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共同研究年次報告書

【海外共同研究】

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Multi-sources images time-series for landscape dynamics analysis in rural areas after Fukushima accident

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Since the accident of the Fukushima power plant in 2011, several evacuation orders have been issued. This kind of fast and massive evacuation proceed is exclusive and can provide unique experimental sites to observe methods of vegetation recovery in agricultural areas. Usually, recovery processes are induced by a gradually abandonment of agricultural practices. These processes are characterized by several steps of landscape transformation from opened areas (agricultural plots) to closed areas (wasteland until forest recovery). Our study focuses on the evolution of the agricultural plots (paddy rice, cropland…) in Namie town in Fukushima prefecture, where decontamination campaigns were limited until now (decontamination visible in 2015). In this context of observatory, multi-source and multi-resolution data can be useful to define landscape trajectories in territories avoided of population and under a limited anthropic influence. To study spatial and temporal characteristics of recovery processes satellite imagery is one of the primary sources of information. In this study, the procedure relies on SPOT and Pleiades times series analysis for crop-mapping. We tested the use of Enhanced Vegetation Index (EVI) and traditional Normalized Difference Vegetation Index (NDVI) to detect vegetation recovery patterns before and after Fukushima accident (from 2009 to 2019). We only focus our research on agricultural plots to spatialized information about scrub invasion, and through wavelet analysis evolving in time.

Keywords:

Contribution of K transporters to Cs uptake and accumulation in rice

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The aim of the project is to develop safe-food or *in-situ* remediation approaches using plants for rehabilitation of soil. In the plants, ^{137}Cs radiological contaminant enters using K^+ transport systems by chemical analogy. To decipher the molecular mechanisms of Cs uptake and accumulation, we have characterized several rice mutants that are affected in different K^+ transport systems in CEA and evaluated those ^{137}Cs accumulations in natural paddy field belong to NARO Agricultural Radiation Research Center in 2018. Among the several candidate lines, HKT2;1 KO lines have been shown to be affected in Cs uptake or accumulation compare to wild-type plants in the paddy field.

In order to validate the results obtained in natural paddy field experiment in 2018, HKT2;1 KO lines were cultivated the same paddy field again and two different K conditions (with or without K fertilizer) were also tested. Around 10 plants of two independent HKT2;1 KO lines were cultivated until harvesting grains (4.5 months, from June to October). Then, the ^{137}Cs contents were measured in grains.

Our results showed the differences in K^+ concentration in presence or absence of fertilizers were not appeared in all tested lines, however, we observed differences in ^{137}Cs contents in grains between HKT2;1 KO lines and control lines again. The mutant line disrupted in the HKT2;1 gene that is expressed in whole plants and highly expressed in the root, shown an increased ^{137}Cs content in grains. To confirm these results, we would like to perform laboratory condition experiments and a new paddy field experiment in 2020.

Keywords: ^{137}Cs, K^+ transport system, HKT2;1

Measurement of emission flux of radioactive cesium to the atmosphere

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Radio-cesium emitted by the TEPCO FDNPP accident has been deposited on soil and vegetation after transpiration in the atmosphere. Re-suspension of the deposited radio cesium is increased atmospheric concentration of its radioactivity even now. Quantitative understanding of re-suspension flux of radio-cesium is essential for the modelling of radio-cesium transition from soil and vegetation via the atmosphere.

For this purpose, we have developed 1-bix and 2-box models to evaluate the emission of radio-cesium by the re-suspension from surface. In these models, the temporal variation of atmospheric concentration of radio-cesium radioactivity is considered to be balance be the difference of emission and deposition in the 1-box model, and by the difference of emission, deposition and vertical diffusion in the 2-box model.

Using the atmospheric concentration data of radio-cesium radioactivity derived from continuous air sample of 12-hour interval at Namie site, Fukushima, in some weeks between March and September, 2019. Dry and wet deposition flux are evaluated from the data of passive sampling of deposited aerosols and rain water at the same site and during same periods. Meteorological parameters were also measured with automated weather station at the site. Vertical diffusion of radio-cesium activity concentration was estimated from two heights (0.6 and 5m) air sampling using 6.5m tower as well as measurement of sensible heat flux measurement by ultrasonic anemometer and two heights temperature measurement.

Using these box models, the resuspension flux coefficient was evaluated and its values were positively correlated with the surface wind speed, indicating that the re-suspension is activated by wind blow at the surface especially in spring (March and May).

.Keywords: Atmospheric radio-cesium, re-suspension, emission flux, box model

Characteristics of chemical compositions of xylem sap in a pine forest under environmental changes

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Trunk xylem is the major conduit for transport of minerals to photosynthetically active leaves from root uptake and assimilation. Studying variations in chemical compositions of xylem sap is thus of basic interest as it reflects the internal minerals uptake and cycling of forest trees. Until now, however, our knowledge of xylem transport in forests under natural conditions and the regulation of this transport are not well understood. We thus depicted the level and behavior of chemical components and stable isotopes in the xylem sap of trunks and branches of different aged trees from a pine forest in northern China. There were no significant differences between the centrifugation and water displacement methods with respect to nitrogen (N) compounds and inorganic ions in the xylem sap. Potassium concentrations obtained by the methods were similar and consistent with the values obtained from earlier publications on woody species. This suggests that the centrifugation method would be a reliable and robust tool for collection of the trunk xylem sap. Additionally, during foliage senescence, dissolved organic N was the dominant component of total N followed by nitrate (NO_3^-) and ammonium (NH_4^+). Potassium and chloride were the predominant cation and anion, respectively, of the xylem sap. More inorganic N components (mainly NO_3^-) were found in young trees than in old trees. The N components in the trunk xylem sap showed obvious diurnal changes. The N isotope (^{15}N) decreases and then increases during transport from the root to the canopy, and was positively correlated with the phloem sap ^{15}N, indicating that the xylem sap in the trunk had a close exchange with the phloem sap during the xylem sap transport. Our study contributes to improve the diagnostic assessments of tree physiological processes and growth in forest trees under environmental changes.

Keywords: Xylem sap, Stable isotope, Sap flow, Plant nutrient transport, Plant nutrient utilization

Determination of Local Ecological Factors on Soil-to-Plant Transfers in Fukushima Forest Ecosystems

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Radiocesium contamination from the March 2011 accident at the Fukushima Da-ichi Nuclear Power Station (FDNPS) was initially deposited on the forest canopies with deposition into the soil occurring over the next few years through litterfall and precipitation. Measurements taken from the Yamakiya site in 2014 show that the contribution of understory plants to the total inventory of radiocesium in aboveground biomass was very low compared to the dominant Japanese cedar trees. However, measurements were not taken in other affected sites within Fukushima prefecture as well as potential change in concentrations of radiocesium in understory biomass since 2014. Data for evaluating the transfer factors was obtained through sampling of soil and understory plants at the Yamakiya, Tsushima, Tomioka, Okuma, and Yokomuki sites inside Fukushima Prefecture. 137Cs content was determined using gamma spectroscopy of the soil and plant samples and to find the concentration of bioavailable 137Cs within the root profile of the understory plants as well as 137Cs concentration within the understory plant itself. The soil and plant 137Cs concentration was used to determine the soil-to-plant uptake factors for the sampled understory species as well as the contribution of the understory plants to the total 137Cs inventory in aboveground biomass. The effect of soil exchangeable [K+], exchangeable [Cs+], exchangeable 137Cs activity concentration, total 137Cs activity concentration, and pH on 137Cs uptake by understory plants was determined through the soil-to-plant uptake factors at the various sample sites. Total 137Cs was a significant contributing factor across all understory plant species in predicting the soil to plant transfer factors. The ability to properly estimate the activity concentration of understory plants using only the one soil factor can contribute to faster estimation of potential 137Cs concentrations in plants or uptakes by herbivores in areas contaminated by 137Cs.

Keywords: Fukushima, Yamakiya, Tsushima, Tomioka, Soil, Understory Plants, ^{134}Cs, ^{137}Cs, potassium, cesium, forest, cesium dynamics, plant uptake, total cesium inventory

Effects of radionuclide exposure on microbiome of wild boar

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Microbiome analysis of the digestive tract of a mammal is variable according to the composition of their diet. The health condition of the digestive tract is vulnerable to specific chemical substances (i.e. ammonium, phenols etc.) that microorganisms produce, and different microbial communities exists in different parts of digestive tract.
Health of mammals is also related to the composition of their microbiome. For example, the mirobiome of the digestive tract is known to have a role in the development of immune system and a normal microbiome help the host to protect from pathogenic microorganisms such as *Salmonella*.
Microbiome of wild boar in the area of Fukushima where radiation level is elevated after the NPP accident can be influenced by the radiation directly or by indirect effects such as human evacuation. As the microbiome of the digestive tract can be an indicator of health condition of wild boar, comparison of the microbiome of digestive tracts of various individuals of wild boar captured in Fukushima was attempted.
Microbiome analysis is attempted based on NGS of the extracted DNA from the content of 7 different parts of digestive tract of 42 individuals of wild boar captured in Jan-Mar 2019. In 2019 DNA extraction by a modified protocol using Quagen DNEasy Power Soil kit to increase purity of DNA was started, and the analyses are ongoing.

Keywords: Fukushima, wild boar, digestive tract, microbiome, DNA

野生イノシシのマイクロバイオームに対する放射線影響

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哺乳類の腸内細菌叢は摂取する餌によって大きな影響を受ける。また，消化管の健康状態が消化管内の微生物によって生産される，例えばアンモニアやフェノール類等特定の化学物質によって影響を受けることが知られている。また，消化管の部位により存在する細菌が異なる。
哺乳類において腸内細菌叢（マイクロバイオーム）が健康状態と関係している。例えば，健康な微生物叢は、免疫の発達においてだけではなくサルモネラ菌などの特定の病原体に対する保護にも役立っているとされている。
放射線の直接的な影響や，住民の避難などによる間接的な影響により，イノシシの腸内細菌叢が変化している可能性がある。イノシシの健康状態と関係していることから，健康状態の指標と考えることができる腸内細菌叢の分析を行い，個体間で比較する。
福島県内で捕獲されたイノシシの消化管内容物中の DNA の塩基配列から微生物組成を分析する。2019 年 1 月から 3 月の間に県内で捕獲された 42 個体のイノシシについて，7 カ所の異なる消化管の部位から採取した。この研究では第一段階としてマイクロバイオーム分析用の DNA 抽出を行った。抽出には Quagen DNEasy Power Soil キットを用いるが，DNA の純度を向上させるために改変したプロトコルを用いた。

キーワード：福島，イノシシ，消化管，マイクロバイオーム，DNA

Radiocesium fate and transport in the soil-water environment in the proximity of the Fukushima Dai-iich Nuclear Power Plant

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The objective of this research was to examine the dynamics of Cs-134 and Cs-137 in the aquatic ecosystems of the contaminated areas adjacent to the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). Samples of water, suspended and bottom sediments from rivers, ponds, and a reservoir were collected. Three rivers (Niida, Abukuma, and Maeda), four ponds (Inkyozaka, Suzuuchi, and Funasawa 1& 2), and Sakashita Reservoir located in the Fukushima contaminated area were the focus of the study. Bottom sediment cores were taken from the Sakashita reservoir to analyze vertical migration of radiocesium (r-Cs). Water samples were collected and filtered using an in-situ filtration system in the field or by using a laboratory filtration system. Dissolved Cs-134 and Cs-137 were removed from water using one of two different sorbents, a cartridge with non-woven cloth impregnated with potassium zinc ferrocyanide or a column filled with granulated Iron Ferrocyanide. Activity concentrations of Cs-134 and Cs-137 were measured via gamma-spectrometry on a high purity Germanium detector, while concentrations of major cations affecting r-Cs behavior were measured via Ion Chromatography and Inductively Coupled Plasma Mass Spectrometry (ICP/MS). Fate and transport of r-Cs in the soil-water environment was assessed and predicted using analytical empirical and semi-empirical models. Analysis of model fit to data is ongoing.

Keywords: radiocesium, sediment, NPP, catchment, Fukushima, soil, vertical migration

Studies on Nickel(II) and Cobalt(III) Complexes of an Isomeric Polyazamacrocyclic Chelator and Its *N*-Pendent Derivative for the Application in Radioactive Waste Decontamination

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Radioisotopes of nickel (r-Ni: ^{59}Ni, ^{63}Ni) and cobalt (r-Co: ^{60}Co) are produced in the structural steels of nuclear reactor vessels and internal components from neutron activation of corresponding naturally occurring stable isotopes. The primary concern with r-Ni or r-Co is in limiting the exposure to personnel working for decommissioning and dismantling nuclear reactors, primarily those are in service for more than 30 yrs. The current work aimed to evaluate the performance of chelators like shape-persistent macrocycles for the selective isolation of r-Ni or r-Co from the waste matrix. Macrocyclic complexes of Ni and Co with the *N*-pendent bis(cyanoethyl) derivative of an isomeric ligand isolated from reduced hexamethyl 14-membered tetraazamacrocycle, namely Me_6[14]ane, using stable chemical analogs, were synthesized. The compounds have been characterized by spectroscopic methods as well as by magneto-chemical, and conductivity data. It is expected that further study on the efficiency of adsorption of the chelators on Ni(II) and Co(III) ions may open a new dimension in radiochemistry.

Keywords: Macrocyclic chelator; complexation; nickel; cobalt

Determination of Radiation Dose to Large Japanese Field Mice in the Vicinity of Fukushima Daiichi Nuclear Power Plant

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Electron spin resonance (ESR) analysis of tooth enamel is recognized as a reliable method for lifetime dose reconstruction, particularly in human tooth enamel. While the use of ESR to reconstruct dose is well understood for human tooth enamel, the reliability and usefulness of dose reconstruction using ESR in mouse tooth enamel has not been as thoroughly studied. This research aims to confirm the minimum detectible dose in the teeth of the Large Japanese Field Mouse for a reliable and useable ESR spectroscopic response. Initial sample preparation methodology followed a basic ESR sample preparation procedure, including manual removal of non-enamel tooth components, followed by a 15% by weight NaOH solution in a sonic bath to dissolve dentin. Preliminary sample preparation methodology revealed a baseline shift in spectrum output, meaning that teeth could not be reliably used to reconstruct dose. A possible source of this shift was attributed to remaining dentin in the samples, or iron components in tooth structure, though iron was not seen in the samples following analysis with energy dispersive x-ray (EDS) analysis. New sample preparation methodology included an extra step using a solution of Titriplex III (EDTA) to remove spurious signal causing the baseline shift, but did not reliably resolve the problem. Because the new procedure did not reliably remove the signal baseline shift, further consideration of causes of the baseline shift will be made, and removal of the shift via mathematical methodology, or further sample preparation methodology, will be attempted. If the baseline shift is successfully resolved, tooth enamel samples will be dosed across a range (below 0.8 Gy, to 2 Gy) to test currently known dose limits in mouse teeth, and to analyze usability of mouse teeth to reconstruct lifetime dose.

Keywords: Electron Spin Resonance, ESR, Electron Paramagnetic Resonance, EPR, Large Japanese Field Mouse, Mouse, Mice, *Apodemus speciosus*, lifetime dose reconstruction, dose reconstruction

Method development and radioactivity analysis of strontium-90 in environmental samples by ICP-MS

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^{90}Sr is a product of nuclear fission. It is present in significant amount in spent nuclear fuel and radioactive waste from nuclear reactors and nuclear fallout from nuclear tests. ^{90}Sr is one of the most dangerous man-made radionuclides with a long half-life of 28 years. It has a high affinity to bone. Tea plants are easy to absorb ^{90}Sr from soils in the growth process, and ^{90}Sr in fallout is prone to contaminate the tea leaves as well. In order to investigate the radioactivity concentration of ^{90}Sr, several kinds of tea samples collected from verious provinces of China were carbonized, ashed, digested and leached, and then samples were measured after ^{90}Sr/^{90}Y was separated by di(2-ethylhexyl)phosphate (HDEHP). The results revealed that the radioactivity concentrations of ^{90}Sr in tea samples ranged from 0.28 to 3.78 Bq/kg, and the possible internal doses of 0.44×10^{-2}～6.00×10^{-2} μSv contributed to each consumer, which were far less than the ICRP annual dose limit of 1 mSv for the public. Besides, we have collected some soil samples in Namie Town, Fukushima prefecture. ^{90}Sr radioactivity concentration were also measured, which ranged from 0.8 to 17.0 Bq/kg.

Keywords: Strontium-90; Tea samples; Soil samples ; Radioactivity; Internal dose

Facilities development for analysis of radionuclides in environmental samples collected from Ruppur Nuclear Power Plant (RNPP), Bangladesh

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Ruppur nuclear power plant (RNPP) is under construction in the study area (Ruppur, Pabna District, Bangladesh), which has schedule for power production in 2023. Most of the local people generally consume groundwater from hand-tube wells, and groundwater also is used for cultivation in the areas of RNPP. Therefore, this study has been designed to collect ground water samples from twelve different sampling points surrounding RNPP site for determining the present status of heavy metals concentration in groundwater and to assess human health risk related to heavy metals contamination in groundwater. The concentrations of Pb, Cd, Cr, Fe, Mn, As, Ni, Cu, and Zn in the groundwater samples were determined using Atomic Absorption Spectrometer (GTA 120-AA240Z, Varian, Australia) at Bangladesh Council of Scientific and Industrial Research (BCSIR). This study revealed that the concentrations of heavy metals in groundwater samples for this study area in decreasing order was as follows: manganese > iron > lead > chromium > cadmium respectively. As, Ni, Cu, and Zn concentrations in the water samples were found to be below detection level. The level of target hazard quotient (THQ) in ingestion in the study area followed the order: Mn > Cr > Pb > Cd > Fe respectively. Subsequently, this study revealed that THQ values for Mn were lower than 1 in five sampling points except 1 sampling point, which indicating that there is no adverse health effects on human body in the study area for the selected metals. THQ values of dermal for all the heavy metals were less than 1. However, hazard index (HI) values for dermal adsorption of Pb, Cd, Cr, Fe and Mn are smaller than 1 indicating that there is no adverse health effects on human body and potential non carcinogenic concern via dermal adsorption of water.

Keywords: Ruppur Nuclear Power Plant, heavy metals, target hazard quotient, hazard index

Studies on the distribution and migration characteristics of actinides in typical karst environment

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Groundwater in karst aquifers constitutes about 25% of drinking water sources globally. Soils on karst landscapes are typically thin, patchy and extremely fragile. It has been observed that the widespread existence of metal binding and transport by natural organic matter (NOM) in karst dripwaters. The vertical distributions of ^{137}Cs, $^{239+240}Pu$ and ^{241}Am activity in soils from Hunan karst environment were investigated. The total inventory of $^{239+240}Pu$, ^{137}Cs and ^{241}Am was 134.5 Bq/m^2, 2342.3 Bq/m^2, and 39.4 Bq/m^2, respectively for the Hunan soil core (0-30 cm). The Pu inventory was apparently higher than the global fallout inventory (42 Bq/m^2) within the 30°-40°N latitude band due to precipitation and vegetation types (forests). In terms of migration, the depth distribution was analyzed in detail and the subsurface Pu activity peak was observed. The scattered high activity values in the deep soil might be derived from bioturbation. The $^{241}Am/^{239+240}Pu$ activity ratios have no obviously change with depth increase, indicating that Am has similar migration behavior in soil with Pu. The activity ratio of $^{241}Am/^{239+240}Pu$ was higher than the global fallout value. This result suggests the underestimation of ^{241}Am activity in global fallout based on the $^{241}Pu/^{239+240}Pu$ activity ratio during the global fallout peak year in the early 1960s. The $^{137}Cs/^{239+240}Pu$ activities ratio in surface soil was 23.48±1.73 which was close to the global fallout value. The $^{137}Cs/^{239+240}Pu$ activities ratios decrease with increase soil depth. This phenomenon may be attributed to different migration mechanisms between plutonium and ^{137}Cs, and further research is needed to elucidate the influencing factors for Pu and ^{137}Cs migration in karst environment.

Keywords: Pu isotopes, $^{241}Am/^{239+240}Pu$ activity ratio, karst environment, global fallout

Tracing the sources of Pu and Hg in the deep-sea sediments using Pu and Hg isotopes

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Radionuclides (e.g., Pu) and heavy metals (e.g., Hg) are important pollutants in marine environments. However, the sources and distribution of these contaminants in the Southern Hemisphere are rarely studied. In this project, we measured $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios, as well as Hg concentration and isotope compositions in deep-sea sediments in the South Pacific for the first time. Extremely low $^{239+240}$Pu inventories in sediments of the South Pacific revealed relatively less fallout compared with that of North Pacific. The ^{240}Pu/^{239}Pu atom ratios in the sediments are typically lower than the global fallout, suggesting mixing with low ratio Pu generated by French Nuclear Tests and/or the United Kingdom Nuclear Tests in Australia. The plausible pathways for Pu transportation were the South Pacific Gyre, based on the observation of latitudinal trends in both the $^{239+240}$Pu inventories and ^{240}Pu/^{239}Pu atom ratios.

Hg in the South Pacific is most likely contributed by global atmospheric circulation, although the contribution from ocean current remains to be studied. Total Hg concentration (THg) and isotope ratios are measured in one sediment core in the Antarctic sea, where local anthropogenic Hg sources are absent. The bottom of the core shows low THg (<10ppb) and significantly negative δ^{202}Hg values that are consistent with the global background of atmospheric Hg(II), suggesting a relatively low anthropogenic influence. The upper section of the core shows much higher THg (up to 45.6 ppb) and δ^{202}Hg values approaching zero, consistent with the increased global anthropogenic Hg emission (e.g., from coal combustion) in recent years, probably transported via long range atmospheric circulation. Future work will compare Pu and Hg isotopes in South Pacific sediments to reconstruct the history of anthropogenic inputs of these contaminants to marine environments, and to assess the influence of human activities at local vs. global scales.

Keywords: South Pacific, Plutonium, Mercury, Isotope, Radionuclide, Heavy metal

Pu and Cs isotopes in the sediment cores of the Northwest Pacific after the Fukushima Nuclear Accident

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Studying the artificial radionuclides in sediment cores is important to understand the history of contamination, material sources, and sedimentary environment. In this study, Pu isotopes (^{239}Pu, ^{240}Pu and ^{241}Pu) and Cs isotope (^{137}Cs and ^{134}Cs) were determined for 4 abyssal sediment cores collected in 2013-2018 in the Northwest Pacific (NP2, 146.5°E, 35°N; NP3,145°E, 31°N) and NP4(148°E, 27°N) and mid-East Pacific Ocean (EP, 155°W, 10°N). $^{239+240}$Pu activities ranged from 0.0018 to 0.72 Bq/kg with the ^{240}Pu/^{239}Pu atom ratios of 0.070-0.246; ^{137}Cs activities ranged from <LOD to 17.9 Bq/kg, and ^{134}Cs activities were <LOD. Generally, ^{239}Pu, ^{240}Pu and ^{137}Cs penetrated to the depth of ~10 cm in the sediment cores, which is caused by particle mixing due to burrowing activities of benthic fauna. With a few exceptions, the activities of $^{239+240}$Pu and ^{137}Cs in the 4 sediment cores decreased generally with the increasing depth. The estimated ^{137}Cs inventory in core NP2 was 104.6 Bq/m^2. The estimated $^{239+240}$Pu inventories (Bq/m^2) of sediment cores NP2, NP3, NP4 and EP were 23.0, 0.87, 0.83 and 8.1, respectively. The "latitude effect" of the global fallout deposition resulted in more deposition in NP2 and the weak scavenging ability of the overlying water column caused much lower deposition in NP3 and NP4. ^{240}Pu/^{239}Pu atom ratio showed Pu in NP2, NP3 and NP4 were from the global fallout and PPG close-in fallout (6% - 46% contribution). ^{240}Pu/^{239}Pu atom ratios of EP ranged from 0.070 to 0.184, suggesting contributions of the source(s) with lower ^{240}Pu/^{239}Pu atom ratio and the global fallout, and was less affected by the PPG close-in fallout. In a comprehensive judgement of ^{240}Pu/^{239}Pu atom ratios and $^{239+240}$Pu activities, we concluded there was no evidence of Pu signal derived from the Fukushima nuclear accident.

Keywords: ^{39}Pu, ^{240}Pu, ^{137}Cs, ^{240}Pu/^{239}Pu atom ratio, sediment core, Pacific Ocean, Fukushima nuclear accident