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MAG34-01

Room:IC

Time:May 23 15:30-15:45

# Radioactive strontium from the Fukushima Nuclear Power Plant accident observed at Tsukuba, Ibaraki, Japan

IGARASHI, Yasuhito<sup>1\*</sup>, Wakari Iwai<sup>1</sup>, Hiroshi Sakou<sup>2</sup>, Kazuma Nabeshima<sup>1</sup>, KAJINO, Mizuo<sup>1</sup>

#### Introduction

After the Chernobyl accident in 1986, the significant nuclear accident did not occur. Because of this concentration levels of the anthropogenic radionuclide in the atmosphere had been at the very low level for these two decades. However, the Fukushima First Nuclear Power Plant of Tokyo Electric Power Company was damaged by the earthquake and tsunami on March 11, 2011, resulted in a serious nuclear disaster, newly added the radioactive material to the atmospheric environment corresponding to some portions of those from the Chernobyl accident. By this large-scale contamination, the concentration level of the anthropogenic radionuclides in the atmosphere over Japan was significantly enhanced. At the Meteorological Research Institute (MRI), Tsukuba, Ibaraki (ca. 260 km far from the accidental site), air sampling and analysis of the radionuclides were continued before and after the accident. Only the result of the gamma-ray emitting nuclides has been so-far reported. Since the analysis of radioactive Sr was started after the accident, this presentation publicizes its air concentration and the month-long total deposition.

#### Outline of the analytical method

By using the high volume aerosol sampler (HV-1000F, Sibata Scientific Technology Ltd.) installed in the MRI observation field in Tsukuba, the collection the aerosol was carried out with a quartz fiber filter (Advantec QR100). Although the usual collection time is one week, since the accident was announced, collection duration was shortened at 6 hour to one day. The flow rate of the sampler was set at 700 L/min, thus the collected volume of the air was about 250, 500, or 1000 cubic meters (corresponding collection time of 6, 12, and 24 hours, respectively). One piece of the filter sample (about 2%) cut by a punch before the following pellet creation was subjected to the radioactive Sr analysis. The radiochemical separation composed of precipitation purifications including a fuming-nitric acid method etc., and, finally Sr was fixed as carbonate salt. In order to see growth of <sup>90</sup>Y and decay of <sup>89</sup>Sr, beta activity was repeatedly measured over the long period of time with a low-background 2pai gas-flow detector. On the other hand, gamma-ray emitting nuclides were determined by using a Ge semiconductor detector, after compressing a filter sample into a pellet with a hydraulic pressing device.

Sampling of the monthly total deposition sample was carried out at the MRI, Tsukuba and Mt. Haruna, Guma, without stopping before and after the accident. The deposition sample was evaporated to dryness and the gamma-ray emitting nuclides were measured by a Ge detector. After the measurement by applying the same chemical procedures as those for the filter sample, radioactive Sr was determined.

#### **Results and discussion**

Temporal changes in air concentration of the gamma-ray emitting nuclides exhibited two high concentration events during March, 2011 in Tsukuba. This features the remarkable advection and diffusion phenomena to the Kanto plain of radioactive plume from the Fukushima nuclear accident. The filter samples which contained significant amount of radioactive Sr were also only samples taken around March 15 and March 21 as expected. Also the served amount of the sample was too small to detect radioactive Sr in other samples. Since <sup>89</sup>Sr with a half-life of 50 days could decay out, it was difficult to detect <sup>89</sup>Sr in the filter samples. The <sup>137</sup>Cs/<sup>90</sup>Sr activity ratio in the detection cases was about 1000, which agrees well with the figure from the emission inventory estimate by Nuclear and Industrial Safety Agency, Japan. Furthermore, between two radioactive plume advection events, there could be a difference in the discharged materials from the accident, which was turned out by a finding that the solubility to acid of radio-Cs on the filter contrasted.

Keywords: Radioactive strontium, Atmospheric samples, the Fukushima accident, Emission inventory

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MAG34-02

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# Physico-chemical characteristics of airborne radio cesium from the Fukushima accident

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Data on the size of radionuclides and their mixing state with other aerosol components have not yet been reported since the occurrence of Fukushima Dai-ichi nuclear power plant accident. The activity size distributions of  $^{134}$ Cs and  $^{137}$ Cs in aerosols collected 47 days after the accident have been measured at Tsukuba, Japan. We found that the activity of these radio cesium reside in the accumulation mode size range and overlapped with the mass size distribution of non-sea-salt sulfate aerosol. From the results, we can regard that sulfate is the main transport medium of these radionuclides, and re-suspended soil particles that attached radionuclides were not the major airborne radioactive substances at the time of measurement. The extraction experiment of radio cesium from the collected aerosol deposits on the filter media by use of water and HCl solution are also presented.

Keywords: radio cesium, size distribution, internal mixture, sulfate, aerosol

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MAG34-03

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# Estimation of released radioactive materials from Fukushima power plant by inverse model

MAKI, Takashi<sup>1\*</sup>, KAJINO, Mizuo<sup>1</sup>, TANAKA, Taichu<sup>1</sup>, Tsuyoshi Thomas Sekiyama<sup>1</sup>, Masaru Chiba<sup>1</sup>, IGARASHI, Yasuhito<sup>1</sup>, MIKAMI, Masao<sup>1</sup>

Huge amount of radioactive materials were emitted in the accident of the Fukushima nuclear power plant which occurred in March, 2011. Although various organizations released the prediction result using their transport models about this event, the most predictions assumed fixed emission amount and the results are not robust. We combined inverse technique (Maki et al., 2011), transport model results with tag-tracer and dose-of-radiation observation data to estimate more precise emission amount in this event.

We used regional chemical transport model named MRI-PM/r (Kajino et al., 2011). The horizontal resolution is 5km and spatial resolution of tag-tracer is 6 hours. We also used 3 tags in vertical (0-100m, 100-200m, 200-400m). The observation data are obtained from Ministry of Education, Culture, Sports, Science and Technology and the number of observation site is 49. The prior information about the emission is obtained by Chino et al., 2011. Now we are trying to use our global aerosol transport model named MASINGAR (Tanaka et al., 2003) to use globally distributed observation data (CTBT and so on).

We could obtain analysis results which are not largely different from observation and prior information by using inverse analysis. We find that the precision and resolution of the analysis depends on quality and quantity of the observation data. We also obtained higher maximum emission value than prior information.

By developing this system, it is expectable for construction of the near real-time forecasting system and the precise analysis system of the total emission amount at the time of the occurrence of an accident of a nuclear power plant etc. To achieve such objectives, it is important to develop more precise transport model including transport and deposition process, to collect as many observation data as possible and to obtain more robust prior information as possible.

Keywords: Inverse model, Fukushima Dai-ichi nuclear power plant, radioactive materials

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MAG34-04 Room:IC Time:May 24 09:00-09:15

# Numerical simulation of atmospheric transport of radionuclides in Meteorological Research Institute

TANAKA, Taichu $^{1*}$ , KAJINO, Mizuo $^{1}$ , MAKI, Takashi $^{1}$ , Tsuyoshi T. SEKIYAMA $^{1}$ , IGARASHI, Yasuhito $^{1}$ , Masaru CHIBA $^{1}$ , MIKAMI, Masao $^{1}$ 

Meteorological Research Institute have been conducted numerical simulation of atmospheric transport of radioactive materials with observation, such as the Chernobyl accident in 1986, 85Kr from nuclear fuel reprocessing plants, 222Rn and its progeny released from ground soil. We are currently conducting the radioactive materials released from the accident of Fukushima Daiichi nuclear power plant after the Tohoku great earthquake. To improve our understanding of the temporal and spatial distributions of transport and depositions quantitatively, we are developing both global and regional scale atmospheric transport models. For the global transport of radioactive materials, we used our global aerosol model named MASINGAR (Model of Aerosol Species IN the Global AtmospheRe). In the setting of the experiment, the horizontal grid is about 0.56° (TL319) and 40 vertical layers (from surface to 0.4hPa). The horizontal wind components are nudged using global analysis of Japan Meteorological Agency (GANAL). For emission flux, we used estimates by Chino et al. (2011) for <sup>137</sup>Cs and <sup>131</sup>I and estimates of Stohl et al. (2011) for <sup>133</sup>Xe. The model treats <sup>137</sup>Cs, <sup>133</sup>Xe, and <sup>131</sup>I as released radionuclides. The model include advective transport, eddy diffusion, convective transport, dry and wet depositions, and radioactive decay. For more detailed distribution and processes of radionuclides, we are developing our regional atmospheric transport model named MRI-PM/r (Passive-tracers Model for Radioactivity). The model incorporates advanced dry deposition parameterization, processes of the mixed-phase cloud microphysical dynamics, the activation of cloud condensation nuclei and ice nuclei, physical and chemical processes of radionuclides, and the interaction with the environment aerosol. This model classifies the aerosol particles into radionuclide primary (PRI), Aitken particle (ATK), particle accumulation mode (ACM), sea salt particles (SS), soil particles (DU), pollen (POL) by category method the aerosol, and considers the elementary processes that include condensation, evaporation, cohesion, activation of cloud condensation nuclei and ice nuclei, dissolution, collision (washout), cloud microphysical processes (conversion processes among following categories; rainout, cloud water, ice clouds, raindrops, snow, hail) and dry deposition. We will show the overview of the current status and challenges of our simulation studies.

Keywords: Numerical simulation, Atmospheric trace substances, Environmental radioactivity

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MAG34-05 Room:IC

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# Intensity of atmospheric radioactivity over East Japan after Fukushima nuclear plant accident

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Fukushima nuclear plant accident has released a huge amount of radioactive matters to the earth environment via the atmosphere and the ocean. Monitoring of atmospheric radioactive matters is significant to understand their emission to the atmosphere and their transport processes as well as estimate the internal exposure by the aspiration.

Several investigators belonging to Japanese Geoscience union, the Geochemistry society of Japan, the Japan Society of Nuclear and Radiochemical Sciences voluntarily took part in the activities for monitoring the radioactive matters released to the environment. Atmosphere team has monitoring atmospheric radioactive matters including Cs-134, Cs-137 and I-131, since late March 2011. The monitoring was conducted at more than 20 sites over Japan till May 2011, it was conducted at 11 sites surrounding the Fukushima nuclear plant till September 2011, and it is continued at 4 sites, Fukushima-city, Koriyama-city, Marumori-town and Hitachi-city, located at a distance of about 80 km from the nuclear plant. The atmospheric samples are obtained with a high-volume air sampler, and gamma-ray intensities from the samples are measured with a cooled Ge-detector.

The monitoring revealed that the distribution and variation of the atmospheric radioactivity intensity. It rapidly decreased in April and May, 2011, and it did not change significantly between June and August. It often showed large, 1-2 orders of magnitude, increase events. These variations can be understand the variation of emission amount of radioactive matters from the Fukushima nuclear plant, and their transport from there to the sampling sites. The atmospheric radioactivity intensity decreased when the radioactive matters from the Fukushima nuclear plant was not transported to the sampling site, but it remains significantly amount (base line level) higher than that before the accident. The baseline level is positively correlated with the amount of the fallen radioactive matter at the sampling sites, indicating that the re-emission of the radioactive matter from soil or trees contributed to the baseline radioactivity level.

Keywords: atmospheric radioactivity

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# Estimation of radionuclides' deposition over the land and ocean using a regional chemical transport model

TAKIGAWA, Masayuki<sup>1\*</sup>, WATANABE, Akira<sup>2</sup>

To reveal the transport and deposition processes of radionuclides that have been emitted from the Fukushima Daiichi nuclear power plant after the nuclear accident caused by the Tohoku earthquake and tsunami, we have conducted numerical simulation of radionuclides (131I and 137Cs) using a regional chemical transport model. Data assimilation using nudging for wind field and relative humidity was applied with the 10-minutes' interval ground-based observation, and the model-calculated meteorological fields with the ground-based observations in Fukushima prefecture showed that the model was capable to reproduce the change of wind direction following to the passage of low-pressure systems, and precipitation periods in March 2011. The transport and deposition process of two major discharges on March 15 was estimated using tag-tracer methods. The airmass from the first discharge released during 7-10 JST was transported southward, and 13% of 137Cs was deposited mainly over Kanto area via dry deposition process. In the afternoon, wind direction was changed following to the approach of low-pressure system, and the airmass from the first discharge was transported northward and 13% of 137Cs was deposited mainly over Naka-dori area in Fukushima prefecture via wet deposition process corresponding with the precipitation since 15-16 JST. The model showed that the deposition over Naka-dori area occurred within 3 hours after the beginning of precipitation, although the intensity of precipitation was less than 0.5 mm/hr during that period. The airmass from the second discharge occurred at 13-17 JST was transported northwestward, and 47% of 137Cs in second discharge were estimated to be deposited over the land. The comparison of accumulated deposition of 137Cs at Azuma-yama mountains supposed that the radionuclides from the Fukushima Daiichi nuclear power plant were trapped within the boundary layer, and the highest concentration existed at around 1000-1500 m above the ground level.

Keywords: radionuclides, chemical transport model, atmospheric chemistry, chemical transport

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MAG34-07 Room:IC Time:May 24 09:45-10:00

### Impacts of the Fukushima Dai-ichi Nuclear Power Plants on the Ocean

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The triple disaster of the March 11, 2011, earthquake, tsunami and subsequent radioactivity releases from the Fukushima Daiichi nuclear power plants are unprecedented events for the oceans. In response, we organized a research cruise in June, 2011 off Japan to study Fukushima derived radionuclides in the waters and biota off Japan. This presentation will provide an overview of these successful sampling activities and our plans for analyses of a wide range of radionuclides. We focus on the cesium-137 and cesium-134 surface distributions and vertical profiles obtained during this cruise. The highest cesium concentrations at that time were not necessarily at the closest sampling point 30 km from the Fukushima NPPs, but 70-100 km off shore. Furthermore, as part of this effort, 24 surface drifters were deployed and subsequently tracked. The trajectories of these drifters indicate that much of the contaminated water was being pulled away from the coast on the northern side of Kuroshio Extension. However, some of the drifters stayed in the coastal region suggesting that some contaminated water may recirculate in this area before being washed off-shore. The absence of drifter crossings across the Kuroshio Extension core suggests that it inhibits the southward spreading of contaminated water, at least over the western Pacific ocean. Measurements of different radioactive contaminants seem to agree with our interpretation based on drifters. These results are discussed in context of prior cesium levels in the waters off Japan and in comparison to radionuclide results from other studies in 2011 off Japan.

Keywords: Fukushima, radionuclides, Kuroshio, ocean circulation

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MAG34-08

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# Concentration of Cs of seawater and zooplankton in the western North Pacific one month after the Fukushima accident

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In March 2011, an accident at the Fukushima Daiichi nuclear power plant (FNPP-AC) was caused by the Tohoku earthquake and tsunami. As a result, gigantic amount of artificial radionuclideds were emitted to the atmosphere, land and ocean. To investigate the transport of radionuclides to the ocean and their dispersion, we conducted a cruise with the R/V Mirai in the western North Pacific about one month after the FNPP-AC and measured caesium (134Cs and 137Cs) in seawater and zooplankton. In addition, a numerical simulation of radionuclide dispersion in the ocean was conducted with a particle-tracking model using surface currents reproduced by the Japan Coastal Ocean Predictability Experiment 2 (JCOPE2) model. A modified one-way nested global?regional air quality forecasting system (AQF) was also applied to simulate the time?space variations in the 137Cs aeolian deposition flux over the western North Pacific. 137Cs concentration in surface seawater ranged from 0.004 to 0.284 Bq kg?1 (average 0.048 Bq kg-1). Samples from stations off Fukushima and Miyagi had higher 137Cs concentrations than those at other stations. 137Cs concentrations north of 40N were relatively higher than those south of 35N. These ranged from a few times to two orders of magnitude higher than those measured before FNPP-AC. 134Cs were detected while not detectable before FNPP-AC and 134Cs/137Cs were estimated to be 1 within measurement error. Because the 134Cs/137Cs ratio of drain water and air from the FNPP was reported to be nearly 1, radionuclides from the FNPP were likely transported to nearly all of the stations. At station K2, the 137Cs concentration in zooplankton from the surface mixed layer was 1.72 Bq kg-ww-1 and that in zooplankton from the subsurface layer was 3.16 Bq kg-ww-1. The corresponding values at S1 were 4.01 Bq kg-ww-1 in the surface mixed layer and 4.31 Bq kg-ww-1 in the subsurface layer. 134Cs was also detected in all zooplankton, with 134Cs/137Cs ratios of nearly 1 within measurement error. The 137Cs concentration of zooplankton around Japan was less than 0.1 Bq kg-ww-1 during the last decade. Thus, the observed concentrations were one to two orders higher than before 11 March. Previously reported values of the concentration factor (CF), the ratio of the Cs concentration of zooplankton to that of ambient, range from 10 to 100, whereas we estimated the CF for zooplankton in this study to range from 200 to 840, an order of magnitude higher than previous observations. The above CF has been estimated under steady state. Thus these might not be applied to our case that Cs concentration of seawater and zooplankton were measured one month after FNPP-AC and both concentrations were still transient. Moreover a possible explanation to this enrichment is that particulate materials with high 134Cs and 137Cs that originated in the FNPP were adsorbed onto the zooplankton, rather than taken up by them, and collected along with the zooplankton. JCOPE2 results showed that plumes of radioactively contaminated water extended north-eastward near the coast to 40N and eastward along the northern flank of the Kuroshio extension about one month after the FNPP-AC. The high 137Cs concentration off Fukushima and Miyagi can be qualitatively explained by dispersion of the water discharged directly into the ocean from the FNPP. In contrast, the JCOPE2 result showed no detectable 137Cs north-east of the FNPP beyond 40N, 150E, despite relatively large values observed there during the cruise, suggesting that radionuclides were transported to distant locations to the north-east by another mechanism. An AQF model indicated that 137Cs emitted from the FNPP would have been deposited over a wide area of the western North Pacific. Therefore, the observed high 134Cs and 137Cs concentration in seawater, SS and zooplankton might be attributable mainly to this aeolian input of particles with high 134Cs and 137Cs concentration.

Keywords: Fukushima Daiichi NPP, Caesium, Western North Pacific, seawater, zooplankton, numerical simulation

<sup>&</sup>lt;sup>1</sup>Japan Agency for Marine-Earth Science and Technology

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MAG34-09

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### Coordinated efforts of modeling radionuclide distribution in the ocean

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After the disruption on the Fukushima-I nuclear power plants, radionuclide falling out from the atmosphere mainly in March spread quickly and widely. A direct flow-out of the power plants in early April gave radionuclide into the coastal region and subsequently the Kuroshio Extension. The Oceanographic Society of Japan has established Working Group and coordinated expertise in oceanic sciences and the related disciplines. Simulation has been carried out by formulating a systematic inter-comparison of ocean models (MIP). Modeling approaches have been examined for entrainment into the Kuroshio, accumulation on bottom sediments and wide spread over the North Pacific. The information collected has been opened to public with appropriate explanations so that thoughtful citizens may be able to understand the fact. Responsibility of the WG is not limited to Japanese but extended to the international community also.

Keywords: radionuclide, marine pollution, modeling, bottom sediments

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MAG34-10

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# Characteristics of the distribution of radioactive materials in Abukuma Mountains, Fukushima Prefecture

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#### INTRODUCTION

Accident of Fukushima-daiichi Nuclear Plant leads to loss of living life in Fukushima and surrounding regions. Science and Technology are imposed of a responsibility to solve the problem. Maps of radioactive materials are produced for extensive area around Fukushima area. The region have an individuality even it is small. To understand behavior of radioactive materials in the environment for coming decades, the monitoring focused on each region based on the understandings of the characteristics of land is necessary.

The radioactivity, dose rate and their spatial distribution in the Yamakiya district in Kawamata Town, Fukushima Prefecture, where is assigned to be the planned evacuation district, are measured to understand the heterogeneous distribution of radioactive materials. Simplified methods are attempted to get the results quickly with wide-area coverage.

#### **METHODS**

#### 1) Dose rate survey by vehicle and by walking

GEORADIS RT-30 (gamma-ray spectrometer) is used to survey dose rate. Synchronized GPS is used to get location with dose rate. Calibration curve is created to convert dose rate inside of a car to one at 1 meter height outside of a car. In walking survey, RT-30 is fitted in the rucksack at 1 meter height and walk about small region.

#### 2) Surface contamination density

NHJ2 by Fuji Electric Co., is used to measure surface contamination density (Bq/cm2). The measured values are compared to the result of measurement by HPGe detector, and confirm the performance of the method.

#### 3) Radioactivity of the soil

HPGe detector provides very accurate result, however, it takes time for measurement. A cork borer is used to get and fill soil sample of 5cm depth to RIA tube, and gamma counter (Aloka ARC-500 and ARC-370M) are used to measure radioactivity for a large amount of soil samples

#### CHARACTERISTICS OF DOSE RATE AND RADIOACTIVITY

#### 1) Dose rate survey in a scale of Abukuma Mountains

Dose rate survey by automobile were carried out on July and August, 2011, around litate and Kawamata districts. Not only main roads but also forestry road were driven to get details of the distribution of dose rate. A figure is attached to show the spatial distribution of dose rate. The figure shows characteristic distribution that may shows the migration of plume and heterogeneous disposition of radioactive materials.

#### 2) Dose rate survey in a scale of small watershed

Walking survey with Gamma-ray spectrometer linked to GPS are carried out in small watersheds in Yamakiya district. The map of dose rate shows heterogeneous distribution patterns that may reflect soil particle size, effect of small topography, and redistribution of radioactive materials.

#### 3) Characteristics of surface contamination density, deposition amount in different land cover

Radioactivity is measured at the ground of coniferous forest (Japanese cedar, red pine), broad-leaved deciduous forest, paddy field, and crop land. Radioactivity at the 0 to 10 cm depth is high at paddy field than cropland. Radioactivity at the litter layer is high in deciduous broad-leaved forest than coniferous forest. Radioactivity at soil layer in 0 to 5 cm depth is high in cedar forest. Multi-site measurements are planned to verify the results.

#### 4) Radioactivity of domestic water

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Shallow groundwater is the main source of domestic water in Yamakiya district. Water from 13 wells are sampled and measured for radioactive materials. As a result, no samples show radioactive cesium over detection limit (<1Bq/L).

#### FINAL REMARKS

Spatial distributions of dose rate and radioactivity are measured for large and small spatial scales. Several simple methods are introduced and confirms the feasibility. At the JpGU meeting, we will present the monitoring results with comprehensive diagrams.

Keywords: accident of Fukushima Daiichi Nuclear Power Plant, Yamakiya district, Kawamata-machi, Fukushima Prefecture, dose rate mapping, radioactivity mapping, ununiformity



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MAG34-11 Room:IC

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# Radioactive fallout: lesson from Chernobyl and what could be experienced for the Fukushima post-accidental situation

MASSON, Olivier<sup>1\*</sup>

Most of northern hemispheric countries have experienced radioactive fallout (from atmospheric nuclear weapon tests, Chernobyl or from Fukushima) but except in the case of the global fallout, various impacts were noticed both at short time scale and in the context of more or less long post-accidental situations. It is probably too early to determine precisely the evolution in the environment of the radionuclides that were released by the Fukushima accident. Thus it could be interesting to share our point of view both in the light of our respective experience of radioactive fallout and deposition and considering the respective environmental features of Europe (France) and Japan. What we have learnt from Chernobyl and what information is still needed and could be experienced from the Fukushima accident or applied to the Fukushima situation, is of great concern in the framework of atmospheric deposition.

Of course, distance from the source is a key (first order) parameter as well as initial conditions that yield to deposition mapping. After initial deposition, contaminated areas act as a delayed and secondary source that can explain the resilience of formerly deposited radionuclides in the atmosphere.

At European scale, Chernobyl provides a lasting step effect in the time series of airborne levels whereas Fukushima did not. This lasting step effect belonged to resuspension mechanisms that became rapidly predominant as the airborne levels drop down radically. Resuspension and biomass burnings provided most of the peaks of activity levels in France. Those mechanisms are encountered in France as well as in Japan but probably with different magnitude and possibly with different consequences regarding the evolution of the background level. A review of those mechanisms and their respective importance will be presented.

Spatially, we observed some deposition heterogeneity that corresponds first to different deposition patterns, especially in mountainous ecosystems. Snow and cloud deposition are among the main typical patterns that distinguish deposition conditions in highlands and lowlands. Occult deposition by cloud/fog water can explain higher levels at altitude locations compared to what could be expected from a rain deposition relationship. In addition, this study could benefit to lowlands and coastal areas where fogs occur, either seasonally or on a regular basis.

Results obtained in samples taken at the summit of a low-altitude mountain will be commented.

Keywords: airborne radioactivity, 137Cs, deposition, resuspension, chernobyl, fukushima

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MAG34-12

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# Spatio-temporal variability of the deposited radioactive materials in forest environments after the Fukushima Daiichi NP

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Soil, vegetation and other ecological compartments are expected to be highly contaminated by the deposited radionuclides after the Fukushima Daiichi nuclear power plant (NPP) accident triggered by a magnitude 9.1 earthquake and the resulting tsunami on Marchi 11, 2011. Study site have been established in Yamakiya district, Kawamata Town, Fukushima prefecture, located about 35 km from Fukushima power plant, and designated as the evacuated zone. The total deposition of radioactive materials at the study site ranged from 0.02to >10 M Bq/m2 for Cs-137. The mature cedar, young cedar, and broad-leaf stands were selected as experimental site for the monitoring of spatio-temporal variability of the deposited radionuclides after the accidental release of radioactive materials. In order to measure the vertical distribution of radioactivity in forest, a tower with the same height of tree have been established at each experimental site. The measurement of radioactivity by using a portable Ge gamma-ray detector (Detective-DX-100, Ortec) and radionuclide analysis of leaf samples at different height revealed that a large proportion of radioactivity was found at the forest were trapped by canopies of the cedar forests. In contrast, in the broad-leaf forest highest radioactivity was found at the forest floor. Furthermore, spatio-temporal variability of radioactivity at the forest floor indicated that huge amount of caesium still remains on the canopy of coniferous forest, and subsequently transfers to forest floor in association with throughfall, stemflow, and litter fall.

Keywords: Fukushima NPP accident, Radionuclides, Forest, Spatio-temporal variability

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MAG34-13 Room:IC

Time:May 24 11:30-11:45

# Heterogeneous distribution of radiocesium in the environment emitted from the Fukushima Daiichi Nuclear Power Plant

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We analyzed <sup>137</sup>Cs in aerosols, rocks, soils and river suspended sediments collected after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. Based on the results of analysis, we discussed the behavior and transportation of radiocesium in natural environments after the accident. First, radionuclides were emitted from FDNPP as hot particles transported by the air, which contained water-soluble fractions of radiocesium. Radiocesium still existed in water-soluble fractions just after deposition on the ground. Subsequently, interaction of hot particles with water (e.g. rainfall) dissolved and strongly fixed radiocesium on rocks and soil particles, which changed radiocesium into insoluble form. The distribution of hot spots was possibly controlled by the initial position of deposition on the ground. Consequently, hot spots were studded on the surface of rocks rather than uniformly distributed. Cesium-137 concentrations of < 2 um fraction of rocks, soils and river suspended sediments were higher than those of the corresponding bulk samples. The distribution of radiocesium in river suspended particles was not homogeneous during transportation by way of rivers, reflecting the heterogeneity of radiocesium in rocks and soils. Leaching experiments demonstrated that radiocesium in rocks, soils and river suspended sediments were fairly insoluble, showing that the adsorption reaction is irreversible. The heterogeneous distribution of radiocesium in aerosols, soils and suspended particles is originated from the presence of hot particles in aerosols. Dissolution of radiocesium in the hot particles in the aerosols and subsequent irreversible adsorption on the soil particle complex are responsible for the preservation of the heterogeneity in soils and also in river suspended particles. The present results showed that their radioactivities are retained by only a part of the constituent in soils and suspended particles originally transported as hot particles in the aerosols.

Keywords: Radiocesium, heterogeneity, soil, Fukushima, FDNPP accident

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### The Cs-134 and Cs-137 concentrations in soil water covered by different vegetations

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The soil water was collected using a suction lysimeter (consisting of a ceramic porous cup) in a flask using a hand pump at the depths of 10, 30, and 50 cm in the grassland and forest sites and at 10, 20, and 30 cm depths in the farmland site. The collected water was filtered using 0.45 micro meter membrane. The Cs-134 and Cs-137 concentrations were determined by a gamma ray spectrometry using a germanium semiconductor detector. Each sample was analyzed for 30,000 seconds at Meteorological Research Institute in Tsukuba.

The concentration of Cs-134 and Cs-137 in the soil water ranged from 0.1 to 2.5 Bg/kg, and the concentration at a depth of 10 cm was higher than that at 30cm and 50 cm depths in the grassland and the farmland, whereas in the forest site, the highest concentration of Cs-137, 1.6 Bg/kg was observed at 50 cm depth in the young forest.

Keywords: cesium, soil water, forest, grassland

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# Continuous monitoring of radiocesium in drainage water from a paddy field during puddling

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Decontamination of agricultural lands contaminated with radiocesium after the accident of Fukushima Dai-ichi Nuclear Power Plant of Tokyo Electric Power Company is urgently requested. Ministry of Agricultural, Forestry and Fisheries (MAFF) has currently been undertaking development of decontamination measures for soils in agricultural lands. In cooperation with MAFF, Institute for Rural Engineering (NIRE) has conducted experiments for decontamination in paddy fields of Iitate Village, Fukushima Prefecture, which is included in the planned excavation zone with 20-50 mSv per year due to fallout from the accident. One of the NIRE experiments was a decontamination method "mixing soils and removal using water", in which muddy water was drained from paddy fields where surface soil and radiocesium was mixed with water. The muddy water was extracted by pump for disposal.

In this experiment, a monitoring equipment made up of a NaI(Tl) detector was developed and applied to an experimental decontamination in paddy fields for continuous monitoring of concentration of radiocesium in muddy water. The detector of monitoring equipment was a 5-inch diameter NaI(Tl) crystal with a photomultiplier tube, that was put into a large plastic bucket. A hole was made 30.5 cm above the bottom of the bucket for regulating the discharge flow from the bucket. Its resolution was about 7%. Signals from the detector were amplified by a gain controller and then sent to a 1,024-channel pulse height analyzer. The energy range was set to 0-2,048 keV so that the targeted nuclides (Cs-134: 605 and 795 keV; Cs-137: 662 keV; K-40: 1,460 keV; Bi-214: 1,765 keV) could be observed. The ratio of emissions of gamma ray from nuclides to detected peak counts, called the detection factor here, was determined by measuring 3.0M KCl aqueous solution (3,700 Bq L<sup>-1</sup>) in the bucket. At depth of 30.5 cm in the bucket, the detected peak count for K-40 was 123 cps, and therefore the detection factor was calculated as 3.2 L<sup>-1</sup>.

The experiment of muddy water removal was executed in two paddy fields, named as Field C1 and C2. The area of each of them was  $420~{\rm m}^2$ . In both C1 and C2, the soil was ploughed together with  $42~{\rm m}^3$  of supplied water, and then the muddy water was discharged by four suction pumps. In Field C2, additionally, the soil was tilled before the water injection, and the soil and water were manually stirred by dragging PVC pipes during the latter part of the pumping. The drained volume of muddy water was  $12~{\rm m}^3$  from Field C1, and  $17.6~{\rm m}^3$  from Field C2. The monitoring equipment was installed at an outlet of one of the pumps. The measurement time was set to ten seconds, and obtained spectra were logged every ten seconds. Radioactivity of Cs-134 and Cs-137 was calculated from the peak counts around 795 keV and 605- $662~{\rm keV}$ , with multiplying the detection factor  $3.2~{\rm L}^{-1}$ . On the other hand,  $300~{\rm mL}$  of the emitted muddy water was sampled every five minutes, and then taken back to the laboratory for measurement of radiocesium concentrations (5-inch NaI(Tl) scintillation counter, 600-second measurement). Results of the continuous monitoring and the laboratory measurement are shown in Fig. 1. The former part of the continuous measurement in Field C1 was unavailable because of malfunction of power supply. The fluctuation of radioactivity obtained by the continuous monitoring was well consistent with that by the laboratory measurement, and revealed that radiocesium concentration in the muddy water reached more than  $20,000~{\rm Bq}~{\rm L}^{-1}$  during the manual stirring although usual radioactivity in the pumped water ranged 5,000- $10,000~{\rm Bq}~{\rm L}^{-1}$ .

Keywords: NaI(Tl) scintillation counter, gamma ray spectrometry, decontamination evaluation, Fukushima Dai-ichi Nuclear Power Plant

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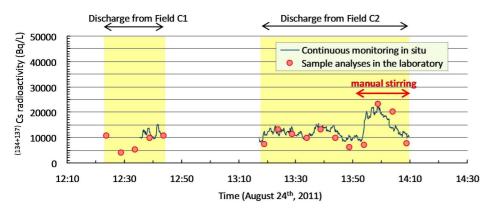


Fig.1 Results of the continuous monitoring of radiocesium in the emitted muddy water by the continuous monitoring equipment and those in the samples measured in the laboratory.