Comprehensive Study on Atmospheric Radionuclides just after the Fukushima Accident by Analyzing used Filter-tapes of Air Monitoring Sites

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The first retrieval of hourly atmospheric radiocesium concentrations during March 12-23, 2011 just after the Fukushima Daiichi Nuclear Power Station (FD1NPS) accident was already published by measuring radionuclides in Suspended Particulate Matter (SPM) on the filter-tapes installed in SPM monitors with beta-ray attenuation method at 40 sites operated by local governments in the air pollution monitoring network of eastern Japan (Scientific Reports, 2014). Since then, hourly atmospheric radionuclides in SPM have been measured at around 100 SPM monitoring stations, and the dataset has been also published on a website (Journal of Nuclear and Radiochemical Sciences, 2015). In this paper, we will introduce new findings by the synthetic analysis of not only those data but also radiation dose rates at many monitoring sites, atmospheric radionuclides independently measured at several sites, and meteorological data by Japan Meteorological Agency. From the FD1NPS, 7 and 6 plumes/polluted air masses with high radionuclides were found to be transported to southern Tohoku region including the Fukushima prefecture, and to the central Kanto region including Tokyo Metropolitan Area located more than 170 km southwest of the FD1NPS, respectively. Many peaks of radiation dose rate were already observed at many monitoring stations, and a one-to-one correspondence could be identified between a peak of radiation dose and a plume/polluted air mass of radionuclides. Many case studies on the transport of plume/polluted air masses have been also made, and which clearly demonstrates that local meteorological conditions such as land and sea breezes, precipitation, and temperature inversion layers near the surface coupled with topography could greatly affect the transport pathways of radioactive materials, their maximum concentrations, and their deposition to the land surface.

Keywords: Atmospheric Cs-137, Suspended Particulate Matter, Fukushima Daiichi Nuclear Power Station, plume, Spatio-temporal variation

Investigation of Physical and Chemical Natures of Particulate Radioactive Matters Emitted from the Fukushima Nuclear Accident

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We have investigated particulate matters containing radionuclides emitted by the Fukushima Daiichi Nuclear Power Plant (FDNPP) Accident found in the environment. In this study, we summarize detailed physical and chemical natures of radioactive particles found by our investigations.
Radioactive particles were sampled from aerosols collected at two institutions in Tsukuba, the Meteorological Research Institute (MRI) and the National Institute of Advanced Industrial Science and Technology (AIST), in March 2011. The detailed sampling procedures were described elsewhere[1)]. We also sampled radioactive particles from a sediment of an outdoor pool and soils collected in the Fukushima Prefecture. They were subjected to a scanning electron microscopy with energy-dispersive X-ray spectroscopy and a gamma-ray spectrometry. The synchrotron radiation (SR) experiments were nondestructively carried out at BL37XU, SPring-8. The SR-X-ray microbeam was used as an analytical probe for chemical composition analysis by X-ray fluorescence spectroscopy (SR-μ-XRF), chemical state analysis by X-ray absorption near edge structure analysis (SR-μ-XANES) and crystal structure analysis by X-ray diffraction (SR-μ-XRD).
Analytical results suggest a presence of three groups (Groups A, B and C) of the radioactive particles having different physical and chemical natures as described below. Group A particles are almost spherical with diameters of 1~5 μm. They were first found in aerosols collected at MRI during March 14th and 15th[1)] 2011. In addition to radioactive Cs, various heavy elements (Rb, Zr, Sn, Ba etc.) are detected in common to Group A. These heavy elements could be derived from fission products of the nuclear fuel. The Group A particles sometimes contain U which might originate from the fuel[2)]. In addition to heavy elements from the fuel, they consist primarily of Si, Fe and Zn associated with construction materials of the reactor. They are insoluble in water and could have a long-term impact on the environment because they are Si-based glassy materials[2-4)]. It is highly possible that they were emitted from the reactor No.2 or 3 of the FDNPP. However it is difficult to assume a single process of preparation and emission for all Group A particles because their chemical composition is inhomogeneous.
Group B particles were found in soils of northwestern region of the FDNPP. It is pointed out that radionuclides emitted from the reactor No.1 fell on this region[3)]. In contrast to Group A, the Group B particles are large (>100 μm) and non-uniform shaped. While their matrix is the Si-based glass like Group A, there is a significant difference in chemical composition between Groups A and B. Several metal elements such as Fe, Mo, Sn and U were concentrated into micro region. Results of the SR-μ-XANES/XRD indicate the presence of non-glass phase in the concentrated region.
Group C particles were sampled from aerosols collected at AIST in March 30th 2011. Although size of these particles are similar to Group A, their shape is not spherical but rugged. In contrast to Groups A/B, Si is not a major component of the Group C particles. There is clear distinction in heavy elemental composition between Group C and Groups A/B. It is expected to that these differences in physical and chemical natures among three groups of radioactive particles were derived from the difference in process of preparation and emission. Therefore this study demonstrated the fact that particulate radioactive matters were emitted from the FDNPP into the environment several times through different processes.

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Keywords: Fukushima Nuclear Accident, Radioactive particles, Aerosol, Soil, X-ray analysis, Synchrotron radiation

Long-term assessment of airborne radio-cesium after the Fukushima nuclear accident: re-suspension from soil and vegetation

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Long-term assessment of ^{137}Cs re-suspension from contaminated soil and vegetation due to the Fukushima nuclear accident in March 2011 and the on-going emission from the premises of the power plant has been conducted using a numerical simulation, a field experiment on the dust deflation at Namie in the restricted habitation area, and air concentration measurements in and out of the area, Namie and Tsukuba, respectively. The analysis period is one year from December 2012, about one and a half years from the accident, up to December 2013. The surface concentration of Cs-137 at Namie was high in the summer (~1 mBq/m^3) and low in the winter (0.1-1 mBq/m^3). The $_{137}$Cs concentration was about one order smaller in Tsukuba (0.01-0.1 mBq/m^3). The differences in the two sites are consistent between the observation and the simulation. Ishizuka et al. (2016) developed a numerical module of ^{137}Cs re-suspension associated with dust deflation based on the flux measurement in Namie. Using the module, the simulated ^{137}Cs from soil had a potential to account for the observed surface concentration in Namie in the winter, but underestimated by 1-2 orders of magnitude in the summer. The Tokyo Electric Power Company assessed the ^{137}Cs emission from the reactor buildings in 2013 as approximately 10^6 Bq/h. By using the emission rate, the simulation substantially underestimated the observation by 2-3 orders of magnitude in Namie. We simulated the re-suspension from vegetation applying a seasonal variation as a function of the green fraction map. With the constant re-suspension rate of 10^{-7} [/h], the simulated vegetation re-suspension quantitatively accounted for the observed surface concentration together with its seasonal variation. Still, so far, the re-suspension mechanism has not been fully understood and thus further investigations for the understanding of the mechanisms and its long-term effects on the environment are needed.

Keywords: Dust deflation module, Three dimensional numerical model, Atmospheric measurement, Budget analysis

Re-suspension processes of radioactive Cs emitted by the FNDPP accident in summer and autumn –possibility of biosphere-atmosphere circulation of radioactive Cs

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Radionuclides emitted in the Fukushima dai-ichi nuclear power plant (FNDPP) accident have been deposited on the soil, ocean and vegetation. Re-suspension of radioactive cesium (Cs) from the soil and vegetation to the atmosphere may be one of significant path in the diffusion of radionuclides after the accident. As well as continuous monitoring of radionuclides in the atmosphere, understanding of the re-suspension processes of radioactive Cs to the atmosphere is essential to predict its environmental influence now and future.
We are monitoring the radioactivity concentration of atmospheric Cs-134/137 at several sites at Tsushima and Yamakiya, Fukushima, where deposition amount of Cs-134/137 is relatively high. Atmospheric suspended particle are collected with high-volume air samplers mounted at these sites, and gamma-ray emission from them were measured with Ge detector. The measured concentrations of atmospheric Cs-134/137 at Tshushima indicated their seasonal variation: they increased in spring, attained their maximum in summer, and decreased in autumn.
SEM observation of atmospheric samples showed that organic particles were dominant in summer and autumn. Fraction of carbon in the samples was positively correlated with activity concentration of Cs-134/137, indicating that these organic particles bring Cs-134/137 in the atmosphere. Chemical analysis of the atmospheric samples shows some organic components from biogenic origin were rich in their fraction of high rad Cs-134/137 activity. Bio-aerosol sampling was also conducted and its preliminary result showed that many biogenic particles such as spores and bacteria found in the samples. Comparison of activity of Cs-134/137 in rainwatrer collected inside and outside broad-leaf forest in Tsushima showed that Cs-134/137 activity was several times higher in rainwater in forest. Atmospheric activity was also increased during rain. These results showed that Cs-134/137 was rich on the surface of vegetation, and that it moved from vegetation to the atmosphere and soil by rain.
Water soluble components and insoluble organic components were extracted from atmospheric particle samples obtained in summer and autumn with ultrapure water and H2O2, respectively. Simultaneously, 40-65% of Cs-134/137 was extracted with pure water and 10-40% of Cs-134/137 was extracted with H2O2. Although water soluble aerosols such as NaCl and sulfates were included in the samples, we suppose that Cs-134/137 extracted by water was contained in the organic particles because Na and S was much less than C in the samples. We cultured poplar shoots in the solution obtained from the extraction of atmospheric samples by pure water during 48 hours, and found that similar amount of Cs-134/137 existed in roots of poplar, its leaves and branches, and remained solution after the cultivation although major part of Cs-134/137 was adsorbed grass beads used in the cultivation. These results strongly suggested that Cs-134/137 is circulating among vegetation, soil, and atmosphere.

Keywords: Fukushima-dai'ichi Nuclear Plant Accident, Radioactive Cesium, Radioactivity in environment, Biosphere-atmosphere Curculation

Effects of radiocesium transfer from the canopy to forest floor on its accumulation in litter and soil layers.

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Radiocesium deposited on forest area is initially intercepted by forest canopy and subsequently deposits on forest floor in association with rainwater and litter-fall. The intercepted radiocesium by the canopy acts as a source of secondary radioactive contamination of forest floor, however those effects on the accumulation of radiocesium to litter and soil layers have not been assessed quantitatively.
We investigated the transfer of canopy-intercepted radiocesium to the forest floor during 4 years following the Fukushima Daiichi Nuclear Power Plant accident. The cesium-137 (^{137}Cs) contents in throughfall, stemflow, and litterfall were monitored in two coniferous stands (plantation of Japanese cedar) and a mixed deciduous broad-leaved forest stand (Japanese oak with red pine). We also measured the gamma count rate of radiocesium at the forest floor using a portable Ge gamma-ray detector.
Total Cs-137 deposition flux from the canopy to forest floor for the mature cedar, young cedar, and the mixed broad-leaved stands were 166 kBq/m^2, 174 kBq/m^2, and 60 kBq/m^2, respectively. These values correspond to 38%, 40% and 13% of total atmospheric input after the accident. The spatial pattern of radiocesium at the forest floor have not changed during monitoring period, suggesting that radiocesium partitioning and leaching by the forest canopy is rather constant over time. We investigated temporal change of radiocesium inventory in litter and soil layer in the study site (Takahashi et al., 2015; NRA, 2015), which was later compared with the radiocesium depositional flux onto forest floor. The radiocesium inventory in litter layer decreased with time in all the forest sites, although the radiocesium continuously deposited on the forest floor (~ 400 Bq/m^2 /day). The radiocesium migration rate from the litter layer to the underlain mineral soil layer was estimated based on the analysis of the measured temporal changes of depositional flux to forest floor and inventory in litter and soil layers.

Keywords: Radiocesium, Forest, Transfer, Litter layer

How much the radiocesium fallout remain in forest surface soils?

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Radioactive contamination in the forested area was caused by the ^{137}Cs discharged from Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. To predict longterm redistribution of fallout ^{137}Cs, we investigated global fallout ^{137}Cs (^{137}Cs-GFO) from nuclear weapon tests in the atmosphere in the 1950s and 60s. We examined concentrations and amounts of ^{137}Cs-GFO of three layers of soil samples (0–5, 5–15 and 15–30 cm in depth) at 316 sites all over Japan, which were collected just before the accident. We determined ^{137}Cs-GFO by NaI well-type scintillation counter with its accuracy verified by measurements using a germanium detector. An average of ^{137}Cs-GFO inventories of forest soils in Japan was estimated to be 1.7 ±1.4 kBq m^{-2} as of Oct. 2008 with highly nation-wide spatial variation. Otherwise district meteorological observatory estimated accumulated ^{137}Cs-GFO deposits to be 2.4 ±0.8 kBq m^{-2}. Since the number of measurement by district meteorological observatory was limited (n = 7), we have to compare these data with regards to spatial variation of the initial deposit possibly caused by climate condition. Climate factors used were precipitation normal value, vertical variation of tropopause height, and special rainfall events within atmospheric nuclear test peak period. The precipitation is positively correlated with ^{137}Cs-GFO deposit. The vertical variation of tropopause height affects the ^{137}Cs concentration of atmosphere in the troposphere. Influence of atmospheric nuclear test on the ^{137}Cs concentration of atmosphere peaked from Dec. 1962 to June 1963. Special rainfall events of this period might have a great impact on the ^{137}Cs-GFO deposit. Winter precipitation of the precipitation normal value (October to February) had been explained well the large amount of the ^{137}Cs-GFO distribution trend with greater accumulation in the north-western part along the Sea of Japan side. We made, you create a general mixed model using winter precipitation as a fixed effect and survey points as random effects (model 1). The tropopause altitude daily observations in 2014 were extrapolated to 0.5 degrees mesh, which turned differential data of the one-day intervals and two day intervals, and the increase in tropopause height was extracted. We added monthly frequency of the increase events as an explanatory variable to model 1 (model 2). The model 2 improved estimation accuracy in Akita pref. and Kitakyushu region underestimated in model 1 and in Okinawa pref. and Tokai region overestimated in model 1. To consider the impact of special rainfall events, the AMeDAS point monthly precipitation (1950-1964) was extrapolated into a 0.1-degree mesh data set using the spline interpolation. Precipitation from Dec. 1962 to Feb. 1963 (a heavy snowfall event) was added an explanatory variable to model 1 (model 3). The model 3 remedied the ^{137}Cs-GFO underestimation in some prefectures (Akita, Saga, Fukui and Nagasaki). Within the frame of the above three models, the ^{137}Cs accumulation in forest soils were compared to the cumulative amount of deposit measured in the district meteorological observatory. All of the three models indicated that there was no significant difference in the amount of the ^{137}Cs between the forest soils and the district meteorological observatories. We concluded that most of the ^{137}Cs-GFO had remained in forest soils during the 60 years.

Keywords: Cs, Forest soil, inventory

The situation of radioactive contamination in crops after five years of FDNPP accident

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Tokyo electric power company’s Fukushima Daiichi Nuclear Power plant (FDNPP) accident affected a large area of Eastern Japan by the fallout of radioactive cesium. In 2011, more than 8000 ha of agricultural field was restricted planting and the area decreased about 2000 ha by the end of 2014. Two major protocols have been applied to the agricultural field, one is decontamination of field mainly by surface stripping method but it results a huge amount of radioactive waste of soil and biomass (more than 2,000, 000 m3). The other method is applying sufficient amount of potassium to the soil before conventional fertilization. The problem is that it is not able to decide the termination of applying excess amount of potassium to the field. Furthermore, as some plant species seems to have higher transfer factor, it makes the radioactive cesium content of the harvest higher than the standard limit in food (100 Bq/kg from April 2012, in Japan). Several countermeasures have been tried to encounter these problems. In the presentation, present situation of Fukushima area which have been affected by radioactive materials and how the agriculture has been reconstructed after the FDNPP accident.

Keywords: Radioactive cesium, Decontamination of radioactive cesium, Potassium

Spatial distribution of dissolved Cs-137 at small Headwater Forested Catchment in Fukushima after Fukushima Dai-ichi Nuclear Power Plant Accident

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Radiocesium migration from headwater forested catchment is important perception as output from the forest which is also input to the subsequent various land use and downstream rivers after Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. In this study, dissolved Cs-137 concentration of stream water, soil water and groundwater were measured. Observations were conducted at headwater catchment in Yamakiya district, located 35 km northwest of FDNPP from April 2014 to November 2015. Stream water discharge was monitored and stream water samples were taken at main channel and sub channel. Stream water discharge was monitored by combination of parshallflume and v-notch weir. Stream water was sampled manually at steady state condition in 3-4 month interval and also intense few hours interval sampling were conducted during rainfall events using automated water sampler. Around the sub channel, it is found that there is a regularly saturated area at the bottom of the slope, temporary saturated area which saturate during the rainy season in summer and regularly dry area. 6 interval cameras were installed to monitor the changing situation of saturated area. Suction lysimeters were installed at three areas (regularly saturated area, temporary saturated area and dry area) for sampling soil water in depth of 0.1 m and 0.3 m. Boreholes were installed at three points along the sub channel. Three boreholes with depth of 3 m, 5 m and 10 m were installed at temporary saturated area, 20 m upstream of sub channel weir. Another three boreholes with depth of 3 m, 5 m and 10 m were installed at dry area, 40 m upstream of sub channel weir. And a borehole with depth of 20 m was installed at ridge of sub catchment, 52 m upstream of sub channel weir. Groundwater was sampled by electrically powered pump and groundwater level was monitored. Also suction-free lysimeter was installed at temporary saturated area for sampling the near surface subsurface water. Soil water samples were collected as much as collected in flask. Stream water and groundwater samples were collected for 40 L each. All the water samples were filtered through 0.45 μm pore-size membrane. Water samples with less than few L were concentrated by evaporative concentration. Water samples with more than 40 L were concentrated using the ammonium molybdophosphate (AMP)/Cs compound method. The Cs-137 concentration was determined using Gamma-ray spectrometry with a germanium semiconductor detector.
Spatial distribution of dissolved Cs-137 concentration in the slope was obtained and the source of Cs-137 concentration in stream water was examined. The Cs-137 concentration in groundwater showed low value of around 0.001 Bq/L. The Cs-137 concentration of soil water showed 0.01-0.1 Bq/L. And Cs-137 concentrations of stream water were in order of 0.01-0.1 Bq/L at steady state condition. Also Cs-137 concentrations in stream water showed temporary increase during rainfall event. The source of dissolved Cs-137 was suggested to be shallow soil water under saturated condition or leaching from the litter might be affecting.

Keywords: Dissolved Cs-137, FDNPP, stream water, soil water, groundwater

Slope-scale Cs-137 wash-off processes estimated with erosion plot observations and laser-scanning

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To predict the fate of Cs-137 deposited on terrestrial due to the accidents of nuclear power plants, understanding of Cs-137 redistribution associated with sediment dynamics is important. Previous studies quantified slope-scale Cs-137 wash-off from various land uses using soil erosion plots and the results were useful for parameterizations of large-scale predictions. However, temporal trends of Cs-137 wash-off has hardly been elucidated because of lack of related long-term observations. It adds uncertainty in predictions of the fate of Cs-137. Previous studies relating to Cs-137 redistribution indicated that Cs-137 concentration of sediments and hence its redistributions depend on erosional processes on slopes, such as rill formation and deposition. Elucidating relation between soil erosion and Cs-137 wash-off processes will improve our understanding of fate of Cs-137. This study presents a Cs-137 wash-off observation and a series of morphological survey on a soil erosion plot established in area affected by the accident of Fukushima Dai-ichi Nuclear Power Plant. Based on these results, we discussed the seasonality of slope scale Cs-137 wash-off. A soil erosion plot with length of 22.13 m and width of 5 m was established on a tobacco farmland in Yamakiya district of Kawamata town on July 2011. The initial Cs-137 deposition was 370 kBq m-2 and the slope gradient was 4.4 degree on the plot. Eroded sediments were collected every one month in winter and every two weeks in other season until August of 2014. Collected sediments were dried, weighed and then served for Cs-137 measurements. Precipitation and surface runoff were also monitored. Surface soil was scanned for ten times using a 3D laser profiler during the observation. Obtained scan data were converted into 1 cm mesh DEM and the differences of the elevation were calculated by subtracting elevation values from those obtained in the previous scanned. During the observation, total amount of eroded sediment was 9.8 kg/m^2 and total Cs-137 wash-off was 107 kBq/m^2. Mean concentration of Cs-137 in eroded sediment was 13 kBq/kg with 38 % of variation coefficient. A decreasing trend in temporal variations in Cs-137 concentration was found but not significant. However, Cs-137 concentration gradually decrease from spring to autumn and it recovered as high as increased again as high as those after winter. Morphology of surface soil showed seasonality. Rill was expanded from spring to summer. There was no significant change from summer to autumn. Soil surface was elevated from winter to spring. Averaged difference of elevation was negatively correlated with amount of eroded sediment. No significant correlation was found between the average difference of elevation and Cs-137 concentration of eroded sediment. However, average difference of elevation during warm season (from spring to autumn) was negatively correlated with Cs-137 concentration of both coarse and fine sediment, whereas the positive correlation was found in winter (from autumn to spring). These relationships suggest the difference of erosion and Cs-137 processes between winter and other seasons. In winter, freezing and thawing cycles provided sediments of high Cs-137 concentration and subsequent snowmelt and rainfall could wash off the detachable sediment. In other seasons, rill was a dominant pathway for transporting sediments with relatively low Cs-137 concentration whereas intensive rainfall event expanded the contributing area of sediment with relatively high Cs-137 concentration. The results of this study suggest that Cs-137 wash-off shows seasonality and

consequently long-term observations will be necessary for more accurate predictions of Cs-137 fate.

Keywords: Laser scanning, Soil erosion, Cs-137

Landscape Controls Fate of Fukushima Contamination

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The Fukushima Daiichi Nuclear Power Plant accident has released massive amount of radiocesium into the terrestrial environment, and the radiocecium have been moved through rainfall and erosional processes. In Japan, intensive field montoring campaign has been started and continued for 4 years. The detailed monitoring of concentration of radiocesium and their flux, which can be applicable for the fate and flux of the radionuclide flow in humid environment espeacially controlled by the landuses of upstream.

Keywords: Cs-137, Fukushima, watershed

Uranium isotope ratio in Fukushima soil samples

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There were huge amounts of radionuclides such as fission products released into atmosphere due to accident of Fukushima Daiichi nuclear power plant (FDNPP). A few studies have been reported about actinide elements released from FDNPP accident as fingerprint of isotope ratio using accelerator mass spectrometry (AMS). Levels of actinides were at much less concentration than ^{134}Cs, ^{137}Cs and ^{90}Sr. However, we need to consider the influence of alpha ray particles on a long-term contamination in environment. We have focused on 236U isotope, and its measurement using a thermal ionization mass spectrometry (TIMS), which may be used as an index of a nuclear accident. We measured highly precise uranium isotope ratios in the soil samples from Fukushima prefecture with high Cs concentration using Isotopx Ltd. Phoenix TIMS.
We also measured Kobe and Okinawa soil samples before the accident as Global Fallout and geological standard sample as JSd-2 and NIST 4350b. Chernobyl and Kosovo samples were measured to notice artificial radioactive materials.
Soil samples were decomposed by a microwave (ETHOS one) digestion method with mixed acids after ashing. Uranium fraction from the dissolved sample was chemically separated by UTEVA-UTEVA resin chromatography to minimize interferences of Fe and Pb prior to isotope ratio measurement using TIMS and MC-ICP-MS.
Results of ^{235}U/^{238}U and ^{234}U/^{238}U ratio by TIMS and MC-ICP-MS couldn't reveal any significant difference between Fukushima soil samples and Global Fallout samples.
^{236}U/^{238}U detection limit of TIMS were considered by a standard solution from U ore and could be considered that was not under the influence of Global Fallout. We could measure ^{236}U/^{238}U ratio in U ore samples from 10^{-10}~1.5×10^{-9} range. The yield for JSd-2 and NIST 4350b were noticed to be $4.2\text{-}7.8\times10^{-8}$ and Kobe and Okinawa as an index of global atomic fallout were $6.5\text{-}9.8\times10^{-9}$. The detailed results will be presented.

Keywords: Fukushima soil sample, Uranium isotope, TIMS, 236U

Development of meta-database of radiation and radioactivity monitoring data for Fukushima Dai-ichi reactor accident

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Great East Japan Earthquake happed 11 March 2011 caused Fukushima Dai-ichi reactor accident resulting significant radiation contamination to the environment. There have been long standing efforts for assessment of early exposure dose of the regident by environmental simulations together with various radiation monitoring data during the sequence. We report the meta-data archive system for the radiation/radioactivity monitoring for Fukushima Dai-ichi reactor accident. This idea will serve meta-database search of various radiation monitoring data and archiving.

Keywords: Radiation, Meta-database, Fukushima Dai-ichi reactor accident

Survey on riverine Cs-137 transport in Fukushima prefecture

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In Fukushima prefecture, river water is widely used for city water, agricultural water, and other purposes. Although the radioactivity in river water already declined enough to drink it, the activity and flux of radiocaesium should be observed in order to evaluate effects on the air dose rate due to deposition while flood events and transfer from agricultural products, wild animals and plants via ecosystem.
Suspended sediments samples were collected at 30 sites in Abukuma river system and Hamadori districts, and the discharge and turbidity were measured at the same sites. And then, particulate radiocaesium activities and fluxes were calculated.
Although the declining tendency of the radioactivity of particulate Cs-137 has been continuing, the declining rate slowed down after one year from the accident. In observation sites on Kuchibuto tributary, in which decontamination works were conducted by Ministry of the Environment, the radioactivity became lower after spring in 2014. The decline was probably caused by the decontamination of agricultural land (After stripping surface soil, soil without contamination was brought in and added).
In September 2015, a heavy rainfall event occurred. In lower reach of Abukuma River, the water discharge was at almost the same level as the event caused by typhoon Roke in September 2011. However, the particulate Cs-137 flux was one order to magnitude lower than that at September 2011, because the Cs-137 activity concentration decreased.

Keywords: Radiocaesium, River

Relationship between radiocesium interception potential (RIP) and other parameters such as cation exchange capacity, organic matter content, particle size, and mineral composition

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The solid-water distribution of radiocesium can be expressed by the distribution coefficient (Kd), but the Kd value is conditional depending on the condition of the solution such as major ion composition and concentration of cesium in the aqueous phase. On the other hand, radiocesium interception potential (RIP) is primarily important to show the solid-water distribution of radiocesium, because of the low total concentration of cesium in aqueous environment, where frayed-edge site is not saturated by the cesium.
In this study, the effects of cation exchange capacity (CEC), organic matter content, particle size, and mineral composition on RIP were discussed. Based on the laboratory studies using suspended sediment (SS) samples collected from various rivers in Fukushima area, we found the results from (i) to (iv):
(i) RIP is positively correlated with CEC, but some RIP values at higher CEC region were lower than those at lower CEC region, because the latter values were obtained from the SS with high organic content.
(ii) RIP is positively correlated with surface area, or negatively correlated with average particle size of SS samples. However, some RIP values with large surface areas were out of the trend written above, possibly because the smaller particles did not contain weathered mica that can strongly adsorb cesium.
(iii) However, the correlation of RIP against mineral composition indicated by the mica/quartz ratio is not very clear, suggesting that the role of bulk mineral content is not very clear. It is possible that the capacity needed for the cesium in water is not that large, which suggests that a small amount of mica-type mineral is sufficient to show the high affinity for cesium.
(iv) Finally, there was clear negative correlation between RIP and the organic content. This effect is explained by the coating of minerals by humic materials, which inhibits adsorption of cesium on the SS samples, as suggested in our previous study (Fan et al., 2014).
We found that the four parameters more or less affect the RIP value. Thus, multiple regression equation is needed to take into account the effects of the four parameters to obtain empirically the RIP value in each environment. We will give the results of the multiple regression equation in the presentation, by which we can discuss important parameters that control adsorption behavior of radiocesium in Fukushima area.

Keywords: Fukushima Dai-ichi nuclear power plant, RIP, cesium, cation exchange capacity

Source Identification and Simulation of Radiocesium Infiltration into Separate Sewer System after Nuclear Power Plant Accident

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Following the Fukushima Dai-ichi nuclear power plant accident, numerous amount of radiocesium was emitted and infiltrates separate sewer system in the surrounding urban areas. To simulate the infitration of radiocesium into separate sewer, we developed the Improved Model Radionuclide Migration in Urban Environments and Drainage Systems (iMUD) bases on model Radionuclide Migration in Urban Environments and Drainage Systems (MUD), which was only applicable for combined sewer system. In this study, we attempted to predict the concentration of radiocesium in the final sludge of wastewater treatment plant (WWTP).
iMUD is a multi compartment model, consits of urban and WWTP sub-model and divides the surface of urban area into five components according to the type of surface layer (roof, paved, soil, tree, wall). Firstly we identified the mechanism of radiocesium infiltration, which after several analysis, the mechanisms are Rainfall-Derived Inflow and Infiltration (RDII) and Human excretion. There after, the model was applied on the two largest urban areas in Fukushima Prefecture, Fukushima and Koriyama. Finally, we compared the predicted values of concentration of radiocesium in the sludge with the three years obeserved data in order to validate the model.
Based on the calculation of Nash Efficiency Coefficient (n), the model showed a satisfactory result, which for Fukushima WWTP case n value of 0.85, and for Koriyama WWTP case, n value of 0.84 were achieved. In addition, R^2 value of 0.85 and 0.86 were achieved for Fukushima and Koriyama respectively. We predicted that the sludge containing radiocesium reaches the standard limit after 3 years for Fukushima and 4 years for Koriyama.

Keywords: radiocesium, separate sewer system, simulation

Fukushima-derived radiocesium in the North Pacific subarctic region and Arctic Ocean in summer 2014

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Accident of Fukushima-Dai-ichi Nuclear Power Plant (FNPP1) on 11 March 2011 resulted in a large amount release of radiocesium (^{134}Cs and ^{137}Cs) into the North Pacific Ocean. Oceanographic observations have revealed that the Fukushima-derived radiocesium in surface seawater is transported eastward along the North Pacific Current. However, penetration of the radiocesium into ocean interior is not understood well. From July to October 2014 we measured vertical profiles of radiocesium from surface to 800 m depth in seawater from the North Pacific subarctic region and Arctic Ocean. The seawater samples (20-40 liter) were collected using a bucket or a pump for surface water and Niskin Sampler for deeper water. Some of them were filtrated and all the samples were acidified with nitric acid on board. In laboratories on shore, radiocesium in the sampled seawater was concentrated onto ammonium phosphomolybdate (AMP). Radiocesium in the AMP was measured using ultra-low-background gamma-ray detectors in Low Level Radioactivity Laboratory, Kanazawa University. Uncertainty of the radiocesium measurement was estimated to be about 8 %. In the Arctic Ocean, about 1.5 and 3.5 Bq/m^3 of ^{137}Cs were observed in surface layer from sea surface to about 200 m depth and subsurface from about 200 m to 800 m depth, respectively. Because these concentrations of ^{137}Cs were observed in the Arctic Ocean before the FNPP1 accident, these could be derived from nuclear weapon testing in the atmosphere mainly in the 1950-60s and release from nuclear fuel reprocessing plants mainly in the 1980-90s. On the other hand, Fukushima-derived ^{134}Cs was detected only at 150 m depth although the concentration was very low (0.07 Bq/m^3). The depth of 150 m corresponded to a salinity minimum layer originated from seawater from the North Pacific. Fukushima-derived ^{134}Cs was not detected in the salinity minimum layer in 2012 and 2013. On the other hand, the ^{134}Cs was observed in surface seawater (less than 0.3 Bq/m^3) in the Bering Sea in 2012, 2013, and 2014. These results imply that transportation of ^{134}Cs from the Bering Sea to the Arctic Ocean takes about 3.5 year after the FNPP1 accident. At stations along 47°N in the subarctic region, activity concentration of ^{137}Cs in surface seawater was lower in the west (less than 2 Bq/m^3) than that in the east centered about 150°W (less than 8 Bq/m^3). The concentration of ^{137}Cs decreased with depth and was about 0.2 Bq/m^3 at 800 m depth both in the west and east. Fukushima-derived ^{134}Cs was detected in seawater from shallower layer than 200 m depth, surface mixed layer, suggesting that ^{137}Cs in layer deeper than 200 m depth was derived not from the FNPP1 accident but the nuclear weapon testing. Activity concentration of Fukushima-derived ^{134}Cs in the surface mixed layer was lower in the west (less than 0.3 Bq/m^3) than that in the east centered about 150°W (less than 2.5 Bq/m^3). The ^{134}Cs-rich surface water was observed at the international-date-line approximately in summer 2012. These results suggest that ^{134}Cs deposited in coastal area near Japan and directly-discharged from FNPP1 had been transported eastward in the surface mixed layer to around 180° by summer 2012 and subsequently around 150°W by summer 2014.
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Keywords: Fukushima-Dai-ichi Nuclear Power Plant, Radiocesium, North Pacific subarctic region

Factors controlling radiocaesium distributions in the western North Pacific in 2011-2013

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The 2011 off the Pacific coast of Tohoku Earthquake and subsequent tsunami on 11 March 2011 caused damages that led to the accident at TEPCO's Fukushima Dai-ichi Nuclear Power Station (FDNPS). Large amounts of radionuclides were dispersed by hydrogen explosions, and radionuclides also leaked from the FDNPS into the terrestrial and marine environments. In the approximately 5 years that have passed since the accident, radiocaesium activities in seawater in the Fukushima coastal area have decreased, but are about 10 times higher than before the accident [1]. There is some controversy about factors affecting radiocaesium in the open ocean, particularly in subarctic area. To improve the accuracy of the diffusion simulation predictions in the ocean, more data are required from many regions regarding temporal changes. This study aimed at elucidating distribution and behavior of dissolved radiocaesium in seawater collected from the western North Pacific during the four sampling periods in 2011- 2013 (Period I; 14 April to 5 May 2011, Period II; 27 June to 4 August 2011, Period III; 4 June to 12 July 2012 and Period IV; 9 July to 29 July 2013). Collected seawater samples were filtered through a 0.2-μm pore size filter and was concentrated by means of improved ammonium phosphomolybdate (AMP) method [2]. In upper-layer seawater (water depth 0-10 m), the dissolved ^{137}Cs activities were relatively high off the Fukushima Prefecture in Periods I and II. In Periods III and IV, the activities at most of monitoring stations were same order. The activity profile patterns at most of stations decreased with increasing water depth were constant at all depth. The activity in middle-layer (water depth 100 or 200 m) was relatively higher than those in upper-layer at KEO in Period II and at JKEO in Period III. At S1, the activity profile patterns were its maximum layer at water depth 100-400 m in Periods III and IV. It seems that maximum layers of ^{137}Cs activity at S1 and KEO had happned because upper-layer seawater including radiocaesium had subducted more deep associated with formation of the North Pacific subtropical mode water. In upper-layer, integrated ^{137}Cs amounts in 35° N-40° N and 140° E-150° E, which zone is located just eastward from the Fukushima Prefecture, were highest compared to those in another zones until Periods II and accounted for more than 50 % of the total ^{137}Cs amounts of each sampling periods. In Periods III and IV, the amounts in divided all zones were small order. It is possible that the amount distributions influenced on several factors such as the location and strength of Kuroshio and Oyashio current and semipermanent eddy.

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Keywords: Fukushima Dai-ichi Nuclear Power Station accident, dissolved radiocaesium, western North Pacific

Behaviour of oceanic ^{137}Cs from the Fukushima Dai-ichi Nuclear Power Plant for four and a half years

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A series of accidents at the Fukushima Dai-ichi Nuclear Power Plant (1F NPP) 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. Additional release pathways by river input and runoff from 1F NPP site with precipitation and were also effective for coastal zone in the specific periods before starting direct release on March 26 2011.
We reconstructed spatiotemporal variability of ^{137}Cs activity in the regional ocean for four and a half years by numerical models, such as a regional scale (horizontal resolution is about 1 km) and the North Pacific scale (horizontal resolution is about 10 km) oceanic dispersion models, an atmospheric transport model and river runoff model.
Direct release rate of ^{137}Cs were estimated for four and a half years after the accident by comparing simulated results and observed activities very close to the site. The estimated total amounts of directly release was 3.7±0.7 PBq. Directly release rate of ^{137}Cs was the order of magnitude of 10^{14} Bq/day and decreased exponentially with time to be the order of magnitude of 10^{9} Bq/day by the end of September 2015. Estimated direct release rate have exponentially reduced with constant rate since November 2011. Apparent half-life of direct release rate was estimated to be 346 days. Simulated ^{137}Cs 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 ^{137}Cs activity in the ocean from 11 to 21 March 2011. Observed data of marine biota should reflect the history of ^{137}Cs activity in this early period. The comparisons between simulated ^{137}Cs activity of marine biota by a dynamic biological compartment and observed data also suggest that simulated ^{137}Cs activity other than attributable to dirent release was underestimated in this early period. We reconstructed the history of ^{137}Cs activity in this early period with direct release, atmospheric deposition, river input, runoff from 1F NPP site with precipitation. River runoff process is still unknown in the early period because there were no observed data. We assumed that 10% of deposited ^{137}Cs on each river basin run off thorough rivers along the Miyagi, Fukushima and Ibaraki coasts. The simulation with overestimated river runoff rate (10 %) suggests that the river flux of ^{137}Cs to the ocean was not effective to the ^{137}Cs activity in the ocean in this early period. We estimated the release rate of ^{137}Cs with rain water runoff from the 1F NPP site from the observed ^{137}Cs activity before 26 March 2011 and precipitation data close to 1F NPP site. Simulation with additional release of ^{137}Cs from the 1F NPP site suggests that additional release from 1F NPP site was effective to the ^{137}Cs activity adjacent to 1F NPP and 2F NPP. Simulated atmospheric depositions of ^{137}Cs on a reginal ocean by 9 regional atmospheric transport models still have huge uncertainties. It is also important to estimate the deposition process on a regional ocean to understand contamination process of marine biota.
In the North Pacific scale, ^{137}Cs activity in the intermediate water increased due to the Subtropical Mode Water (STMW) formation. ^{137}Cs is a useful tracer to detect the STMW formation. Not only the direct release but also the atmospheric deposition are essential for the distribution of ^{137}Cs activity in the North Pacific. Five-member, ensemble simulation with high resolution can represent the increase of ^{137}Cs activity in the intermediate water.

Keywords: Fukushima Dai-ichi Nuclear Power Plant Accident, Oceanic despersion, Cesium-137, Regional Ocean Model, Ocean General Circulation Model

