Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

MAG38-01 Room:501 Time:May 2 09:00-09:15

Cs-bearing spherical particles emitted from an early stage of the FDNPP accident

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We found radioactive Cs-bearing, spherical particles from the filters collected in March 14 and 15, 2011, just after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, in Tsukuba. These particles mainly consist of Fe and Zn but contain detectable amounts of Cs using a scanning electron microscope (SEM) and energy-dispersive X-ray spectrometer (EDS). They are several micro meter and are hardly water soluble. They are mostly spherical, suggesting they formed through rapid cooling of radioactive materials. These particles were only found in the filters collected on March 14 and 15, 2011, and these filters had many spots of radioactive materials when measured using an imaging plate (IP). To date, we have identified six such Cs-bearing particles in the filter.

The finding of such Cs-bearing spherical particles suggests the following implications; understandings of the accident and health effects for the radioactive materials emitted at the early stage of the accident and estimations of the current and future environmental radioactivity contaminated by the particles.

Reference: Adachi K., Kajino M., Zaizen Y., and Igarashi Y., Emission of spherical cesium-bearing particles from an early stage of the Fukushima nuclear accident, Scientific Reports, 2013, 3, Article number: 2554.

Keywords: Cesium, electron microscope, aerosol, radioactive material

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MAG38-02 Room:501 Time:May 2 09:15-09:30

Measurement of Cs-137 in atmopshieric aerosols in Fukushima prefecture and the surrounding area

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A large amount of radioactive materials were released in the environment by the accident at the Fukushima Daiichi Nuclear Power Station. We have been collecting air-dust using high volume air sampler at Fukushima city (Fukushima Pref.), Marumori town (Miyagi Pref.) and Hitachi city (Ibaraki Pref.) since the accident. We identified the radioactivities of 134Cs and 137Cs in filters using HPGe detector. We will discuss time variations of radioactive cesium concentration.

Keywords: Atmospheric observation, Air dust, Radioactivity Measurement, Cs-137 concentration

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MAG38-03 Room:501 Time:May 2 09:30-09:45

Estimate of possible sources of high Cs-137 in atmospheric aerosols measured in south Miyagi during 2 years (2012-2013)

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A volunteer team organized by the Japan Geoscience Union has started an intensive field study to monitor radioactive materials in the atmosphere, which were released by the Fukushima Daiichi Nuclear Power Plant (FD1NPP) accident, and by re-suspension of radioactive materials from soils and forests in a regional scale in and surrounding Fukushima area since April 2011. At present, the continuous measurement has been made at Marumori town in Miyagi prefecture, Fukushima city and Koriyama city in Fukushima prefecture, and Hitachi city in Ibaraki prefecture. In this paper, a case study on high concentrations of atmospheric radiocesium frequently measured at Marumori will be reported. At the Marumori town office in south Miyagi, atmospheric aerosols have been collected since December 2011, on a quartz fiber filter every several days by using a high volume air sampler, and radioactive materials in the aerosols were measured with a Ge detector. Forward trajectory analysis by a Lagrangian model was made to trace air masses started from the FD1NPP for 48 hours. The atmospheric concentration of Cs-137 at Marumori was in a level of 10^{-4} Bq m^{-3} until April 2012, and then gradually decreased to the level of 10^{-5} Bq m^{-3} in the latter half of 2013. High concentrations of Cs-137 more than 10^{-4} Bq m^{-3} were measured in the winter and early spring of 2012 and 2013 when the wind speed was high and relative humidity was low. It strongly suggests that the possible source of high Cs-137 could be re-suspension of radioactive materials from soils. In September and November 2012 and from May to August 2013, however, high concentrations more than 10^{-4} Bq m^{-3} were also frequently measured, and the highest concentration of $4.6x10^{-3}$ Bq m^{-3} was measured in a sampling period of 16-20 August 2013. On 19 August, unusual high Cs-137 concentration of $7.1x10^{-1}$-$8.7x10^{-1}$ Bq m^{-3} and $5.8x10^{2}$ Bq m^{-3} was measured at a monitoring post of Koriyama in Futabamachi 2.8 km north of the FD1NPP, and in front of a building inside the FD1NPP, respectively. According to the forward trajectory analysis, the air masses started from the FD1NPP at 09:00 and 12:00 on August 19 2013 arrived at Marumori on the afternoon of 15:00 and 18:00, respectively. It indicates that radioactive materials released from the FD1NPP were directly transported to Marumori about six hours later. The transport pathways similar to those on August 19 were also shown by the forward trajectory analysis in the other periods when the high Cs-137 concentrations were measured except for winter and early spring. These results clearly demonstrate that radioactive materials were still released into the atmosphere from the FD1NPP. We acknowledge the staff members of the Marumori town office for continuous sampling of atmospheric aerosols for these two years.

Keywords: atmospheric aerosols, radiocesium, source estimate, forward trajectory analysis

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MAG38-04 Room:501 Time:May 2 09:45-10:00

Study on the carrier of airborne radiocesium collected for six month in Tsukuba after the Fukushima nuclear accident

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To obtain the knowledge on the physico-chemical properties of airborne radionuclides, we had been collected size-resolved aerosol in Tsukuba, Japan, since April 28, 2011, although the data obtained do not include the first radioactive plumes that reached to Tsukuba on March 15, 2011. From the initial result, we proposed a hypothesis that the sulfate aerosol was the potential carrier of the ^{134}Cs and ^{137}Cs that had undergone the middle- to long-range transport from the damaged reactor. We further inferred that re-suspended soil particles that attached radionuclides were not the major airborne radioactive substances from late April to May, 2011 (Kaneyasu et al., 2012).

Nevertheless, there are some issues to be addressed on the nature of airborne radionuclides. Those are, a) until when the sulfate aerosol acted as a carrier of the radiocesium released from the reactor, or the other substances acted as carriers instead, and b) what is the carrier substance when the re-suspension or re-emission of became the dominant source in the airborne radiocesium.

In this study, we address these subjects by analyzing the long-term aerosol samples collected later than those presented in the previous study. The temporal change in the activity size distribution of radiocesium for six month will be discussed. In addition, the carrier substance of radiocesium in the coarse mode size range aerosol is investigated by use of the autoradiograph and scanning electron microscope to the aerosol sample collected in 2011 summer.

Keywords: radiocesium, size distribution, re-suspension, electron microscope, autoradiograph

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MAG38-05 Room:501 Time:May 2 10:00-10:15

Resuspension of radioactive cesium from soil and forest

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Radionuclides emitted from the Fukushima dai-ichi nuclear power plant (FNDPP) accident 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 signi?cant path in the diffusion of radionuclides after the accident. Therefore, the quantitative understanding of these re-suspensions is important to understand future transition of radionuclides. Identification of aerosol species which bring Cs-134/137 is necessary to understand the mechanism of re-suspension, and its efficiency.

We have measured atmospheric concentration of radiation by Cs-134/137 in Namie high school Tsushima-branch where is away 30km from FNDPP. Relationship between Cesium radioactivity and aerosol size destribution show that multiple re-suspension mechanisms contribute and their contribution varies with the season. The mechanisms of re-suspension will be shown and discussed.

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MAG38-06 Room:501 Time:May 2 10:15-10:30

Evaluation of radioactivity resuspension by dust emission using a size-resolved 1-D vertical model in Namie, Fukushima

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Radioactive materials released into the atmosphere by the Fukushima Daiichi Nuclear Power Plant Accident in March 2011 were deposited over a wider area. Those materials adhered to the soil particles (dust particles) and its resuspension by strong winds is apprehensive about as secondary emissions. We have proposed a size-resolved, one-dimensional resuspension scheme to calculate the concentration of radioactivity in the atmosphere, in the last annual meeting. The results underscore the importance of taking into account soil texture when calculating the concentrations of resuspended, size-resolved atmospheric radioactivity. However, various assumptions were incorporated into both the scheme and evaluation conditions. In this study, we made analyses of soil particle size distribution and soil radioactivity at a school ground in Tsushima District, Namie Town, Fukushima, which was heavily polluted by the accident. The model results were compared with in situ observational data of the size spectrum of atmospheric radioactivity. We validated the applicability of the scheme and the behavior of resuspended radioactive aerosols.

Keywords: Secondary emission, Radioactive aerosol, Dust, Fukusihma accident

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MAG38-07 Room:501 Time:May 2 10:30-10:45

Simulation of I-131 in the atmosphere emitted from the Fukushima Daiichi Nuclear Power Plant

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A large amount of radioactive materials was released into the atmosphere after the accident of the Fukushima Daiichi Nuclear Power Plant (FD1NPP). Inhalation of iodine 131 is important for internal exposure, but the observation of iodine is quite limited especialy in the early phase of the accident. We have conducted the simulation of radionuclides using a regional chemical transport model for March 2011. Calculated accumulated deposition of iodine 131 and caesium 137 was compared with the estimation using aircraft monitoring by MEXT and DOE (Torii et al., 2013). The model well captured the meridional gradient in the ratio of iodine 131 to caesium 137 around FD1NPP. The ratio of iodine 131 to caesium 137 is larger than 15 in the south of FD1NPP, and relatively small (around 0.7) in the northwest. This result implies that the regional model and the source term estimated by JAEA can generally reproduce eventual releases which cause large depositon ofer the land in March 2011.

Keywords: numerical simulation, atmospheric environment

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MAG38-08 Room:501 Time:May 2 11:00-11:15

The applicability of lichens as indicator of radiocaesium fall-out following the Fukushima Daiichi nuclear accident

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Lichens are symbiotic organisms consisted of fungi and algae. A number of studies was carried out after the nuclear weapons tests and Chernobyl accident, and demonstrated that lichens were useful for indicator of radioactive fallout because (i) they spread in almost all terrestrial habitats e.g. on rocks, tree barks, and soils, (ii) they could take up large amount of radionuclides directly from their thallus due to lack of root system and retain them, and (iii) they were long-lived. It is necessary to understand the behavior of radiocaesium released into the environment from the Fukushima Daiichi nuclear power plant (FNPP) on March 2011, because it is considered to migrate in the ecosystem over a long period. For this purpose, some indicators of initial amount of deposited radiocaesium are required to be compared. Though, the amount of deposited radiocaesium on the topsoil gradually decreases by weathering, while lichens are expected to retain radiocaesium for long time. However, very little work is currently available on the concentration of radiocaesium in lichens and there is no experience of applying lichens to indicator of fall-out in Japan.

In this study, an applicability of lichens as an indicator for amount of deposited radiocaesium was discussed based on the following investigations related to the Fukushima Daiichi nuclear accident. The lichens were widely collected from the area in Fukushima prefecture (mainly west side) and Kanto region affected by the accident since December 2012. Lichen species were focused on parmelioid lichens which were widely distributed around FNPP. (1) After the lichens were removed from barks and dried, the concentrations of ^{134}Cs and ^{137}Cs in the lichens were measured with a CsI scintillation detector or a Ge semiconductor detector and compared to amount of ^{137}Cs deposited on the topsoil on June 2011 and air dose rate. (2) The retention capability of radiocaesium was evaluated by comparing radiocaesium concentrations in lichens to those of barks of lichen habitat.

The radiocaesium concentrations in lichens tended to be higher than those of barks, indicating that parmelioid lichens had retention capability of radiocaesium than tree barks. It was observed that the radiocaesium concentrations in lichens increased with increasing the amount of ^{137}Cs deposited on the topsoil and air dose rate. These results suggested the applicability of parmelioid lichens as an indicator of radiocaesium fall-out in Fukushima.

Keywords: Fukushima daiichi nuclear accident, Parmelioid lichens, radiocaesium

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MAG38-09 Room:501 Time:May 2 11:15-11:30

Estimation of radioactive cesium translocation by litterfall, stemflow and throughfall in the forest of Fukushima

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The accident of Fukushima Daiichi nuclear power plant after the earthquake and Tsunami in March 11th 2011 caused large amount of radioactive cesium (Cs) deposition onto the forest in surrounding areas. Deposited radioactive Cs that were caught by the tree canopy, reaches to the forest floor via various several pathways. To estimate the annual flux of radioactive Cs translocate to forest floor, we investigated the component and amount of those which move from tree canopy based on the measurements of litterfall, stemflow and throughfall.

Field study was conducted in a forest at the upstream part of the Kami-Oguni River catchment, northern part of Fukushima Prefecture. Three plots (2 deciduous-pine (*Pinus densiflora*) mixed stands and 1 Japanese cedar (*Cryptomeria japonica*) plantation) were set in the forest. Five litter traps were set in each plot and collected every month from October 2012 to September 2013. Litter samples were sorted among tree species and also branches, seeds and barks. Throughfall and stemflow were collected every 1 or 2 months. Water samples were filtered and particulate matters were collected for radioactive Cs measurement. Radioactive Cs concentration of all samples were measured by germanium semiconductor detector and NaI(Tl) scintillation counter. Both concentrations of ^{137}Cs and ^{134}Cs were measured but only data for ^{137}Cs were discussed in this report.

The concentration of ^{137}Cs in leaf litter samples varied from non-detected level to above 30 kBq/kg. The ^{137}Cs concentration was highest in pine needles and followed by cedar. Leaf litters of deciduous tree species showed significantly lower concentration compared to those of evergreen trees. This was because deciduous trees were before leafing stage at the time of the accident. However, significant levels of ^{137}Cs in the leaves even of deciduous trees suggest that ^{137}Cs have been translocated from some part of tree body. On the other hand, deposited ^{137}Cs at the time of the accident still remains on the leaves of evergreen tree. Amount of ^{137}Cs translocated from canopy to forest floor in cedar plantation was about 3 times higher than that of deciduous-pine mixed forest. This was due to higher ^{137}Cs concentration and larger litter biomass of cedar.

^{137}Cs concentration of throughfall and stemflow were comparable. Since the amount of throughfall was larger than that of stemflow, significant amount of ^{137}Cs moved to the forest floor by throughfall. Higher ^{137}Cs translocation occurred according to the high precipitation. ^{137}Cs concentration fluctuated depending on the season, but there was no apparent tendency to decrease between 2013 and 2012. Since the concentration of ^{137}Cs in open rainwater was below the detection limit, it is suggested that ^{137}Cs is still supplied constantly from the tree canopy and source limitation is not occurring from leaves and trunks, despite the fact that it has past more than one and half year from the fallout.

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Effect of Radiocesium Transfer on Ambient Dose Rate in Forest Environment

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We investigated the transfer of canopy-intercepted radiocesium to the forest floor following the Fukushima Daiichi nuclear power plant accident. The cesium-137 (Cs-137) contents of throughfall, stemflow, and litterfall were monitored in two coniferous stands (plantation of Japanese cedar) and a deciduous broad-leaved forest stand (beech with red pine). We also measured an ambient dose rate at different height in the forest by using a survey meter (TCS-172B, Hitachi-Aloka Medical, LTD.) and a portable Ge gamma-ray detector (Detective-DX-100T, Ortec, Ametek, Inc.).

In decreasing order of total Cs-137 deposition from the canopy to forest floor were the mature cedar stand, the young cedar stand, and the broad-leaved forest. The ambient dose rate in forest exhibited height dependency and its vertical distribution varied by 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 rates at the canopy (approx. 10 m-) decreased earlier than physical attenuation of radiocesium, whereas those at the forest floor varied among three forest stands. These data suggested that an ambient dose rate in forest environment can be variable in spatially and temporally reflecting the transfer of radiocesium from canopy to forest floor.

Keywords: Fukushima Daiichi NPP accident, Cesium-137, Forest environment, Canopy interception, Transfer, Ambient dose rate

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MAG38-11 Room:501 Time:May 2 11:45-12:00

Three different structures of radionuclide ratios on the surface soil in the northwestern area from the FDNPP

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The Fukushima Dai-ichi Nuclear power plant (FDNPP) accident caused radioactive contamination on the surface soil at Fukushima and its adjacent prefectures. Substantial contamination has been found in the northwestern area from the FDNPP, according to the airborne monitoring survey and the ground base survey by MEXT, Japan. Radionuclide ratios would have characteristic information on emission source because each nuclearreactor at the FDNPP had different amount of radionuclide and different activity ratio. The activity ratios can be used to make emission source and transport process in the contamination more obvious . We address the issue of radioactive contaminated process, we have measured radionuclides on the surface soil at the town of Namie in the northwestern region from the FDNPP, in the viewpoint of activity ratio.

This study focused on the gamma-ray emitting radionuclides of ^{134}Cs, ^{137}Cs, and ^{110m}Ag. The activities were decay-corrected as of 11 March 2011 when all nuclear reactors scrammed. Data of activity ratios by our results and the Japanese official report classified the investigated northwestern region into 3 groups. Ratios of 0.02 for ^{110m}Ag/^{137}Cs and 0.90 for ^{134}Cs/^{137}Cs were observed northern area of inside 15 km from the FDNPP. On the other hand, two kinds of ^{110m}Ag/^{137}Cs ratios of 0.005 and 0.002 were distributed broadly in the area 60 km away from the plant. The ^{134}Cs/^{137}Cs ratio was 0.98 there.

The activity ratio in the northern area from the FDNPP corresponds to those of nuclear fuel in Unit 1 according to estimation using the ORIGEN code. The ^{134}Cs/^{137}Cs in the northwestern area from the FDNPP agrees with that of Unit 2 and 3. The ^{110m}Ag/^{137}Cs ratios of 0.005 and0.002 are 1/5 ? 1/10 of the Unit 2 and 3. Official report has announced that discharges of radionuclides from Unit 2 and 3 occurred on 14th March. It is known that contamination in the northwestern area from the FDNPP took place on 15th March. Ag has higher boiling point than Cs. Reactor core would be cooled down to lower temperature below the boiling point of Ag at the timing when contamination occurred. Thus, Ag with higher boiling point was not much released than Cs with lower boiling point. The ^{110m}Ag/^{137}Cs ratio has served to identify the specific sources of contamination in the northwestern area from the FDNPP.

Keywords: Fukushima Nuclear Power plant Accident, 110mAg/137Cs ratio, Surface soil

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MAG38-12 Room:501 Time:May 2 12:00-12:15

Depth profiles of ^{129}I and ^{137}Cs in soil before and after the FDNPP accident

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Massive nuclear fission products such as radioiodine and radiocesium were deposited on the land surface of Fukushima via radioactive pollution plumes derived from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. In order to evaluate inventory and penetration of accident-derived ^{129}I and ^{137}Cs in the land surface, depth profiles of ^{129}I, ^{129}I / ^{127}I atomic ratio and ^{137}Cs in 30-cm-long soil cores before (May 2008) and after (November 2012) the accident were compared at two sites (Iw-2 and Iw-8) on the western area within 10 km from the FDNPP.

Total ^{129}I inventories in soil core at two sites after the accident were estimated to be 0.74 - 1.96 Bq m^{-2}, 14 - 34 times higher than those before the accident (53.6 - 57.0 mBq m^{-2}). Average ^{129}I / ^{127}I ratios ((1.4 - 6.2) $\times$ 10^{-7}) in soil core after the accident were consistent with the ^{129}I / ^{127}I ratio of the radioactively-contaminated surface soils in Fukushima (1.5 $\times$ 10^{-8} - 7.2 $\times$ 10^{-6}, Miyake et al., 2012). We also estimated that total ^{137}Cs inventories after the accident were 0.60 - 3.15 MBq m^{-2}, 280 - 470 times higher than those before the accident (2.1 - 6.7 kBq m^{-2}). Average ^{134}Cs / ^{137}Cs activity ratios (1.07 - 1.08) in soil core fell within the activity ratio in Unit 1 - 3 (0.94 - 1.08) of the FDNPP calculated by ORIGEN2 code (Nishihara et al., 2012). These results suggested that accurate total inventories of accident-derived ^{129}I and ^{137}Cs in soil could be determined by deduction of those backgrounds at almost same site, thus, the FDNPP accident caused ^{129}I deposition of 0.69 - 1.90 Bq m^{-2} and ^{137}Cs deposition of 0.59 - 3.14 MBq m^{-2} on the western area within 10 km from the FDNPP. Moreover, deposited ^{129}I and ^{137}Cs at Iw-2 (4.2 km west from the FDNPP) were respectively, 2.9 and 5.3 times higher than those at Iw-8 (8.4 km west from the FDNPP).

Depth profiles of ^{129}I concentration, ^{129}I / ^{127}I atomic ratio and ^{137}Cs concentration before the accident were essentially declined from upper layer with depth at two sites. On the basis of the highest values in these profiles, background levels were determined to be 420 $\pm$ 11 Bq kg^{-1} for ^{129}I, 1.6 $\pm$ 0.1 $\times$ 10^{-8} for ^{129}I / ^{127}I and 48 $\pm$ 2.5 Bq kg^{-1} for ^{137}Cs. After the accident, significant elevated values of ^{129}I (40.2 - 130 mBq kg^{-1}), ^{129}I / ^{127}I ((0.9 - 9.3) $\times$ 10^{-6}) and ^{137}Cs (44.6 - 255 kBq kg^{-1}) were found in the uppermost layer at the two sites, then these profiles exponentially declined with depth. Approximately 90% of deposited ^{129}I and ^{137}Cs at two sites were absorbed upper 37.4 - 50.5 kg m^{-2} (4.1 - 4.3 cm) and upper 13.3 - 21.3 kg m^{-2} (1.0 - 3.1 cm) in depth, respectively. In addition, since the relaxation mass depths (h_0) of ^{129}I were 9.2 - 12.8 kg m^{-2} greater than those of ^{137}Cs (6.8 - 11.7 kg m^{-2}) at two site, radioiodine was considered to penetrate slightly deeper than radiocesium in upper layer of both sites as Kato et al. (2012) found at 40 km northwestern site from the FDNPP. This is not contradicting to increasing tendency of ^{129}I / ^{137}Cs activity ratio with depth at both sites. Based on the fact that both ^{129}I and ^{129}I / ^{127}I in soil after the accident declined to a background level under 84.8 kg m^{-2} in depth at Iw-2 and under 133 kg m^{-2} in depth at Iw-8, about 8 - 9% of accident-derived ^{129}I were likely to penetrated 37.4 - 84.8 kg m^{-2} (4.3 - 8.6 cm) in depth at Iw-2 and 50.5 - 133 kg m^{-2} (4.1 - 10.2 cm) in depth at Iw-8.

Keywords: FDNPP accident, Radioiodine, Radiocesium, AMS, Gamma-ray analysis, Soil profile

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MAG38-13 Room:501 Time:May 2 12:15-12:30

Distribution of ^{129}I in the environment released from the FDNPP accident and estimation of ^{131}I/^{129}I ratio

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Radioiodine is one of the most important radionuclides released from the Fukushima-Daiichi Nuclear Power Plant (FDNPP) accident. ^{131}I (half-life: 8 d) has a short half life time. Because of the difficulty of measuring ^{131}I at this time, it is expected to estimate ^{131}I precipitation from ^{129}I (half-life: 1.57×10^7 y) with the long half-life in the surface soil. We have measured ^{129}I concentrations in the surface soil at Fukushima. ^{129}I/^{127}I ratios were measured by accelerator mass spectrometry (AMS) at the MALT, the University of Tokyo (Matsuzaki et al., 2007). Stable iodine of ^{127}I was determined by inductively coupled plasma mass spectrometry (ICP-MS). We already got a result that the average ^{129}I concentration was $(2.74 \pm 1.35) \times 10^8$ atoms/g prior to the FDNPP accident as ^{129}I background at Fukushima. After the accident, average isotopic ratio of ^{131}I/^{129}I at Fukushima is estimated to $(4.02 \pm 0.81) \times 10^{-2}$ as at March 11, 2011. The results of calculation about ^{131}I/^{129}I ratio made by the ORIGEN2 code are 3.18×10^{-2} for the Unit 1, 4.57×10^{-2} for the Unit 2 and 4.81×10^{-2} for the Unit 3 (Nishihara et al., 2012). In this presentation, we report the distribution of ^{129}I in terrestrial environment at Fukushima and ^{131}I/^{129}I ratios by region.

Keywords: FDNPP accident, Radioiodine, ^{131}I/^{129}I, AMS

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MAG38-14 Room:501 Time:May 2 12:30-12:45

Desorption behavior of intrinsic cesium in smectite: Effect of aggregation on the cesium fixation in clay particles

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The radiocesium from the Fukushima Daiichi nuclear power plant accident is retained at the surface soils around the power plant. The expandable fine grained clay minerals such as smectite and vermiculate are the candidates for the host phases of radiocesium. The sorption mechanism of cesium in the clay minerals is expected to be cation exchange reaction in the interlayer of the clay minerals. Therefore, the retained Cs must be desorbed to the solutions in the presence of high concentrations of major cations. On the other hand, some natural observations after the Fukushima accident have shown that the radiocesium in the contaminated soils or sediments is merely desorbed to the water even in saline solutions (e.g. Aoi et al 2013 JPGU meeting). The purpose of the study is to reproduce the unexpected fixation of cesium in clay minerals from the laboratory experiment by using standard well characterized smectite (Kunipia-F). The desorption behavior of intrinsic trace Cs (10 nmol/g from LA-ICP-MS) in smectite by major cations were systematically examined. The results of the present study showed that the aggregation of smectite by the presence of the divalent cations or high concentration of monovalent cations lead to the fixation of cesium in the clay aggregates.

Keywords: cesium, smectite, desorption, fixation, aggregation

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Evaluation of the migration of radiocesium based on chemical speciation

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Chemical form of radiocesium is fundamental information for evaluation of its migration in the environment. After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, we analyzed ^{137}Cs in aerosols, rock, soil, leaves, river suspended sediment and river water collected in Fukushima. Here, we review the migration of radiocesium in the environment based on our up-to-date data.

Many particles with high radioactivity were found in aerosols collected in March, 2011, where 50% to 90% of radiocesium was water-soluble. This means that radiocesium was still present mostly in a water-soluble fraction of aerosols before deposition and just after deposition on the ground. However, it was found that little amount of radiocesium was contained in a soluble fraction in soil and weathered rock samples by leaching experiments with water at various pH conditions. Possibly, such a soluble fraction of radiocesium was strongly fixed on rock and soil particles after dissolution in water (e.g. rainfall) on the ground. At the moment, chemical species of radiocesium would have changed from soluble to insoluble form. This strong fixation of radiocesium in soils can be explained by formation of inner-sphere complex in phyllosilicate minerals of clay minerals, which was confirmed by extended X-ray absorption fine structure (EXAFS) analysis. Field-scale observation reflected well the strong adsorption of radiocesium because most of the radiocesium stayed within 5 cm from the surface in soil layers.

In particular, in river and ocean systems, whether radiocesium is particulate or dissolved form is closely related to uptake by organisms and incorporation into food chain in ecosystems. We have monitored radiocesium concentrations in the Abukuma River system since summer in 2011. Total ^{137}Cs concentration in river water including both dissolved and particulate fractions decreased drastically from summer to winter in 2011, and then gradually decreased with time except at heavy rainfall events. From the strong fixation of radiocesium on soil particles, it was expected that radiocesium was predominant in particulate matter in river systems. More than 70% of radiocesium was particulate form, where the contribution of silt size (3 ? 63 μm) fraction was the largest. However, radiocesium in dissolved fraction suggested an increase at estuary. This implies desorption of radiocesium from particulate matter because of an increase in salinity.

We made adsorption experiments to determine distribution coefficient, K_d, between fluvial sediment and river water, and further desorption experiments to examine the reversibility of adsorption-desorption process. Kd values determined by adsorption and desorption experiments were consistent, indicating that radiocesium adsorption was a reversible process. In addition, when artificial seawater was used for desorption experiment, the resulting K_d value was lower than that obtained using river water. This clearly demonstrated the influence of ionic strength on adsorption-desorption process through competition of cesium ions with other ions (e.g., K^+, Na^+ and Ca^{2+}), which is consistent with the field observation as noted above. Furthermore, we applied generalized adsorption model (GAM) to predict the distribution of radiocesium between particulate matter and water in the Abukuma River system. As a result, it was demonstrated that GAM can predict the apparent K_d values calculated from ^{137}Cs concentrations in fluvial sediment and river water as well as lower K_d values at estuary.

Keywords: Fukushima, Radiocesium

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Radiocesium wash-off associated with soil erosion from various land uses after the Fukushima Dai-ichi NPP accident

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Soil erosion is the initial process which drives radiocesium into the aquatic systems and therefore the quantification of radiocesium wash-off associated with soil erosion is indispensable for mitigating the risks. This study presents two year ' s observation of soil erosion and radiocesium wash-off to quantify differences in radiocesium behavior in various land uses. Seven runoff plots were established in four landscapes; uncultivated farmland (Farmland A1, Farmland B1), cultivated farmland (Farmland A2, Farmland B2), grassland (Grassland A, Grassland B) and Japanese cedar forest (Forest) in Kawamata town, an area affected by the Fukushima Dai-ichi Nuclear Power Plant accident. The discharged sediments were collected approximately every two weeks. In laboratories, collected sediments were dried and weighed for calculating soil erosion rates (kg m^{-2}) and served for measurements of radiocesium concentration (Bq kg^{-1}) with HPGe detectors. The erosivity factor of the Universal Soil Loss Equation (R-factor: MJ mm ha^{-1} hr^{-1} yr^{-1}) was calculated based on the data of precipitation. Standardized soil erosion rates (kg m^{-2} MJ^{-1} mm^{-1} ha hr yr), observed soil erosion rates divided by R-factor, was 1.8×10^{-4} in Farmland A1, 6.0×10^{-4} in Farmland A2, 1.5×10^{-3} in Farmland B1, 8.3×10^{-4} in Farmland B2, 9.6×10^{-6} in Grassland A, 5.9 × 10-6 in Grassland B and 2.3×10^{-6} in Forest. These erosion rates were basically proportional to their vegetation cover of soil surfaces except for cultivated farmlands. Concentrations of Cs-137 in eroded sediments basically depended on the local deposition of Cs-137 and varied enormously with ranging several orders of magnitude in all the landscapes. For the observation period of time decreasing trends in concentrations of Cs-137 in eroded sediments were not obvious. To compare these results with those of Chernobyl, we calculated normalized solid wash-off coefficient (m^2 g^{-1}) with dividing the mean total concentration of Cs-137 in sediments by local deposition of Cs-137 (Konoplev et al., 1992). The coefficient was 4.4 × 10-5 in Farmland A1, 1.3×10^{-5} in Farmland A2, 6.4×10^{-5} in Farmland B1, 1.0×10^{-5} in Farmland B2, 2.2×10^{-5} in Grassland A, 1.0×10^{-5} in Grassland B and 8.2 × 10^{-5} in Forest. High erodibilities and relatively low values of normalized wash-off coefficients in cultivated farmlands can be attributed to the mixing of surface soil by ploughing. These values almost corresponded to those of Chernobyl. It was found that the total solid wash-off coefficient of radiocesium from farmlands is high and for 2 years period of time after the accident reaches 10%. Generally high precipitation in the region and steep slopes promote higher wash-off of radiocesium as compared to the Chernobyl case. Also, normalized wash-off coefficients exhibited relatively less volatility than erodibilities in the landscapes. These results suggest that soil erosion management is crucial for mitigating risks of radiocesium.

Keywords: soil erosion, erosion plot, Cs-137

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MAG38-17 Room:501 Time:May 2 14:45-15:00

The distributed models to predict interannual changes in inventory and discharge of rCs from river basin

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Radioactive materials emitted from Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in March 11, 2011, are spreading to wide area and deposited on the ground. Abukuma mountains where vast amount of radioactive nuclides are deposited, is mostly covered by forest. Transition of radioactive nuclides arises with hydrologic and material cycles in forested mountain watershed, and the redistribution will proceed for a long time. Monitoring of the distribution and time changes in radioactive materials are necessary. At the same time, the prediction of long term behavior of radioactive materials is necessary to make use of restoration of contaminated area. The purpose of the study is to calculate erosion rate in wide area, and predict long term change in the inventory of radioactive cesium, especially cesium 137, by distributed parameter model.

Spatial resolution of the distributed model is 25m, same as aerial monitoring of dose rate and inventory maps published by MEXT. The area of calculation is the extent of 36 river catchment within the 80 km zone from FDNPP including Abukuma River Basin.

Members of USLE (Universal Soil Loss Equation) to calculate erosion rate are derived from observation in USLE plots established in different land cover in Yamakiya District, Kawamata Town, Fukushima Prefecture, by team Tsukuba University.

Land use type for each grid cels is derived from present vegetation map prepared by Ministry of Environment. Gridded land use map with 25m resolution is created from shape file of the vegetation map. Topographic parameters are extracted from 25m resolution DEM re-constructed from 10m DEM by GSJ (Geospatial Information Authority of Japan). Vegetation cover ratio map is created from MODIS NDVI datasets with 250m resolution processed by Tokyo University of Information Sciences.

Erosion rates on each grid cell are calculated and make distribution map. Erosion rate is high in crop land, and low in forested area. Average erosion rate in crop land is about 1.4 ton/ha/year, and the one in forested area is about 0.1 ton/ha/year.

The model that calculate the transition of cesium-137 is developed and the changes in the inventory from 2011 to 2041 are calculated. The erosion rate is annual value, so time step is set to one year. The eroded sediment is transported to down slope. Sediment Delivery Ratio (SDR), the ratio of transported sediment over total sediment, should be determined, however, the proper SDR is knot known, so SDR=1 is adopted in the calculation and maximum transportation rate is assumed.

The amount of cesium-137 is calculated by introducing Sc. Sc is the ratio of effluent cesium-137 (Bq/kg) over inventory (Bq/m2). Sc is determined by observations at the USLE plots of different land use. Out flowing cesium-137 is calculated by erosion rate multiplying by Sc.

The movement of debris along the slope is generally very slow, however, after the debris reach to the valley bottom, where saturation usually occur at the precipitation events, sediment is removed by flowing water. DEM is used to calculate Topographic Index (TPI) to designate the area of stream flow generation. When sediments reach to the area, cesium-137 flushes to the outlet of the watershed. In this calculation, all the cesium-137 is considered to be removed to the cell, and flushes to the outlet.

The calculation shows the average inventory of cesium-137 is about 10% lower than the one that only radioactive decay is considered. Total amount of discharge of cesium-137 at Iwanuma point, Abukuma River, is the order of 10ˆ13 Bq in both case in the first year after the deposition of radioactive materials. Discharge of cesium-137 sharply decrease in the first years, after the sharp drop, discharge decreased in exponential form.

The result of the study is based on the empirical model, however, it considers the established knowledge in the field of stream flow generation. The results reflect the actual condition of cesium-137 transition.

Keywords: Universal Soil Loss Equation, erosion rate, radioactive cesium, inventory change, distributed model, FUKUSHIMA

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Cs-134 and Cs-137 radioactivity of riverine suspended solids in the Abukuma River after the heavy rain in June 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 a combination with wind direction and precipitation. 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 transport of Cs-134 and Cs-137 in the Abukuma River running through Fukushima and Miyagi Prefecture in Japan, 15 months after the Fukushima Dai-ichi NPP accident. Field experiments were carried out at Shirakawa (upper), Motomiya, Data (middle) and Iwanuma (lower) during June 19-21, 2012. We also carried out the research at the Uta, Niida, Natsui and Same Rivers. 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 continuous centrifugation. The radioactivity of Cs-134 and Cs-137 was measured with gamma-ray spectrometry after drying them by freeze-dry method.

Total radioactivity of Cs-134 and Cs-137 in river waters was 0.091-3.83 Bq/l in high flow conditions by heavy rain. 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 radioactivity of Cs-134 and Cs-137 increased from 500 Bq/kg-ss in the upper site (Shirakawa) to 3470 Bq/kg-ss in the lower site (Iwanuma). The Cs-137 radioactivity was 3200 Bq/ kg-ss in the Uta River, 42440 Bq/ kg-ss in the Niida, 850 Bq/ kg-ss in the Natsui River and 550 Bq/ kg-ss in the Same River. These results indicate that the input of radiocesium associated with suspended particles from the watershed to the river water is controlled by the accumulation of radiocesium on the ground surface in the river watershed and transport processes of suspended solids in the river systems.

Keywords: river water, radioesium, particulate forms, migration, heavy rain event

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Transportation of radiocesium through rivers in Fukushima

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Due to Fukushima Daiichi Nuclear Power Plant accident, radioactive materials including Cs-134 and Cs-137 were widely distributed in surrounded area. The radiocesiums have been transported in river networks. This study showed the monitoring results of radiocesium concentration in river waters and suspended sediments in Abukuma river basin and smaller coastal river catchments.

The monitoring started at 6 sites from June 2011. Subsequently, additional 24 monitoring sites were installed between October 2012 and January 2013. Flow and turbidity (for calculation of suspended sediment concentration) were measured at each site, while suspended sediments and river water were collected every one or half month to measure Cs-134 and Cs-137 activity concentrations by gamma spectrometry.

Activity concentrations of Cs-134 and Cs- 137 on suspended sediments were generally decreasing at all sites. The decreasing rate changed lower at about one year later from the accident. Activity concentration in river waters also showed the same tendency although there are only few data within 1 year from the accident.

Activity concentrations measured at the same day are proportional to the mean catchment inventory. Therefore, the activity concentration can be normalized by the mean catchment inventory. The normalized activity can be fitted to following double exponential function:

[At] = 1.551 exp (-5.265 t) + 0.069 exp (-0.266 t), where t is the time from the accident [year].

Radioceasium flux at a monitoring site was measured from the flow and turbidity data and the radioceasium concentration. Suspended sediment concentration (SSC) could be estimated from the turbidity data. Suspended sediment flux was calculated by multiplying the SSC by flow rate. Then, multiplying the suspended sediment flux by radioceasium concentration gave the radioceasium flux. The highest radioceasium flux occurred in Sep. 2011 due to the typhoon roke. Then, the radioceasium flux declined, however the flux increased in the summer and autumn of 2013 due to typhoon events.

There is no time evolution of Kd between suspended sediments and river water. Instead, Kd was varied spatially. Although the reason of the spatial variation is not clear for now, geology of the catchment (i.e. mineral composition of suspended particles) seems to relate the variation.

Keywords: Radioceasium concentration, suspended sediment

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A sediment transport model for analyzing the environmental dynamics of radionuclides in estuarine and coastal oceans

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Several oceanic dispersal modeling have been conducted by multiple institutions on dissolved radionuclides leaked at the Fukushima Dai-ichi Nuclear Power Plant (FNPP). Among others, we developed a multi-nesting oceanic model at the lateral grid resolution down to 1 km and performed the comprehensive dispersal reanalysis of the direct release of ^{137}Cs from FNPP occurred in March and April 2011 (Uchiyama *et al*., 2013, *J. JSCE*). The model reveals that the current field on the continental shelf off Fukushima varied with surface wind stress and largely confined in the narrow coastal strip by about 30 km offshore. The spectral coherence analysis suggests that predominant alongshore transport of nuclides is caused by coastal jets on the shelf, presumably as forced shelf waves associated with the alongshore component of the wind stress. The coastal dispersal of the radionuclides is affected not only by direct release but also by atmospheric fallout (deposition) and discharge from the rivers. The last process introduces a time lag behind the direct release with hydrological process because the nuclides mostly attach to suspended particles (sediments) that are transported quite differently to the dissolved matter in the ocean.

In the present study, an Eulerian sediment transport model as an active tracer conservation equation with a prescribed settling velocity added to the vertical advection term, a wave-enhanced bed boundary layer model and a simple stratigraphy model proposed by Blaas *et al*. (2007) are implemented into ROMS (Shchepetkin and McWilliams, 2005, 2008). Three classes of sediments, viz., fine sand, silt and clay fractions, are considered here. The modeling procedure is approximately the same as Uchiyama *et al*. (2013), whereas the third embedding is done at the horizontal resolution dx of 250 m within the existing 1-km domain to develop the triple nested configuration forced by the assimilative JCOPE2 reanalysis (Miyazawa et al., 2009) as the outer-most boundary conditions. Thus the grid refinement occurs from JCOPE2 (dx ~ 10 km) to ROMS-L1 (dx = 3 km), to ROMS-L2 (dx = 1 km), and finally to ROMS-L3 (dx = 250 m). Sediments are taken into account in ROMS-L3 model carried out for March through August 2011. The bed skin stress is evaluated by a combined wave-current stress model of Soulsby (1995) with the wave field computed by a SWAN spectral wave modeling at dx = 1 km embedded in the JMA GVP-CWM spectral wave reanalysis. The bathymetry is provided by the 50-m resolution dataset compiled by Japan's Cabinet Office. The initial distributions of fractions of the marine bed sediment classes are estimated with an optimally interpolated field of the observations reported by Miyagi and Fukushima Prefectures (1991, 2013). Daily discharges of 6 major rivers and 14 minor rivers in the L3 domain are provided from the hydrological surface water model HYDREEMS conducted in CRIEPI. An empirical, mean relation between river discharge and sediment flux based on Takekawa *et al*. (2013) is employed for estimating the section-averaged sediment flux at each river mouth. Fraction of sediment classes in the river water is estimated from a USLE based river model conducted by JAEA (2013). The passive tracer is additionally considered to track dissolved ^{137}Cs released from FNPP as the direct release, whereas its absorption and desorption to the sediments (i.e., suspended ^{137}Cs) are not considered yet.

We intend to talk at the conference on initial dispersal of dissolved ^{137}Cs at dx = 250 m, extent of the land-derived sediments from each river mouth, resuspension and recirculation of the deposited bed sediments during storm conditions, in conjunction with corresponding oceanic states. We will further touch on potential distribution of suspended and dissolved ^{137}Cs if absorption and desorption occur.

Keywords: multi-class sediment transport model, radioactive cesium 137, multiple nesting approach, ROMS (Regional Oceanic Modeling System)

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Distribution of radionuclides in the surface seawater developed by aerial radiological survey

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This study investigated the distribution of anthropogenic radionuclide in the surface seawater derived from the Fukushima Daiichi Nuclear Power Plant (FNPP1) observed by aerial radiological survey as an initial attempt. The aerial radiological survey over the coastal region was performed by the U.S. Department of Energy National Nuclear Security Administration (DOE/NNSA) within a 30 km radius of the FNPP1 on 18 April 2011. We found good correlations between the in-situ activities of radionuclide (131I, 134Cs, 137Cs) in the surface seawater and gamma-ray peak count rates by aerial radiological surveys (correlation coefficients for 131I, 0.89; 134Cs, 0.96; 137Cs, 0.92). Based on these relations, we find that the area with high concentrations extend south-southeast from the FNPP1. The maximum concentrations of 131I, 134Cs, and 137Cs reached 329, 650, and 599 Bq L-1, respectively. The131I/134Cs ratios in surface waters of the high activities area on 18 April were about 0.6-0.7. Considering the radioactive decay of 131I (half-life: 8.02 d), we determine that the radionuclides in this area are due to direct release from FNPP1 to the ocean. These also confirm that the aerial radiological survey might be very effective to investigate the surface distribution of anthropogenic radionuclides in the surface seawater. Furthermore, the model reproduced the distribution pattern of the FNPP1 derived radionuclides, although simulated results by regional ocean model are underestimated.

Keywords: Airborne surveys, Ocean, Anthropogenic radionuclide, Gamma-ray peak count, Regional Ocean Modeling System, Fukushima Daiichi Nuclear Power Plant

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Approach taken by oceanography specialists toward building emergency system and analyzing radiocesium in bottom sediment

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Eastern Japan along the Pacific coast has been damaged seriously and is still trying to recover after the nuclear power plant accident in Fukushima due to the magnitude-9 earthquake on March 11, 2011. In addition, we should prepare ourselves for another accident in future. The necessary system is to predict and monitor radionuclide distributions immediately following a possible accident, even if it is a rare case. We have started a plan of testing an emergency system based on ocean simulation models. The other actions include monitoring and modeling of radiocesium concentration, which still keeps a high level in the bottom sediments. 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, while symposia have been held from time to time. 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, with a particular attention to re-suspending sediments.

Keywords: radionuclide, emergency system, sediments

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Long-term behavior of Cs-137 activity in the ocean following the Fukushima Daiichi Nuclear Power Plant Accident

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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.

We reconstructed spatiotemporal variability of Cs-137 activity in the ocean by the comparison model simulations and observed data. We employed a regional scale and the North Pacific scale oceanic dispersion models, an atmospheric transport model, a sediment transport model, a dynamic biological compartment model for marine biota and river runoff model to investigate the oceanic contamination.

Direct releases of Cs-137 were estimated for two years and six months after the accident by comparing simulated results and observed activities very close to the site. The estimated total amounts of directly released was 3.6±0.7 PBq. Directly release rate of Cs-137 decreased exponentially with time by the end of December 2012 and then, was almost constant. The daily release rate of Cs-137 was estimated to be 3.0 x 10^10 Bq/day by the end of September 2013. The activity of directly released Cs-137 was detectable only in the coastal zone after December 2012. Simulated Cs-137 activities attributable to direct release were in good agreement with observed activities, a result that implies the estimated direct release rate was reasonable, while there is no observed data of Cs-137 activity in the ocean from 11 to 21 March 2011. Observed data of marine biota should reflect the history of Cs-137 activity in this early period. We reconstructed the history of Cs-137 activity in this early period by considering atmospheric deposition, river input, rain water runoff from the 1F NPP site and absorption in sediment. The comparisons between simulated Cs-137 activity of marine biota by a dynamic biological compartment and observed data also suggest that simulated Cs-137 activity attributable to atmospheric deposition was underestimated in this early period. In addition, river runoff model simulations suggest that the river flux of Cs-137 to the ocean was effective to the Cs-137 activity in the ocean in this early period. The sediment transport model simulations suggests that the inventory of Cs-137 in sediment was less than 10% of total released Cs-137. Sediment is not dominant sink of Cs-137 in the ocean.

Keywords: Fukushima Daiichi NPP accident, Regional Ocean Model System, Cesium 137