

Environmental Radioactivity Research
Network Center
(ERAN)

FY2021 FINAL REPORT
【International】

NO	氏名	所属機関名	研究課題名	共同研究者	受入れ研究者	所属
I-21-01	Mikhailova Rena	Russian Institute of Radiology and Agroecology	Simulation of the absorbed dose rate for Japanese Cedar trees depending on the litter and soil water content		Yuichi Onda	CRIED
I-21-02	Xinchao Sun	Tianjin University	Isotopic evaluation on relative contribution of canopy microbial nitrification to throughfall nitrate in temperate forests		Yuichi Onda	CRIED
I-21-03	Nick Beresford	UK Centre for Ecology & Hydrology	Scoping study to establish a 'Radioecological Observatory' site linked to other worldwide sites	Yuichi Onda Mike Wood Peter Martin	Yuichi Onda	CRIED
I-21-04	Joske Ruytinx	Vrije Universiteit Brussel	Building a culture collection of <i>Suillus luteus</i> , an ectomycorrhizal fungus associated with pine trees		Kenji Namba	IER
I-21-05	James Beasley	University of Georgia	Influences of Rooting Activity by Wild Boar on ¹³⁷ Cs Bioavailability		Yoshifumi Wakiyama Kenji Namba	IER IER
I-21-06	Debashis Palit	University of Chittagong, Bangladesh	Study of naturally occurred radioactivity in the soil and rocks of Chittagong Hill Tracts area of Bangladesh	Shahadat Hossain	Rahman Ismail Md. Mofizur	IER
I-21-07	Suman Barua	University of Chittagong, Bangladesh	Assessment of radiological contamination of soils due to open-beach ship recycling activities in Bangladesh	Shahadat Hossain	Rahman Ismail Md. Mofizur	IER
I-21-08	Rashedul Islam Ripon	Faculty of Science, Mawlana Bhashani Science and Technology University, Bangladesh.	Design, synthesis and characterization of cobalt(III) and nickel(II) complexes with biogenic chelator triazoles, pyrrole, pyrazole, imidazole and their derivatives.	Mohammad Kahdemul Islam Foni Bushon Biswas	Ismail Md. Mofizur Rahman	IER
I-21-09	Tapashi Ghosh Roy	University of Chittagong	Synthesis, characterization, and radiometric investigations on nickel(II) complexes of a new N-alkyl pendent azamacrocyclic chelator.	Zinnat A. Begum Saswata Rabi	Rahman Ismail Md. Mofizur	IER
I-21-10	Roman Bezhenar	Institute of Mathematical Machines & Systems Problems of National Academy of Sciences of Ukraine (IMMSP)	Investigation of expected radioactive contamination and doses to human in result of potential release of dumped water from Fukushima storage tanks into marine environment by using box modeling	Vladimir Maderich	Hyoe Takata	IER
I-21-11	Olena Burdo	Institute for Nuclear Research National Academy of Sciences of Ukraine	Establishment of FISH probes for dicentric analysis of wild rodents in Chernobyl	Tomisato Miura Yohei Fujishima	Hiroko Ishiniwa	IER
I-21-12	Makhsun	National Nuclear Energy Agency of Indonesia	Monitoring of radon gas in the ground as an early warning system of earthquake	Untara	Shinji Tokonami	IREM
I-21-13	Ceiga Nuzulia Sofyaningtyas	National Nuclear Energy Agency of Indonesia	Determination Cs-137 and Cs-134 in Food Samples	Kusdiana Eka Djatnika Nugraha	Shinji Tokonami	IREM
I-21-14	Dwi Ramadhani	National Nuclear Energy Agency of Indonesia (BATAN)	Evaluation of the Serum Catalase Activity in the Individuals Exposed to High Levels of Radon Concentration: A Pilot Study	Sofiaty Purnami	Tomisato Miura	IREM
I-21-15	IIN KURNIA	Center for Radiation Safety Technology and Metrology	Detection of 8-oxo-dG level in serum and γ -H2AX, micronuclei, and apoptosis caspase-3 in lymphocyte of Indonesian Nuclear Reactors	Yanti Lusiyanti Teja Kisananto Devita Tetriana	Tomisato Miura	IREM
I-21-16	Elistina	National Nuclear Energy Agency	Determination of Pu-239/240 in water	Kusdiana Eka Djatnika Nugraha	Naofumi Akata	IREM
I-21-17	Mathew James Bruce Swallow	Mount Royal University	Development and implementation of methods to assess radiocesium transfer in forest soil ecosystems	Yusuke Unno Akira Takeda	Naofumi Akata	IREM
I-21-18	Fei Chen	National Center for Occupational Safety and Health	Gross alpha and gross beta radioactivity concentration in environmental water	Quiang Xiong Lu Zhang	Hirofumi Tazoe	IREM
I-21-19	Shan Xing	China Institute for Radiation Protection	Method development and analysis of refractory radionuclides in seawater samples	Xiongxin Dai Taeko Shinonaga	Hirofumi Tazoe	IREM
I-21-20	Md. Safiur Rahman	Bangladesh Atomic Energy Commission	Capacity building for development of nuclear techniques for analysis of radionuclides in environmental samples collected from Ruppur Nuclear Power Plant (RNPP), Bangladesh	Tasrina Rabiya Choudhury	Hirofumi Tazoe	IREM
I-21-21	Sirtom BURANURAK	Khon Kaen University	Assessment of natural radioactivity and radiological health impact on paleontologists and workers in dinosaur fossil excavation sites containing with high-background radiation		Shinji Tokonami Kranrod Chutima	IREM IREM
I-21-22	Olivier EVRARD	CEA (French Atomic Energy Commission)	High-resolution reconstruction of sediment and radiocesium source contributions in a lake draining the main Fukushima radioactive pollution plume	Anthony FOUCHER Irène LEFEVRE	Seiji Hayashi Hideki Tsuji Yoshifumi Wakiyama	NIES NIES IER
I-21-23	Zhiyong Liu	Medical College of Soochow University	Characteristics of radioisotope fingerprints of forest soils in Changbai Mountain, Northeast China	Li Zhihong Han Xiaoxiao	Jian Zheng	QST
I-21-24	Shaoming Pan	Nanjing University	Simultaneous determination of ²³⁷ Np and Pu isotopes in seawater by SF-ICP-MS with a single column chromatographic separation	Shuai Zhang	Jian Zheng	QST
I-21-25	Wang Zheng	Tianjin University	Tracing the sources of Pu and Hg in the deep-sea sediments using Pu and Hg isotopes		Jian Zheng	QST
I-21-26	Hai Wang	University of South China	Studies on the distribution and migration characteristics of Np and Pu in typical karst environment	Renjuan Liu	Jian Zheng	QST
I-21-27	Wu MEN	Third Institute of Oceanography, Ministry of Natural Resources	Analytical method of ¹³⁵ Cs and ¹³⁷ Cs in large volume of seawater by ICP-MS spectrometry	Fenfen WANG	Jian Zheng	QST

Simulation of the absorbed dose rate for Japanese Cedar trees depending on the litter and soil water content

氏名 : Mikailova Rena 受入

研究者 : Yuichi Onda

1. 成果

After the accident at the Fukushima NPP, a lot of radioactive materials released into the atmosphere were intercepted by the forested areas. Though the majority of the territories were under remediation, there still are zones with high levels of radioactivity. The purpose of the research was to calculate the absorbed dose rates for the Japanese cedar trees in order to predict possible consequences for the forest ecosystems located in the vicinity of the accident and determine the most significant ecological factors for dose rate formation. Calculation of the dose rates for trees was performed via a dosimetric model [1], modified and parameterized according to the collected data [2-7]. The vertical profile of a forest ecosystem is displayed as a set of layers with certain parameters: layer thickness (m), layer density (kg/m³), the concentration of radionuclides in the layer (Bq/m³). The model allowed estimating the dose rate resulted from the energy absorbed by the media (in our case – forest ecosystem). The modelling was carried out using Mathcad 15.0 software (PTC Inc.) and data processing was performed in MS Excel. To simulate the absorbed, dose rates it was necessary to consider moisture in litter and soil layers. To determine the water content calculations included data on the precipitation rate and correlation of soil and litter moisture and rainfall. The research by the specialists of Tsukuba University allowed tracing the water content in soil due to measurements of soil moisture and registering the precipitation data from March to July of 2018. The measurements showed good correlations with the effective rainfall index with the $T_{1/2} = 3$. The approximation allowed to make a retrospective evaluation of soil moisture during the period from 2011 to 2017. The moisture content allowed performing the retrospective estimation of the dose rate for the Japanese cedar stand. The estimated dose rate for the Japanese cedar stand in 2011 was 32 μ Gy/day in the upper canopy layers and 39 μ Gy/day at a height of 0.9 m. By 2017, the values of these indicators decreased to 10 and 13 μ Gy/day, respectively. The discrepancy between the calculated and measured data is possible due to the inhomogeneity of the contamination densities, which affects the results of monitoring studies and the parameterization of the model. In the first years after the accident, ¹³⁴Cs was the main dose-forming radionuclide; by the end of 2013, the dose rates from ¹³⁷Cs and ¹³⁴Cs became equal. By 2017, ¹³⁷Cs had become the main source of dose exposure due to the shorter half-life of ¹³⁴Cs. The model allows studying the influence of environmental factors

on the formation of the radiation exposure on the components of the tree layer of the forest. Figure 6 shows the dose rate at a height of 1 m, calculated under the condition of constant soil moisture (40%) and litter moisture of 20% and 80%. At high humidity, the screening capacity of the soil and litter increases by 1.5 times compared to the “low moisture” scenario. The research results allowed us to make a conclusion that significant fluctuations in the dose rate of trees are possible due to the change of environmental factors. Using the model, it was possible to estimate the variability of the dose rate depending on precipitation and water content in soil and litter.

2. 論文

- ① Mikailova, R., Onda, Y., Fesenko, S., and Kato, H.: Absorbed dose rate assessment for the Japanese cedar stand affected after the Fukushima NPP accident., EGU General Assembly 2021, online, 19–30 Apr 2021, EGU21-10529, <https://doi.org/10.5194/egusphere-egu21-10529>, 2021.
- ② Mikailova R.A., Fesenko S.V., Onda Yu., Kato H., Takahashi J., Spiridonov S.I. Retrospective evaluation of radiation dose rates to coniferous forest after the Fukushima-1 NPP accident. // Technogenic Systems and Environmental Risk: Book of abstracts of IV International (XVII Regional) Scientific Conference / Ed. by A.A. Oudalova. – Obninsk: IATE NRNU MEPhI, 2021. – P. 174-176.
- ③ Mikailova R.A., Fesenko S.V., Onda Yu., Kato H., Takahashi J., Spiridonov S.I. Evaluation of the influence of environmental factors on the formation of radiation doses of woody plants in the region of the Fukushima-1 NPP // Radioecological Consequences of Radiation Accidents – to the 35th anniversary of the Chernobyl accident: Proceedings of the International Research and Practice Conference, Obninsk, April 22-23, 2021 / Eds. N.I. Sanzharova, V.M. Shershakov. Obninsk: RIRAE, 2021. – P. 102-105

I-21-02

Isotopic evaluation on relative contribution of canopy microbial nitrification to throughfall nitrate in temperate forests

氏名 : Xinchao SUN
受入研究者 : Yuichi Onda

1. 成果

Forest canopies play a significant role in regulating carbon and water exchanges with the atmosphere, with profound effects on climate. However, the contribution of tree canopies in altering the chemical composition of precipitation and, consequently, the nutrient cycling within a forest has been less investigated. In particular, it is unclear whether the deposition of reactive nitrogen species (N) to canopies is retained, re-emitted and/or altered by chemical or biological reactions and what portion and chemical form of deposited N eventually reaches the soil as washed out N compounds. The application of the dual isotope approach in nitrate, in bulk precipitation and throughfall has provided another important step towards a better understanding of the importance of N and of its cycling in forests. Therefore, this study combined methods of stable isotopes and forest hydrology with the objectives: 1) to examine the changes in concentrations and the isotopic composition of atmospheric N during canopy passage, in addition to their impact factors; 2) to estimate the proportions of biologically and atmospherically derived N in the canopy. We found that a seasonal partitioning between biologically and atmospherically derived nitrate was observed, with a higher proportion ($\approx 30\%$) in TF derived from canopies nitrification in September, indicating processing of N within canopies should not be neglected and needs further exploration, with the combination of multiple isotope tracers. These findings can contribute to achieve an integrated mechanistic understanding of interactions between atmospheric nitrogen deposition and forest canopies, and better assess forest health and function in the context of environmental changes. A more in - depth analysis of these processes will require a larger number of replicates, while further research will be needed to confirm the observed seasonal changes at larger spatial and temporal scale and better quantify the relative contribution of canopy biotransformations to forest N cycling.

2. 論文

No

I-21-03

Scoping study to establish a ‘Radioecological Observatory’ site linked to other worldwide sites

氏名 : Nick A. Beresford

受入研究者 : Yuichi Onda

共同研究者 : Yuichi Onda · Mike Wood · Peter Martin

1. 成果

The contaminated areas following the Fukushima Dai-ichi Nuclear Power Plant accident in 2011 are some of the most radioactively polluted sites in the world. The accident resulted in anthropogenically-derived radionuclide depositions across the Fukushima Prefecture and resulted in large scale evacuations of the human population. Government sponsored decontamination activities and natural radionuclide decay are lower ionising radiation exposures below the government set threshold for human occupancy in some evacuated areas, many parts remain uninhabitable. Our project’s initial aim was to select an extensive area to establish a Radiological Observatory similar to that established in the Chernobyl Exclusion Zone. Within the observatory, there should have been ecological similar areas (ie., tree stand species, tree stand density, soil characteristics) with varying levels of radioactive contamination. However, a small part of the entire evacuated area (approximately 1 ha) was selected to conduct preliminary observations and deploy observatory devices. This preliminary site is located in Namie Town (37°33′12.13″N, 140°50′05.5″E) of the Fukushima Prefecture approximately 23 km northwest of the FDNPP and received about 4730 kBq m⁻² of total radiocesium (134Cs and 137Cs) deposition. This preliminary area contains forests, several streams, and has brown forest soils. Forest stands consisted of Japanese Cedar of approximately 3,300 trees ha⁻¹. Near the site, multiple radiological studies have been conducted and once data is accessible this area will provide an excellent site to study radioecology in a terrestrial ecosystem. At the preliminary site, we deployed wildlife monitoring surveillance systems that consisted of (I) typical game trail cameras and (II) Wi-Fi based surveillance cameras. These monitoring systems provide essential information to determine species inhabiting the site throughout the year. Eight wildlife cameras (Browning-Trail Cameras Strike Force) were deployed across the observatory to monitor wildlife activity. Data was stored on SD cards and each camera required monthly maintenance. We developed a new Wi-Fi capable system that allows global access to a 24/7 surveillance system that also acquired audio files. Once deployed, wildlife activity is video recorded, stored online, and is soon available for select users to download. Such data availability allows for easy communication between international collaborators. The Wi-Fi capable system used solar power. Monitoring of the preliminary site found that, while both systems successfully

recorded wildlife activity, the game-trail cameras were optimal for Radiological Observatory purposes. The Wi-Fi based system required extensive maintenance, expense, and was not operational in the autumn-winter seasons due to poor sunlight conditions. The number of solar panels needed to be increased in summer due to the increased tree canopy limiting sunlight. These negative conditions outweigh the ability to access audible cameras 24/7 and directly download monitoring observations. We recommend the use of typical wild-life cameras that have been optimized for wildlife monitoring. Consideration of colocating sound recorders with the game-trail cameras should be considered to enable audio recordings of wildlife.

2. 論文

I-21-04

Building a culture collection of *Suillus luteus*, an ectomycorrhizal fungus associated with pine trees

氏名 : Joske Ruytinx

受入研究者 : Kenji Namba

1. 成果

【Objective】

To investigate the role of mycorrhizal fungi in the transfer of rCs to red spruce in a symbiotic culture, we found a diversity of rCs concentrations, transfer coefficients, and accumulation transfer coefficients in *Suillus luteus* and *S.guranulatus*, and attempted to culture the fungi in the laboratory.

【Methods】

Mushrooms and 100 cm³ of soil at 5 cm depth were collected in Tsushima, Namie Town on 10/22/2020 and 10/15/2021, in Kanayama Town on 9/9/2021 and in Fukushima University Campus on 2021/11/1. Soil and dust were removed from the collected mushrooms, and small pieces were cut from fresh sections and inoculated onto Modified Fries agar medium. The remaining mushrooms and soil samples were dried and measured with a germanium semiconductor detector until the radioactivity error for ¹³⁷Cs and ⁴⁰K was less than 5%.

【Results】

(1) ¹³⁷Cs and ⁴⁰K concentrations of *Suillus luteus* and soil in Tsushima, Namie Town

There was no significant difference in ¹³⁷Cs concentrations between 2020 and 2021 in *S. luteus* collected in Tsushima. In relation to the ¹³⁷Cs concentration in 5 cm of soil surface layer, mushrooms with concentrations around 100 kBq/kg DW were found for soil 2.4-218 kBq/kg DW of soil, while mushrooms with concentrations over 200 kBq/kg DW showed concentrations proportional to those of soil at a range of TF=5-7. The relationship between the concentrations of ⁴⁰K and ¹³⁷Cs in the *S. luteus* collected in Tsushima showed that the ratio of ¹³⁷Cs concentration to ⁴⁰K activity (1.5-5.8 kBq/kg DW) was about 100 in four individuals, but in the others, the ¹³⁷Cs concentration was almost always below 70 kBq/kg regardless of the ⁴⁰K concentration (1.1-4.9 kBq/kg DW). The ¹³⁷Cs concentration was almost always below 70 kBq/kg regardless of the ⁴⁰K concentration (1.1-4.9 kBq/kg DW).

(2) ¹³⁷Cs and ⁴⁰K concentrations of *S.luteus* and *S.granulatus* and soil in Fukushima University Campus and Kaneyama Town

The ¹³⁷Cs concentrations in *S.luteus* collected in the Fukushima University campus were 0.1-1.8 kBq/kg DW (TF=0.2-10), while 0.5-7.5 kBq/kg DW for *S.granulatus* (TF=1.2-51). The ¹³⁷Cs concentration in *S.luteus* did not seem to be related to the ⁴⁰K concentration, but

in *S.granulatus*, the lower the 40K concentration, the higher the 137Cs concentration. However, this relationship was not observed in *S.granulatus* in Kanayama Town.

【Discussion】

The two species of the genus *Suillus* collected in this study are mycorrhizal fungi symbiotic with *Pinus densiflora*. These mushrooms showed some diversity in terms of 137Cs transfer coefficient and the relationship between K concentration and 137Cs concentration in the mushrooms. When these mushrooms make symbiosis with *P.densiflora*, it will be interesting to see how the differences in the accumulation capacity of *Suillus* spp. for 137Cs affect *P. densiflora*. In this study, we are developing strains of these fungi, and hope to conduct cultivation experiments using them in symbiosis with red pine.

2. 論文

I-21-05

Influences of Rooting Activity by Wild Boar on ¹³⁷Cs Bioavailability

氏名 : James C Beasley

受入研究者 : Yoshifumi Wakiyama · Kenji Namba

1. 成果

Studies to determine the distribution of radiological contamination within the area impacted by the Fukushima-Daiichi accident are a high priority because that accident represents one of the most significant releases of anthropogenic radiological contamination in history. Recent research has shown that some wildlife species, including wild boar, are increasing in number within evacuated areas, despite the radiological contamination. Increases in wild boar populations are of growing concern because wild boar cause extensive damage to property and agriculture through rooting, where boar overturn surface soil in search of food items, effectively tilling areas to a depth of 120 cm. Rooting by wild boar has the potential to alter the soil chemistry, arthropod communities, and vegetation composition, and thus wild boar likely play an important role in the redistribution of contaminants within soil profiles. Redistribution of contaminants to the soil surface likely increases their availability to plants and may facilitate the transport of contaminants more broadly within the landscape through erosion. Impacts of wild boar rooting on contaminant bioavailability likely are particularly pronounced in areas contaminated from the Fukushima-Daiichi accident, given their increase in abundance in these areas. Despite their global distribution and potential role in the movement of contaminants, there are currently no studies assessing the impact of wild boar rooting on contaminant bioavailability. Therefore, our objective in this study was to determine whether rooting by wild boar alters Cs-137 bioavailability. We targeted areas within the difficult to return zone for this research, as these areas have the highest densities of boar and greatest levels of environmental contamination. We hypothesize wild boar will alter the distribution of Cs-137 within the soil profile, increasing the bioavailability of this contaminant to biota and the potential for redistribution through erosion. We collected preliminary samples in Okuma town by the support of ERAN in 2020. These samples have yielded promising data in support of our hypotheses, but additional sample collections were needed in 2021 to verify our results. In 2021, we collected soil core samples from 5 sites in Namie town. We targeted former agricultural areas and grassland sites, as well as recent rooting and old rooting [i.e., > 6 months old (which has been subjected to erosion processes)] to investigate the influence of habitat and weathering on accumulation of Cs-137 contamination. In addition, we collected multiple soil cores within each rooted area (3 replicates per site), as

well as control samples from nearby locations to determine differences in the vertical distribution of bioavailable Cs-137 in the surface and sub-surface soil layers for all sampling sites. Radiocesium activity was determined for all collected samples using an Auto-Gamma Counter. Our results indicated that rooting disrupts the vertical distribution of Cs-137 within soil profiles. Almost all Cs-137 in soil cores at the control site was contained in the surface (depth up to 5 cm), whereas areas disturbed from wild boar rooting had more widely dispersed Cs-137 within soil profiles that was present at greater depths (depth of 0–20 cm) than control sites. Our results suggest wild boar rooting may alter bioavailability to plants in impacted areas. This research contributed to a clearer understanding of the environmental impact of wild boar rooting which can better inform appropriate management actions given the growing population of wild boar within the difficult to return zone.

2. 論文

I-21-06

Study of naturally occurred radioactivity in the soil and rocks of Chittagong Hill Tracts area of Bangladesh

氏名 : Debashis Palit

受入研究者 : Rahman Ismail Md. Mofizur

共同研究者 : Shahadat Hossain

1. 成果

Introduction

Chittagong Hill Tracts area is a hilly area situated in between 21025' N to 23025' N latitude and 91054' E to 92050' E longitude in the southeastern part of Bangladesh. Weathering and erosion of both igneous and metamorphic rocks of this area transform rocks into soil and sand which bear natural radionuclides from the uranium and thorium series as well as potassium. Again, this area is having a border with Myanmar and India and is not very far from China. As because neighboring India and China have nuclear power, their activities also can increase the background gamma radiation level. But there is no study of measurement of the radioactivity of this area as a whole. So, our aim is to study the background radiation level of the whole area.

Sample Collection and Measurement of Radioactivity level

The samples were collected from the undisturbed area. The samples were then dried at 1000 C for 24 h in an oven and weighed. The dried samples were then grinded into fine powder. The powdered samples were sieved through 1mm mesh to keep uniform grain size. About 1Kg of the homogenous soil sample then be poured and sealed in an airtight Marinelli beaker and was stored for 4 weeks before gamma-ray analysis. Gamma spectrometric analysis was performed by using a gamma-ray spectrometer with a coaxial HPGe γ -ray spectrometer at Atomic Energy Centre Chittagong.

Results and Discussion

The degree of retention of stable radionuclides by soil varies with the physical and chemical characteristics of the element involved, the physical and chemical form of the nuclide, the concentrations of the nuclide and of ions which compete with nuclide ions for retention sites, soil pH, soil organic matter content, soil mineral type, concentrations of chelating agents or other compounds which form complexes with the nuclide, and the time allowed for equilibrium of the nuclide between solid and liquid phase. If we compare the concentrations for all the soil samples of the three districts, we shall find that the ^{40}K concentrations varied between 74 ± 17.02 and 610 ± 73.2 Bq kg⁻¹, ^{232}Th ranged from 23 ± 2.99 and 151 ± 16.61 Bq kg⁻¹, and ^{226}Ra ranged from 3 ± 0.36 and 21 ± 2.94 Bq kg⁻¹. The average concentration of ^{40}K , ^{232}Th and ^{226}Ra is 242.5 ± 30.82 Bq kg⁻¹, 68.68 ± 8.99 and

11.62 ± 1.92 Bq kg⁻¹, respectively. The anthropogenic radionuclide ¹³⁷Cs was also analysed in this study by assuming that it could be deposited in soil of Bangladesh as a result of atmospheric fallout following the Chernobyl disaster on 26 April 1986 and other previous atmospheric tests of nuclear devices by the neighbouring countries. No ¹³⁷Cs was detected in any of the samples.

The higher concentration of ²²⁶Ra is related to its original parent concentrations, the environmental mobilities of ²³²Th and ²³⁸U, their decay schemes, and the half-lives of the generated decay chain radionuclides. Although thorium has low mobility in the environment, and its level is little affected by weathering, uranium has a high solubility in oxidized environments and may be mobilized from the solid phase.

Conclusion

Wide variation in specific activity within each district is observed. This non-uniform behaviour of radionuclides may be attributed to their uneven and irregular distribution in the earth crust, differences of geological structures and also to various topographical and agricultural activities.

2. 論文

1. E. Debanath¹, R. K. Shil,² S. Rabi, D. Palit, B. K. Dey and T. G. Roy, "Vanadium(IV) and vanadium(V) complexes with hexamethyl tetraazamacrocyclic ligands: Synthesis, characterization and antimicrobial studies", *Inorganic and Nano-Metal Chemistry*. (Published online) (2022).

2. S. Rabi, Lucky Dey, D. Palit, B. K. Dey, I. M. M. Rahman, E. R. T. Tiekink and T. G. Roy, Crystal structure of [meso-5,7,7,12,14,14,-hexamethyl-1,4,8,11-tetraazacyclotetradecane]nickel(II) diperchlorate dimethylsulphoxide di-solvate, *C₂₀H₄₈Cl₂N₄NiO₁₀S₂*. *Z. Kristallogr. New Crystal Structure*, 236(6), 1243-1245 (2021).

3. L. Dey, S. Rabi, D. Palit, S. K. S. Hazari, Z. A. Begum, I. M. M. Rahman, T. G. Roy. Syntheses, characterization, and antimicrobial studies of Ni(II), Cu(II) and Co(III) complexes with an N-pendant azamacrocyclic chelator. *Journal of Molecular Structure*, 1240(130579), 1-9, (2021).

I-21-07

Assessment of radiological contamination of soils due to open-beach ship recycling activities in Bangladesh

氏名：Suman BARUA

受入研究者：Rahman Ismail Md. Mofizur

共同研究者：Shahadat Hossain

1. 成果

Introduction “Shipbreaking” is the dismantling process of end-of-life ships to recover steel and other valuable materials for reintroducing them into the economic flow. The current study explores the spatial variation in the naturally occurring radionuclides (NORMs) in soils of the shipbreaking and recycling industrial (SBRI) areas in Bangladesh.

The research question is: “Do ship-breaking activities affect the NORMs in soil?” Experimental Soil samples collected from nine locations (one control, S1, and eight SBRI, S2 to S9) are processed and analyzed in high-purity germanium (HPGe) semiconductor detector. Activity concentrations (in Bq kg⁻¹) of ²²⁶Ra, ²³²Th, ⁴⁰K was used to assess the health hazards via radiological indices, such as radium equivalent activity (Raeq), absorbed gamma dose rate (DR), annual effective dose rate (Eaed), external and internal hazard index (Hex and Hin), and excess lifetime cancer risk (ELCR).

Results and discussion NORMs evoke occupational and public health concerns due to chemotoxicity and radiotoxicity. The Raeq, Eaed, Hex, and Hin values for all studied locations, including the control, were lower than the respective recommended values (Table 1). However, the DR and ELCR were above the recommended limits, representing radiological risks for the SBRI workers. Spatial variations in soil NORMs were observed among the SBRI. However, NORMs in SBRI soils were lower than the control, which indicates a minimal impact of SBRI activities on the NORMs in soil.

2. 論文

I-21-08

Design, synthesis and characterization of cobalt(III) and nickel(II) complexes with biogenic chelator triazoles, pyrrole, pyrazole, imidazole and their derivatives.

氏名 : Rashedul Islam Ripon

受入研究者 : Ismail Md. Mofizur Rahman

共同研究者 : Mohammad Kahdemul Islam · Foni Bushon Biswas

1. 成果

Objectives a) Synthesis and characterization of new cobalt(III) and nickel(II) complexes of biogenic chelators. b) Carrying out a study on their antimicrobial activities. c)

Study on chelation efficiency of biogenic chelators on radioisotopes of Ni and Co.

Synthesis of the complex Cobalt (II) chloride dihydrate (0.82 g; 0.0034 mole) and 1.6 g of the sodium salt of saccharin (0.0078 mol) were dissolved in 100 mL of water. The solution was heated gently on a water bath until the volume is reduced to 50 mL. It was then allowed to stand overnight, whereupon rose red crystals was separated. The mixture was cooled in an ice bath, and the crystals were collected on a filter paper, washed with cold water, and dried. Finally tetra aqua bis(saccharinato) cobalt(II) dihydrate was obtained. The starting tetra aqua bis(saccharinato) cobalt(II) dihydrate (0.18g; 0.37 mmol) and 2-guanidinobenzimidazole (0.2162g; 2 mmol) were dissolved in 15 mL methanol with stirring at 40o -50oC temperature. After that light brown powder was obtained. The light brown powder was collected by filtration. It was then washed with acetone and dried in air. [Co(sac)₂(2GB)₂]: Color, Brown. M.W., 769.44. FT-IR (cm⁻¹): ν N-H, 3332; ν C=O, 1673; δ ring(N-H) 1641; ν C-N, 1149.1; ν C=C 1471; ν S=O, 1278, ν Co-N, 540. ¹H-NMR (δ, ppm in DMSO-d₆): 2.47 (s, 1H), 6.91 (s, 1H), 7.16 (s, 1H (ring NH)), 7.65 (m, 4H). UV-Vis (λ max in nm): 432, 301, 264 (in DMF).

Summary of Findings a) New cobalt(II) and nickel(II) complexes: [Ni(sac)₂(2GB)₂], [Co(sac)₂(2GB)₂], [Ni(2MB)₂(H₂O)₂], [Co(2MB)₂(H₂O)₂], [Co(sac)₂(2,2-PB)₂] have been synthesized and characterized by ¹H-NMR, UV-Vis and FT-IR spectroscopy measurements. b) The complexes might adopt an octahedral structure (proposed). c) The ligand is found to act as a very efficient chelator of Ni(II) and Co(III) during complex formation.

N.B: Sac: Saccharine; 2GB: 2-Guanidinobenzimidazole; 2MB: 2-mercaptopyrimidine; 2,2-PB: 2,2-pyridylbenzimidazole.

2. 論文

Synthesis, characterization, and radiometric investigations on nickel(II) complexes of a new N-alkyl pendent azamacrocyclic chelator.

氏名 : Tapashi Ghosh Roy

受入研究者 : Rahman Ismail Md. Mofizur

共同研究者 : Zinnat A. Begum · Saswata Rabi

1. 成果

Radioisotopes of Ni (r-Ni) are produced in the structural steels of nuclear reactor vessels and internal components from neutron activation of corresponding naturally occurring stable isotopes. The macrocyclic chelators can be used to isolate Ni from the waste matrix. Moreover, the application and importance of macrocyclic chemistry in coordination and analytical chemistry are proliferating. In this connection, this current research plan was aimed to investigate the ability of the chelators for the selective isolation of r-Ni from the waste matrix. So, a new N-alkyl pendent azamacrocyclic chelator and its Ni(II) complexes have been prepared and characterized. During the complex formation process, the new N-alkyl pendent macrocycle was found to act as a very efficient chelator of Ni(II) ions. The newly formed compounds have been characterized by modern analytical tools, such as IR, ¹³C-NMR, ¹H-NMR, mass spectroscopy, molar conductivity data, and magnetic measurements. Most of these compounds were found to act as potential antibacterial and antifungal agents. It is expected that future studies on the efficiency of adsorption of these types of chelators on Ni(II) ions can play a vital role in the development of radiochemistry.

2. 論文

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I-21-10

Investigation of expected radioactive contamination and doses to human in result of potential release of dumped water from Fukushima storage tanks into marine environment by using box modeling

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受入研究者 : Hyoue Takata

共同研究者 : Vladimir Maderich

1. 成果

The accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in 2011 led to an uncontrolled release of a significant amount of radioactive material. Additional release of contaminated water was prevented but resulted in a large amount of contaminated water stored near the NPP. In April 2021 around 1.26 million m³ of contaminated water was stored in tanks containing radionuclides with long and intermediate half-life. The water was processed to reduce its radioactive content; nevertheless, decontamination systems cannot remove all radionuclides from the water and the capacity of the tanks is almost exhausted. As a result, the Japanese authority is now planning for a release of the contaminated water to the Pacific Ocean. This study is aimed to make an assessment of the radiological impact to human and environment. For this purpose the compartment model POSEIDON-R is applied to compute the concentration of activity in the water, bottom sediments, and biota in the north-western Pacific with focus on the area around the FDNPP. The findings are then used to compute the dose to marine organisms and the human dose from seafood consumption. Two release scenarios are considered, assuming a 10 and 40 year release period for all collected activity. The total activity is estimated for 10 dominant radionuclides (³H, ¹⁴C, ⁶⁰Co, ⁹⁰Sr, ⁹⁹Tc, ¹⁰⁶Ru, ¹²⁵Sb, ¹²⁹I, ¹³⁴Cs, ¹³⁷Cs) using available data from present radionuclide concentrations and tank volumes. According to the simulations, ³H concentration in the seawater is the highest among all radionuclides due to its high activity content in the tanks. The highest concentrations in the marine organisms come from ¹⁴C because of its high bioaccumulation rate, while the highest annual dose rate to humans is expected from ¹²⁹I due to its high dose coefficient. Calculated concentrations of activity in marine products will not exceed food safety limits in Japan even in the 4x4 km coastal box around FDNPP. The computed maximum annual dose rate to humans from all radionuclides together is less than 1 μSv. Here ¹²⁹I and ¹⁴C are responsible for respectively 82% and 16% of the dose, while the other 8 radionuclides contribute only 2%, half of which is from tritium. For boxes located at larger distances from the FDNPP the dose rate drops by several orders of magnitude. Absorbed doses to non-human biota are expected in the order of 0.05 to 20 μGy per year and

dependent on the aquatic organism and release scenario. The doses are well below any radiation limits for non-human biota, meaning that no deleterious effects from ionising radiation to the sea organisms is expected.D6

2. 論文

Bezhenar R., Takata H., de With G., Maderich V. Planned release of contaminated water from the Fukushima storage tanks into the ocean: Simulation scenarios of radiological impact for aquatic biota and human from seafood consumption. *Marine Pollution Bulletin*. 2021. Vol. 173. 112969.

Establishment of FISH probes for dicentric analysis of wild rodents in Chernobyl

氏名：Olena Burdo

受入研究者：Hiroko Ishiniwa

共同研究者：Tomisato Miura・Yohei Fujishima

1. 成果

In Chernobyl where the nuclear accident occurred 35 years ago, a large amount of radioactive materials remain in the environment. Dicentric chromosome analysis, a type of DNA lesion, is important and robust method to evaluate effect of radiation on animals. The frequency of dicentric chromosomes reflect the sum of internal and external exposure doses, therefore it is used as a biological dosimeter. However, dicentric chromosome analysis requires a great deal of time and skilled technique. Especially, most of chromosomes are acrocentric that are extremely short arms, makes analysis much difficult and many rodent species have this type of chromosomes. Bank vole, *Myodes glareolus* ($2n = 56$), is common rodent in Chernobyl also has 54 or 53 acrocentric chromosomes in female or male, respectively. The aim of this study is to establish the fluorescence in situ hybridization (FISH) method that visualize the centromeres of chromosomes for bank vole, simplify the procedure, and shorten the analysis time.

Five bank voles were captured in Ukraine using livetraps baited with oil-fried bread pieces. The livers were removed from euthanized bank voles and immediately stored in 70 % ethanol. The livers were incubated in lysis buffer containing proteinase K at 56 degrees with slow shaking and dissolved. Proteins were removed from the lysate by mixing with phenol and chloroform then DNA were purified by ethanol precipitation. Finally, about 45 micro-g/0.1 mg of genomic DNA of bank voles from each tissue were obtained.

As for the next step, we will make probe using extracted DNA and establish the primary cell culture from bank vole or related species to check whether probe works.

2. 論文

I-21-12

Monitoring of radon gas in the ground as an early warning system of earthquake

氏名 : Makhsun

受入研究者 : Shinji Tokonami

共同研究者 : Untara

1. 成果

Radon is one of many geophysical and geochemical phenomena that can be considered to be an earthquake precursor. The successful prediction of earthquakes is yet to be accomplished, in terms of their magnitude, location and time, and much effort has been spent on this goal. Radon emanation from grains depends mainly on their ^{226}Ra content and their mineral grain size, its transport in the earth being governed by geophysical and geochemical parameters while exhalation is controlled by hydrometeorological conditions. The stress-strain developed within the Earth's crust before an earthquake leads to changes in gas transport and a rise of volatiles from the deep earth up to the surface, resulting in anomalous changes in radon concentration.

This work aims to develop a monitoring system that conduct the systematic radon concentration measurements as early warning system of earthquake. This Research includes measurements of radon concentration in the soil at a depth of 80 cm along Opak and Progo faults, Yogyakarta and Cimandiri fault, Sukabumi. In addition this research carried out the laboratory tests on the homemade prototype of radon monitoring system.

The tests were carried out by monitoring the level of radon concentration in a bore well continuously for approximately three months. The monitoring measured the concentration of radon every hour, sent the data to the internet and storing it in a data logger. The tests were also carried out on solar panels as a source of electricity that used to feed the energy for the prototype device.

The measurements results of radon concentration in the soil in Opak fault ranged from $1,150 \pm 245 \text{ Bq/m}^3$ to $10,100 \pm 693 \text{ Bq/m}^3$, while thoron concentrations ranged from from $977 \pm 338 \text{ Bq/m}^3$ to $8,160 \pm 903 \text{ Bq/m}^3$. The measurements results of radon concentration in the soil in Progo fault ranged from $432 \pm 165 \text{ Bq/m}^3$ to $17,100 \pm 896 \text{ Bq/m}^3$, while thoron concentrations ranged from $488 \pm 277 \text{ Bq/m}^3$ to $4,340 \pm 672 \text{ Bq/m}^3$.

The results on laboratory test of homemade radon monitoring system show that the apparatus could work as expected and there are no obstacles or any damages. The prototype could measure the concentration of radon every hour and send the data to the internet and save it in a data logger. Solar panels and power storage devices could meet energy needs for device operations

2. 論文

Heavy Metal Assessments of Soil Samples from a High Natural Background Radiation Area, Indonesia, *Toxics* 2022, 10(1), 39.

Determination Cs-137 and Cs-134 in Food Samples

氏名 : Ceiga Nuzulia Sofyaningtyas

受入研究者 : Shinji Tokonami

共同研究者 : Kusdiana · Eka DjatnikaNugraha

1. 成果

Introduction:

Since the Fukushima Daichii nuclear accident 10 years ago, several radionuclides such as Cs-137 and Cs-134 have been released into the environment and even outside Japan. Radionuclide Cs-137 and Cs-134 are two of the fission products of the nuclear fission process in a nuclear facility. Cs-137 and Cs-134 has high toxicity and long half-life. When it enters the human body, Cs-137 and Cs-134 will attack the bones or muscles and can cause cancer. It becomes important to know the content of Cs-137 and Cs-134 which may be present in the environmental samples, especially in food consumed by the population. This is in line with Indonesian government regulations regarding food safety requirements in Indonesia, including foods imported from Japan, by setting limits for radioactive contamination in foodstuffs. For this reason, research to determine the content of Cs-137 and Cs-134 in food samples is necessary.

Method: Using particle size 100-600 μm KNiFC-PAN Resin (EICHROM, USA).

Procedures:

1. Resin characterization using mixed standard solution (Merck, Germany).

Mix standard solution containing stable Cs, Fe, Na, K, Mg, Ca, Sr (20 mg kg⁻¹ each) streamed through 2 ml bed of KNiFC-PAN resin by 1-3 ml min⁻¹ of flow rate using a peristaltic pump. Every 7 ml of the output were collected and measured using FAAS (Flame Atomic Absorption Spectrophotometer).

Result: Chemical Recovery of Cs = 99.93%

2. Resin Characterization using food samples.

1000 mg kg⁻¹ of Cs carrier was added into 10 g of dried pineapple sample. The destruction was carried out using HNO₃ and H₂O₂. The solution then streamed through the resin. FAAS measurements were carried out to compare the Cs concentration in the solution before and after passing through the resin. (Table 1.)

Table 1. Cs Concentration in Food Samples Measured by FAAS

Sample blank -> Before resin: - ; After resin: 0.0461 mg/kg

Sample+Cs carrier -> Before resin: 20,32 mg/kg ; After resin: 1,493 mg/kg

2. 論文

Evaluation of the Serum Catalase Activity in the Individuals Exposed to High Levels of Radon Concentration: A Pilot Study

氏名 : Dwi Ramadhani

受入研究者 : Tomisato Miura

共同研究者 : Sofiati Purnami

1. 成果

In Indonesia, Tande-Tande sub-village in Botteng Utara (Northern Botteng) village, Mamuju, West Sulawesi, is known to have a high indoor radon concentration. The indoor radon concentrations in this location can exceed 1,644 Bq/m³. People in this area are exposed chronically to high levels of radon. Radon is an odorless and colorless noble gas derived from the uranium decay chain (U-238), which results directly from the decay of radium (Ra-226). Alpha and beta radioactive isotopes are among the radon decay products. Alpha particles, which are made up of two protons and two neutrons, have the capability to ionize and damage the DNA contained in living cells. Ionizing radiation in the form of alpha particles can cause DNA damage from chromosomal aberrations (CA), double strand DNA breaks and generate reactive oxygen species (ROS). As a form of free radical, ROS includes molecules such as singlet oxygen, superoxide anion, hydrogen peroxide, and a hydroxide radical. Several mechanisms exist in the human body for controlling and inhibiting ROS generation. One of them is by producing enzymatic-antioxidant enzymes such as superoxide dismutase (SOD), glutathione peroxidase (GPX), and catalase (CAT). Interestingly, our preliminary study showed that SOD and GPX activities in Tande-Tande sub-village inhabitants did not differ significantly from those in Topoyo inhabitants. Thus, to explore more information on the effects of radon exposure in this area, we assessed the frequencies of CAT concentrations in the sera of Tande-Tande sub-village inhabitants and compared the results to those obtained from populations who live in areas with low indoor radon concentrations. Fifty seven adult subjects from Tande-Tande sub-village, Mamuju and fifty three healthy adult subjects from normal background radiation areas (NBRAs) were enrolled in this study. Blood samples were obtained from the antecubital vein. Sera then were collected by centrifugation at 3600 r.p.m for 15 min and keep at 4° C. The catalase concentration in serum samples then were measured using Catalase ELISA Kit in our study. The median CAT concentration per total protein in the Tande-Tande sub-village inhabitants (0.159) was statistically lower compared to median value found in Topoyo village inhabitants (0.256; $p < 0.001$; Fig. 1). No significant correlation between CAT concentration per total protein and indoor radon concentration, annual effective dose, and cumulative dose in Tande-Tande sub-village inhabitants ($p = 0.262$; $p = 0.141$; and $p = 0.064$, respectively), as well as in Topoyo village inhabitants except in CAT

concentration per total protein with annual effective dose ($p=0.044$). Interestingly, the significant correlation between CAT concentration per total protein and indoor radon concentration, annual effective dose, and cumulative dose were found when all data from both groups were pooled ($p<0.0001$; $p<0.0001$; and $p=0.001$, respectively).

2. 論文

I-21-15

Detection of 8-oxo-dG level in serum and γ -H2AX, micronuclei, and apoptosis caspase-3 in lymphocyte of Indonesian Nuclear Reactors

氏名 : IIN KURNIA

受入研究者 : Tomisato Miura

共同研究者 : Yanti Lusiyanti · Teja Kisnanto · Devita Tetriana

1. 成果

Detection of 8-oxo-dG level in serum and γ -H2AX and Annual Occupational Dose, of Indonesian Nuclear Reactors Background: Reactor workers are exposed to long-term low levels of ionizing radiation, which makes them vulnerable to DNA damage. There are potential occupational health hazards from radiation exposure. Purpose : Detection of 8-oxo-dG level in serum and γ -H2AX and Occupational Dose in lymphocyte of Indonesian Nuclear Reactors Materials and Methods: 28 blood samples will be taken from 28 consist reactor workers and other radiation workers(man), follow up from 2020 subject before. and they will be analyzed for the presence of 8-oxo-dG level in serum and γ -H2AX, in lymphocyte, 8-0xo dG will be detected by Elisa and γ -H2AX expression as a biomarker of DNA DSB damage assay were carried out by immunofluorescence microscopy. Result : There was a significant correlation between concentration of 8-oxo-dG level in serum to age and working duration of workers ($p < 0,05$) and no significant correlation between of 8-oxo-dG to expression of γ -H2AX and annual occupational dose (Fig, a,b,c,d) a b c d *Because of some major situation as Covid 19 Pandemic the Caspase-3 could not be detected and still the validation data of micronucleus expression

2. 論文

Have not Published

Determination of Pu-239/240 in water

氏名 : Elistina

受入研究者 : Naofumi Akata

共同研究者 : Kusdiana · Eka Djatnika Nugraha

1. 成果

Plutonium is a radionuclide particularly difficult to measure in environmental and need radiochemical method for determination of 239 , 240 Pu and 241 Pu. During the analysis, tracer 242 Pu was use to estimate the chemical yield of the plutonium isotope. After suitable pre-treatment of the samples, the plutonium nuclides in solution were co-precipitated with ammonium hydrogen phosphate and calcium nitrate. Recent developments in ion exchange resin and extraction chromatography resins such as Eichrom[®] UTEVA and TRU have resolved the analytical problems but drawbacks such as low recovery and spectral interferences still occasionally occur. The methods able to separate from one same sample, both natural and anthropogenic radionuclides. The method is also suitable for measuring 239 , 240 Pu in water samples at the μ Bq/l level. Considering the high radio-toxicity of Pu, accurate methods are required to measure activities in the environment. In the publication 239 Pu and 240 Pu, due to the two radionuclides' energy, these are very close together, so the result measurement is a mixture of the two radionuclides, with a composition of 60% 239 Pu and 40% 240 Pu. In that way, radiochemistry analysis have been proposed for the determination of 239 , 240 Pu in environmental samples. Besides, environmental samples often contain other α -emitter radionuclides (U, Am, Th and their daughter's products), at much higher activities than Pu.

The radiochemical protocols follow three main steps : sample digestion, chemical separation and measurement. All reagents were of analytical grade. Pre packed columns of UTEVA and TRU resins. Preparation 2 L of water sample was placed into a glass beaker. Then the sample was evaporated until the remaining volume was 500 mL. The evaporated sample was then acidified to pH 2 using concentrated nitric acid. Then 242 Pu tracer, U tracer and Am tracer were added to the sample. The beaker glass containing the sample was then carried out preparation stage until the dark red color changed to a dark yellow color which indicated the reduction of Fe⁺³ to Fe⁺². Actinides separation for this, ion exchange resin and extraction chromatography resins UTEVA to separate U from Am and Pu, TRU to separate Am and Pu. Uranium separation ; the resin to be used is UTEVA (Uranium & Tetravalents Actinide) which can be used to separate uranium, zirconium, and actinide (IV) elements such as Np, Th and Pu. Americium separation ; the resin to be used is TRU (Trans Uranian) which can separate iron and actinide groups. The conditioned resin was then added to the eluent from the elution of the sample using UTEVA resin containing Pu and Am. Plutonium

separation ; the resin used to separate plutonium is TRU resin. The resin that was used to elute the americium. The three eluent are evaporated and form a precipitate dissolved with HNO₃ then input to vial and added cocktail. The final solution was measured using an ultra low-level liquid scintillation spectrometer, Quantulus 1220 Wallac (Perkinelmer, USA).

In this experiment, LSC by radiochemical method for element separation can be successfully applied ^{239,240}Pu as well as ²³⁸U and ²⁴¹Am. The result of the analysis of plutonium radionuclide samples using standard tritium. The counting efficiencies are nearly for ^{239,240}Pu and ²⁴¹Pu. The proposed procedure has been tested for the simultaneous determination of ²³⁹Pu + ²⁴⁰Pu and ²⁴¹Pu in water samples. Through this research project, it can be obtained that measurement result for the radiochemical analysis methods and can be used to estimate activity of ^{239,240} Pu in water samples. This activity is expected to be able to understand environmental radioactivity.

2. 論文

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**Development and implementation of methods to assess radiocesium
transfer in forest soil ecosystems**

氏名 : Mathew James Bruce Swallow

受入研究者 : Naofumi Akata

共同研究者 : Yusuke Unno · Akira Takeda

1. 成果

Initially the plan for this project was for Dr. Swallow to travel to Japan in the fall of 2021 and then January 2022 to conduct the research at Hirosaki University. These plans were drastically altered due to the pandemic resulting in Dr. Swallow not being able to travel to Hirosaki. However, the research team worked diligently since the start of 2022 to ensure progress was made. Consequently, this annual report will reflect the progress under these challenging circumstances. The method to assess radiocesium mobility in soils has fully developed in Canada. Currently running experiments show that the method is able to effectively manipulate water transmission from soil in relation to predetermined levels of soil water activity (-0.5 MPa, -1.0 MPa and -2.0 MPa). In the near future these samples will be tested to see if the manipulated levels of soil water activity result in corresponding increase in water soluble elements being transported out of the soil. Ultimately, this work will generate soil nutrient isotherms that indicate the level of nutrient availability in relation to soil water activity. While this work is being done outside of scope of this ERAN proposal, it will demonstrate that the method should work as a means to determine radiocesium mobility. Current plans are for Dr. Swallow to travel to Hirosaki University in July 2022 and replicate the method on soils from Fukushima. An additional objective was to determine if it is possible to remove radiocesium from soil by creating Prussian blue colloids in-situ and then removing both the Prussian blue colloids and radiocesium. Work done at Mount Royal University in Canada by Dr. Swallow shows that it is possible to generate Prussian blue colloids in-situ and remove the colloidal components from soil. Yet it is still unknown if there is a corresponding removal of radiocesium. On that end, Dr. Akata has coordinated with Dr. Takeda and Dr. Unno to begin work in Japan. This work is ongoing and will continue with the support provided by ERAN this year.

2. 論文

Gross alpha and gross beta radioactivity concentration in environmental water

氏名：Chen Fei

受入研究者：Hirofumi Tazoe

共同研究者：Quiang Xiong · Lu Zhang

1. 成果

In this work, the water samples were collected in 2012 from Ukedo River near Fukushima, and gross-alpha, gross-beta, tritium, and γ radionuclides were determined. The purpose of this work is to estimate whether the samples were still harmful to the environment and human body.

The methods were all the standard methods used in China.

From Table 1, we can see the gross alpha and gross beta activity of all samples were at very low level, most of the samples were below the MDL (minimum detection limit) .

From Table 2, we can find the tritium level of all the samples were at environmental background level.

Besides, 12 radionuclides were measured using γ spectrometer, only Cs-137 was found in R3 , R6-12, R14 at the level of 0.32-1.95 Bq/kg.

At last, it can be concluded the received water samples from Ukedo River were at low levels of radioactivity, resulting in little impact on the environment and human health.

2. 論文

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Method development and analysis of refractory radionuclides in seawater samples

氏名：Shan Xing

受入研究者：Hirofumi Tazoe

共同研究者：Xiongxin Dai・Taeko Shinonaga

1. 成果

Introduction: Pu isotopes are anthropogenic radionuclides with high radiological toxicity and persistence in the environment. They are considered to be one of the major contributors to the internal dose in people. For the analysis of Pu isotopes at femtogram levels or lower in sparse samples, highly sensitive analytical techniques are required and it is very challenging to obtain reliable Pu data. We are accordingly developing the analytical methods for Pu isotopes.

Method

The separation method for Pu isotopes was developed using

- co-precipitation method
- anion exchange and extraction chromatography
- ICP-MS/MS equipped with a high sensitivity sample introduction system

Results and Discussion

Ø Comparison of the removal efficiency of U interference at different NH₃ flow rates
The results showed that the overall interference from uranium in count rates was reduced by a factor of $< 2.4 \times 10^{-7}$, shifting U⁺ to U(NH_m)_n⁺ and UH(NH_m)_n⁺ under the combined condition: 0.4 mL/min NH₃ – 6.4 mL/min He.

Ø Effect of collision conditions on Pu detection

The results showed high ²³⁹Pu sensitivity: 13,900 Mcps / (mg/L) due to ion focusing effect of Pu by NH₃ and He in the collision/reaction cell. The optimum parameters of AA, He, NH₃ and KED were 1.6 V, 6.4 mL/min, 0.4 mL/min and -11.2 V, respectively.

The detection limits for the method were estimated to be 0.2 fg for ²³⁹Pu, 0.05 fg for ²⁴⁰Pu and 0.04 fg for ²⁴¹Pu.

Ø Applications

The method was applied to determine ultra-trace Pu isotopes in 1g sediment samples for sediment dating.

The method enables an estimation of < 1 mSv dose in urine samples.

Acknowledgements

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are gratefully acknowledged. References

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[2] Y. Wu, Y. Xu, S. Xing, et al., Rapid determination of ultra-trace plutonium in large volume of urine samples by tandem quadrupole ICP-MS. *Spectrochimica Acta Part B: Atomic Spectroscopy* 184 (2021) 106259.

Capacity building for development of nuclear techniques for
analysis of radionuclides in environmental samples collected from
Ruppur Nuclear Power Plant (RNPP), Bangladesh

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受入研究者 : Hirofumi Tazoe

共同研究者 : Tasrina Rabiya Choudhury

1. 成果

Soil samples were collected from ten sampling points surrounded the Rooppur Nuclear Power Plant to determine the metal concentration like As, Pb, Rb, Sr, Zr, Cu, and Zn. The metals concentrations in soil samples were determined by the Energy Dispersive X-ray Fluorescence (EDXRF) technique. The average metal concentrations in the study area were found in the following descending order: Zr (334.84 mg/kg) > Rb (179.35 mg/kg) > Pb (172.77 mg/kg) > Sr (138.86 mg/kg) > Zn (120.54 mg/kg) > Cu (16.96 mg/kg) > As (8.50 mg/kg) respectively. Moreover, the sampling sites were organized according to the total metal concentration as S7 > S3 > S4 > S5 > S11 > S2 > S9 > S1 > S10 > S8 > S12 > S6 respectively. This study showed that the average concentration of As, Sr, and Cu surpassed the suggested standard and average shale value (ASV) limits. In this study, the identification of the soil quality associated with the ecological and human health risk was evaluated using some important indices. The geoaccumulation index (I_{geo}) deliberated that most of the sites were not contaminated except for S3 and S7, which were classified as strong contamination and strong to extreme contamination. The enrichment factor (EF) revealed that three sampling points of S3, S4, and S7 were enriched with Pb, which was consistent with the contamination factor (CF) suggested that the study area was contaminated by only Pb and Zn. It might happen due to excess battery rechargeable vehicles in the study area. However, this study revealed that the pollution load index (PLI) for most of the sampling points was lower than 1 (PLI < 1) suggesting good soil quality in the study area. The finding of PLI was similar to the potential ecological risk assessment, which signified that the study area was not in a harmful situation as it did not surpass the limit of 100. Subsequently, the assessment of human health risk suggested that both adults and children would not be exposed to the non-carcinogenic and carcinogenic risks as these remained under the corresponded threshold limits (1 and 10⁻⁶ to 10⁻⁴ respectively). However, this study suggests an action plan for the continuation of further investigation for monitoring the soil quality change due to the nuclear power plant's present ongoing construction work and future operation in the study area

2. 論文

Multipotential Trace Metal Concentrations in Soil Associated with the Ecological and Human Health Risk near the Rooppur Nuclear Power Plant, Pabna, Bangladesh

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**Assessment of natural radioactivity and radiological health impact
on paleontologists and workers in dinosaur fossil excavation
sites containing with high-background radiation**

氏名 : Siritorn Buranurak

受入研究者 : Shinji Tokonami · Kranrod Chutima

1. 成果

In 2021, the project entitled “Assessment of natural radioactivity and radiological health impact on paleontologists and workers in dinosaur fossil excavation sites containing high-background radiation” received a great opportunity to be granted by the ERAN FY2021. It is the collaborative project between the IREM-HiroDai and the Physics-KKU, aiming to 1) quantify the natural radioactivity in fossil excavation sites and 2) assess the radiological risks caused by inhalation of radon-thoron gases and their progenies. The idea of this project was inspired by the exploratory report informing that the Korat plateau (i.e., where is considered the largest sandstone-type uranium resource in Thailand) has geologically similar to the Colorado Plateau of the United States, where radioactive fossils had been discovered. Even though the exploration of radioactive materials has been previously reported in some areas of Thailand, the study focusing on the assessment of radioactivity in fossil excavation sites and quantification of radioactivity concentration in dinosaur fossil bones has never been presented. For this study, the USB-Inspector (S.E International, Inc., USA) was used to demonstrate radioactivity levels over dinosaur excavation sites. Optically Stimulated Luminescence Dosimeters (OSLDs) were used for seasonal monitoring of gross-gamma radiation, while CR-39 plastic nuclear track detectors were used to monitor radioactive gas to estimate lung cancer risks preliminarily. The overall gamma radiation obtained from both excavation sites was in the range of 0.036 – 0.220 μ Sv/h and was found to be consistent with the seasonal gamma doses observed by the OSLDs, while seasonal radon gas concentration was in the range of 3.06 – 74.01 Bq/m³. Although the radon levels measured from both sites were found to be less than 148 Bq/m³ of the maximum level allowed by the IAEA standards, some principle safety plans are recommended for safety reasons – for example, using a working-time limit to reduce radiation exposure, using a face shield or mask to prevent radioactive dust from getting into the respiratory system, and improving an airflow system to dissipate the radon concentration.

2. 論文

I-21-22

High-resolution reconstruction of sediment and radiocesium source contributions in a lake draining the main Fukushima radioactive pollution plume

氏名 : EVRARD Olivier

受入研究者 : Seiji Hayashi · Hideki Tsuji · Yoshifumi Wakiyama

共同研究者 : Anthony FOUCHER · Irène LEFEVRE

1. 成果

In 2021, because of the travel restrictions due to the covid-19 pandemic, fieldwork in Japan was not possible for the French partners. As an alternative output, the full database of radionuclide activities measured in sediment lag deposits collected along rivers draining the main radioactive plume of Fukushima Prefecture was published in full open access and presented in a specifically dedicated data paper (also available in open access): Evrard, O., Chartin, C., Lacey, J.P., Onda, Y., Wakiyama, Y., Nakao, A., Cerdan, O., Lepage, H., Jaegler, H., Vandromme, R., Lefèvre, I., Bonté, P. (2021). Radioactive dose rates and fallout radionuclide activities in sediment deposits along rivers draining the main Fukushima plume, Japan. PANGAEA, <https://doi.pangaea.de/10.1594/PANGAEA.928594> Evrard, O., Chartin, C., Lacey, J. P., Onda, Y., Wakiyama, Y., Nakao, A., ... & Bonté, P. (2021). Radionuclide contamination in flood sediment deposits in the coastal rivers draining the main radioactive pollution plume of Fukushima Prefecture, Japan (2011–2020). *Earth System Science Data*, 13(6), 2555-2560. In addition, two sediment cores were kindly collected in June 2021 (in midstream and downstream positions) by the Japanese partners from NIES in the Mano Dam Reservoir, in Fukushima Prefecture, and sent for analysis to France (because of the border control procedures, the sediment cores arrived in France in September 2021). The analyses led by Thomas Chalaux, a PhD student who started his thesis in October 2021, included fallout radionuclides by gamma spectrometry, XRF core scanner, elemental XRF (to calibrate the relative signals obtained with the XRF core scanner), laser granulometry, visible colorimetry, Carbon/Nitrogen total contents and stable isotopes (d13C and d15N). Currently, a very high resolution sediment source tracing technique based on the high resolution XRF records is being developed to investigate the changes in sediment source contributions before and after the 2011 accident and the impact of decontamination on source changes. Preliminary results confirm that the entire period between 2011 and 2021 is covered by the sediment archives, which will allow to reach the objectives.

2. 論文

Evrard, O., Chartin, C., Laceby, J.P., Onda, Y., Wakiyama, Y., Nakao, A., Cerdan, O., Lepage, H., Jaegler, H., Vandromme, R., Lefèvre, I., Bonté, P. (2021). Radioactive dose rates and fallout radionuclide activities in sediment deposits along rivers draining the main Fukushima plume, Japan. PANGAEA, <https://doi.pangaea.de/10.1594/PANGAEA.928594>

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Characteristics of radioisotope fingerprints of forest soils in Changbai Mountain, Northeast China

氏名 : Liu Zhiyong

受入研究者 : Jian Zheng

共同研究者 : Li Zhihong · Han Xiaoxiao

1. 成果

$^{239+240}\text{Pu}$, ^{237}Np and ^{241}Am in soils and sediments in the Northeast China were analyzed by sector magnetic field electromagnetic inductively coupled plasma mass spectrometry (SF-ICP-MS, ELEMENT 2), and ^{137}Cs , ^{238}U , ^{232}Th , ^{226}Ra , ^{210}Pb were analyzed by anti-Compton γ -ray spectrometer. Interestingly, the activity of these radionuclides in different soils and sediments varies greatly. In the surface samples, the $^{239+240}\text{Pu}$ activity in Zhalong Wetland (2.09-10.84 Bq/kg) was higher than that of other sample types (0.05-2.32 Bq/kg), ^{137}Cs also showed high activity in wetlands. The highest ^{237}Np activity (2.93 mBq/kg) appeared in lake sediments, and the highest ^{241}Am activity (1.48 Bq/kg) appeared in river sediments. The inventories of ^{137}Cs , ^{237}Np , $^{239+240}\text{Pu}$, and ^{241}Am in Zhalong Wetland were 823.9-2468.7, 0.110-0.785, 57.2-1286.7, and 310.9-443.5 Bq/m², respectively, which is very rare in the environment, much higher than other soils. In addition, the convective-dispersion equation model was used to exhibit the convection/dispersion phenomenon of radionuclides in different soils, and it was found that the dispersion process of ^{137}Cs and $^{239+240}\text{Pu}$ is dominant, while the convection process of ^{237}Np is dominant. $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio showed that the source of $^{239+240}\text{Pu}$ in Northeast China is mainly global fallout. However, the $^{237}\text{Np}/^{239}\text{Pu}$ atomic ratio and $^{241}\text{Am}/^{239+240}\text{Pu}$ activity ratio varied widely, mainly because non-isotopic deposition rates vary. This work revealed the background value and source of artificial radionuclides in sediment core in Zhalong Wetland and Chagan Lake, soil core in paddy fields sediment and dry fields soil, surface sediment samples in lakes, rivers, and reservoirs, and surface soil in forest in Northeast China, clarified the differences in the enrichment of radionuclides in the wetland system, and further proposed the importance of studying the distribution pattern and environmental behavior of radionuclides in different environments such as wetlands.

2. 論文

I-21-24

Simultaneous determination of ^{237}Np and Pu isotopes in seawater by SF-ICP-MS with a single column chromatographic separation

氏名：Shaoming Pan

受入研究者：Jian Zheng

共同研究者：Shuai Zhang

1. 成果

FY2021 ERAN Final Report (I-21-24) Simultaneous determination of ^{237}Np and Pu isotopes in seawater by SF-ICP-MS with a single column chromatographic separation Shaoming Pan (Nanjing Univ. China), Shuai Zhang (Nanjing Univ. China), Jian Zheng (QST) ^{237}Np is a potential geochemical tracer since it has a similar behavior to ^{137}Cs in the marine environment and a long half-life (2.14×10^6 y). This project aims to develop a new method for simultaneous analysis of ^{237}Np and Pu isotopes in small volume seawater by SF-ICP-MS. $\text{Fe}(\text{OH})_2$ co-precipitation was used for the preconcentration of ^{237}Np and Pu isotopes from 15L seawater sample, and $\text{CaF}_2/\text{LaF}_3$ co-precipitation and two anion exchange AG MP-1M resin column chromatographic were used to remove the matrix and to separate and purify target radionuclides. Following the optimization of the redox system, the concentration efficiency (%) of ^{237}Np and $^{239+240}\text{Pu}$ were 99.1 ± 1.0 (n=3) and 97.8 ± 2.2 (n=3), respectively, in the $\text{Fe}(\text{OH})_2$ co-precipitation procedure. 20mL conc. HBr with small amount of HF (0.1M) can recover both Pu (0.860 ± 0.002 , n = 3) and Np (0.915 ± 0.019 , n = 3) efficiently from two AG MP-1M resins, which indicate that ^{242}Pu can be used as yield tracer for the simultaneous determination of ^{237}Np and Pu isotopes. The high U decontamination of 4.2 or 3.5 $\times 10^6$ was achieved, which made the U interference negligible. This method to effective detection of ultra-trace level ^{237}Np and Pu isotopes in seawater. Further experiments will be performed to validate and evaluate the method by analyzing the standard reference material IAEA-443. Meanwhile, a paper entitled "Vertical distribution characteristics of Pu isotopes, ^{137}Cs and ^{237}Np in sediments of Lake Xingkai and their response to catchment environmental changes" has been submitted for publication. And another paper entitled "Method development for rapid analysis of ^{237}Np and Pu isotopes in small volume seawater (15L) by SF-ICP-MS" is in preparation.

2. 論文

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

氏名 : Wang Zheng
受入研究者 : Jian Zheng

1. 成果

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 $^{239+240}\text{Pu}$ activities and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios, as well as Hg concentration and isotope compositions in deep-sea sediments in the South Pacific for the first time. The purpose of this project is to understand the contributions of anthropogenic activities and natural processes to these two common contaminants in marine environments that are relatively remote from direct pollution. We found extremely low $^{239+240}\text{Pu}$ inventories in the South Pacific sediments, suggesting relatively less fallout compared with that of North Pacific. The $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in the South Pacific sediments (0.097 ± 0.018) are well below the global fallout value (0.174 ± 0.014), suggesting that global fallout is unlikely the dominant source of Pu in the South Pacific. We suggest that French Nuclear Tests and/or the United Kingdom Nuclear Tests in Australia, which generated relatively low $^{240}\text{Pu}/^{239}\text{Pu}$, are the main sources of Pu. Using an isotopic mass balance model, we estimated that the contribution of French Nuclear Test ranges from ~10% to ~90%. The plausible pathways for Pu transportation were the South Pacific Gyre, based on the observation of latitudinal trends in both the $^{239+240}\text{Pu}$ inventories and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios.

We also studied Hg isotopes in two sediment cores from the Southern Ocean near the Ross Island, Antarctica. Ocean plays a critical role in the global mercury (Hg) cycling, due to the in-situ formation of neurotoxic methylmercury (MeHg) in seawater and its bioaccumulation in marine food web, but the source and transformation of Hg in ocean are not fully understood. We measured Hg concentrations and stable isotopes in two deep-sea sediment cores (SX-09 and SX-10) in the North Ross Sea to investigate the source of Hg to the Southern Ocean and the impact of the glacial-interglacial climate change on marine Hg cycling. Sedimentary Hg isotope data suggests significant changes in Hg sources with time. In SX-10, atmospheric deposition via long-range transport was the dominant Hg source before the Last Glacial Maximum (LGM), as evidenced by slightly positive mass independent fractionation (MIF, $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$) and negative mass dependent fractionation (MDF, $\delta^{202}\text{Hg}$), which is similar to those of modern open-ocean sediments. In contrast, the SX-09 core shows

strongly negative $\Delta 199\text{Hg}$ but positive $\Delta 200\text{Hg}$ before LGM, which are similar to the Hg isotope signatures of Antarctic snow. We suggest that the SX-09 core was dominated by Hg input from glacial materials transported from the Antarctic icesheet during the expansion of glacial. However, after LGM, $\Delta 199\text{Hg}$ and $\Delta 200\text{Hg}$ in both cores shifted consistently towards zero together with increases of Hg concentration and $\delta 202\text{Hg}$, which may have recorded enhanced volcanic Hg emission (near-zero MIF) and terrigenous input (negative MIF) from Antarctica due to the retreat of glacial. Our results show that Hg isotopes in the Southern Ocean can record the historical Hg cycling and its responses to climate changes. In the year of 2022 we expect to publish the above results as 2-3 papers.

2. 論文

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I-21-26

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

氏名 : Hai Wang

受入研究者 : Jian Zheng

共同研究者 : Renjuan Liu

1. 成果

Studies on the distribution and migration characteristics of Np and Pu in typical karst environment 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 drip waters. The distribution and transfer of ^{237}Np and Pu isotopes in the karst environment remains largely unknown. The radioactivity of ^{137}Cs , $^{239+240}\text{Pu}$ and ^{241}Am in soils and lake sediment from Hunan province were investigated. The $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio were 0.177 ± 0.003 in surface soil and 0.182 ± 0.011 in lake sediment and were consistent with the characteristic value of global fallout. Vertical distributions of $^{137}\text{Cs}/^{239+240}\text{Pu}$ and $^{241}\text{Am}/^{239+240}\text{Pu}$ indicated the migration velocity was $\text{Am} \approx \text{Pu} > \text{Cs}$. Future studies will continue the investigation the regional distribution, vertical distribution profile and migration mechanism of ^{237}Np and Pu and aim to provide technical support for environmental impact assessment in karst areas during nuclear emergency response.

2. 論文

Distributions of fallout ^{137}Cs , $^{239+240}\text{Pu}$ and ^{241}Am in a soil core from South China

I-21-27

Analytical method of ^{135}Cs and ^{137}Cs in large volume of seawater by ICP-MS spectrometry

氏名：Wu MEN

受入研究者：Jian Zheng

共同研究者：Fenfen WANG

1. 成果

^{135}Cs is a promising geochemical tracer to study the seawater mixing and exchange. This project will establish an analytical method for Cs isotopes (^{135}Cs and ^{137}Cs) in large volume of seawater : 1.The separation and purification techniques of Cs isotopes in the large volume of seawater will be achieved. 2.The measuring techniques using triple-quadrupole inductively coupled plasma – mass spectrometry also will to be developed. At present, the recoveries of ~95.5% were obtained in the series of experiments using 10ml AMP resin to concentrate 60L of seawater sample under the flow rate of 60-80ml/min. The AMP leaching experiment is in progress. Due to the COVID-19, the remaining part of the experiments and measurements have not been implemented in QST yet. In 2021, I filed to find a partner to complete the remaining experiments in China. So the remaining part of the experiments is at a standstill. I hope to complete it once the epidemic is over. Keywords: ^{137}Cs , ^{135}Cs , seawater, analytical method, ICP-MS

2. 論文

no