

## One-year, regional-scale simulation of radiocaesium-137 radioactivity in the ocean following the Fukushima Daiichi Nucle

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A series of accidents at the Fukushima Dai-ichi Nuclear Power Plant 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. A 1-year, regional-scale simulation of <sup>137</sup>Cs activity in the ocean offshore of Fukushima was carried out, the sources of radioactivity being direct release, atmospheric deposition, and the inflow of <sup>137</sup>Cs deposited on the ocean by atmospheric deposition outside the domain of the model.

The rates of direct release of <sup>131</sup>I, <sup>134</sup>Cs, and <sup>137</sup>Cs were estimated for 1 year after the 1F NPP accident by comparing simulated results and measured activities. The estimated total amount of directly released <sup>131</sup>I, <sup>134</sup>Cs, and <sup>137</sup>Cs by the end of February 2012 were 11.1 PBq, 3.52 PBq, and 3.55 PBq, respectively. Tsumune et al. (2012) previously estimated the total amount to be 3.51 PBq by the end of May 2011. The total amount of directly released <sup>137</sup>Cs activity increased by 0.04 PBq between June 2011 and February 2012. We used an atmospheric transport model with atmospheric release rates to estimate atmospheric deposition onto the ocean.

We analyzed <sup>131</sup>I/<sup>137</sup>Cs activity ratios to investigate the contributions of each source of <sup>137</sup>Cs (Tsumune et al., 2012) and compared simulated results and measured activities. The fact that simulated <sup>137</sup>Cs activities attributable to direct release were in good agreement with measurements suggests that the estimated direct release rates were reasonable. Employment of JCOPE2 instead of HYCOM for nudging improved both the offshore transport result and the reproducibility of <sup>137</sup>Cs activities 30 km offshore. Simulated <sup>137</sup>Cs activities attributable to atmospheric deposition were underestimated relative to observations. The rate of atmospheric deposition onto the ocean was underestimated compared to measurements because of a lack of measurements of deposition itself when atmospheric deposition rates were estimated. Measured <sup>137</sup>Cs activities attributable to atmospheric deposition helped to improve the ability of simulated atmospheric deposition rates to reproduce observations. Simulated <sup>137</sup>Cs activities attributable to inflow of <sup>137</sup>Cs deposited onto the ocean outside the domain of the model were in good agreement with measurements in the open ocean in the model domain after June 2012.

Although the contribution of inflow increased with time and was dominant by the end of February 2012, the activity associated with directly released <sup>137</sup>Cs decreased exponentially with time and was present only in the coastal zone by the end of February 2012.

Keywords: Fukushima Daiichi Nuclear Power Plant, Accident, Regional Ocean Model, Radiocaesium, Release amount, Ocean dilution

## Horizontal distribution of radiocaesium in the northwestern North Pacific after the accident of TEPCO's Fukushima NPS

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The accident of TEPCO's Fukushima Dai-ich Nuclear Power Station (Fukushima NPS) was caused by the 2011 off the Pacific coast of Tohoku Earthquake and tsunami on March 11, 2011 and large amount of radioactive material were discharged by the hydrogen explosion and leaked from Fukushima NPSs to the ocean. To investigate the transport of radionuclides to the ocean and their dispersion, we conducted two cruises with the R/V Mirai in the western North Pacific about one and three months after this accident and measured caesium (Cs-134 and Cs-137) in seawater. During two cruises (from 14 April to 5 May and from 27 June to 3 August, 2011), 20-L seawater in the surface and shallow layer (< 200 m) were collected along the cruise track with an underway surface pump and a multiple water-sampling system, analysed by the ammonium phosphomolybdate/Cs compound method and measured by low back ground gamma spectrometry using Ge detectors.

Cs-137 in surface seawater ranged from < 0.002 to 0.284 and <0.001 to 0.561 Bq/kg during April and May and during June and August, respectively. The radiocaesium could not be detected in seawater of the Kuroshio extension current. Samples from stations off Fukushima and off Miyagi had higher Cs-137 (0.11 ~ 0.28 Bq/kg) than those at other stations in April, however, the highest Cs-137 was observed at JKEO (38.5N, 148.25E) and Cs-134 could be detected in the 200 m depth at JKEO and off Fukushima in June. At S1 (30N, 145E), is located in the south of the Kuroshio current, Cs-134 in the 200 m depth was observed in April, that was not detected in June. On the other hand, at K2 (47N, 160E), the activities of Cs-134 and Cs-137 were almost 0.01 Bq kg/kg in the surface seawater, and those of Cs-134 were less than the limit of detection (< 0.002 Bq/L) at the depth of 200 m in both April and June. The obvious variation of these activities were not observed around the observed northern east area. These results suggest that the dispersion of radiocaesium in the marginal area were effected of the atmospheric diffusion and the leakage of contaminated water, and that in open sea were effected of only the atmospheric diffusion after a half year from the accident.

Keywords: radiocaesium, seawater, North Pacific, The accident of TEPCO's Fukushima NPS

## Surface pathway and subduction of Fukushima radioactivity in the North Pacific Ocean in March 2011 - June 2012

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<sup>134</sup>Cs and <sup>137</sup>Cs were released to the North Pacific Ocean by two major likely pathways, direct discharge from the Fukushima NPP1 accident site and atmospheric deposition. High density observations of <sup>134</sup>Cs and <sup>137</sup>Cs in the surface water were carried out at 322 stations by 17 cruises of cargo ships and several research vessel cruises since March 2011 till March 2012. Main body of radioactive surface plume of which activity was exceed 10 Bq m<sup>-3</sup> had been travelling along 40 oN, and reached International Date Line on March 2012. A feature was that the radioactive plume was confined along 40 oN when the plume reached International Date Line. Although activities of <sup>134</sup>Cs and <sup>137</sup>Cs in the surface water decreased rapidly, a maximum of activities of <sup>134</sup>Cs and <sup>137</sup>Cs in seawater at about 300 meters depth was observed at 40 oN 165 oE in June 2012. We can say that the observed maximum of <sup>134</sup>Cs and <sup>137</sup>Cs was formed by subduction in winter 2011/2012 because a density of Central Mode Water of 26.3 kg m<sup>-3</sup> corresponded to about 300 meters depth in this region. Water column inventory of <sup>134</sup>Cs from surface to 1000 meters depth was 2710 +/- 210 Bq m<sup>-2</sup> in October 2011 at 40 oN 165 oE and 80 % of water column inventory of <sup>134</sup>Cs existed shallower than 200 meters, however only 20 % of water column inventory of <sup>134</sup>Cs was shallower than 200 meters while 80% was deeper than 200 meters in June 2012 due to subduction in winter 2011/2012.

Keywords: Fukushima, radioactivity, plume, subduction, radiocaesium, subduction

## Possible southward transport of the directly-discharged Fukushima-derived radiocesium across the Kuroshio Extension

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The massive Tohoku earthquake and consequent giant tsunami of March 11, 2011 resulted in the global release of radiocesium (Cs-134 and Cs-137) in the environment from the Fukushima Dai-ichi nuclear power plants (FNPPs). In the North Pacific Ocean, a large portion of the Fukushima-derived radiocesium has been settled both through atmospheric deposition and direct discharge. The direct discharge of radionuclide-contaminated waters brought a serious contamination of radiocesium (about ten million times higher than the background level) in coastal seawaters near the FNPPs just after the accident. Some observations in open oceans clarified that radiocesium activity in surface water of the North Pacific became more than ten times higher than that before the accident due to the atmospheric deposition. About two years after the accident, the radiocesium activity in seawaters decreased remarkably while that in sediment samples on coastal seafloors is still high. Therefore it is necessary to address risks to marine ecosystem and public health for a long time. Meanwhile evaluations of the total amount and behavior of the Fukushima-derived radiocesium in the vast North Pacific Ocean are essential for an estimation of the total amount of the released radiocesium and a prediction of spreading process of the radiocesium in the future, respectively. We present here the Fukushima-derived radiocesium in seawaters at stations in the northwestern North Pacific Ocean hundreds km away from FNPPs in January and February 2012. Surface and deeper samples (0 - 800 m) were collected into 20-L cubitainers using a bucket and a conductivity-temperature-depth rosette with water samplers. The samples were filtrated and acidified by nitric acid on board. Radiocesium in the seawater sample was concentrated onto ammonium molybdophosphate (AMP). Radiocesium, cesium-134 (half-life 2.07 years) and -137 (half-life 30.04 years), in the AMP/Cs compound was measured using a gamma-spectrometry with well-type Ge detectors. Cesium-134 was observed in surface waters from all the stations between 20°N and 42°N about one year after the disaster (0.2 - 18 Bq/m<sup>3</sup>). This suggests that the Fukushima-derived radiocesium has been settled at all the stations because cesium-134 activity in the North Pacific before the accident was below detection limit. The cesium-134 activity in surface waters of the subarctic (north of 39°N approx.) and subtropical (south of 35°N approx.) areas were less than 4 and 1 Bq/m<sup>3</sup>, respectively. Relative high activities of cesium-134 (8 - 18 Bq/m<sup>3</sup>) were found in the transition area between the subarctic and subtropical areas, which is due to an eastward transport of the direct-discharged radiocesium from FNPPs along the North Pacific Current. Cesium-134 activities in the winter mixed layer from surface to 150 or 200m depth approx. were constant and these below the mixed layer were not detected. At a station located just south of the Kuroshio Extension, which is boundary between the transition and subtropical areas, the activity in the mixed layer was less than 1 Bq/m<sup>3</sup> while there was a cesium-134 maximum (5 - 9 Bq/m<sup>3</sup>) just below the mixed layer (200m and 300m depths). Water density (sigma-theta) of the maximum ranged from 25.2 to 25.4, which corresponds to densities of surface waters in the transition area in winter. Thus the subsurface cesium-134 maximum at the station in the subtropical area probably originated from the direct-discharged radiocesium in the transition area. Furthermore in deeper layers (400m and 600m depths) at the station low activities of cesium-134 were detected significantly. These results suggest a southward transport of the directly-discharged radiocesium from the transition to the subtropical areas along isopycnal layers across the Kuroshio Extension during the one year after the accident. In our presentation results from other stations, including results of cesium-137, will be discussed.

Keywords: Fukushima Dai-ichi nuclear power plants, radiocesium, North Pacific

## Transport of Fukushima-derived radiocaesium to the ocean interior by sinking particle

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The 2011 Tohoku-Oki Earthquake, occurred on 11 March 2011, and the tsunami it caused seriously damaged the Fukushima Daiichi Nuclear Power Plant (FNPP1). Large quantities of artificial radionuclides were emitted from FNPP1. At two stations in the western North Pacific, K2 in the subarctic gyre and S1 in the subtropical gyre, time-series sediment traps were collecting sinking particles when the FNPP1 accident occurred. Radiocesium (<sup>134</sup>Cs and <sup>137</sup>Cs) derived from FNPP1 accident was detected in sinking particles collected at 500 m by late March 2011 and at 4810 m by early April 2011 at both stations. The sinking velocity of <sup>134</sup>Cs and <sup>137</sup>Cs was estimated to be from 8 to 36 m/day between the surface and 500 m and >180 m/day between 500 m and 4810 m. <sup>137</sup>Cs specific activity varied from 0.14 to 0.25 Bq/g dry weight. These values are higher than those of surface seawater, suspended particles, and zooplankton collected in April 2011. The total <sup>137</sup>Cs flux by late June at K2 and by late July at S1 was from 0.5 to 1.7 Bq/m<sup>2</sup> at both depths. Compared with <sup>137</sup>Cs input to both stations by April 2011, estimated from the surface <sup>137</sup>Cs activity and mixed layer depth and by assuming that the observed <sup>137</sup>Cs flux was constant throughout the year, the estimated removal rate of <sup>137</sup>Cs from the upper layer (residence time in the upper layer) was from 0.3 to 1.5%/year (68 to 312 years). The estimated removal rates and residence times are comparable to previously reported values. Based on preliminary results of <sup>134</sup>Cs analysis on sinking particle collected at 4810 m of K2 between August 2011 and June 2012, the maximum <sup>134</sup>Cs flux and concentration were observed between May and Jun 2011 and decreased gradually thereafter. However <sup>134</sup>Cs was still detected and the ratio of <sup>134</sup>Cs to <sup>137</sup>Cs was close to one in sinking particle collected in April 2012. Total <sup>134</sup>Cs flux at 4810 m of K2 by April 2012, at about one year after FNPP1 accident, was estimated to be higher than 2.5 Bq/m<sup>2</sup>. Assuming that the <sup>134</sup>Cs inventory (atmospheric <sup>134</sup>Cs input) at K2 was 450 Bq/m<sup>2</sup>, removal rate of <sup>134</sup>Cs from the upper layer (residence time in the upper layer) was > 0.6%/year (< 180 years) and comparable to previous estimate. At 4810 m of S1, highest <sup>134</sup>Cs specific activity was found in December 2012 and about half a year later than that at 4810 m of K2. Although flux collected at 4810 m of S1 were not always sufficient for analysis, the <sup>134</sup>Cs was detected by early February 2012. In June-July 2012, seafloor sediments at K2 and S1 were collected. Analysis of radiocesium in the seafloor sediment and sinking particle collected by sediment trap is still ongoing. During 2013 JPGU meeting, Fukushima derived radiocesium flux in sinking particle and inventory of radiocesium in the seafloor sediment will be discussed.

Keywords: Fukushima Daiichi Nuclear Power Plant, Artificial radiocaesium, Western North Pacific, Sinking particle, Sediment trap, 2011 Tohoku-Oki Earthquake

## Horizontal distribution of Fukushima-derived radiocesium in zooplankton in the north-western Pacific Ocean

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The magnitude of the 9.0 Tohoku earthquake and the ensuing tsunami on March 11, 2011, inflicted heavy damage on the Fukushima Dai-ichi nuclear power plant (FDNPP). Fission products were emitted, falling over a broad range in the northern hemisphere, and water contaminated with radionuclides leaked into the ocean. In this study, we described the horizontal distribution of the Fukushima-derived radiocesium in zooplankton and in surface seawater in the western North Pacific Ocean (500-2100 km from the FDNPP) 10 months after the accident. <sup>134</sup>Cs and <sup>137</sup>Cs were detected in all zooplankton and seawater samples. Because of its short half-lives, <sup>134</sup>Cs detected in our samples could only be derived from the FDNPP accident. The radiocesium concentrations in zooplankton were high at around 25N while those in surface seawater were high at around the transition area between the Kuroshio and the Oyashio Currents (36-40N). We analyzed the structure of the zooplankton communities but could not find out evidence which biological factors (taxa composition and relative biomass of carnivores) influenced <sup>137</sup>Cs concentration in bulk zooplankton. Zooplankton communities contaminated with the Fukushima-derived radiocesium included many kinds of diel vertical migratory species. These migrants were exposed to higher concentrations of radiocesium while feeding at night in the ocean surface; however, exposure to contamination was lower as these species swam in the deeper mesopelagic layer during the day. In the subtropical region, 20-60% of krill and 3-36% of copepods on abundance basis were diel vertical migrants. On the other hand, 80-100% of krill and 77-88% of copepods were the migrants in the transition and the subarctic regions. Accumulated radiocesium in the migrant bodies are transported and may be taken into the mesopelagic food web.

Keywords: North Pacific ocean, zooplankton, FDNPP accident, <sup>134</sup>Cs, <sup>137</sup>Cs

## Radiocesium stored in bottom sediments after the nuclear power plant accident due to the M9 earthquake

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Eastern Japan along the Pacific coast has been damaged seriously and is still on the way of recovery after the nuclear power plant accident in Fukushima due to the magnitude-9 earthquake on March 11, 2011. The radiocesium concentration went down greatly by the summer of 2011 in sea water, while it still keeps a high level in the bottom sediments. In particular off the coast from Miyagi to Ibaragi, some spots with high concentration have been found in the region shallower than 200-m depth. The dedicated members of the Oceanographic Society of Japan have been making estimations and discussion to find which processes are responsible for the high concentration. A symposium is held in March with widely opened discussion. We have so far reached the tentative conclusion that any process could be a possible one for the present condition among absorption/adsorption by plankton, detritus and disturbed sediments, direct adsorption of seawater cesium and inflow of suspended solids from rivers. The further collection and analyses of samples are required to confirm the actual processes, and therefore, the monitoring plan is proposed with effective analytical methods.

Keywords: radionuclide, earthquake, sediments



## Regional-scale transfer of fallout radiocaesium in river networks impacted by the Fukushima Daiichi Nuclear Power Plant

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The Fukushima Daiichi Nuclear Power Plant accident which followed the magnitude 9.0 earthquake and tsunami on 11 March 2011 resulted in the release of Cs-134 and Cs-137 into the surrounding environment, where highly elevated levels are reported. As a result, there is considerable concern about the redistribution of these radioactive contaminants from the atmosphere to vegetation, soil and aquatic systems. This study reports on the magnitude of fluvial transfer of Cs-134 and Cs-137 through river networks located across the fallout region. Initially six nested river monitoring stations were established within the Abukuma River basin from June 2011. Subsequently, an additional 24 stations were established between October and December 2012, which included a further 12 stations within the Abukuma basin and another 12 sites monitoring the smaller coastal catchments north and south of the power plant. Combined, these 30 sites provide a regional-scale measure of fallout radiocaesium transfer by river networks. The catchment areas range from 8 to 5,172 km<sup>2</sup> and span a large range in average catchment inventories of Cs-134 (16-2030 kBq/m<sup>2</sup>) and Cs-137 (19-2380 kBq/m<sup>2</sup>) based on MEXT inventory mapping. Flow and turbidity (converted to suspended sediment concentration) were measured at each station while bulk suspended sediment samples were collected at regular intervals using time-integrated samplers to allow measurement of Cs-134 and Cs-137 activity concentrations by gamma spectrometry. Preliminary monitoring data will be presented that shows highly elevated but generally declining activity concentrations of Cs-134 and Cs-137 on suspended sediment and provides a measure of the regional-scale variability in fluvial radiocaesium fluxes. While most radiocaesium is likely to remain in the soil profile, fluvial redistribution may contaminate downstream floodplains that were subject to low fallout and deliver significant quantities of highly contaminated fine sediment to the coastal zone.

Keywords: Fukushima, Radiocaesium, Sediment, Rivers, Contamination



## Observation of flux and outflow-property of radioactive cesium in paddy field; A case study in Kawamata city, Fukushima.

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Dynamics of radioactive cesium derived from Fukushima Dai-ichi NPP accident on land have big impacts on land utilization and migration to water system. There are many paddy fields in Fukushima prefecture. However, the distribution, effect of decontamination and dynamics of radioactive cesium in paddy field have been poorly understood. The outflow of soil attached by radioactive cesium from paddy field can be affected by many factors such as water management, irrigation/non-irrigation periods and inventory of radioactive cesium. For the dynamics of radioactive cesium, input of radioactive cesium through water intake is also one of important processes. To better understand the dynamics of radioactive cesium in paddy field, examination plots were established at planned evacuation zone in Kawamata city, Fukushima prefecture, and the input and output of radioactive cesium were studied through irrigation/non-irrigation period. In addition, the relation between concentration of radioactive cesium in suspended soil obtained in paddy field and the inventory of radioactive cesium was investigated.

Two plots were established. The one was cultivated normally (normal plot, 510 m<sup>2</sup>), and the other was decontaminated by scraping of surface soil with 5-10 cm thickness before cultivation (decontaminate plot, 731 m<sup>2</sup>). To measure the initial concentration and inventory of radioactive cesium, soil core (14 cm depth) was collected from twelve points from each plot, and the core was sliced into 2 cm thickness. The flow volume and suspended soil concentrations in influent/effluent water were monitored by parshall flume/water-level gauge and turbidity probe, respectively. Suspended soil in the water was trapped by time-integrated suspended soil sampler and collected every one or two weeks. The soil core and suspended soil were dried (105 degrees Celsius, 24 hours) and disaggregated by grinding. The activity of radioactive cesium was measured by germanium semiconductor detector. Particle size distribution was measured by laser diffraction particle size analyzer.

The outflows of soil and radioactive cesium showed statistically significant correlations with rainfall in both irrigation/non-irrigation periods, although clear relation was not observed for all data through the two periods. These results indicate that the amount of radioactive cesium migrated from paddy field can be estimated from rainfall by dividing the period into irrigation/non-irrigation periods. Difference in the slopes of the relations indicated that the outflow of radioactive cesium due to rainfall easily occurred during non-irrigation period. The output of radioactive cesium was higher than their input. Although more investigations were necessary concerning on inventory of radioactive cesium at catchment of irrigation water and water management in paddy field, the effective half-life of radioactive cesium in paddy fields can be shorter than the decay half-life. The concentration of radioactivity of cesium in suspended soil showed significant correlation with inventory, indicating that the concentration of radioactivity of cesium in runoff soil is possible to be estimated from their inventory.

Keywords: Fukushima prefecture, Paddy field, Radioactive cesium, Soil, Flux, Inventory

## Characteristics of radioactive Cs in the sediment from the storage reservoir in Iitate village, Fukushima prefecture

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The storage reservoir sediment contaminated by radioactive Cs, obtained from Hiso district, Iitate village, Fukushima prefecture Japan, were characterized in detail. The 30 cm sediment core from the reservoir were cut into 1 cm on site. The water contents, radioactive Cs concentrations, mineral composition and densities from each sample were measured. The clay fraction was obtained from the parts of the sediments which contain high radioactive Cs concentrations. The clay mineralogy was analyzed by X-ray diffraction of the chemical treated oriented specimen. The desorption experiments of radioactive Cs from the clay fraction were conducted by washing with high Mg concentrations solutions. The radioactive Cs concentrations before and after the experiments were measured.

The radioactive Cs was strongly sorbed by clay minerals containing vermiculite and mica and hardly desorption in the conditions of natural water. The transport of the radioactive Cs in the surface condition must be accompanied with the movements of the clay minerals. The high accumulation of Cs-137 was observed at top 9cm layer in the sediment. The Cs-137 concentration steeply decrease with depth after 9 cm depth. At the surface 9cm layer, the 137Cs concentrations possessed periodicity and were inversely correlated with the densities. In addition, the 137Cs concentrations has inverse correlation with the records of the precipitation in the area. These behavior indicate that the contaminant sediments come in the storage reservoir from the catchment area by the erosion accompanied with precipitation. Therefore, the storage reservoirs play a role for the sink of the radioactive Cs in the area.

Keywords: radioactive Cs, Fukushima Daiichi Nuclear Power Plant, clay minerals, storage reservoir sediment

## Effects of rain events on transport of radiocesium in the Abukuma River during 2011-2012

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About 15 PBq of both Cs-134 and Cs-137 was released from the Fukushima Daiichi Nuclear Power Plant (NPP) after the 2011 Tohoku earthquake and tsunami. Surface deposition pattern of Cs-134 and Cs-137 occurred at Fukushima, Tochigi and Gunma Prefecture by combination with wind direction and precipitation. Therefore, it is important to elucidate the short-term to long-term impacts of the Fukushima Daiichi NPP accident on ecosystems of river watershed environments. This study was conducted to investigate the effects of rain events on transport of Cs-134 and Cs-137 in the Abukuma River running through Fukushima and Miyagi Prefecture in Japan, 15 months after the Fukushima Daiichi NPP accident.

Field experiments were carried out at Shirakawa (upper), Motomiya, Date (middle) and Iwanuma (lower) in the Abukuma River during June 19-21, 2012. Typhoon Guchol struck Japan on June 20. Fukushima Prefecture had rainfall of 77-136 mm during June 19-21. The suspended particles were separated using centrifugation and filtration with No. 5A filters and a pore size of 450 nm membrane filters. The radioactivity of Cs-134 and Cs-137 in the filtered river waters was measured as dissolved forms of radiocesium with gamma-ray spectrometry using ammonium molybdophosphate (AMP)/Cs compound method. The Cs-134 and Cs-137 were measured using gamma-ray spectrometry with a low BKG Ge detector for 1-3 days. The suspended solids were also measured using gamma-ray spectrometry after drying them at room temperature.

Total radioactivity of Cs-134 and Cs-137 in river waters was 0.016-0.27 Bq/l at normal flow conditions on April 18 and June 19 in 2012, but it increased to 3.83 Bq/l in high flow conditions by heavy rains occurring with the typhoon. The particulate fractions of Cs-134 and Cs-137 were 77-89% at the normal flow condition, but were close to 100% after the typhoon. The variations of radiocesium concentration may correlate with the water level at each site. These results indicate that the pulse input of radiocesium associated with suspended particles from land to coastal ocean occurred by the heavy rain event.

Keywords: river water, radiocesium, dissolved forms, particulate forms, migration behavior, raine events

## The behaviour of radio-Cs in the Abukuma riverine system emitted from the FDNPP

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As a consequence of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, a huge amount of radio-Cs was discharged into the environment. Especially for the Abukuma riverine system, which is the largest river in the Tohoku-area and can be a dominant water resource of this region, the watershed of this river has been seriously contaminated. To observe the fate/behaviour of radio-Cs in this region, we analysed Cs-137 in aerosol, soil, river suspended solid and sediments and water samples. In addition to the analyses of Cs-137 in natural samples, the adsorption/desorption experiments were conducted for analogous to natural system.

From the observations of riverine samples in the period from June/2011 to Dec./2012, it was found that Cs-137 dominantly existed in particle fraction rather than dissolved fraction. This is the opposite result of the situation of Ukraine in one to two years after the Chernobyl accident, and might be due to the differences of the content of organic matter in soil between Japan and Ukraine. This possibility is supported by our results of adsorption/desorption experiments of Cs and humic-acid on clay minerals. The concentration of Cs-137 in particulate fraction (Bq/L) in river water was decreased with date. However, the concentration in a suspended solid (Bq/g) was not decreased. So, it can be said the decrease of the concentration for Cs-137 (Bq/L) is apparent one rather than decontamination of river water. The concentration of dissolved Cs-137 (Bq/L), in Dec./2012, is one to two order of magnitude smaller than that in June/2011. This result suggested that the more soluble fraction was leached immediately after the accident, and a small amount of Cs-137 has been leached from soil steadily. Actually, we recognise the existence of Cs with some chemical species from the results of long term leaching-experiment of surface soil and aerosol. The difference of chemical species can show the specific leaching/desorption behaviour of Cs in the environment.

Using our results for riverine system and flow rate of Abukuma river, the amount of drained off Cs-137 from the river to the ocean was estimated as 13 TBq in the last two years. By the result of long term leaching-experiment of surface soil with seawater, it is estimated that about 20% of discharged Cs-137 to the ocean would be leached in the sea water.

Keywords: FDNPP, Cs-137, Abukuma Riverine system

## Transfer of fallout radiocesium in various terrestrial environment in Yamakiya district, Kawamata Town

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Radioactive contamination has been detected in Fukushima due to the nuclear accident at Fukushima Daiichi Nuclear Power Plant (NPP) following the earthquake and tsunami on 11 March 2011.

Following comprehensive investigation (FMWSE project; <http://fmwse.suiri.tsukuba.ac.jp/>) was conducted to confirm migration of radionuclides through natural environment including soils and rivers funded by MEXT, Japan. Experimental catchments have been established in Yamakiya district, Kawamata Town, Fukushima prefecture, located about 35 km from Fukushima power plant, and designated as the evacuated zone. Approximate Cs-137 fallout in this area is 200-1000k Bq/m<sup>2</sup>.

The main finding is as follows:

- 1) Migration of radionuclides to soil water, stream water and ground water was confirmed low at present. On the other hand, concentration of radiocaesium was found approximately 50 kBq/kg in the suspended sediments flowing down the river.
- 2) Amount of sediments deposited in the tank placed at the end of downstream within the USLE plot was confirmed together with the concentrations of radiocaesium.
- 3) In forests, distribution of radiocaesium was able to be confirmed to a certain extent by placing towers in the Japanese cedar forest and broad-leaved forest. To date, since a large amount of radiocaesium is considered to be found in coniferous tree canopies, these data are expected to be applied to future decontamination. Moreover, further investigation is necessary on the chemical state of radiocaesium which falls down to the forest floor as throughfall and on the actual condition of downward migration from forest floor to deeper soil layers.

Keywords: Radionuclides, Transfer, Soil Erosion, Forest, Cs-137

## Transfer of the Fukushima reactor accident-derived radionuclides in forest environments

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The Fukushima Daiichi nuclear power plant accident resulted in extensive radioactive contamination of the forest environment in Fukushima and the neighboring prefectures. In this study, we analyzed radiocesium concentrations in rainwater, throughfall, stemflow, and litterfall to characterize the transfer of the deposited radiocesium in various forest stands (evergreen conifers and broad-leaved forests), in Tochigi (Cs-137 fallout < 10 kBq/m<sup>2</sup>) and Fukushima (Cs-137 fallout = 300-600 kBq/m<sup>2</sup>) prefectures. Furthermore, in-situ measurement of radiocesium were conducted to delineate spatio-temporal variability of radiocesium in the canopy and forest floor. The result of this study demonstrated that a large proportion of radionuclides which deposited on forest were initially trapped by canopies, and subsequently transferred to forest floor in association with throughfall, stemflow, and litterfall. In the deciduous broad-leaved forest, the highest radioactivity was found at the forest floor; however, 25-40% of the total deposited radiocesium remained in the canopy of evergreen coniferous forests one year after the reactor accident.

Keywords: Fukushima Daiichi Nuclear Power Plant, Radiocesium, Forest environment, Canopy interception, Transfer

## Investigation of an environmental fate of radiocaesium in the Fukushima forests

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In this paper, we introduce an outline of the research plan of the "Long-term assessment of Transport of radioactive contaminant in the environment of Fukushima (Fukushima-Trace Project)" in the Fukushima Environmental Safety Center, Japan Atomic Energy Agency, and present a preliminary results of an investigation of an environmental fate of radiocaesium in the Fukushima forests, by an application of systems analysis.

Keywords: radiocaesium, environmental fate, Fukushima, forest, systems analysis



## Dynamic of radiocesium from conifer needles to male flowers of sugi

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A large amount of radioactive substances were released into air through the severe accident of Fukushima Daiichi Nuclear Power Plant (NPP) in March 2011. They deposited on forest environments and absorbed and transported in trees, so they were detected in leaves developed even after 2012, when the release of radioactive substances from Fukushima Daiichi NPP should be stopped. In the spring of 2012, radioactive substances including radiocesium were detected in pollen of sugi (*Cryptomeria japonica*) in Fukushima. Since a large quantity of sugi pollen dispersing in spring has been recorded in Japan, it is considered that the secondary radioactive dispersion might be occurred through pollen. In order to estimate the secondary dispersion of radiocesium from sugi forests, it is important to understand the transfer of radiocesium from needle leaves to pollen through male flowers. We measured the radiocesium concentrations in sugi needle leaves, male flowers, and pollen, and then evaluated the transport characteristics.

Sugi needles with male flowers were collected from 114 sites in Tohoku, Kanto and Koshinetsu districts from November to December 2012. Three samples (one sample from one individual) were collected at one site. Male flowers were collected from the samples and the needles were separated into three different parts corresponding to each elongation year; 2012, 2011, and before 2010. They were washed with tap water and rinsed with distilled water, and then oven-dried at 80°C for 48 hours. They were enclosed into U-type containers without pulverization for male flowers and with pulverization for needles. By gamma ray spectrometry using High purity germanium (HPGe) radiation detector, cesium 137 and 134 (Cs-137, 134) were detected, and the radioactivity per dry weight (Bq/kg) were calculated. Gamma ray spectrometry of the pollen samples were conducted by using a well type of HPGe radiation detector. The distribution of radioactive substances in needles and male flowers were detected by an autoradiography method of Imaging Plate (IP).

Needles at the part of before 2010 resulted in the highest concentrations of Cs-137, 134 compared with other parts. Autoradiograph images showed spotted distribution of high radioactivity. Therefore, it is considered that the radioactive fallouts from Fukushima Daiichi NPP accident attached and remained on sugi needles without washout. Male flowers showed higher radiocesium concentrations than needles elongated in 2012, and the top of needles, which sustained male flowers, also contained higher concentrations of radioactivity than the other part of needles elongated in 2012. These results suggested that radiocesium is easy to transport to the needle top including male flowers.

Keywords: Fukushima Daiichi Nuclear Power Plant accident, sugi, needle, male flower, Cs-137, Cs-134

## Migration dynamics of $^{137}\text{Cs}$ deposited on the forested ecosystem in Fukushima after the nuclear power plant accident

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A massive amount of radioactive substances, including cesium-137 ( $^{137}\text{Cs}$ ), emitted from the disabled nuclear power plant, has been deposited on the forested areas in the northeastern region of Honshu Island, Japan after the Fukushima Daiichi nuclear power plant accident. Forest ecosystems in these regions are particularly important, not only for the forest products industry but also for source areas of drinking water and for residential environments. To clarify the migrating mechanisms of  $^{137}\text{Cs}$  deposited on the forested ecosystem, we initiated intensive field observations in a small catchment that included forest headwaters and farmlands in the northern part of Fukushima Prefecture. The following expected major pathways of  $^{137}\text{Cs}$  export and diffusion were investigated: 1) transportation of dissolved and particulate or colloidal forms via hydrological processes within a forested catchment and export dynamics through the stream, and 2) diffusion through the food web in terrestrial and aquatic organisms of forests. Preliminary findings indicated the following: 1) Most of the  $^{137}\text{Cs}$  was discharged as suspended matter. High water flow generated by storm acted to accelerate the transportation of  $^{137}\text{Cs}$  from the forested catchments. Thus, the estimation of  $^{137}\text{Cs}$  export requires precise evaluation of the high flow acceleration during storm events; 2) Because litter and its detritus may form the biggest pool of  $^{137}\text{Cs}$  in the forested ecosystem,  $^{137}\text{Cs}$  diffusion occurs more rapidly through the detritus food chain than the grazing food chain. Most predators have already ingested  $^{137}\text{Cs}$ , particularly in aquatic environments. An urgent question that needs to be addressed is when and how  $^{137}\text{Cs}$  diffuses through grazing food chains and how rapidly this process occurs. To elucidate or to be able to predict these phenomena, the mechanisms of  $^{137}\text{Cs}$  release from litter and soil organic matter need to be clarified.

Keywords:  $^{137}\text{Cs}$  deposition, forested ecosystem, hydrological process, food web

## Atmospheric radioactive cesium in the broad-leaf and Japanese cedar forest and canopy

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Radionuclides emitted in the Fukushima dai-ichi nuclear power plant (FNDPP) accident have been deposited on the soil, ocean and vegetation. Especially, a significant amount of them has been attached on Japanese cedar trees because they are dominant evergreen trees which extend leaves in Fukushima mountainous region when the accident occurred. Re-suspension of radioactive cesium from these trees to the atmosphere may be one of significant path in the diffusion of radionuclides after the accident.

We have measured atmospheric concentration of radiation by Cs-134/137 in two forest sites: broad-leaf tree (BT) site and young Japanese cedar (JC) site at Kawamata-town near Fukushima to study re-suspension of radioactive Cs in the forest. Atmospheric suspended particle are collected with high/low-volume air samplers mounted at the forest floor and canopy, respectively, and gamma-ray emission from them were measured with Ge detector.

The measured concentration of atmospheric Cs-134/137 was about 2-times larger at the forest floor than that at the canopy both in BT and JC sites, indicating that particles including Cs-134/137 are produced or emitted in the forest. The ratio of atmospheric Cs-134/137 radiation concentrations to surface density of Cs-134/137 is significantly higher in JC site than that in BT site. Processes of re-suspension of radioactive cesium in these forests will be discussed.

Keywords: Fukushima daiichi nuclear plant accident, environmental radioactivity, re-suspension

## Predicted spatio-temporal dynamics of radiocesium deposited onto forests following the Fukushima nuclear accident

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Radiocesium (<sup>134</sup>Cs and <sup>137</sup>Cs) released from the Fukushima Dai-ichi nuclear power plant to the atmosphere contaminated a large area of Japan's land surface, the majority of which is covered by forest (Hashimoto et al. 2012b). The availability of countermeasure options for forest areas is limited to restriction of access and removal of contaminated materials (Hashimoto et al. 2012a). Even considering only the most heavily contaminated forests ( $\geq$  <sup>134</sup>, <sup>137</sup>Cs 1000 kBq m<sup>-2</sup> in 2011), however, the total volume of radioactively contaminated materials is estimated to be 33 million cubic meters and 21 Tg (dry matter) (Hashimoto et al. 2012b). To develop effective countermeasures to mitigate the impacts of radioactive contamination of forests, detailed monitoring of the radiocesium migration and re-distribution is essential. In addition, it is useful to be able to predict the future dynamics of the radiocesium between forest components; modelling is the best tool for this prediction. Here we simulated the dynamics of radiocesium deposited on Japanese forest ecosystems in 2011 using a model that was developed for tracking radionuclides in forest ecosystems after the Chernobyl accident in 1986 (RIFE1 model). The fate of the radiocesium was simulated using the initial conditions observed following the Fukushima accident. In addition, the simulation results were incorporated with a spatial distribution map of deposited radionuclides that was based on an air-borne survey. The simulation demonstrated that in the first two years after initial deposition radiocesium is retained primarily in the soil surface organic layer. Over a period of five to ten years radiocesium is predicted to move from the surface organic soil to the mineral soil, which will eventually become the largest reservoir of radiocesium within forest ecosystems. Spatial analysis clearly shows the reduction in the extent of contaminated areas which will occur as a result of natural decay of radiocesium, as well as the spatial distribution of radiocesium in each forest component. Considering the heavier rainfall and warmer conditions in Japan than in the countries contaminated by the Chernobyl accident, migration of radiocesium from organic to mineral soil may be faster than predicted. Although the uncertainty of our simulations should be taken into account, they provide a basis for understanding and anticipating the future dynamics of radiocesium in Japanese forests following the Fukushima accident.

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Keywords: forest, radiocesium, decontamination, model, regional prediction, soil

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