount of FNPP1 derived radiocaesium in the STMW was 4.2 ±1.1 PBq in October–November 2012. However, there is no estimation of the amount of radiocaesium in the CMW. Therefore, it is impossible to discuss about the mass balance of radiocaesium injected into the North Pacific Ocean. In this study, we conducted the optimum interpolation (OI) analysis to estimate the inventory of radiocaesium in the ocean interior as well as surface sweater by using the measured activities. Furthermore, transport speed of radiocaesium in the surface layer in the North Pacific Ocean were also estimated. The data used in this study were derived from all of the available data reported by such as the Tokyo Electric Power Company (TEPCO), the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT), and voluntary cargo ships. The data analysis period was until December 2015 after the FNPP1 accident. It was found that the radiocaesium across the North Pacific Ocean were reached to 180˚ E around 40˚ N latitude at July, 2012 by OI analysis. The transport speed was estimated to 8.5 cm s^{-1} . These were reached to the coastal site of America continent and the activities were increased after the year of 2014. The transport speed across 70˚ W (40˚N latitude) was decreased to 5.2 cm s^{-1}.
We estimated the inventory of radiocaesium in the surface seawater (depth; 0-100m) during the periods from August to December, 2012, based on the OI analysis. Amount of ^{134}Cs inventory was estimated to 4.7 PBq with decay-corrected to 1 October 2012 (7.9 PBq at the time on 11 March 2011). (In the case of ^{137}Cs, the inventory was estimated to 12.5 PBq with decay-corrected to 1 October 2012 and 13 PBq at the time on 11 March 2011 which includes pre-Fukushima ^{137}Cs). These correspond to 43-53% of the injected ^{134}Cs in the North Pacific Ocean. It was reported that the 4.2±1.1 PBq of ^{134}Cs were distributed in the STMW (Kaeriyama et al., 2016). Taking into account these estimation, FNPP1 derived radiocaesium existed in the CMW in the North Pacific Ocean would be about 3-6 PBq.

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