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

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

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

Japan Geoscience Union Meeting 2014
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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

Japan Geoscience Union Meeting 2014
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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

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

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

Japan Geoscience Union Meeting 2014
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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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MAG38-10 Room:501 Time:May 2 11:30-11:45

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

Japan Geoscience Union Meeting 2014
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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

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

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

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

MAG38-15 Room:501 Time:May 2 14:15-14:30

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

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

MAG38-16 Room:501 Time:May 2 14:30-14:45

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

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

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

Japan Geoscience Union Meeting 2014
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MAG38-18 Room:501 Time:May 2 15:00-15:15

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

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

MAG38-19 Room:501 Time:May 2 15:15-15:30

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

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

MAG38-20 Room:501 Time:May 2 15:30-15:45

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)

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

MAG38-21 Room:501 Time:May 2 15:45-16:00

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

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

MAG38-22 Room:501 Time:May 2 16:15-16:30

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

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

MAG38-23 Room:501 Time:May 2 16:30-16:45

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

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

MAG38-P01 Room:Poster Time:May 2 16:15-17:30

Characteristics of radioactive Cs in reservoir sediment in Iwaki, Fukushima prefecture

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Large amount of radioactive elements, mainly Cs, were emitted from Fukushima Daiichi Nuclear Power Plant (FDNPP) because of Tohoku Earthquake occurred in March, 2011 and Fukushima prefecture and prefectures of the neighborhood were contaminated. Nuclear Regulation Authority, Japan (2013) reported that air dose rates evaluated based on the airborne monitoring results clearly show larger declines than those calculated based on the physical half-life of radioactive Cs. The reasons for such larger declines may include the effects of natural environmental erosion, such as rainfall. We have applied the sediment trap to sample the reservoir sediment. Sediment trap can observed the erosion continuously. Our purpose is to examine the characteristics of Cs contaminated soil continuously from summer to winter in 2013 in detail in Iwaki city, Fukushima prefecture.

Keywords: Radioactive Cs, Sediment, Erosion, Soil, Clay mineral

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

MAG38-P02 Room:Poster Time:May 2 16:15-17:30

Deposition and Migration of Radioactive Cs in the Matsukawa Ura and Feeder Rivers, Fukushima, Japan (Preliminary report)

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Radionuclides were released into the environment by the associated accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP). Radioactive Cs that are released from FDNPP and is deposited on the land will migrate to the ocean finally through the surface flow. In this study, we were intended to determine the actual transport of radioactive material in the system of river - estuary - ocean as a model area the feeder rivers and Matsukawa Ura located in Soma City, Fukushima Prefecture. Sediment sampling were continuously obtained from Matsukawa Ura and feeder rivers (Uda River, Koizumi River, Ume River and Nikkeshi River) from September 2013. The radioactivity of the Gamma ray nuclide was measured using a Ge semiconductor detector. Radioactive Cs activity in the Ume River and Nikkeshi River, which are located on the south side were higher than that in the Koizumi River and Uda River, located on the north side, because that reduced rainfall led to the increases in radioactive Cs concentration, except for the Nikkeshi River effected by heavy rain. Thus, it is thought there is a strong correlation between precipitation and radioactive Cs inventory of Matsukawa Ura, and the river flow in brackish area is dominant by the increasing precipitation which led to the increasing of flow rate, result in the river bed sediment inflowing to Matsukawa Ura. So it suggests that radioactive Cs activity has decreased because of increasing precipitation. In the Nikkeshi River, radioactive Cs activity was increased and sediment was changed to fine grain size at the same time after heavy rain as compared with before. This is considered that fine particles have been transported due to salt water intrusion during returning from overflow to the calm water after the heavy rain event. Transport situation of radioactive material in the river - estuary - ocean system revealed that physical and chemical process contributes significantly influence on it such as water flow and dynamics of fine sediment.

Keywords: Radioactive Cs, Matsukawa Ura, Brackish water area

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

MAG38-P03 Room:Poster Time:May 2 16:15-17:30

Rapid determination of Radiostrontium in seawater sample using DGA Resin

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A large amount of radionuclides were dispersed to the environment as a result of the accident at the Fukushima Daiichi Nuclear Power Plant in March 2011. Assessment of Sr-90, one of the major fission products, is crucial from the perspective of its bioavailability depending on behaviour similar to that of Ca, although few reports exist, so far. Traditional analytical procedures applies harmful huming nitric acid or large scale ion chromatography in order to separate between Sr and Ca prior to beta counting. In this study, rapid and robust purification technique for the daughter radionuclides yttrium-90 of Sr-90 using DGA chelating resin (Eichrom) without separation of Sr from Ca. DGA resin shows high distribution coefficient in high hydrochloric and nitric acid concentrations. Furthermore, we optimize the preconcentration method of Sr in seawater.

Keywords: Sr-90, Yttrium, Fuskuhima, Nuclear power plant, seawater

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

MAG38-P04 Room:Poster Time:May 2 16:15-17:30

Synchrotron radiation X-ray analyses of the radioactive single airborne particle emitted by the Fukushima nuclear accide

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The Fukushima Daiichi nuclear power plant (FDNPP) accident released radioactive materials into the air environment over the entire Northern Hemisphere in March 2011. In order to elucidate environmental transfer of the radioactive materials from the FDNPP accident, a large number of studies have been carried out until today. However, we still do not know the exact physical and chemical properties of the radioactive materials. Such knowledge is necessary to construct the numerical models to estimate the geographical distributions and to evaluate the human exposures during and after the FDNPP accident. Therefore, we studied the radioactive materials which were released in the air environment by a FDNPP accident based on the multiple SR (synchrotron radiation) X-ray analyses of the single airborne particles with strong radioactivity trapped in Tsukuba, Ibaraki Prefecture at the time of the FDNPP accident. The samples were the radioactive single particles collected on quartz fiber filter at the Meteorological Research Institute, Tsukuba using a high-volume aerosol sampler on March 14-15. We selected the radioactive single particles out of this filter using micromanipulator and transferred to the KaptonR tape on an acrylic plate for SR X-ray analyses.

SR experiments were performed at the beam line BL37XU of SPring-8. The monochromatic SR X-ray beams were focused to about 1 μm (horizontal) x 1 μm (vertical) by K-B mirror. Two excitation X-ray energies were selected depending on the target elements for analysis: i.e., 15.0 keV (low-energy mode) and 37.5 keV (high-energy mode). SR X-ray fluorescence (XRF) imaging was applied to obtain elemental distribution, and X-ray absorption near edge structure (XANES) analysis was used for chemical state analysis, and X-ray powder diffraction (XRD) analysis was carried out to obtain crystal structural information of the particles.

We have successfully analyzed three radioactive single airborne particles. XRF analysis has revealed the existence of Cs in all of them. We were able to detect various elements shown below depending on the excitation energy. In addition, XRF imaging shows that each element exhibited uniform distribution in the particles.

High-energy mode: Cs, Ba, Te, Sn, Mo, Zr, Rb, Zn, Fe

Low-energy mode: Fe, Mn, Cr, Zn, Ti

Each particle showed different chemical compositions. XANES analysis of Sn,Mo,Zn,Fe in the particles showed that these metallic elements existed in high oxidation states in glass matrix. Furthermore, XRD analysis shows that the particle was amorphous because no diffraction line was observed. These results suggest that the detected elements are components of materials constituting the reactor and fission products. It is presumed that the reactor materials including the nuclear fuel melted at a high temperature and quenched by releasing to the air environment as a glassy materials.

Keywords: Fukushima Daiichi nuclear power plant, Synchrotron radiation X-ray analysis, airborne particle, strong radioactive particle

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

MAG38-P05 Room:Poster Time:May 2 16:15-17:30

An approach to chemical reactions in the atmosphere

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1. Introduction

We discuss an approximate approach to simulate time series reactions in the atmosphere. At first, we write a reaction at definition time-t, as A+B=C. Next, we suppose that densities of the compounds are written by Gaussians. The Gaussian is a solution for general small particles diffusion processes. The time-t is discrete about the interval is dt. If 2 particles of compound A and B are interacted within the interval, the reaction reaches equilibrium, and a compound C is generated.

2. Descriptions

Considering properties of the atmosphere, we adopt Gaussian having different parameters for the horizontal and vertical directions.

$GA\{A\}(r,z)=QA\{A\}\exp\{-\alpha A\ (r-rA)^2-\beta A(z-zA)^2\}$, (1)

The suffix A corresponds to compound A. The Q is density and the unit is [M/volume] of compounds. In case of uncertain compounds chemically, it is replaced by [kg/volume]. A vector r is for x- and y-coordinates, and z is for z-coordinate. The function exp(whose arguments is 3-dimensional distance) is a kind of the volume. Eq. (1) is a relation of [M]; that is, a reaction equation, which is defined at any time.

The α and β (which are positive) are diffusion parameters and they depend with elapsed time from the generation. The dependency is very complex and the evaluation is difficult. In the puff-model approach, it is calculated by many turbulence parameters. However; we wonder that model is significant in case of very diffused case. We wish to adopt Lagrangian particles (L-particles), where alpha-beta-parameters are not, and effects of the turbulence are expressed by random numbers.

L-particles are a finite volume of the air, and have no shape. Therefore; we redefine it to be Gaussian. The multiply of Gaussians is a Gaussian; it is an appropriate function to express reactions.

Under the representation, alpha-beta-parameters are fixed coefficients to define a unit volume. They are a kind of mesh intervals. The re-defined Gaussians are moved by meteorological fields, as if they were L-particles. The Gaussian is like as a mesh-unit in Euler approach, which has a finite volume. They are in a space, and are moved by wind fields; however, they are not arranged orderly in Euler approach. Here, if the arrangement is introduced as following;

A transformation between L-particle and Euler-mesh:

Q(mesh coordinates)=Integral{GA(r,z)G(on mesh)dv},

{GA(r,z)}–>{Q(on mesh)}.

The transformation seems to be usable to evaluate diffused mist.

3. Reactions

In an interval time, chemical equilibrium is,

Keq=[C]/([A][B]). (2)

For every times,

QA(t+dt)=QA(t)-QC(t), QB(t+dt)=QB(t)-QC(t), (3)

rA (t+dt)=rA (t)+{u,v}Adt+Rand(), (4)

ZA (t+dt)=ZA (t)+{w}Adt+Rand(), (5)

Where, a vector {u,v,w} is wind speeds. Rand() is normal distributed random numbers.

In another reaction, A+B=C+D, we get,

Keq=([C][D])/([A][B]), (6)

Since the distributions of C and D are same at the first step,

$GC=GD=(KeqGAGB)^{0.5}$. (7)

4. Progress of the research

We try to simulate some reactions in the atmosphere now.

Keywords: atmospheric reaction, SPM, L-particle

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

MAG38-P06 Room:Poster Time:May 2 16:15-17:30

Secular distribution of radioactive strontium concentration in the atmosphere after after the accident of FD-NPP

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1.Introduction

On March 12, 2011, a large amount of radioactive nuclides have been released into the environment by the nuclear accident at the Fukushima Daiichi Nuclear Power Station. Measurement about radioactive nuclides will give us much information about the accident circumstance. Furthermore, radioactivities in the air dust are critical for estimation of internal exposure. There are many measurements results of I-131, Cs-134, Cs-137 in environment samples. However, in other nuclides, such as the pure beta emitter nuclide Sr-90 has not been measured sufficiently. Sr-90 is considered one of the harmful radioactive nuclides. Therefore, measurement of Sr-90 in the air dust is important for calculating exposure. We developed a new simple and quick strontium isolation technique using solid-phase extraction for determination Sr-90 in the air dust by liquid scintillation counter (LSC).

2. Method

In this study, we used 3M EmporeTM Strontium Rad Disk to extract strontium ion from air dust samples. This filter can collect Sr2+ ion efficiently. However, it is known that this filer also catches Pb2+. Natural radioactive nuclide Pb-210 seriously will be interferences in Sr identification in beta ray counting. In this study, cation exchange with EDTA adopted for Sr isolation. We made test experiments with radioactive Sr tracer and obtained that the chemical yield was about 90 %. The time for chemical operation was about 3-4 hours. To determine Sr-90, Cherenkov radiation of Y-90 has been measured by LSC, 1220 QUANTULUSTM Ultra Low Level Liquid Scintillation Spectrometer. With Sr-90 standard solution, we obtained that the Y-90 Cherenkov light detection efficiency was 68.7% and the Sr-90 detection limit was 0.004 Bq. With sequential measurement, the growth curve of Y-90 was described to determinate activity of Sr-90.

3.Results

We measured Sr-90 in the air dust samples of Fukushima, Hitachi, Kawasaki and Osaka. We chose some air dust samples that have high Cs-137 activity for Sr-90 measurement. Strontium isolation with solid phase extraction was performed. In Hitachi, the Sr-90 activity concentration in air is decreased with time and the ratio of Sr-90/Cs-137 is about 10-3. It is possible that after April, Sr-90 has been the same behavior of Cs-137. We observed a long time variation of Sr-90 air concentration in Hitachi and Fukushima and found that the Sr-90/Cs-137 activity ratio increased over time. We are going to discuss about behaviors of the Sr-90 and Cs-137 in the atmosphere.

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

MAG38-P07 Room:Poster Time:May 2 16:15-17:30

Correlation between Atmospheric Re-entrainment of Radioactive Cs and Meteorological Phenomena Conditions.

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1.Introduction

Massive earthquake attacked the eastern Japan on March 11 2011. It triggered the Fukushima Daiichi Nuclear Power Point accident, where large amount of radioactive substances were released. Released radioactive substances are diffused with atmospheric diffusion process, and eventually deposit on the ground surface and vegetation. Deposited radioactive Cs are released again from the ground surface and vegetation.

Today's main factor of atmospheric radiation concentration fluctuation is atmospheric Re-entrainment of radioactive Cs. Re-entrainment mechanism of radioactive Cs is a complex and unprecedented problem. We must consider an interdisciplinary study on deposited radioactive Cs for long-term estimation.

We infer that so Cs has a property that is taken in by clay minerals in soil that one of carriers of radioactive Cs is soil particles. The purpose of this study is to make clear how long does atmospheric radiation concentration increase by its re-entrainment, under what meteorological phenomena conditions.

2.About sampling

Since December 2012, we have been observing atmospheric radiation concentration of radioactive Cs by High-Volume Air Sampler on ground at Namie high school. It collects aerosols by passing through quartz filter. Wind velocity is measured at three altitudes by Three Cup Anemometer..Soil moisture is measured by Moisture Meter of Time Domain reflectometry system.

3.Correlation between seasonal re-entrainment of radioactive Cs and meteorological phenomena conditions

4.Investigation of direct transport by back-trajectory analysis

Keywords: Radioactive Cs, Atmospheric Re-entrainment, Fukushima Daiichi Nuclear Plant accident, Environmental Radioactivity

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

MAG38-P08 Room:Poster Time:May 2 16:15-17:30

Estimate of relationship between composition of aerosol and radioactive cesium observed in Namie Town, Fukushima Pref.

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Radionuclides emitted from the Fukushima dai-ichi nuclear power plant (FNDPP) 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 where is away 30km from FNDPP. We have set seven high-volume air samplers (HV) at the site and one operated for 24 hours day by day.

Then gamma-ray emission from HVsamples was measured with Ge detector.

In this way we have gotten atmospheric concentration of radiation which interval is one day.

While sampling, we measure atmospheric concentration of aerosol: black carbon, sulfate, and the number of particle which have size dependence using Electrical Low Pressure Impactor (ELPI).

We have analyzed the aerosols which had collected on HV filter with chemical analysis such as chromatograph.

We examined for correlation between the results of analysis and atmospheric concentration of radiation. And we examined what factor affects atmospheric concentration of radiation, and where the factor comes from using Positive Matrix Factorization (PMF). The PMF is multivariate analysis which estimates factor profile and factor contribution from observed value. The analysis needs only the observed value and number of factor (i.e. need not source profile), so there is possibility of finding the unexpected source.

This study used the date of March and August , 2013 .

Keywords: Fukushima daiichi nuclear plant accident, environmental radioactivity