

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

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

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

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

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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

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:

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

$$\{GA(r,z)\}\rightarrow\{Q(\text{on mesh})\}.$$

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,

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

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

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

Where, a vector $\{u,v,w\}$ is wind speeds. $\text{Rand}()$ 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

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 filter 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.

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

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 significant 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