Japan Geoscience Union Meeting 2015
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MAG38-01 Room:301B Time:May 26 09:00-09:15

Hourly atmospheric radionuclides after the Fukushima accident by analyzing filter-tapes of SPM monitoring sites

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The current estimates for the internal radiation doses from inhalation by the Fukushima Daiichi Nuclear Power Station (FD1NPS) accident on March 11, 2011 have large uncertainty, because no observed data has been found of continuous monitoring of radioactive materials in the atmosphere in the Fukushima prefecture (FP) just after the accident. And most of the atmospheric transport models have simulated the surface deposition of radionuclides released by FD1NPS accident, to compare with many observed deposition datasets in eastern Japan. To retrieve the atmospheric transport of radioactive materials released from the FD1NPS, we collected the used filter tapes installed in Suspended Particulate Matter (SPM) monitors with beta-ray attenuation method operated by local governments in the air pollution monitoring network of eastern Japan. Then, we measured hourly Cs-134 and Cs-137 concentrations in SPM at 40 monitoring sites in the FP and Tokyo Metropolitan Area (TMA) located more than 170 km southwest of the FD1NPS, after more than one year. The period for measurements was during March 12-23, 2011, when atmospheric, aquatic, and terrestrial environments were seriously suffered in most of eastern Japan by a large amount of radioactive materials released from the FD1NPS. In this paper, a comprehensive study will be reported for the first time on a spatio-temporal variation of atmospheric Cs-137 concentrations in the FP and the TMA. Major results are as follows; (1) Nine major plumes with Cs-137 concentrations higher than 10 Bq m^{-3} were found, of which 5 and 4 plumes were transported to the FP and TMA, respectively. (2) High radioactive materials from the FD1NPS were transported five times (March 12-13, 15, 18, 19, and 20-21) to the northern part of Hamadori located in the east coast of the FP, and which was little known until this study. Hence, the time-integrated atmospheric Cs-137 concentrations were highest among the FP and TMA during the period. (3) Two plumes were transported to Nakadori, located in the central part of the FP, on March 15, and March 20-21. The polluted air masses with high Cs-137 were observed in Nakadori for more than half a day from the evening of March 20 to the morning of March 21, and which was not recognized until now. (4) The radiation dose rate measured at some monitoring posts in Nakadori did not increase even when the plume (March 20) passed by. It was already too high to detect a new plume, due to the groundshine caused by the deposition of a large amount of radionuclides on the grounds by precipitation. (5) Two plumes transported to the TMA on March 16 and March 20 were newly found, in addition to the well-known two major plumes on March 15 and March 21, 2011. (6) A local area of relatively high Cs-137 deposition density in the TMA by precipitation on the morning of March 21, 2011, was consistent with an area where the time-integrated atmospheric Cs-137 concentrations were also high due to the transport of a plume on the morning of March 21, 2011. In the Fukushima prefecture, however, the correlation was not so clear. We thank to all the local governments in eastern Japan who kindly allowed us to measure radionuclides in the SPM collected on the used filter-tapes in the SPM monitors. We also thank Prof. S. Wakamatsu (Ehime University), and many other persons who cooperated to store the filter tapes. We sincerely acknowledge Y. Katsumura and M. Ishimoto in The University of Tokyo for supporting the measurements. Part of this study was financially supported by the Ministry of Education, Culture, Sports, Science and Technology, and the Ministry of Environment, Japan.

Keywords: Atmospheric Cs-137, FD1NPS, Spatio-temporal variation, SPM

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A new insight into the deposition mechanism of airborne radionuclides from the Fukushima accident

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Introduction

On the radiocesium emitted from Fukushima Daiichi Power Plant (FDNPP) and transported to Eastern Japan, airborne (MEXT, 2012) and car-borne (Tanigaki, 2014) surveys have been conducted to map the horizontal distribution of contaminated area.

In the mapped ^{134,137}Cs distribution by the airborne surveys, the contamination in the Nikko Mountain area, located 120 km north of Tokyo, is prominent. From the previously reported results of transport/dispersion/deposition modeling studies, this radioactive contamination appeared to have occurred in the transport event in March 15, 2011.

We thus analyzed the formation mechanism in this area by the on-hoot measurements of ambient gamma dose rate (GDR) in air at multiple mountains conducted from 2012 to 2014.

Instrumentation and Measurements

Altitudinal distributions of GDR in air were measured in the Nikko Mountainous area at the northern rim of the Kanto Plain, Japan, using a portable CsI (TI) scintillation gamma-ray detector (Gamma RAE II R, RAE Systems) carried along the mountain trails. The horizontal position of the observer was pinpointed by a global positioning system.

Results and Discussion

In the Nikko Mountain area, the altitudinal distribution of ambient GDR exhibited maxima at about 900-2,000 m above sea level (ASL).

Meteorological sounding data indicated that the corresponding altitudes were within the cloud layer. A visual-range monitor deployed in an unmanned weather station at 1,292 m ASL also recorded low visibility on the afternoon of March 15. In the gridded data products of Japan Meteorological Agency Meso-scale Model, cloud over (%) of low struts started to cover over the Nikko area at about 1500 JST, as seen in Figure 1. Radar-AMeDAS (Automated Meteorological Data Acquisition System) analysis shows this area was not affected by precipitation until 2300 Japan Standard Time on March 15, 2011. The mechanism anticipated to have caused the concentrated deposition in a particular altitudinal range was cloud deposition (or fog/occult deposition). Atmospheric aerosol particles often act as cloud condensation nuclei (CCN) so that cloud (fog) droplets form on them. With cloud/fog deposition, these droplets, and with them the original aerosol particles are intercepted by vegetation and deposited on the ground.

Airborne radiocesium, one of the major gamma-ray emitting radionuclides deposited onto the land and marine environments after the FDNPP accident, exists in the aerosol phase. Kaneyasu et al.(2012) reported that sulfate aerosols are a potential carrier of airborne radiocesium. Sulfate aerosol is one of the representative species acting as CCN. Therefore, cloud or fog droplets activated from CCN, including radiocesium and other gamma-ray emitting radionuclides, have greater deposition velocities than that of sulfate aerosol itself. This leads to the transfer of radionuclides from the atmosphere to the forest and ground surface being far more effective. Recently, Katata et al.(2014) incorporated a simple fog deposition scheme into their transport/disposition model and reproduced the deposition pattern in the mountain area qualitatively.

Conclusion

The proposed mechanism of the altitude-dependent radioactive contamination in Nikko Mountain area was the cloud (fog/occult) deposition process of the radionuclides contained in aerosols acting as cloud condensation nuclei.

Acknowledgement

The figure of cloud cover was produced by Dr. Y. Takane of AIST, Japan.

References

Kaneyasu, N. et al. (2012) *Environ. Sci. Technol.*, 46 (11), 5720-5726

Katata, G., et al. (2014) *Atmos. Chem. Phys. Discuss.*, 14 (10), 14725-14832

MEXT (2012) http://radioactivity.nsr.go.jp/ja/contents/7000/6289/24/203_0928.pdf

Tanigaki, M. (2014), *In* Radiation Monitoring and Dose Estimation of the Fukushima Nuclear Accident; Takahashi, S., Eds.; Springer: XV; pp.67-77

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Keywords: radionuclides, ambient gamma dose rate in air, deposition mechanism, altitudinal distribution, cloud deposition

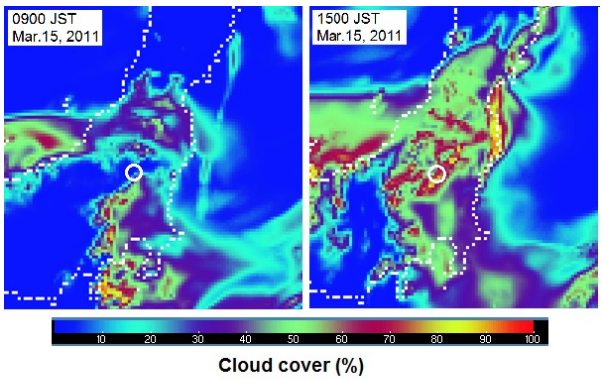

Figure 1 Cloud cover of low stratus in the gridded data products of Japan Meteorological Agency Meso-scale Model (MSM) on March 15, 2011. White circles indicate the location of Nikko Mountain area.

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MAG38-03 Room:301B Time:May 26 09:30-09:45

Seasonal variation of the atmospheric 137Cs & 134Cs - Concentration and aerosol size transporting radioactive Cs ?

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By the FDNPP accident on March 2011, large amounts of radioactive cesium (radiocesium) were released to the atmosphere. Total release amount of ^{137}Cs is estimated to 1.3×10^{16}(Bq) by Chino et al.(2012) and were deposited on the ground, vegetation and ocean. A half-life of ^{137}Cs and ^{134}Cs is 30.2 and 20.6 year respectively , and for the radioactive cesium which was diffused and deposites to repeat resuspention. The influence on a polluted area is expected for an extended period and also feared of risk to health

But the physical and chemical form of radiocesium have a lot of undefined , the resuspention system for now and the seasonal variation of the concentration and aerosol size transporting radioactive Cs is not understood enough. The purpose of our research is to understand the seasonal variation of the concentration and aerosol size transporting radioactive Cs with a long term monitoring atmospheric radiocesium.

We observed at 4 sites , Site-A(Kawatmata ceder forest) , Site-B(Tsushima school play ground) , Site-C(Tsushima broad leaf forest) , Site-D(Tsushima decontaminated playground). Site-A and Site-D was decontaminated in August to December 2012 and December 2011 to February 2012 respectively , therse sites are lower spatial dose. But Site-B and Site-C is not decontaminated, spatial dose is higher. This report used the date from December 2012 for research resuspention of radiocesium.

Sampling of techniques in Site-A and Site-C was mainly size distribution by impactor , sometimes only total concentration by High volume air sampler(HV). In Site-B observed size distribution and total concentration(short time cycle) at the same time.In site-D observed only total concentration.

Seasonal variation of total radiocesium have the same trend in all sites, lower concentration in winter and high concentration in summer. Thus in summer, there is 2 peaks , high concentration in May to July and August to September.

There is the seasonal variation of size distribution of transpoating radiocesium. Particles of 10.3˜0.69μm is mainly in summer and august , particles on backup filter(0.39μm and bounced larger particles) was high in winter and spring. This result suggests the change of process for resuspention in each seasons.

The spatial dose between site-A and site-B have a big difference , but the distance is very close, only about 0.75km departed. From this difference , we can expect the scale of broadness. Fraction(site-B divided site-D) is 1.33 in summer and august and 2.16 in winter and spring respectively. This result indicates the transpoting scale is larger at the season of high concentration and large particles are dominant (in summer and august). Oppositely at the seasons of low concentration and large particles are not dominat (in winter and spring) , transporting scale is very local .

Keywords: radioactivity, cesium, atomosphere, resuspention, FDNPP, ISET-R

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MAG38-04 Room:301B Time:May 26 09:45-10:00

Identification of aerosol contributing atmosperic re-suspension of radiocesium

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Coming soon

Keywords: radiocesium, atmospheric resuspension, aerosol

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Impact of the regional climate on the transport of radioactive materials

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Radioactive materials, which are released from Fukushima-Daiichi unclear power plants (FDUPP) in March 2011, is transported to Kanto and Tohoku areas. From many previous studies, the transport process of the radioactive materials has been clear. However the relation between the regional climate and transport process still is not sufficient to reveal. We investigated the mechanism of the transport to Kanto area using a regional model considering the regional climate in March. From the results, it is found that the radioactive materials, which are observed on 21th March 2011, is transported by the northeasterly in the lower layer from FDUPP to Kanto area. Generally, the northeasterly often forms corresponding to the low pressure system and the stationary front, which often form along the southern coastal area of Kanto area and causes the warm front with southerly warm wind. It is found that the cold air of the northeasterly is formed by cold air surges accompany with winter monsoon and moves to south corresponding to the temperature gradient in meridional direction near the surface. Therefore the northeasterly also has the property of gravity current. The wind field could be greatly influenced by the local temperature distribution because the cold air surges are often found in the off-shore of the Fukushima Prefecture. We will report the detailed transport process by numerical experiences applying the past events.

Keywords: Radioactive materials, Regional climate, Regional model

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MAG38-06 Room:301B Time:May 26 10:15-10:30

Distribution of radiocesium fallout on forest area throughout Japan after decades from former atmospheric nuclear tests

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To predict the movement of radioactive contamination caused by Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident is a strong concern, especially for the forest and forestry sector. To learn from the precedent, we investigated soil samples collected systematically from 316 forest sites in Japan just before the accident, which retain the global fallout ^{137}Cs (^{137}Cs-GFO) from the nuclear test bomb during the 1950s and 60s. We measured the radioactivity of ^{137}Cs-GFO in three layers of soil samples (0-5, 5-15 and 15-30 cm in depth) at each site. We divided 316 sampling sites into 10 groups separated by one longitudinal line and four transversal lines on the islands of Japan, then analyzed rainfall and geomorphological effects on ^{137}Cs-GFO inventories. In addition to the analysis of ^{137}Cs-GFO above, we examined the behavior of ^{137}Cs discharged from FDNPP (^{137}Cs-Fk) within the whole trees to study a possibility of biological effect on ^{137}Cs transport to soils from trees. We measured the radioactivity of ^{137}Cs-Fk of above- and belowground tree parts of three 26 year-old *Quercus serrata* and associated soils at a contaminated area in Fukushima in April, 2014.

We estimated an average of ^{137}Cs-GFO inventories of forest soils in Japan to be 1.7 $\pm$ 1.4 kBq/m^2 as of 2008. ^{137}Cs-GFO inventories varied largely from 0-7.9 kBq/m^2 around the country. We found high accumulation of ^{137}Cs-GFO in the north-western part facing to the Sea of Japan. We detected significant rainfall effects on the high accumulation due to winter rainfall. The vertical distribution of ^{137}Cs-GFO showed that 44% of ^{137}Cs-GFO remained within the 5 cm of soil from the surface whereas the rest of 56% was found in the layer of 5-30 cm in depth, indicating that considerable downward migration of ^{137}Cs-GFO occurred during these fifty years in forest soils in Japan. However, multiple linear regression analysis by geomorphological factors related to soil erosion, such as inclination angle or catchment area calculated from Digital Elevation Model, showed almost no significant effects on the distribution of ^{137}Cs-GFO.

The radioactivity of ^{137}Cs-Fk concentrations of fine roots collected from the 0-10 cm layer were 1600-2400 Bq/kg, which were comparable to those of one-year old branches (1400-2200 Bq/kg). The radioactivity of the fine roots was 7 times higher than that found in the soil of 50-100 cm layer (220-350 Bq/kg). This difference the radioactivity of the fine roots among the soil layers was remarkably small when compared with the 1000 times or more difference of radioactivity of soils in the same layers (one outlier sample in the 40-60 cm layer was excluded). The findings indicated that ^{137}Cs-Fk circulated through the whole tree within three years after the accident. Considering root litter fall inside the soils we estimated that contaminated ^{137}Cs on trees at the above ground part could be transported to soils through roots.

We clarified that ^{137}Cs-GFO has been held at deposited site and migrated downward gradually in soil. There are two possible major driving forces to be considered to explain the downward migration of ^{137}Cs-GFO. One is the migration of ^{137}Cs associated with vertical water movement and the other one is the transport of ^{137}Cs by root litter fall or root exudate. Further research is needed to analyze these processes to obtain reliable prediction of future distribution of ^{137}Cs-Fk.

Keywords: radioactive contamination, Fukushima Dai-ichi Nuclear Power Plant, 137Cs, forest soil, trees, secondary migration

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MAG38-07 Room:301B Time:May 26 10:30-10:45

Three-year monitoring study of radiocesium transfer and dose rate in forest environments after the FDNPP accident

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We investigated the transfer of canopy-intercepted radiocesium to the forest floor during 3 years following the Fukushima Daiichi nuclear power plant (NPP) 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 (Japanese oak 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.).

Total Cs-137 deposition flux from the canopy to forest floor for the mature cedar, young cedar, and the mixed broad-leaved stands were 157 kBq/m^2, 167 kBq/m^2, and 54 kBq/m^2, respectively. These values correspond to 36%, 39% and 12% of total atmospheric input after the accident. 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 slightly differed among three forest sites. In order to clarify the difference of decreasing trend of ambient dose rate, we investigated the relationship between radiocesium transfer flux from the canopy to forest floor and reduction of ambient dose rate at 1-m height above forest floor. We also evaluated effects of forest decontamination on ambient dose rate in coniferous stands.

Keywords: Fukushima Dai-ichi Nuclear Power Plant, Radiocesium, Forest environments, Transfer, Ambient dose rate

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MAG38-08 Room:301B Time:May 26 11:00-11:15

Analysis of each radionuclides discharged by the Fukushima Daiichi Nuclear Power Plant measured by airborne surveys

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Many radioactive substances were released by the Fukushima Daiichi nuclear power plant accident occurred on March 11, 2011 in the atmosphere. A lot of short half-life nuclides which are ^{131}I, ^{132}Te (^{132}I) and ^{133}I, etc., in addition to longer half-lived nuclides such as ^{134}Cs and ^{137}Cs. The estimated release amount of these nuclides from the reactor 1st to 3rd unit is reported, but it's found to be quite different in the short half-lived nuclides by the reactor units. Because the radioactivity ratio of ^{134}Cs and ^{137}Cs was slight different between the reactor units, it can be considered that the valuable source is obtained by the measurement of ^{134}Cs/^{137}Cs ratio in the environment around the Fukushima Daiichi nuclear power station at the present stage when the nuclides with short half-lives had already decayed. We have measured high-resolution gamma-ray spectrum using an unmanned helicopter equipped with $LaBr_3$(Ce) detector in a 3-km range from the power station which was near to the release source of the radioactive cesium. Because the $LaBr_3$(Ce) detector has high resolution of gamma rays, the discrimination of many nuclides is possible. In addition, there is extremely much number of the data provided by the distribution measurement with the unmanned helicopter. Because a new map was illustrated by the analysis of the ^{134}Cs/^{137}Cs ratio, we report the outline.

Keywords: unmanned helicopter monitoring, radionuclide distribution

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MAG38-09 Room:301B Time:May 26 11:15-11:30

Radiocesium transfer by water and sediment discharge through river networks after the Fukushima NPP accident

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Our research team has been monitoring the environmental consequences of radioactive contamination just after the Fukushima Daiichi NPP accident in Yamakiya-district, Kawamata town, Fukushima prefecture. Research items are listed below.

1. Radiocesium wash-off from the runoff-erosion plot under different land use.
2. Measurement of radiocesium transfer in forest environment by hydrological pathways such as throughfall and overlandflow on hillslope.
3. Monitoring of dissolved and particulate radiocesium concentration in river water, and stream water from the forested catchment.
4. Measurement of radiocesium content in drain water and suspended sediment from paddy field.
5. Continuous monitoring of suspended sediment and river water for 30 locations in abukuma catchments and coastal catchments.

Our monitoring result demonstrated that the Cs-137 concentration in eroded sediment from the runoff-erosion plot has been almost constant for the past 3 years, however the Cs-137 concentration of suspended sediment from the forested catchment showed slight decrease through time. On the other hand, the suspended sediment from paddy field and those in river water from large catchments exhibited rapid decrease in Cs-137 concentration with time. The decreasing trend of Cs-137 concentration were fitted by the two-component exponential model, differences in decreasing rate of the model were compared and discussed among various land uses and catchment scales . The declining trend such analysis can provide important insights into the future prediction of the radiocesium wash-off from catchments with different land uses.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident, Radiocesium, River system, water and sediment discharge, transfer

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MAG38-10 Room:301B Time:May 26 11:30-11:45

Radioactive Cs-137 discharge by Dissolved water, Suspended Sediment and Coarse Organic Matter from Headwater Catchment

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The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, following the earthquake and tsunami disaster on March 11, 2011, resulted in a substantial release of radionuclides to the environment. Most of the area in which the radionuclides were deposited was forested area. Therefore it is very important to quantify the discharge of radionuclide from the forested catchment in headwater region which will be the input of the radionuclide to downstream various land use.

The concentration of dissolved ^{137}Cs in groundwater and stream water in the headwater catchments in Yamakiya district, located ˜35 km north west of Fukushima Dai-ichi Nuclear Power Plant (FDNPP), was monitored from June 2011 to December 2014. Also ^{137}Cs concentration in suspended sediments and coarse organic matter such as leafs and blanches were monitored. Groundwater and stream water were sampled at intervals of approximately 2 months at each site. Intensive sampling was also conducted during rainstorm events. Compared with previous data from the Chernobyl NPP accident, the concentration of dissolved ^{137}Cs in stream water was low. In the Iboishi-yama catchment, a trend was observed for the concentration of dissolved ^{137}Cs in stream water to decline, which could be divided into two phases by October 2011 (after 200 days after the accident).

The highest ^{137}Cs concentration recorded at Iboishi-yama was 1.2 Bq/L at the peak on August 6, 2011, which then declined to 0.021-0.049 Bq/L during 2013 (in stream water under normal water-flow conditions). During the rainfall events, the concentration of dissolved ^{137}Cs in stream water increased temporarily. The concentration of dissolved ^{137}Cs in groundwater at a depth of 30 m at Iboishi-yama displayed a decreasing trend from 2011 to 2013, with a range from 0.039 Bq/L to 0.0025 Bq/L. The effective half-lives of stream water in the initial fast flush and secondary phases were 0.1-0.2 y and 1.0-2.2 y, respectively, in the three catchments. The trend for the concentration of dissolved ^{137}Cs to decline in groundwater and stream water was similar throughout 2012-2013, and the concentrations recorded in deeper groundwater were closer to those in stream water. The declining trend of dissolved ^{137}Cs concentrations in stream water was similar to that of the loss of canopy ^{137}Cs by throughfall, as shown in other reports of forest sites in the Yamakiya district.

The ^{137}Cs concentration in suspended sediments showed 10000-45000 Bq/kg and ^{137}Cs concentration in organic matter showed 1000-12000 Bq/kg. The ^{137}Cs concentration in organic matter showed declining trend through the observation period and the trend was similar to that of dissolved ^{137}Cs concentration in stream water. In contrast ^{137}Cs concentration in suspended sediment did not showed a clear declining trend in every catchment (before the decontamination works). After the decontamination works in Iboishi-yama catchment, the ^{137}Cs concentration in suspended sediment declined largely to 200-300 Bq/kg. The ^{137}Cs concentration in organic matter and dissolved ^{137}Cs concentration in stream water did not declined largely.

In conjunction with discharge data and turbidity data, ^{137}Cs discharge flux were calculated for three components (dissolved water, organic matter, suspended sediments). As a result, it is shown that more than 98% of the ^{137}Cs discharge was due to the discharge by suspended sediments. It is also suggested that dissolved ^{137}Cs concentration have relationship with ^{137}Cs concentration of organic matter and so it is still important to clarify the process of ^{137}Cs migration through dissolved water and organic matter.

Keywords: Cs-137, Dissolved water, Suspended sediment, Coarse organic matter, Headwater catchment, Discharge

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MAG38-11 Room:301B Time:May 26 11:45-12:00

Transport of radiocesium in the Niida River, Fukuhsima Prefecture in 2011-2014

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Surface deposition of ^{134}Cs and ^{137}Cs reveals considerable external radioactivity above 3000k Bq/m^2 in a zone extending northwest from the NPP after the Fukushima Daiichi Nuclear Power Plant (NPP) accident. Therefore, it is important to elucidate the short-term to long-term impacts of the accident on ecosystems of river watershed environments. This study investigated the transport of ^{134}Cs and ^{137}Cs in a small river, Niida River running through IIdate Viledge, in Fukushima Prefecture, Japan at normal and high flow conditions during 2011-2014.

Field experiments were conducted at a fixed station (Kinouchi bridge) in the lower Niida River during the period of May 2011-December 2014. The 20 L of surface river water samples were collected at the station using buckets. The radioactivity of ^{134}Cs and ^{137}Cs in the river waters before and after the filtration was measured with gamma-ray spectrometry using ammonium molybdophosphate (AMP)/Cs compound method.

Total radioactivity of ^{137}Cs (dissolved and particulate phases) in the river waters ranged from 0.11 to 4.18 Bq/L during May 2011-December 2014. Highest value was found in May 2011. Total ^{137}Cs radioactivity indicates the decreasing trend with increasing time at normal flow condition. However the higher radioactivity was observed after rain events. The ^{137}Cs radioactivity increased by 1.83 Bq/L after the heavy rain event by Typhoon Guchol in June 2012, and 1.68 Bq/L by Typhoon Jelawat in October 2012.

Percentage of ^{137}Cs associated with riverine suspended solids was 47-48% at normal flow condition in July and September 2011, but after December 2011 ranged from 75 to 93% at normal flow condition and 86-91% at high flow condition due to rain events. The radioactivity of particulate phase of ^{137}Cs ranged from 20 to 42 Bq/g-riverine suspended solids. Rain events are mainly contributed to the transport of radiocesium in the Niida River.

Keywords: Cs-134, Cs-137, rain event, riverine suspended solids

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MAG38-12 Room:301B Time:May 26 12:00-12:15

Radiocesium discharge from Niidagawa river basin after the accident of Fukushima Dai-ichi Nuclear Power Plant

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Redistribution of radiocesium through a river system poses potential risks on residents within the river basin. Niidagawa river basin on north eastern part of Fukushima prefecture is indeed a case in point. The river basin includes highly contaminated area on its upstream part and the river goes through residential area of its downstream. Quantitative evaluation of radiocesium redistribution associated with discharged sediment is required for mitigating radiological risk on residents and ocean environment. Previous studies on the environmental monitoring of various contaminants, as well as radiocesium, suggested that large portion of discharge of such contaminants should occur during intensive rainfall events. This study aims to quantify radiocesium discharge within the Niidagawa River Basin and to reveal its discharge processes. For quantifying radiocesium discharge, we installed sets of time-integrative suspended sediment sampler, turbidity sensor and water level sensor on three points in summer 2014. The points are named as Sakegawa-bashi (N37^{o}38'33", E141^{o}00'20"), Notegami-kita (N37^{o}39'16", E140^{o}47'47") and Warabi-daira (N37^{o}36'49", E140^{o}48'04") and specific initial Cs-137 deposition are 752, 810, 1462 kBq/m^2, respectively. We have collected suspended sediments trapped in the samplers approximately every 2 months and served them for radiocesium measurements with HPGe gamma detectors in laboratory. Radiocesium discharge (Bq/hour) were calculated by multiplying radiocesium concentrations of suspended sediment (Bq/kg) with time-series data of turbidity (kg/m^3) and water discharge (m^3/hour). Mean Cs-137 concentration of suspended sediment (n = 3) at Sakegawa-bashi, Notegami-kita and Warabi-daira were 13, 14 and 31 kBq/kg, respectively. Those concentrations appeared to agree with initial Cs-137 deposition for each point basically. No common temporal trend of Cs-137 concentration was found among Cs-137 concentrations of these three points. Estimated total Cs-137 discharges at these points were in magnitude of 10^{11} Bq. Estimated Cs-137 wash-off rates (%), calculated by dividing specific Cs-137 discharge (Bq/m^2) with initial Cs-137 inventory, were in magnitude of 0.1 %. For evaluating Cs-137 discharge during intensive rainfall events, we focused on three large rainfall events during 8-11th August (59 mm), 5-8th October (109 mm) and 13-16th October (69.5 mm). Ratios of Cs-137 discharges during these events to total Cs-137 discharge through all observation period approximately were approximately 0.3 at Sakegawa-bashi and approximately 0.7 at both of Notegami-kita and Warabi-daira. Discharge of Cs-137 from smaller catchment area appeared to be more vulnerable to intensive rainfall events.

Keywords: Radiocesiumu, River, Fukushima Dai-ichi Nuclear Power Plant, Suspended sediment

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MAG38-13 Room:301B Time:May 26 12:15-12:30

Investigation of the River Particulate Matters in Fukushima and Chernobyl Related to Cesium Behavior

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The behavior of the radioactive cesium (radio-Cs), which was emitted by nuclear power plant accident, was different between in the Abukuma River (Fukushima) and in the Pripyat River (Chernobyl). Previous studies showed that dissolved Cs was dominant in the Pripyat River (approximately 70%). Conversely, Cs in the particulate matters (PMs) was dominant in the Abukuma River (dissolved Cs is approximately 30%). this difference of radio-Cs behavior was related to the blocking effect by the coating of natural organic matter (NOM) on clay minerals in the aquatic environments. However, the mechanism of the blocking effect by NOM has not been investigated for environmental samples yet in terms of morphological image and characterization.

Scanning transmission X-ray microscopy (STXM), X-ray microscopy using soft X-ray region, is very confortable to confirm the blocking effect by NOM. However, STXM had not been installed into synchrotron radiation facilities in Japan until 2012. Therefore, compact STXM (cSTXM) was newly designed and developed at BL-13A in KEK-Photon Factory (PF) until 2014. Its size, similar to laptop (A4 paper size), is much smaller than conventional STXM (Bruker's STXM). Piezo electronic driven stages were installed to all axis stages in the cSTXM. This system can be fast and precise sufficient to control the nano-scale imaging. As to spatial resolution using 30 nm outer most zone width FZP, we can attain resolution about 37 nm at sample measurement position. All detection was conducted by counting system using avalanche photo diode (APD) or photo multiplier (PMT). Soft X-ray region allow us to measure the light elements such as carbon, nitrogen, and oxygen.

As to samples, suspended water was collected from in the Abukuma River and the Pripyat River, which was filtered initially by 3.0 μm membrane filter and subsequently by 0.45 μm membrane filter using a pressurized pumping system. Finally, fine PMs ($>$0.45 μm) were obtained by this filtration. Before STXM measurement, these PMs were dispersed into water by supersonic wave for 5 min. The water droplet including the PMs was dropped on a 50-nm-thick Si3N4 membrane and air-dried. In addition to the sampling, the concentration of dissolved organic carbon (DOC) was measured in these rivers. In this study, we analyzed these PMs obtained from the two rivers using scanning transmission X-ray microscope (STXM) to investigate (i) the chemical distribution images of NOM (mainly composed by carbon) and clay minerals (mainly composed by aluminum) and (ii) the functional group images of carbon. In addition, (iii) the characteristic of NOM was measured by STXM-near edge X-ray absorption fine structure (NEXAFS) extracted from target area of NOM adsorbed on clay minerals.

Two distribution images (carbon and aluminum) showed that clay minerals were covered with NOM in the Fukushima and Chernobyl samples. By functional group mapping of carbon, it was found that distribution of each functional group was similar among different functional groups (e.g. aromatic, phenolic, carboxylic) in the PMs obtained from the two rivers. NEXAFS spectra of carbon were also similar to all samples, suggesting that humic substances (mainly composed of humic and fulvic acid) are the main cause of the blocking effect in these rivers. Based on these results, it is suggested that the blocking effect could occur in the Abukuma and Pripyat Rivers, which is caused by humic substances covering with clay minerals. Therefore, it is concluded that the degree of the blocking effect by NOM related to radio-Cs behavior was dependent on the concentration of DOC in the river (in the River Abukuma: 0.9-1.3 mg/L, in the River Pripyat: 18 mg/L). In addition to the studies on the rivers, investigation results for PMs obtained from Fukushima pond (DOC value is between the Abukuma and Pripyat Rivers) will be explained in this presentation.

Keywords: Natural organic matters, river particulate matters, Cs behavior, Scanning transmission X-ray microscopy (STXM), Fukushima, Chernobyl

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MAG38-14 Room:301B Time:May 26 12:30-12:45

Identification of radioactive microparticles in the vicinity of the Fukushima Dai-ichi Nuclear Power Plant

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The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) disaster in 2011 discharged a large amount of radionuclides to the environment. Previous studies already disclosed the source term and inventory of radionuclides. However, physical and chemical states of radionuclides in the environment have yet to be revealed. Numerous environmental samples have been analyzed to investigate distribution of radionuclides by digital autoradiography using an imaging plate (IP). These studies pointed out that the spot-type contaminations were appeared and very common not only for the samples collected during the early stage (collecting until April 2011) of the FDNPP disaster but also for those collected later. In the FDNPP disaster, major forms of radioactive materials have been assumed to be gaseous. This is because the disaster underwent no reactor explosion nor fire of the reactor core, which is quite different from the situation in the Chernobyl NPP (ChNPP) disaster in 1986. In addition, it has been revealed that the emitted nuclear fuel (uranium) and 90Sr was very low. Therefore, elucidation of the cause of the spot-type contamination is crucial, which could contribute to know about physical and chemical state of radionuclides in the environment, as well as disclose their release processes from the FDNPP. Present study thus aiming at elucidation what makes the spot-type contamination in the IP analysis.

Surface soil samples were collected in the area about 20 km northwest from the FDNPP in June 2013. Spot-type contaminations were detected by IP analysis. Then, the radioactive particles were isolated by repeated sorting process and measured by the γ-spectroscopy with the high purity germanium semiconductor detector. The particles isolated were finally observed by SEM-EDS to determine the elemental composition and the morphology.

So far identified are four particles, and determined radionuclides by γ-spectroscopy were ^{134}Cs and ^{137}Cs. The morphology of the particles exhibited two types; spherical and fragmental. Thus, the spot-type contamination was characterized as due to such specific radioactive particles. The particles consist of Si, O, Zn, Fe, etc. and Cs (of which activity matched with the results by γ-spectroscopy) were also easily detected by SEM-EDS, suggesting glassy molten material. However, other fission products were not found in the particles. Tremendous amount of radioactive particles were collected around the ChNPP, and they were termed hot particle. Dominant components of the hot particle are nuclear fuel and typical fission products such as ^{90}Sr, ^{153}Eu as well as ^{134}Cs and ^{137}Cs. In contrast elemental composition of the present particles were dominated by Si. Uranium was found for only one of the present particles, but its concentration was very low and localized within the particle. In conclusion, characteristics of the present particles are quite different from the Chernobyl hot particles, but very similar to those reported for the particles isolated from HV filter samples in Tsukuba (Adachi et al., 2013, Abe et al., 2014).

Keywords: FDNPP disaster, Radioactive particle, Consist elements, Isotope ratio

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MAG38-15 Room:301B Time:May 26 14:15-14:30

Desorption behavior of cesium (133 and 137Cs) from the clay minerals distributed around the Power Plant

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In 11 March 2011, Tohoku region Pacific Ocean earthquake occurred and Fukushima Daiichi nuclear power plant caused a steam explosion. AAnd a large amounts of radioactive material was were released around nuclear power plant. Among the released radioactive material, from the total emissions and half-life, a major cause of soil pollution around nuclear power plant has been said to beis radioactive cesium (Cs).

Extensive radioactive Cs around nuclear power plants have been identified retained in the fine materials of the soil surface and. the The layered clay minerals such as smectite, vermiculate and illite contained in the universal soil is are pointed out that considered to be the main uptake medium Csa for radioactive Cs. Soil of Fukushima Prefecture has Abukuma granite and host rock, smectite, vermiculite, the presence of illite has been confirmed is the layered clay mineral is its weathering products.Host rock of soil of Fukushima Prefecture is in Abukuma granite.having a layered clay mineral which is a weathering product, smectite, vermiculite, illite presence has been confirmed. Layered These clay minerals has possess a the layered crystal structure and hydrated cations held in the interlayer, which can be exchanged with the foreign cations in solution. For a particularly high affinity for the Cs+ to these clay minerals,Therefore, Cs emitted by the nuclear accident is expected that it isto be firmly held between the layers of the layered clay minerals.

However, if the concentrations of the foreign main cations in the solution is are high concentration, the retained Cs+ by exchange with firmly held Cs+ is a also other cations may be leachedin the clay minerals is possibly released to the solutions. Natural water the clay particles are contacted in naturegenerally contains a wide variety of the major cations at different concentrations. Thus radioactive Cs which is adsorbed on the natural soil it is feared eluting natural environment. To the understanding of the dynamics of radioactive Cs in the environment, the understanding of Cs leaching desorption behavior by major cation from natural soil is essential. This study, by using a soil clay distributed around the Fukushima Daiichi nuclear power plant, was intended to systematically verify the desorption behavior of Cs from the soil clay minerals distributed around the Fukushima Daiichi Power Nuclear Plant by adding the majorin cation.

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MAG38-16 Room:301B Time:May 26 14:30-14:45

A study of Cs-adsorption behavior of clay minerals using autoradiography and considering the actual environments

KOGURE, Toshihiro[1*] ; MUKAI, Hiroki[1] ; HIROSE, Atsushi[2] ; MOTAI, Satoko[1] ; TANOI, Keitaro[2] ; NAKANISHI, M.tomoko[2]

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Though there are a number of studies to analyze the soil contamination with radioactive cesium released from Fukushima nuclear accident, microscopic analyses to investigate, for example, the adsorption sites of radioactive cesium in the actual soil are few. Recently we found, using autoradiography with special imaging plates (IPs) and various electron microscopic techniques, that weathered biotite originated from Abukuma granite in Fukushima is a major adsorbent in the soil collected from Iitate village (Mukai et al., 2014). On the other hand, the adsorption experiments performed in the laboratory to determine the Cs-adsorption ability of various clay minerals indicated that weathered biotite or vermiculite did not distinctively adsorbed Cs, compared to the other clay minerals, even at similar contamination level to that in Fukushima (e.g., http://reads.nims.go.jp/). This discrepancy suggested that actual adsorption of Cs in the soil was controlled by kinetics among multi-minerals and solution, rather than by simple equilibrium between mono-mineral and solution. Moreover, Cs adsorption experiments to minerals should be performed at a sub-ppm level in the minerals, employing the actual soil in Fukushima. Considering these ideas, we conducted a new Cs-adsorption experiment, in which various clay minerals were immersed together in dilute Cs-137 radioisotope solutions and the amount of Cs-137 adsorbed in each mineral was measured by autoradiography using an IP. It was found that a large quantity of Cs-137 predominantly fixed by the weathered biotite collected in Fukushima, compared to those by the neighboring clay minerals (unweathered biotite, illite, montmorillonite, kaolinite, halloysite, allophan, imogolite, etc.). Moreover, it was also observed that, with the increase of the immersion time, Cs-137 initially adsorbed in the other clay minerals moved to weather biotite, indicating that mineral species fixing the Cs-137 were changing with time. This phenomenon might be related to the fixation of radioactive cesium in the soil of Fukushima, which was reported recently.

Keywords: Fukushima nuclear accident, radioactive cesium, clay minerals, weathered biotite, adsorption, autoradiography

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MAG38-17 Room:301B Time:May 26 14:45-15:00

Characterization of mineral phases in radioactive particles collected from Fukushima contaminated soil using SR-u-XR

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MACHIDA, Akihiko[2] ; KURAMATA, Chisaki[1] ; KIKUCHI, Ryosuke[1] ; KIRAZAWA, Hideaki[3] ; KOGURE, Toshihiro[1]

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The behavior of radioactive cesium is one of important concern about Fukushima nuclear accident. Clay minerals are considered as major radioactive cesium adsorbent in soil from lab experiments reported by previous studies. However, it has been unclear what adsorbs radioactive cesium, because the amount of radiation in actual soil is too low to detect. Recently, Mukai et al. (2014) has detected the radioactive particles in actual soil using autoradiography with imaging plates (IPs). They characterized and classified these radioactive particles into three types using SEM-EDS: 1) weathered biotite, 2) aggregate of fine clay minerals, and 3) organic matter containing clay minerals particulates. On the other hand, it is not enough to identify mineralogical characteristic of particles adsorbed radioactive cesium using only SEM-EDS. Then, we identify and characterize mineral phases of radioactive soil particles using Synchrotron micro X-ray Diffraction (SR-μ-XRD) at BL22XU of SPring-8. After radioactive soil particles were separated by autoradiography and divided into above three types from SEM-EDS observation, each particle mounted on a kapton pin using micro manipulator for the XRD measurement. The incident X-ray with wavelength λ= 0.8273 $\mathring{A}$ at 15 keV were collimated to a diameter of 40 or 60 μm. Angle dispersive diffraction patterns were recorded on an imaging plate. Two-dimensional X-ray diffraction images on the IP were integrated as a function of 2θ to obtain conventional one-dimensional diffraction profiles. Spotty diffraction patterns of particles, characterized as 1) weathered biotite by SEM-EDS observation, show feature of single crystal. Broad peaks at ˜10 $\mathring{A}$, ˜14 $\mathring{A}$ and ˜7 $\mathring{A}$ appear at the low angle diffraction patterns of these weathered biotite particles. It indicates that these particles are weathered biotite with varied weathering degree from biotite, via vermiculite, to kaolinite. There is no clear relationship between the diffraction patterns of weathering biotite and amount of radiations estimated from luminescence on IP autoradiography. 2) The diffraction pattern of an aggregates minerals shows ring pattern ˜14.0 $\mathring{A}$ indexed smectite, 3) on diffraction patterns of a few organic matter containing particles, there are no clear diffraction pattern of clay minerals.

Keywords: Fukushima nuclear accident, radioactive cesium, autoradiography, clay mimerals, weathered biotite, SR-u-XRD

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MAG38-18 Room:301B Time:May 26 15:00-15:15

Distribution of radioactive cesium in contaminated Fukushima soil particles using FIB and IP autoradiography

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Recently radioactive particles in the contaminated soil collected from Fukushima have been specified by imaging plate (IP) autoradiography and characterized using electron and X-ray microanalyses (Mukai et al., 2014). These particles are several tens of micrometers in size and, in the next step, it is of importance that how radioactive cesium is distributed in the particles to discuss the states and dynamics of Cs in the soil. For this purpose, IP autoradiography itself is no longer useless because its spatial resolution is around a few hundred of micrometers at best. To overcome this limitation, we successfully applied micro-processing using the focused ion beam (FIB) technique along with IP autoradiography.

According to the results by Mukai et al. (2014), three types of the radioactive soil particles: (1) weathered biotite, (2) organic particle containing fine mineral particulate, and (3) aggregate of fine clay minerals, were investigated. The weathered biotite with plate-like morphology was divided into small pieces of a few micrometers using FIB and they were separated to be distinguished by IP autoradiography, using a micromanipulator. The autoradiography detected radiation from all pieces, indicating that radioactive cesium is distributed rather homogeneously in the biotite. Probably a solution containing radioactive cesium penetrated inside the crystal, through dense cleavage spaces which are well developed in the weathered biotite. On the other hand, one or two of several pieces from the organic particle were radioactive, meaning that the cesium is localized in the particle, probably at the mineral particulates and/or organic matter in the particles. Finally, the aggregate of clay minerals were crushed and spread on IP using the micromanipulator. Most fragments were radioactive, suggesting that the cesium is widely distributed in the aggregate.

Keywords: Weathered biotite, Fukushima nuclear accident, Radioactive cesium, Autoradiography, FIB

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MAG38-19 Room:301B Time:May 26 15:15-15:30

Measurement of radiocesium in spare seawater samples collected in marine monitoring for the Fukushima accident in 2011

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In March 2011 radionuclides, including radiocesium, were released from damaged Fukushima Dai-ichi Nuclear Power Plants (FNPP1) to the environment. It has been suggested that a large portion of radiocesium was transported into the North Pacific Ocean. However, its transportation process and total amount in the open are still unclear. Just after the accident, from 23th March 2011, Ministry of Education, Culture, Sports, Science and Technology (MEXT) started marine monitoring in a coastal area between 30 and 50 km approximately away from the FNPP1. Results of radionuclides (radiocesium, radiostrontium, and radioiodine) measurements had been published by MEXT (after April 2013 those are available on a web page of Nuclear Regulation Authority). Number of these observational data, however, is not enough to discuss transportation process of the Fukushima-derived radionuclides in the ocean mainly because of urgent and quick measurements during the marine monitoring. For example, radiocesium in the seawaters had been measured using a simple method by July 2011. As a result, about three months from May to July 2011 was a data-blank period in the open ocean within approximately 200 km from the FNPP1 because most of the measurement results were under detection limit. During the marine monitoring in 2011, spare seawater samples were also collected in some cruises conducted by research ships of the Japan Agency for Marine-Earth Science and Technology (JAMSTEC). We obtained these spare samples from JAMSTEC and measured radiocesium in them using a high-sensitivity method and present here spreading process of the Fukushima-derived radiocesium from May to July 2011 in the open ocean. The spare seawater samples (10 or 20 liters each) were collected during seven cruises using Niskin samplers. Radiocesium in the spare samples from four cruises (YK11-E02, NT11-E01, MR11-E02, and KR11-E04) was concentrated by evaporation or addition of ammonium phosphomolybdate and measured with low-background Ge-detectors at the Low Level Radioactivity Laboratory, Kanazawa University or Mutsu Institute for Oceanography, JAMSTEC. Uncertainty of the measurements, which was derived from sample preparation and analysis, was about 8%. In the early May 2011, high activity concentration (more than 500 Bq/m^3) of ^{134}Cs, which was released from the FNPP1 to the North Pacific though direct discharge of contaminated water in April 2011, was observed within about 50 km from the shore in off of Fukushima and Miyagi Pref. This north/southward spreading could be explained by prevailing north/southward coastal current in off of Fukushima Pref. On the other hand, a high activity concentration observed at a station located in 37.5N/142E about 100 km away from the FNPP1 implies eastward spreading of the Fukushima-derived ^{134}Cs due to a mesoscale eddy. In the early June 2011, the activity concentration decreased in off of Fukushima Pref. within about 50 km from the FNPP1 while the high activity concentration water had been spread to the Sendai Bay (Miyagi Pref.) and Kashima-Nada (Ibaraki Pref.). In addition, the high activity concentration water was transported eastward to about 200 km offshore (around 142.5E) along the Kuroshio Extension Current between 36.5 and 37N approximately. The southward and eastward wide-spreading in June could be derived from erosion of a warm mesoscale eddy off Ibaraki Pref. by the end of May 2011. In the early July, the high activity concentration of ^{134}Cs was observed only in the Sendai Bay and Kashima-Nada, except at stations nearby the FNPP1, suggesting that a main body of the ^{134}Cs contaminated water due to the direct discharge had been transported to outside of about 200 km from the FNPP1. Water-column inventory of the Fukushima-derived ^{134}Cs will be also presented in our presentation. This work was partially supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grant Number 24110005.

Keywords: accident of Fukushima Dai-ichi Nuclear Power Plants, radiocesium, marine monitoring

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MAG38-20 Room:301B Time:May 26 15:30-15:45

Estimation of total released 134Cs and 137Cs derived from TEPCO-FNPP1 accident into the North Pacific Ocean by

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The oceanic distribution of ^{134}Cs and ^{137}Cs released from the Tokyo Electric Power Company-Fukushima Daiichi Nuclear Power Plant (TEPCO-FNPP1) accident were investigated by using the optimal interpolation (OI) analysis. In this study, we conducted the OI analysis during the period from the end of March to the end of May 2011. A data set reported in research papers and documents was used for the analysis.

During the end of March to early April, extremely high ^{134}Cs and ^{137}Cs activities in seawater occurred along the coast near the TEPCO-FNPP1. The high activities spread to the region of 165^{o}E with a latitudinal center of 40^{o}N in the western North Pacific Ocean. Atmospheric deposition is also cause to high activities in the region between 180^{o}and 130^{o}W in the North Pacific Ocean. The total inventory of FNPP1-released ^{134}Cs in the North Pacific Ocean is estimated to be 15.2$\pm$1.8 PBq. In these, about half (8.3$\pm$1.8 PBq) of the total released amount is concentrated in the coastal region near the TEPCO-FNPP1. Considering that the direct released ^{137}Cs in the seawater was 3.5$\pm$0.7 PBq (Tsumune et al., 2012), atmospheric deposition contribute to the ^{134}Cs inventory in the near the TEPCO-FNPP1. The FNPP1-released ^{134}Cs inventory in the coastal region decreased exponentially with half-time of 4.2$\pm$0.5 days after cease of the direct discharge (6 April 2011) and become to about 2$\pm$0.4 PBq at the middle of May 2011. Considering that the ^{134}Cs/^{137}Cs activity ratios for the FNPP1 accident were very close to one and extremely uniform during the first month, the total amount of ^{137}Cs released by the TEPCO-FNPP1 accident reached to 20% of a current North Pacific inventory of bomb-derived ^{137}Cs injected in the 1950s and early 1960s.

Keywords: TEPCO-FNPP1, ^{134} Cs, ^{137} Cs, North Pacific Ocean, Optimal Interpolation analysis, Inventory

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Modeling dispersal of land-derived suspended radionuclides in the Fukushima coast

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Several oceanic dispersal modeling have been conducted on dissolved radionuclides leaked at the Fukushima Dai-ichi Nuclear Power Plant (FNPP). These models normally consider scenarios where the direct release of radionuclides from the FNPP and atmospheric deposition as the secondary source. In the present study, we view freshwater discharge from the rivers as a missing piece for the inventory of the radionuclides in the ocean. The land-derived input introduces a time lag behind the direct release through hydrological process because these radionuclides mostly attach to suspended particles (sediments) that are transported quite differently to the dissolved matter in the ocean. Therefore, we develop a sediment transport model consisting of a multi-class non-cohesive sediment transport model, a wave-enhanced bed boundary layer model and a stratigraphy model proposed by Blaas et al. (2007) incorporated into ROMS. A 128 x 256 km domain with the grid resolution of dx = 250 m centered at the FNPP is configured as a test bed within the existing dx = 1 km domain (Uchiyama et al., 2012, 2013). Three classes of sediments, viz., fine sand, silt and clay fractions, are considered here. The bed skin stress is evaluated by a combined wave-current stress model of Soulsby (1995) with the wave field computed with a SWAN spectral wave model at dx = 1 km embedded in the JMA GVP-CWM wave reanalysis. A total of 20 rivers inclusive of the 6 major rivers located in the domain are considered as point sources of the sediments. The daily-averaged freshwater discharges from the 20 rivers are evaluated with a surface runoff model HYDREEMS (Toyoda et al., 2009). Sediment volume fluxes from the rivers are then calculated with an empirical L'-Q' (discharge to sediment flux) relation proposed by Takekawa et al. (2013). Fraction of three sediment classes in the riverine discharge is determined empirically based on the outcome of a USLE-based river sediment modeling conducted by JAEA.

The developed model successfully reproduces the dispersal of the land-derived sediments and their recirculation processes associated with resuspension and deposition in the Fukushima coast for 4 months after the accident. The discharged sediments can be transported about 50 km from the shore with prominent patchiness of deposition and erosion near the mouth of each river. For instance, the offshore region of the mouth of the Niida River is evaluated to be erosion dominated, consistently with the measurement. Misumi et al. (2014) estimate suspended 137Cs concentration in the bed with considering static adsorption and desorption of 137Cs between the seawater and the bed sediments based on the dissolved 137Cs model result of Tsumune et al. (2012). The inferred bed 137Cs agrees well with the observation in the shallow area, whereas substantially underestimated in the offshore area at depth deeper than 200 m. They attribute the reason of the underestimate to the 137Cs due to sediment transport that is omitted in their model. We carefully diagnose our model results and find that although the clay-class sediments reach the deeper area, the time-integrated deposition is merely about 0.002 kg/m2 that is considered to be a minor fraction. Therefore, it is suggested that detritus and debris of organic matters rather than land-derived minerals likely cause the offshore 137Cs deposition. We further examine nearshore dispersal patterns and quantify the inventory of 137Cs attached to the sediments originated from the land by applying the empirical power law for 137Cs concentration as a function of specific surface area of the suspended particles proposed by Onda et al. (2014).

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

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134Cs and 137Cs in the North Pacific Ocean derived from the TEPCO Fukushima Dai-ichi Nuclear Power Plant accident

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We collected 2 - 10 litre surface seawater samples at more than 300 stations and water column samples were also collected at 24 stations.

The measured 137Cs concentration in a seawater sample at north 5 and 6 canal of the FNPP1 site reached 68 MBq m-3 on 6 April. It decreased to 1000 ? 10000 Bq m-3 during the period from August 2011 to July 2012 which means that direct discharge rate became very small, about ca. 100 GBq day-1. After July 2012, the activities of 137Cs in surface water at near FNPP1 site were still kept around 1000 Bq m-3 which corresponds about 10 GBq day-1.

After the FNPP1 accident, both 134Cs and 137Cs are observed in a wide area in the North Pacific Ocean and 134Cs activity and 137Cs activity ranged from 1000 +- 71 to less than 0.4 Bq m-3 and from 1080 +- 60 to 1.2 +- 0.2 Bq m-3, respectively. A zonal speed of FNPP1 derived radiocaesium in surface water at mid latitude in the North Pacific Ocean was 7 km day-1, 8 cm s-1, until March 2012 just after one year the accident (Aoyama et al., 2014). It after March 2012 till August 2014 was also estimated to be ca. 3 km day-1, 3.5 cm s-1 which showed apparent decrease of zonal speed. In 2013 and 2014, a maximum of Fukushima origin 137Cs activity in surface water was already close to pre-Fukushima level and observed at the eastern part of the North Pacific Ocean.

Until the end of 2011, a main body of Fukushima derived radiocaesium were existed at surface mixing layer, however, after winter cooling occurred subsurface maximum of Fukushima derived 134Cs and 137Cs due to subduction were observed at about 300 to 400 meters at 35 deg. N to 40 deg. N along 165 deg. E due to subduction. Southward transport due to subduction was also observed at 24 deg. N, 165 deg. E, too.

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Temporal change of Fukushima-derived Cs-137 in the sediments in the waters off Fukushima and nearby Prefectures

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The ^{137}Cs derived from the Fukushima Daiichi nuclear power plant accident in seawater has been transported to the open ocean with currents and its concentration has decreased exponentially from about 200 Bq/L to several mBq/L, a little higher than the pre-accident level, in the outside of 30 km radius of the nuclear power plant. The nuclide in the sediments, however, has not shown such a steep temporal change. Thus, its distribution pattern and temporal change should be systematically studied for a long time in order to access quantitatively the impact of the accident to the coastal environment. In addition, it is necessary to explore the mechanism for temporal change of the distribution in the sediments to predict the environmental recovery in the area. Soon after the accident, Marine Ecology Research Institute has been monitoring the water off Fukushima and nearby Prefectures for the radioactivities under the contract with Ministry of Education, Culture, Sports, Science and Technology (currently with the Nuclear Regulation Authority). We will introduce the distributions of Fukushima-derived ^{137}Cs in the bottom sediments and their temporal change, and factors controlling the changes.

Methods

In the coastal waters off Miyagi, Fukushima, Ibaraki and Chiba Prefectures, bottom sediments were retrieved using a multiple corer. From May to June 2011, sediments were collected at 12 sites six times. After that, the sites were increased. Now the sampling is done 4 times a year at 32 sites. Surface 3 cm of cores were used to study the surface distributions of ^{137}Cs. Vertical profiles of ^{137}Cs in the sediments were also studied at selected sites. Additional parameters such as sediment grain size, contents of organic material and elemental composition were also measured.

Results

The concentrations of ^{137}Cs in the surface sediments shows wide variation ranging from 0.8 to 540 Bq/kg-dry; Almost all of the data exceeded the level of pre-accident 5yr-average (0.87 +/- 0.41 Bq/kg) in the waters off Fukushima Pref. Proximity of the sampling site did not necessarily correspond to the higher values.

The temporal change of surface concentrations do not indicate steep decrease as that of seawater as a whole and the concentration at each sampling site varies so that it is not evident to see the temporal trend. However, geometric means calculated from Sept., 2011 to Nov. 2014 reveal the exponential decline trend from 47 Bq/kg to 17 Bq/kg.

Spatial variation of surface ^{137}Cs concentration and its temporal change would be ascribed to several factors. The former may be related to grain size distribution of the sediment, chemical composition, riverine input from the land, and pathway of polluted water after the accident, and the latter desorption of Cs from the sediments, resuspension and subsequent lateral transportation, and bioturbation. The temporal change of vertical distributions of ^{137}Cs do not show significant increase of the nuclide in the deeper layer, suggesting that bioturbation may not be the main factor for the surface decrease.

The detail of the spatiotemporal changes of ^{137}Cs in the sediments and the relevant factors will be presented in the talk.

Acknowledgements

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Keywords: Fukushima nuclear power plant accident, Cs-137, bottom sediment

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Temporal variation of ^{137}Cs in zooplankton and its primary factor in the waters off Fukushima and nearby prefectures

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The East Japan earthquake on 11 March 2011 and the ensuing tsunami resulted in the release of large amounts of radionuclides from the accident of Fukushima Dai-ichi Nuclear Power Plant (FDNPP) into the surrounding environment. Cesium has similar chemical properties with potassium, which is the essential element for living organisms, and is easy to be incorporated into living organisms. The half-life of radioactive cesium (^{134}Cs and ^{137}Cs) is 2 and 30 years, respectively. For the relatively long half-life of ^{137}Cs compared with ^{134}Cs, continuous monitoring of the levels of ^{137}Cs contamination is indispensable.

Zooplankton play a key role in marine biogeochemical cycle as secondary producers in the marine food web because they are major food for fishes and organisms of higher trophic levels. Therefore it is important to have information on their ^{137}Cs concentration and behavior of ^{137}Cs between zooplankton and surrounding environment such as seawater, surface sediment, and suspended marine particle.

Methods

In this study, zooplankton samples were collected at seven to eight sampling locations in the waters off Fukushima and nearby prefectures during May 2012 to January 2014. The ^{137}Cs concentrations in the samples were measured with Ge detectors and the compositions of zooplankton species were analyzed by microscopic observation. The zooplankton samples were obtained by horizontal towing at a depth range of 0-80 m using a large ring net. Seawater and sediment samples were also collected at the corresponding locations with zooplankton samples.

Results

Concentrations of ^{137}Cs in zooplankton ranged from 0.26 to 184 (Bq/kg-dry) during the sampling period (May 2012-January 2014). The concentration peaks did not appear concurrently at each station. In January 2014, concentrations of ^{137}Cs in zooplankton ranged from 6.59-40.3 (Bq/kg-dry) and they were still one or two orders of magnitude larger than those detected before the accident of FDNPP (0.09-0.4 Bq/kg-dry, Kaeriyama et al., 2008). The taxonomic composition varied seasonally and geographically but Maxillopoda were generally dominant throughout the study.

^{137}Cs in zooplankton are thought to be derived from surrounding environment such as seawater, surface sediments, suspended particle, and food. Since the ^{137}Cs concentration in surface sediments has not decreased so fast as that in seawater with time, ^{137}Cs in surface sediments might be incorporated into zooplankton by resuspension of surface sediments. Assuming that aluminum in zooplankton is only derived from surface sediments, the contributions of surface sediments to ^{137}Cs in zooplankton were estimated as 10%. ^{137}Cs in zooplankton in unit volume of seawater (μBq/m^3) were not correlated with both of ^{137}Cs in ambient seawater (Bq/L) and zooplankton biomass (mg-dry/m^3). On the other hand, the timings of high relative abundance of Appendiculata, Osteichthyes and phytoplankton corresponded to the timings of relatively high concentrations of ^{137}Cs in zooplankton. Therefore it is implied that variability in ^{137}Cs concentrations in zooplankton attributed to the composition of plankton species rather than concentration of ^{137}Cs in surrounding environment and plankton biomass.

Acknowledgements

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Reference

Kaeriyama, H., Watabe, T., and Kusakabe, M. (2008). ^{137}Cs concentration in zooplankton and its relation to taxonomic composition in the western North Pacific Ocean. Journal of environmental radioactivity, 99(12), 1838-1845.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident, ^{137}Cs, Zooplankton, community composition