Radioactive particles in residual soil after strong acid leaching

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In March 2011, the Fukushima Dai-ichi Nuclear Power Plant (F1NPP) accident has been occurred, and consequently large amount of radionuclides discharged to the environment. Especially Cesium-137 (^{137}Cs), which is radioisotope of cesium and it has 30 a of half-life, were remained in the environment for a long time. Understanding of the physical and chemical properties of the emitted radioactive cesium is important to accurately evaluate the possible human health impacts and to assess the long-term distributions of these radionuclides after deposition in residential areas, agricultural fields, mountains, and aquatic environments. Especially, the chemical form in soils is interesting at the point of decontamination of residence area and reconstruction of agricultural field. This study also analyzed chemical form of radiocesium in the soil collected at Motomiya city.
Soil sample was collected at the Motomiya city, Fukushima Prefecture, located of 40 km west from the F1NPP in October 2012. Concentration of ^{137}Cs in soil was 8 kBq/kg (In March 11, 2011). Sequential extraction was performed (Five fraction was obtained, 1) water dissolve, 2) cation exchangeable, 3) organic form, 4) concentrated acid leachable, 5) residue.), and approximately 50% of ^{137}Cs was remained in soil. Several number of spot type contamination were detected with autoradiography using an imaging plate. A spot was isolated from soil, and it was spherical particle. Constitution elements are Fe, Zn, Si, O, as well as Cs, which was detected using an energy dispersive X-ray spectrometry. These features are very similar to Cs-bearing radioactive particles isolated from air dust filter collected at the Tsukuba city by Adachi et al. (2013) and Abe et al. (2014). Therefore the particles distributed widely. In addition, dominant composition of particle is silicate of approximately 80 %. This result is consistent with Satou et al. (2015) and Yamaguchi et al. (2016). A silicate usually indicates acid resistance. These results suggested that the particulate form radiocesium is stable in environment, and it could be not remove by acid chemical treatment.

Keywords: Radioactive particle, Silicate compound, Acid resistance

Long-term prediction models of atmospheric concentration of Cs-137 and their comparison with AIC

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In the Fukushima Daiichi nuclear power plant accident, a large amount of radioactive nuclides were released. Ceasium-137 is one of the most significant radionuclides which were released in large amount to the atmospheric and has a half-life of thirty years; the Cs-137 pollutes the atmosphere for a long term. Some models which can predict the concentration of Cs-137 for long term have been proposed.
In this study, first, I evaluate the seasonal variation in the concentration change of Cs-137. Analyzing the residuals between the measured data and the models, we reveal periodic fluctuations with the period of one year. We found that the changes of the concentration is in the form the sine curve of the one year period, which reaches at the maximum value in the summer and minimum in the winter. Adding such a sine curve in long-term models, we propose a new model considering seasonal variation. Second, we compare the accuracy of the models. Some models to reproduce the atmospheric concentration of Cs-137 have been proposed, but they are not evaluated quantitatively with regard to which model is the best. We evaluate some models using Akaike's Information Criterion(AIC). AIC evaluate some models on consistency and number of free parameters. At first, we evaluate which is good, considering seasonal variation or not. As a result, model considering seasonal variation is better. Second, I evaluate some proposed models. As a result, the best model is $C(t)=Aexp(-\lambda_{decay}t)t^{-\alpha}$ (then C(t) is concentration of Cs-137, λ_{decay} is decay constant of Cs-137, A and αare free parameter). This is conclusion that $C(t)=Aexp(-\lambda_{decay}t)t^{-\alpha}$ and considering seasonal variation is the best model.

Keywords: activity concentration, aerosol, resuspention

Resuspension processes of Fukushima radioCs: Could fungal spore play a significant role?

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The authors have studied for atmospheric re-suspension of the radioactive cesium (Cs) originated from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in contaminated area by the accident to assess the atmospheric effect of the accident. As a result the authors came to know that 1) in summertime, Cs concentration increases (see Fig. 1) in such typical Japanese village-vicinity mountain area and that 2) radioactive particles in summer seem to be dust from its appearance and their optical micrograph, but on the contrary, we realized that most has biological origin from electron-microscopic view (Fig. 2). Taking into consideration the fact that true fungi concentrate Cs due to misidentifying Cs as potassium, we can assume that the fungal spore as the major contributing factor for resuspension.
Supposed that only fungal spore can carry ^{137}Cs, estimating the ^{137}Cs amount per spore under various assumptions; the value would be 5×10^{-10}-3×10^{-7} Bq/fungal spore particle, in that case, the spore needs to be released at a rate of 9×10^{3}-5×10^{5} particle/m^2/sec from forests to support the present ^{137}Cs in the air. This value is surprisingly 1-3 digits larger than the forest maximum value (387 particle/m^2/sec) of the fungal spore emission rate given in Table 2, Sesartic & Dallafior (2011), suggesting a potentially large environmental impacts of the spore. Actually, number concentration of bio-aerosol would be reaching $5\text{-}8\times10^{5}$ particle/m^3 in our preliminary observation during past summer in 2015 in a forest in Fukushima Prefecture, revealing that more bio-aerosol release could be occurring from Japanese forests than our expectation. Further, based on this postulation that fungal spore would be a major source of atmospheric Cs especially during summer, ^{137}Cs concentration would be ranging 2.5×10^{-4}-0.15 Bq/m^3 air in the forest, which almost fits to the actual ^{137}Cs concentration level shown in Fig. 1.

Keywords: the Fukushima Daiichi Nuclear Power Plant accident, Radioactive cesium, Resuspension, Bioaerosol, Dust

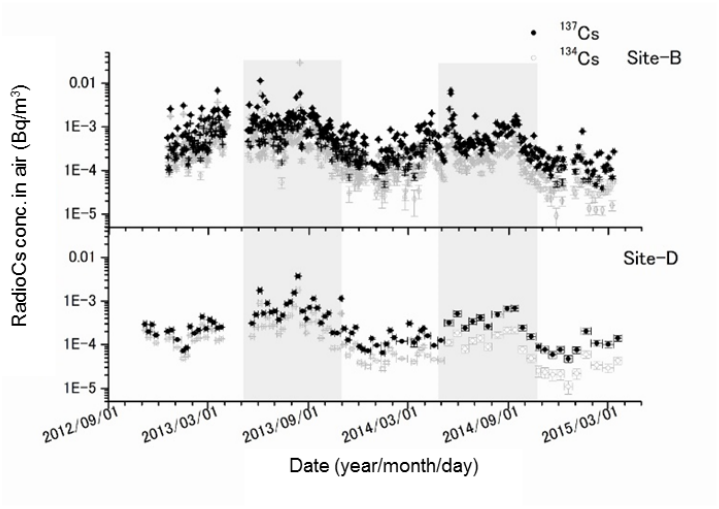

Fig. 1: Temporal trends in atmospheric radioCs concentrations in contaminated area in Fukushima Prefecture during fall of 2012 to spring of 2015 Shaded: summer season

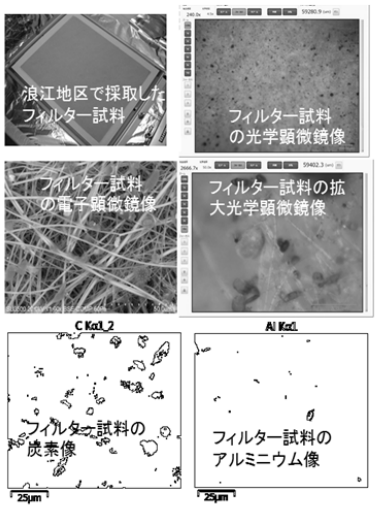

Fig. 2: Photos of a filter specimen sampled in summer; optical microscopic views, and electron microscopic views The bottom panel shows carbon distribution (left) and aluminum one (right).

Correlation-study about the ambient dose rate and the weather conditions

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The long-term radiation risks are believed to be heavily affected by the resuspension process. We therefore focus on the surface-atmosphere exchange process of released radioactive materials in this study. Radioactive materials were deposited on the soil and float in the air, and such complicated process are influenced by the weather conditions deeply. We need to reveal the correlation between the weather conditions and the ambient dose rate.
In this study, we study the correlation between the weather conditions and the ambient dose rate with the correction of the decrease due to the radioactive decay. We found that there is a negative correlation between the ambient dose rate and the soil water content by the correlation coefficient. Using this result, we reconstruct the ambient dose rate from the weather conditions by the multiple regression analysis and found that the reconstructed data agree with the observation very well. Using Kalman filter, which can be sequentially updates the state estimate, we obtained such a good agreement.

Keywords: Fukushima Accident, Radioactive nuclides, Karman Filter

Can we estimate vertical profile of radiation caused by deposited radioactive materials from atmospheric electric field measurement?

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The 2011 off the Pacific coast of Tohoku Earthquake generated large tsunami resulting in serious damage to the Fukushima Daiichi nuclear power plant (FDNPP). The damage caused radioactive materials be discharged to the environment. After the discharge, transported radioactive materials were deposited around east Japan. The radioactive deposition modulated atmospheric electricity such as the decrease of atmospheric electric field (AEF) for several months. From the ground-based observations of AEF, detectable modulated area on the ground was estimated only within a few hundred kilometers from FDNPP. In addition, we estimate the modulated height above the ground by applying the observed data to a global electric circuit model, so that the modulated height was up to approximately 1 km, which agrees with aerial radiation monitoring. Therefore, the ground-based AEF observation might contribute to estimating the vertical profile of radiation.

Keywords: Radiation, Atmospheric Electric Field, Conductivity

Stochastic modeling of the migration of Cs-137 in soil considering a power law tailing in space

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We develop a theoretical model to reproduce the measured data of Cs-137 in the soil due to the Fukushima Daiichi NPP accident. The Advection Diffusion Equation (ADE) is proposed by He and Walling (1996) and has been used to predict migration in soil. This model shows Gaussian diffusion process called normal diffusion. However, We found that the concentration of Cs-137 has a discrepancy from ADE model, specifically in a deep part because the depth profiles have a power law tailing. The diffusion comes off ADE is called anomalous diffusion. Therefore, we improved ADE model in the following aspect. When Cs particle (or Cs solution) migrate in the soil, the diffusion coefficient should be the results of many processes in the soil. These processes include the effect of various materials which constitute the soil (clay, litter, sand), or the variations of pore size in the soil. Hence we regard the diffusion coefficient as the stochastic variable, we derive the model. Specifically, we consider the solution of ADE to be the conditional probability $C(x, t \mid D)$ in terms of the diffusion coefficient D and calculate $C(x, t) = \int_{0\sim\infty} C(x, t \mid D) * f(D) * dD$, where $f(D)$ is the probability density function of D. This model has a power law tailing in space like the space-fractional ADE.

Keywords: Cs-137, Anomalous diffusion, Power law tailing in sace

Estimation of caesium runoff after the Fukushima accident with the consideration of the fractal mountain geometry

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In Fukushima Daiichi NPP accident, a lot of radioactive substance was released into a wide range of Fukushima Prefecture. It is necessary to estimate how long and how much radioactive contaminants will be discharged from the forest to the rivers and eventually to the sea. In this study, we tried a new method of prediction caesium (Cs-137). We apply the fractal geometry of the surface of mountains, thereby estimate the amount of runoff of surface soil with Cs-137 attached. It is a method to calculate z (height) coordinates of arbitrary points. We calculate the height of the location that we want to know from average of neighborhood points with the white noise. We can simulate a ridgeline of mountain, which similar to the real mountain. In the study, I used the midpoint displacement expanded into three dimensions. The universal soil loss erosion (USLE) model was used to calculate the volume of soil erosion of the simulated ground. And we calculate erosion of Cs-137 from the soil erosion volume. We compared this data of Cs-137 erosion and erosion of real ground. We found that our result agrees with the data roughly. So we concluded this method used the fractal simulation is useful.

Keywords: discharge, runoff, radionuclide

Uptake of radioactive Cs by soybeans depending on the level of K application

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[Introduction]
After the Nuclear Power Plant accident, radioactive contamination of farmland caused contaminated crop production. It is known that radiocesium (radio Cs; Cs-134 + Cs-137) behaves similarly to potassium (K) in the environment; when the concentration of exchangeable K in soil is low, plants uptake radio Cs easily. Because soybeans (*Glycine max*) require substantial K, soybeans more easily absorb radio Cs from soil than other crops. Inhibition of absorption of radio Cs using K fertilization is required for safe crop production.
In this study, we investigated the time-dependent change of radio Cs concentration in soybeans, and the exchangeable K and radio Cs concentration when changing K fertilizer application rate. This information may be applied to develop an optimal method for inhibition of radio Cs absorption.
[Materials and Methods]
The study site is located in Fukushima Prefecture. Soybeans were seeded in June and harvested in October, 2015. Soil samples were collected before seeding and at five-leaf, full flowering, full seed, and full maturity stage from directly under the plants. Plant samples were also collected at each of these growth stages. Plant samples were separated into leaves, petiole, stems, pods, and beans and the growth amount, yield, and radio Cs concentration were measured for each part. The exchangeable K concentration, radio Cs concentration and exchangeable radio Cs concentration were also determined.
We conducted an exchangeable K level test and K sustained effect test. The basal fertilizer was N-P-K = 3-12-0 kg/10 a in all tests. In the exchangeable K level test, five levels of exchangeable K were set: no K fertilizer (3 years) and 15, 30, 50, and 70 mg K_2O/100 g. The K fertilizer was applied at seeding. Non-planted plots dressed with the same levels of K fertilizer were also established. We also investigated the sustained effect of K, in which the concentration of exchangeable K was set to 70 mg after harvesting in 2014, and no K fertilizer was applied to the same sections in 2015. All tests were conducted in triplicate.
[Results and Discussions]
The average concentration of radio Cs in soil was 3,391 ±240 Bq/kg at harvest.
In the Exchangeable K level test, there was no major difference in bean yield between exchangeable K levels. The grain radio Cs concentration and transfer factor from soil to beans (TF) decreased for applications greater than 30 mg K_2O (relative to no K fertilizer and 15 mg K_2O). The highest exchangeable radio Cs in soil was found before seeding in all treatments. For all treatments, the exchangeable radio Cs content throughout the year was lower when exchangeable K levels were higher. The no K fertilization, 15 and 30 mg K_2O plots had higher exchangeable radio Cs content than non-planted plots. This suggests that when soil K_2O content is below 30 mg, soybean planting increases exchangeable radio Cs in soil.
Because there was sustained K effect of 2013 and 2014, planted plots had different soil K contents before seeding. However, the exchangeable K concentration was similar in all non-planted plots, and at levels over 30 mg K_2O/100 g, it increased after fertilization to five-leaf. Exchangeable K at harvest was comparable between K levels regardless of planting status. Thus, the no K fertilization and 15 mg K_2O/100 g have higher radio Cs concentrations and TF because of a lack of exchangeable K and an increasing exchangeable radio Cs.

The concentration of grain radio Cs in the K sustained effect test was lower than in the no K fertilization plots. This suggests that the exchangeable K concentration before seeding was higher, presumably because sufficient K content had been ensured. But the soil K_2O content decreased with time-dependent, so there is a risk that radio Cs concentration of grain will increase next cultivation.

Keywords: Radioactive Cs, Soybean, Potassium

Vertical profiles of 134Cs and 137Cs in 1980, 2002, 2011, 2012 and 2015 along 165 deg. E in the North Pacific Ocean

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134Cs and 137Cs, hereafter radiocaesium, were released to the North Pacific Ocean by two major likely pathways, direct discharge from the TEPCO Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident site and atmospheric deposition off Honshu Islands of Japan, east and northeast of the site. Activities of radiocaesium released by the FNPP1 accident were measured along 165 deg. E in 2011, 2012 and 2015. In this presentation, we present long term behavior of FNPP1 released radiocaesium in the ocean interior of the North Pacific Ocean based on the observations and model simulations through 2015. We also discuss about 137Cs profiles observed in 1980 and 2002 in the same region which derived from atmospheric nuclear weapons tests conducted in late 1950s and early 1960s.

In 2002, the 137Cs profile along 165°E in the North Pacific Ocean is characterized by several subsurface cores with high 137Cs, including two 137Cs concentration maxima at 20°N, 165°E, one at 250 m and one at 400–500 m depths. The shallower maximum is in the density range of subtropical mode water (STMW) and the deeper one is in the density range of central mode water (CMW). The main 137Cs cores, therefore, were formed by movements of STMW and CMW in the interior ocean during the past four decades in 2002. The 137Cs has been transported from subarctic region to subtropics and tropics as a result of subduction.

In October 2011, 134Cs activity derived from FNPP1 accident showed a maximum of 24.4 ±1.77 Bq m−3 at 26 meters depth at 40°N, 165°E and 80 % of 134Cs inventory existed shallower than 200 meters depth while 134Cs activity showed a maximum of 9.18 ±0.71 Bq m−3 at 301 meters depth at 39°N, 165°E and only 20 % of 134Cs inventory existed shallower than 200 meters depth in June 2012.

In June 2012, 134Cs activity also showed a maximum at subsurface at 29°N, 165°E. This subsurface maximum, which was also observed along 149°E, might reflect the southward transport of FNPP1-derived radiocaesium in association with the formation and subduction of STMW. In June 2012 at 34°N–39°N along 165°E, 134Cs activity showed a maximum at around potential density= 26.3 kg m−3, which corresponds to CMW. 134Cs activity was higher in CMW than in any of the surrounding waters, including STMW. These observations indicate that the most effective pathway by which FNPP1-derived radiocaesium is introduced into the ocean interior on a 1-year time scale is CMW formation and subduction. In June-July 2015 at 36°N–44°N along 165°E, there are only very week signal of subduction of FNPP1-derived radiocaesium which means subducted radiocaesium might move eastward from this region.

Keywords: radiocaesium, North Pacific Ocean, subduction, Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident

Sensitivity analyses of sediment and ^{137}Cs behaviors in reservoirs during rainfall events

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Investigations and simulations are important to understand sediment and radioactive cesium migration in reservoirs contaminated by the accident at the Fukushima Dai-ichi Nuclear Power Plant. In the previous study, we presented one- and two-dimensional simulations of sediment and radioactive cesium migration in the Ogaki Dam reservoir located in the middle reach of the Ukedo River and found that the reservoir played an important role to delay and buffer the movement of radioactive cesium in heavy rainfall events and that the buffer effect depended on particle sizes of suspended sediment and the water level in the reservoir. In this study, to understand the sensitivity of intensity and duration of flood events to discharges of sediment and radioactive cesium during flood events, we performed sensitivity analyses by using the FLESCOT code, a three-dimensional finite volume model developed by the Pacific Northwest National Laboratory. It considers turbulent water flow and transports of multi-size sediment and radioactive cesium both in dissolved and particulate forms. The results showed that the discharge proportions depended on sediment size, event intensity and event duration. The proportions of sediment/^{137}Cs discharges increase, as event duration is shorter/heavier. The silt component is a main carrier of radioactive cesium in larger events, while the clay-sorbed and dissolved forms are dominant in smaller events. In heavier events, resuspention of bed sediment took place.

Keywords: Fukushima Daiichi NPP, Reservoir, Sensitivity analysis, FLESCOT, Cesium, Sediment

Study on transport behavior of cesium in the ground water via fractured rock

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Radioactive cesium released by the accident of Fukushima Daiich Nuclear Power Plant was deposited widely around Abukuma mountain region of eastern Fukushima Prefecture.
Fractured granite is widely distributed at the Abukuma Mountains. Many of the deposited Cs adsorbed on the soil of a few ~ 10cm from the surface. However, in some area, the basement fractured rocks are exposed directly on the ground, or distributed at the shallower depth from the top. In these area it can't be denied the possibility that Cs moves quickly via fracture in the basement rock. Especially, as for the Cs recharged at the mountain area, the concentration in the ground water of the spring and/or wells at the discharge area could increase with time at later time as the half-life time of the 137Cs is about 30 years. It is one of the biggest concern for residents who are already considering return to their hometown, or already use the spring and wells. However there has been seldom investigated cases of long-term behavior of the Cs released by the accident in the fractured rock.
For this reason, we have carried out the investigation of ground water analysis, dating of ground water and measurement of Cs concentration to predict the future exposure dose at discharge area. We have conducted geological, hydrogeological and hydrochemical investigation, and ground water simulation at around Odaka-ku, Minami-soma city, Fukushima Prefecture, where Cretaceous granite of Abukuma Mountain area at west part and Neogene sedimentary formation is distributed at the eastern lowland. Thus the general trend ground water flow is from west to east. The Futaba fault is situated between the Abukuma granite and Neogene sedimentary formation.
From the results of the geological investigation, Futaba fault has mylonite fabric which suggests the flow barrier function to ground water flow initially, though, brittle fracture of later stage is observed in the mylonite. And small amount of ground water flow from the fracture. This suggests Futaba fault doesn't have the flow barrier function. Regarding the hydrochemical analysis, the ground water sampled from the Futaba fault and artesian well indicate relatively longer residence time. Estimated ground water age is 25 to 30 years from the result of the tritium analysis etc. Moreover, Cs concentration in the ground water from the artesian well is below the detection limit. From the result of the ground water flow simulation which is modeled granite, Futaba fault and sedimentary formation with assigned hydraulic conductivity from data base, the hydraulic head distribution varies with the assigned hydraulic conductivity (higher or lower permeability) of the Futaba fault. For the origin of the ground water at the artesian well, it is unlikely from the Abkuma Mountain, but possibly from the sedimentary formation east side of the Futaba fault.
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Keywords: Fukushima, Fractured Rock, Radioactive Cesium, Transport Behavior

