

放射能環境動態・影響評価
ネットワーク共同研究拠点 (ERAN)

2020 年度 年次報告書
【海外共同研究】

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I-20-14	Dwi Ramadhani	National Nuclear Energy Agency of Indonesia	A Preliminary Study to Assess The Activity of Antioxidant Enzymes in Population Exposed to Very High Levels of Radon Concentration	三浦 富智	IREM
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受付番号	申請者氏名	所属機関名	研究課題名	受入研究者	受入機 関
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I-20-19	Chanis Pornnumpa	Kasetsart University	EVALUATION OF THE CONVERSION FACTOR OF RADON AND THORON PROGENY DETECTOR IN DWELLING USING TRACK ETCH DETECTORS	Chutima Kranrod 玉熊 佑紀	IREM
I-20-20	Zhou shumin	East China University of Technology	Study on the migration law of radioactive aerosol	Chutima Kranrod 玉熊 佑紀	IREM IREM
I-20-21	Wu men	Third Institute of Oceanography	Analytical method of ^{135}Cs and ^{137}Cs in large volume of seawater by ICP-MS spectrometry	鄭 建	QST
I-20-22	Hai Wang	University of South China	Studies on the distribution and migration characteristics of Np and Pu in typical karst environment	鄭 建	QST
I-20-23	Wang Zheng	Tianjin University	Tracing the sources of Pu and Hg in the deep-sea sediments using Pu and Hg isotopes	鄭 建	QST
I-20-24	Shaoming Pan	Nanjing University	Method development for simultaneous analysis of ^{237}Np and Pu isotopes in the seawater by SF-ICP- MS	鄭 建	QST
I-20-25	Xiongxin Dai	China Institute for Radiation Protection	Source identification and environmental impact assessment of actinides around Fukushima: Development of ultra-sensitive methods in	鄭 建	QST

【I-20-01】

Atmospheric deposition and canopy exchange processes at different topographic gradients in a forested watershed

Principal Researcher : Xinchao Sun

Host Researcher : Yuichi Onda

1. Report (about one page of A4 paper)

Forest canopies create a high spatial variability of both canopy interception (Ei) and acidic and base cationic deposition. However, spatial characteristics of these hydrochemical fluxes along a slope gradient in forest watersheds remain unclear. Here we monitored bulk precipitation (BP), throughfall (TF), stemflow (SF), and associated acidic and base cationic fluxes, in addition to acid neutralizing capacities (ANC) and alkalinity (ALK) in an upper slope plot (UP) and a lower slope plot (LP) in a subtropical broadleaved forest, Southeast China. In addition, a canopy budget model was applied to distinguish the relative contributions of dry deposition (DD) and canopy exchange (CE) to net TF + SF (NTF) flux. Results showed that annual rates of TF, SF and Ei to BP in UP were 85.2%, 2.6%, and 12.2%, respectively. Corresponding values in LP were 88.6%, 2.2%, and 9.2%. There were no significant differences among these rainfall partitioning components between UP and LP. Additionally, annual volume weighted mean concentrations of NO₃--N in TF and SF were significantly lower than in BP, whereas no significant differences were found for SO₄²⁻-S among BP, TF and SF. Unlike annual TF NO₃--N fluxes were similar in UP and LP, annual TF SO₄²⁻-S fluxes significantly increased along the UP to LP. Net TF + SF (NTF) fluxes were negative for NO₃--N and SO₄²⁻-S at the two slope plots, illustrating retained/uptake in the canopy. Dormant season NTF S fluxes were close to zero, indicating that dry deposition does not appear to play a major role, whereas exchange with foliage surfaces should be the dominant factor controlling the spatial patterns of TF N and S fluxes in the watershed. In addition, annual volume weighted mean concentrations of base cations in TF and SF were significantly higher than those in BP in both UP and LP. Annual fluxes of TF or SF base cations had no significant differences between UP and LP, resulting in a comparable of the NTF fluxes. However, the relative contributions of DD and CE to NTF were different at the

two slope plots. NTF K⁺ flux was dominated by CE regardless of slope position. Unlike K⁺, NTF Ca²⁺ and Mg²⁺ fluxes were dominated by DD in UP, while CE in LP. In addition, ANC and ALK of TF and SF were significantly higher compared to BP, suggesting rainwater beneath the canopy became less acidic and may mitigate in part the risk of soil acidification, especially in the dormant season. These findings can provide insights into the spatial variability of chemical characteristics of hydrochemical fluxes, and improve our understanding of the quantities and controls of external and internal nutrient inputs occurring within the canopy in forest watershed.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

- (1) Xinchao Sun, Zhao Zhang, Yanhong Cao, Li Liu, Feilong Hu, Xiaoqiang Lu *. Canopy modification of base cations deposition in a subtropical broadleaved forest: Spatial characteristics, canopy budgets and acid neutralizing capacity. *Forest Ecology and Management*, 2021, 482, 118863.
- (2) Xiaoqiang Lu#, Yan Liu#, Li Liu, Zhao Zhang, Feilong Hu, Xueyan Liu, Xinchao Sun*. Rainfall partitioning and associated nitrate and sulfate fluxes along a slope gradient in a subtropical broadleaved forest. *Journal of Hydrology*, 2020, 591: 125584.

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【I-20-02】

Developing deeper insights into water cycling of forests using radiocesium as a tracer

Principal Researcher : Delphis Levia
Host Researcher : Yuichi Onda • Hiroaki Kato

1. Report (about one page of A4 paper)

1. Research description

Our research examines the vertical flux (leaching and washing-off) and cycling of radiocesium via the branchflow and stemflow within forest stands, and its transport in the aftermath of the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. In particular, the research investigated and compared the distribution of radiocesium concentration (specifically Cs-137) transported by branchflow from upper and lower portions of the canopy and deposited by stemflow to the trunk-base. Furthermore, a stable isotope approach was initiated as to define the mixing process of intercepted rainfall, a signature of evaporation loss, and elucidate the transport of radiocesium via branchflow and stemflow on its routing through the canopy. The research was conducted in a coniferous forest (*Cryptomeria japonica* (L. f.) D. Don, young Japanese cedar stands, mean height of 10.9 m, mean DBH of 0.560 m) and a mixed deciduous broadleaved forest (*Quercus serrata* Murray, Japanese oak stands, mean height of 14.3 m, mean DBH of 0.789 m). The forest plots were located in Yamakiya, Fukushima Prefecture, with a radial distance of approximately 40 km from FDNPP. In addition, individual experimental tree stands were selected. Cedar: Cedar A, Cedar B and Cedar C and oak: Oak 1 and Oak 3. Branchflow was collected from branches of younger foliage, mixed foliage and dead foliage for cedar stand, while branches of uppermost foliage and mid-level foliage for oak stand. Stemflow was collected from upper-tree stemflow at 1 m below the last branch, and at 1.37 m from forest floor for both cedar and oak stands. Cedar and oak stands were partitioned into canopy layers (as described above) and then comprised into two compartments: canopy and trunk.

2. Research outcome

Significant vertical variability of Cs-137 concentration, depositional flux and volume generated in the branchflow and stemflow was observed among the sampling periods and season and also differed within the tree layers and between the two forest stands. The oak stand exhibited higher Cs-137 concentrations than the cedar stand. Branchflow of the dead foliage in the cedar canopy remained a significant contribution to the leachable Cs-137, reflecting the initial fallout that was intercepted by the upper canopy.

For seasonal changes, the distribution of Cs-137 concentration via branchflow and stemflow in the cedar and oak stands was most pronounced during winter season, with larger Cs-137 concentrations from the trunk than the canopy. This emphasized the importance of winter leaching, especially from the tree trunk, although a lower volume was generated.

Overall, the Cs-137 depositional flux from the oak stand was 7.5 times greater than for cedar. The Cs-137 depositional flux generated by branchflow and stemflow for the cedar stand and oak stand were 375.2 and 2812.8 Bq/m²/year, respectively. 71% of Cs-137 depositional flux originated from the canopy of cedar stands and 48% from the canopy of oak stands. Results also indicated that Cs-137 concentrations were strongly governed by leaching and sequestration processes along the preferential flowpaths within the canopy, as intercepted rainwater interacted with foliage and outer bark. Furthermore, the isotopic composition of branchflow was generally enriched in δ 18O and δ D compared to open rainfall and throughfall, whilst oak stands more enriched than cedar stands.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

Zul Hilmi SAIDIN, Yuichi ONDA, Hiroaki KATO, Janice E. HUDSON, Kazuki NANKO, Delphis F. LEVIA, 2020. Vertical variability of radiocesium cycling via branchflow and stemflow through the canopy of cedar and oak stands in the aftermath of the Fukushima Dai-ichi Nuclear Power Plant accident. [in preparation, to be published in Chemosphere].

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【I-20-03】

Contribution of K transporters to Cs uptake and accumulation in rice

Principal Researcher : Nathalie LEONHARDT

Host Researcher : Jun Furukawa

1. Report (about one page of A4 paper)

The aim of our project is to develop in situ remediation approach using higher plants for rehabilitation of soil, a promising strategy for cost effective treatment of contaminated sites. Plants take up their essential elements from the soil through changes in the physico-chemical properties of the rhizosphere (acidification, excretion of chelators) and via transport systems of the essential elements (K, P, Fe,..etc). The radiological contaminants enter also using similar transport systems by chemical analogy (Cs/K). Also, using rice mutants affected in different transporters involved in the mobilization or transport of cesium, we will evaluate their capacity to increase or decrease the ^{137}Cs uptake in plants in order to generate new cultivars that can be used for phytoremediation or safe-food strategies.

In this experiment, we focused on OsHKT2;1 mutant line(oshkt2;1), which increased Cs uptake. Two mutant lines obtained from Tos17 mutant lines (ND4057(control and mutant), NC2534(control and mutant)) have used in our experiment. Seedlings transferred to paddy field (Fukushima) and cultivated for 5-6 months. Samples collected and separated into up ground part and grains. ^{137}Cs measured by Germanium counter, and K and Na contents measured by ICP-AES.

Field experiments performed 2018, 2019 and 2020. Three independent experiments (2018-2020) have showed same tendency in ^{137}Cs contents of grains. (^{137}Cs contents of leaves are under measurement in NARO's Fukushima institute). K and Na contents of 2018 sample showed Na contents differences in the plants but not in K. (2019-2020 samples are under measurement in NARO's Fukushima institute). We will obtain all measurement data within 2021, and then we want to understand the Cs absorption control mechanism by integrating field data and laboratory data.

Published Papers

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【I-20-04】

**Establishment of Radiocesium Concentration Ratios for Japanese Eels
(*Anguilla japonica*) and Conger Eels (*Conger myriaster*) in Fukushima**

Prefecture

Principal Researcher : Thomas Johnson
Co-researcher : Meghan Dieffenthaller
Host Researcher : Toshihiro Wada

1. Report (about one page of A4 paper)

コロナ禍で申請者のトマスジョンソン教授（コロラド州立大学）が来日できなかったため、受け入れ研究者の和田が福島県相馬市松川浦および周辺水域において予備的な調査を行った。

淡水域～海域に生息する魚類について、環境水と魚体中の放射性セシウム濃度比（CR）を明らかにし、特に汽水環境での魚類の放射性セシウム汚染メカニズムの解明を目指すことを目的として汽水性の潟湖である松川浦にて調査をおこなった。松川浦湾口部外側に位置する新港（St. 1）、松川浦北部（St. 2）、松川浦南部（St. 3）、流入河川の日下石川（St. 4）にて、8月および9月に魚類等の採集調査および水質調査を行った。塩分が大きくなる各調査地点において、淡水～海域に生息する様々な魚類等（マアナゴ、ニホンウナギ、クサフグ、マハゼ、イシガニ）が採集された。マハゼの Cs-137 濃度の平均は 1.64 Bq/kg-wet であったが、個体差（0.90-2.66 Bq/kg-wet）が認められた。これらの濃度は、各地点の堆積物中の Cs-137 濃度（51-530 Bq/kg-dry）に比べて低い値であった。

Published Papers

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【I-20-05】

Radiocesium solid-liquid distribution and retention in soils and sediments

Principal Researcher : Ralf Sudowe

Co-researcher : Anna Deak

Host Researcher : Alexei Konoplev

1. Report (about one page of A4 paper)

Monitoring of dynamics in radioactive contamination of the ponds and dam reservoirs in the FDNPP exclusion zone is essential not only in the context of radiation safety, but also for a better understanding of the main mechanisms responsible for persistence, mobility, and bioavailability of Fukushima-derived r-Cs in the freshwater environment. These water bodies are valuable and convenient subjects for the study of the surrounding freshwater environment with all its features and complexities.

Water and sediment samples from reservoirs/ponds within the exclusion zone were collected and analyzed. The contaminated water was filtered through membrane filters to separate particulate and dissolved r-Cs and then dissolved r-Cs was caught up by an ANFEZH (iron ferrocyanide) sorbent dissolved cesium. Soil/sediments size distribution was obtained and analyzed. The dynamics of the r-Cs distribution coefficient in the sediment-water system were analyzed.

Accumulated suspended material in ponds and dam reservoirs under study was mainly presented by clay and silt fractions. The typical XRD peak patterns of soil and sediment particles confirmed the presence of micaceous clay minerals, which selectively adsorb and fix radiocesium, in sediments and catchment soils.

The highest levels of the dissolved ^{137}Cs were observed from June to October, depending on the water body. Additional dissolution of ^{137}Cs in the summer can be attributed to the temperature dependence of ^{137}Cs desorption from FES and its remobilization by ammonium. The ^{137}Cs exchangeable distribution coefficients $K_{d^{\text{ex}}}$ were found to be linearly proportional to the inverse effective concentration of major Cs competitive cations $([\text{K}^+] + 5[\text{NH}_4^+])^{-1}$, as would be expected from the theory of radiocesium selective

sorption on the FES of micaceous clay minerals. The exchangeable RIPex(K) was estimated to be 1650–2250 mEq/kg, which is within the range of values measured in the laboratory and available in the literature.

Despite ^{137}Cs contamination in the ponds being relatively persistent, a decline in both particulate and dissolved ^{137}Cs activity concentrations was revealed. The reduction rate of particulate ^{137}Cs activity concentrations was much higher than that for dissolved ^{137}Cs . Thus, the apparent distribution coefficient $K_d(^{137}\text{Cs})$ in the sediment–water system decreased with the rate constant 0.12–0.18 yr⁻¹. We hypothesized that the $K_d(^{137}\text{Cs})$ decline may be associated with the decomposition of CsMP and subsequent ^{137}Cs leaching from them since the rate of K_d decline in the ponds corresponds to the roughly estimated rate of ^{137}Cs leaching from CsMP in a freshwater environment based on the latest available data of laboratory experiments.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

Konoplev A, Wakiyama Y, Wada T, Udy C, Kanivets V, Ivanov V, Komissarov M, Goto A, Nanba K (2021) Radio cesium distribution and mid-term dynamics in the ponds of the Fukushima Dai-ichi nuclear power plant exclusion zone. *Chemosphere*, 265: 129058.

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【I-20-06】

Influences of Rooting Activity by Wild Boar on ^{137}Cs Bioavailability

Principal Researcher : James Beasley

Host Researcher : Yoshifumi Wakiyama • Kenji Nanba

1. Report (about one page of A4 paper)

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 where rooting occurs to a depth of 120 cm. Rooting by wild boar has the potential to alter the soil chemistry, arthropod communities, and vegetation composition, and 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. Our objective in this study is to determine whether rooting by wild boar alters the bioavailability of ^{137}Cs . We hypothesize wild boar rooting alters the distribution of ^{137}Cs within the soil profile, increasing the bioavailability of this contaminant to biota and the potential for redistribution through erosion.

To evaluate the effects of rooting on ^{137}Cs distribution in soil, we initiated collection of soil samples within the difficult to return zone, in areas with and without boar rooting during 2020. However, due to the COVID-19 pandemic Dr. Beasley was unable to travel to Japan to carry out this research. Preliminary samples were collected and analyzed by Dr. Wakiyama at the Institute of Environmental Radioactivity, and we have purchased additional supplies to expand data collection this summer. These preliminary data support our hypothesis, and suggest wild boar rooting appears to disrupt the vertical migration of ^{137}Cs within soil, although additional sampling is needed to validate this observation. During summer 2021 we intend to travel to Japan as soon as the travel restriction is lifted to collect and analyze the remaining samples for this research. We will target both former agricultural areas as well as forested sites to investigate the influence of habitat and soil type. In addition, we will collect samples from recent (<1-2 weeks) and old (e.g., >3 months) rooting to study the effects of

weathering on the redistribution process. For all sampling sites, we will collect soil cores to determine the vertical distribution of ¹³⁷Cs in the surface and sub-surface soil layers. Radiocesium activity will be determined for all collected samples using an Auto-Gamma Counter at the Institute of Environmental Radioactivity.

Although we only have preliminary results to this point, these initial results suggest increases in wild boar populations have the potential to alter the distribution of contaminants within soil profiles. Upon completion of sampling in fall 2021 this study will produce the first results to date on the effects of disturbances from wild boar on changes to the distribution of ¹³⁷Cs. Given the growing population of wild boar within the difficult to return zone, a clear understanding of the impacts of this species on the local environment is necessary to better inform appropriate management actions. Thus, the proposed research will produce unique insights that could be relatable to ongoing management of this species in areas impacted by the Fukushima Daiichi nuclear accident.

Published Papers

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One paper is currently being finalized, and will be submitted to a major international journal within the next 1-2 months.

In addition, the following paper was recently published where the PI and host researcher are co-authors.

Gerke, H.C., T.G. Hinton, T. Takase, D. Anderson, K. Nanba, and J.C. Beasley. 2020. Radiocesium concentrations and GPS-coupled dosimetry in Fukushima snakes. *Science of the Total Environment* 734:139389

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【I-20-07】

Effects of radionuclide exposure on the microbiome community within
gastrointestinal tract of wild boar

Principal Researcher : Diana Lafferty

Co-researcher :

Host Researcher : Kenji Nanba

1. Report (about one page of A4 paper)

哺乳類に共生あるいは寄生して生活する微生物叢、マイクロバイオームの大部分は、宿主の消化管 (GIT) に存在する。これらは、腸内マイクロバイオーム [GMB] とよばれる。GMB は宿主の行動、生理、繁殖、健康、生存、そして最終的には進化に深く影響する無数の役割を担っている。野生動物の GMB を形成する多様な要因（食性、系統、生息地の質、性別、年齢など）についての理解は進んでいるが、放し飼いにされている野生動物の GMB に対する低線量放射線の影響についてはあまり知られていない。

この研究では、低線量放射線被曝のレベルの違いが、イノシシ (*Sus scrofa*) の GMB 構成種や組成にどのように影響するかを明らかにすることを目的としている。その前提として、イノシシの GMB の基本的な情報も明らかにする必要がある。そこで、イノシシの腸内細菌叢における性別および腸管の位置による分類学的および系統学的 GMB 組成変化を特徴づけ、定量化する。イノシシの捕獲場所の放射線量率および筋肉中の放射能濃度から生涯被曝線量を求める。そして、イノシシの 3 つの年齢階級における GMB コミュニティ組成を生涯被曝線量率と比較して特徴づける。2018 年 1-2 月に帰還困難区域内外で採集した合計 42 個体について分析を行った。主な結果の概要は以下の通り。

- ・福島の帰還困難区域で捕獲されたイノシシは、帰還困難区域外で捕獲されたイノシシ ($0.2 \pm 0.1 \mu\text{Gy}/\text{h}$) と比較して、捕獲時の放射線量率が大幅に高かった ($5.1 \pm 6.6 \mu\text{Gy}/\text{h}$)。
- ・GMB の組成と構造は、性別による違いはなかった。
- ・腸の上部（胃、十二指腸、空腸、回腸）では多様性が小さく、下部（盲腸、結腸、遠位結腸）では統計的に類似していた。
- ・標高と地理的距離に関連した GMB の変化がみられた。これは、標高と空間的な近さ（地理的距離が 40km 未満）の両方が、GMB 組成に大きく影響し、離れた場所に生息するイノシシ（距離が 40km 以上）は、全く異なる分類学的系統の GMB であることを示していた。

- ・1歳のイノシシのGMBは、慢性的な低線量放射線への曝露によって有意に異なるが、高齢のイノシシ（亜成体および成体）では線量による違いは見られず、免疫系が発達途上の若い動物は放射線の影響を受けやすいことが示唆された。

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

まだありません。

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【I-20-09】

**Synthesis and characterization of nickel(II) and cobalt(III) complexes
of aryl-substituted multi-azamacrocyclic chelator for application in
radioactive waste decontamination**

Principal Researcher : Tapashi Ghosh Roy

Co-researcher : Zinnat Ara Begum

Host Researcher : Ismail Md. Mofizur Rahman

1. Report (about one page of A4 paper)

Different radioisotopes of nickel(II) and cobalt(III) are produced in the structural steels of nuclear reactor vessels and internal components from neutron activation of corresponding naturally occurring stable isotopes. The shape-persistent macrocycles can be used for the selective isolation of r-Ni from the waste matrix. However the field of macrocyclic chemistry is growing very rapidly because of its application and importance in the area of coordination and analytical chemistry. The present research plan aimed to evaluate the capability of chelators for the selective isolation of r-Ni or r-Co from the waste matrix. In this context, new aryl substituted multi-azamacrocyclic chelator and its Co(III) and Ni(II) complexes have been synthesized and characterized. The new macrocycle is found to act as very efficient chelator of Ni(II) and Co(III) during complex formation. The compounds have been characterized by spectroscopic methods as well as by magneto-chemical, and conductivity data. It is expected that, future studies on the efficiency of adsorption of the chelators on Ni(II) and Co(III) ions can play a vital role on the development of radiochemistry.

Keywords: Macrocyclic chelator; complexation; nickel; cobalt; radiometric studies.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

Title: Syntheses, Characterization and Antimicrobial Studies of Ni(II), Cu(II), and Co(III) Complexes with an N-Pendent Azamacrocyclic Chelator

Authors: Lucky Dey, Saswata Rabi, Debashis Palit, Saroj K. S. Hazari, Zinnat A. Begum, Ismail M.M. Rahman, Tapashi G. Roy *

Journal Name: Journal of molecular structure

Status: Comments of Reviewers have arrived just now from Editor (Answers on queries will be provided soon for taking further action on it)

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【I-20-10】

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

Principal Researcher : Debashis Palit

Co-researcher : Shahadat Hossain

Host Researcher : Ismail Md. Mofizur Rahman

1. Report (about one page of A4 paper)

Introduction

Chittagong Hill Tracts area is a hilly area situated in between 21°02' N to 23°02' N latitude and 91°05' E to 92°05' 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 has 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

Due to current pandemic situation, the whole area was locked down, University is closed till today. So, we could not collect the requisite amount of sample, only nine samples were collected from Rangamati district. The samples were collected from the undisturbed area. It is then dried grinded, sieved and kept closed in a Marinelli beaker for 4 weeks. The radioactivity level of radioactive K, Th and Ra isotopes are measured by an HPGe detector.

Results and Discussion

The result of the natural radioactivity of the measured sample is given on Table 1

Table 1: Radioactivity level of soil samples of Chittagong Hill Tracts

Sl. No.	Sample location	K-40(Bq/Kg)	Th-232(Bq/Kg)
	Ra-226(Bq/Kg)		
1	Rm-1		190 ± 19

98 ± 10.78		3 ± 0.36
2	Rm-2	115 ± 14.95
89 ± 8.9		5 ± 0.6
3	Rm-3	270 ± 45.9
109 ± 16.35		11 ± 1.43
4	Rm-4	235 ± 25.85
60 ± 7.2		10.1 ± 2.32
5	Rm-5	235 ± 32.9
50 ± 6.5		12.9 ± 2.45
6	Rm-6	320 ± 38.4
23 ± 2.99		6.7 ± 1.21
7	Rm-7	390 ± 46.8
34 ± 5.78		5 ± 0.85
8	Rm-8	340 ± 40.8
41 ± 4.51		10 ± 1.7
9	Rm-9	390 ± 46.8
40 ± 5.2		10 ± 1.7

From Table 1 it is clear that among the collected sample the highest value of 40K is $390 \pm 46.8 \text{ Bq/Kg}$, ^{232}Th $109 \pm 16.35 \text{ Bq/Kg}$ and ^{226}Ra is $16.1 \pm 2.9 \text{ Bq/Kg}$. All this values in the data are below world average¹ except some values for ^{232}Th in some places. But with this small number of samples with a limited area we cannot conclude about the background radioactivity level of any area.

Conclusion

We could not complete our project because we were not able to collect the requisite amount of sample due to current pandemic situation. We hope to complete the rest of work this year if the situation permits.

Reference

¹UNSCEAR, Sources and effects of ionizing radiation: United Nations Scientific Committee on the Effects of Atomic Radiation, Vol.II. 2000

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

We could not publish any paper from this ERAN funded project. But we have several papers on our other works.

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【I-20-11】

**Low dose radiation effect on the brain development in racoon and
large Japanese field mice from Fukushima area after Fukushima
Daiichi Nuclear Power Plant accident.**

Principal Researcher : FENG RU TANG

Host Researcher : Tomisato Miura

1. Report (about one page of A4 paper)

Abstract:

The brain cellular and blood vessel changes including newly generated neurons in the subgranular zone of the dentate gyrus, astrocytes, microglia, oligodendrocyte progenitor cells and blood vessel in the Raccoon and large Japanese mice from Namie Town, Japan from contaminated (Namie, Fukushima) and reference (clean) areas were investigated using respective markers including doublecortin (DCX), Glial fibrillary acidic protein (GFAP), Ionized calcium binding adapter molecule 1 (IBA1), IB4, and platelet derived growth factor receptor beta (PDGFR1). More samples are still needed to make solid conclusions.

Methods:

The brains from Raccoon and large Japanese mice from Obori, Omaru, Namie, Kakura and Kiyohashi, Ishikari, Hirosaki, Japan were harvested and fixed with 4% paraformaldehyde and cut with 40 μ m, free-floating sections were immunostained with DCX, GFAP, IBA1, IB4, and PDGFR1

Results and conclusion:

Newly generated neurons in the subgranular zone of the dentate gyrus, astrocytes, microglia, oligodendrocyte progenitor cells and blood vessel in the Raccoon and large Japanese mice were well stained as indicated in Fig. 1.

As only 3 raccoons and 4 large Japanese mice were obtained from intermediate (0.65-1.81 μ Sv/h) or high (>3 μ Sv/h) dose rate regions, more animals are still needed for statistical analysis.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

Nil

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【I-20-12】

Evaluating radiocesium mobility from forest soils to soil ecosystems and soil fauna

Principal Researcher : Mathew Swallow

Host Researcher : Naofumi Akata

1. Report (about one page of A4 paper)

This project was implemented to determine radiocesium mobility in forest soils affected by the FDNPP accident. The method being developed aims to mimic the movement of radiocesium in the soil water induced by the evaporative flux of plants. The method will also allow for the collection of mobile radiocesium to understand the colloidal fraction which facilitates the transport of radiocesium within the soil. Unfortunately, travel restrictions due to COVID-19 limited the project to development of methodology and beginning testing in Canada on Canadian forest soils. However, despite the delay caused by the pandemic significant progress has been made in the project's development.

To date, we have successfully developed a treatment cell and methodology to ensure delivery of water solely through suction caused by matric forces of the soil driven by evaporative suction in the wicking material. We have done a small pre-trial test using 100 gram samples of dry mineral soil from a local forest treated with 5 ml of 5 ppm cesium chloride solution. These samples were subjected to 3 different wet/dry cycles (1, 5 and 10 times) to test ageing of cesium within the soil and to elicit a change in cesium availability.

Results show that the ageing treatment appears to increase the release of colloidal organic matter accumulation in the wicking material, though unfortunately due to local equipment malfunctions the wicks themselves have not yet been analyzed for the presence of cesium. However, the higher accumulation of colloidal organic material within the wicks of aged soils suggests that any radiocesium bound within this fraction would be mobile and freely migrate towards the rhizosphere of active plants. After entering the rhizosphere, these organic colloids would be at risk of being decomposed leading to the potential for transfer of radiocesium into the soil foodweb and plant tissues.

The pre-trial has shown that the wicking methodology can capture mobile colloids yet there

are still many challenges needed to complete this work. For one, the wicking material in its current form only elicits a moderate soil suction of approximately -200 kPa to -300 kPa, which is well below the generally accepted suction threshold of -1.5 MPa for terrestrial plants. Modification of the method is currently underway to lower the suction potential of the wicking to levels more commonly seen in terrestrial plants. We are also developing the potential for the system to function as a technique for in situ remediation treatment of radiocesium affected soils. The development efforts will continue and hopefully the easing of travel restrictions to Japan will allow for Dr. Swallow to travel to Hirosaki and Rokkasho to work with team members on the project on soils affected by the FDNPP accident.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

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【I-20-13】

Trend of radiocesium and relate radionuclides concentration in surface-water and ground-water after FDNPP accident

Principal Researcher : Rawiwan Kritsanauwat
Host Researcher : Hirofumi Tazoe • Naofumi Akata

1. Report (about one page of A4 paper)

It is important to monitor the radionuclide concentration in the aquatic environment such as river, groundwater, lake water in the situation of accident of nuclear facilities. In this study, we have developed a simple enrichment system for radioactive cesium (Cs-134, Cs-137) and radioactive strontium (Sr-89, Sr-90), which are important man-made radionuclides for environmental monitoring to avoid ingestion of contaminated water. In order to concentrate radionuclides at the same time, we constructed a two-stage concentration system in which consists of two chromatographic columns. The product KNiFC PAN Resin by Prussian blue was used as an adsorbent for radioactive cesium. This cesium adsorbent can collect cesium under both acidic and neutral pH conditions. A research group at Hirosaki University has established a method for radioactive cesium in river water using this resin (Hegedus et al., 2020). In order to quantify low-concentration radioactive cesium, it is necessary to focus on a small amount of resin that can be measured with a well-type germanium semiconductor detector. According to previous work, a column filled with 1 mL of KNiFC PAN resin was used. Radioactive cesium in 2 L of river water sample could be quantitatively collected by passing through a column at a flow rate of 2 mL/min by peristaltic pump. Since adsorption of Sr to KNiFC PAN resin is negligible, a cation exchange column (Bio-Rad AG1-X8, 100-200 mesh, 5 mL) was combined with KNiFC PAN resin packed column to concentrate Sr directly. The divalent cations containing Sr were quantitatively concentrated and could be eluted with 30 mL of 3M HNO₃.

Regarding radioactive cesium, the resin was transferred to a measuring tube and measured with a well-type high-purity germanium semiconductor detector for 3 days. As a result, it was possible to quantify with minimum detectable activity concentration of 5 mBq/L. Regarding radiostrontium, although radiocesium has been removed, other artificial radionuclides and

natural radionuclides coexist even after cation exchange, so further chemical separation is required. When Sr-90 is directly separated by ICPMS or radiation measurement, it can also be separated using Sr resin. When quantifying with the progeny nuclide Y-90, after 2 weeks or more have passed since the cation exchange, Y-90 in radiation equilibrium with Sr-90 is chemically separated by DGA Resin and radiation measurement is performed (Tazoe et al., 2016).

Reference

1. Hegedus, M., Shiroma, Y., Iwaoka, K., Hosoda, M., Suzuki, T., Tamakuma, Y., Yamada, R., Tsujiguchi, T., Yamaguchi, M., Ogura, K., Tazoe, H., Akata, N., Kashiwakura, I., Tokonami, S. Cesium concentrations in various environmental media at Namie, Fukushima Journal of Radioanalytical and Nuclear Chemistry. 323(1), 197-204 (2020).
2. Tazoe, H., H. Obata, T. Yamagata, Z. Karube, H. Nagai, M. Yamada: Determination of strontium-90 from direct separation of yttrium-90 by solid phase extraction using DGA Resin for seawater monitoring. Talanta, 152, 219-227 (2016).

Published Papers

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【I-20-14】

A Preliminary Study to Assess The Activity of Antioxidant Enzymes in Population Exposed to Very High Levels of Radon Concentration

Principal Researcher : Dwi Ramadhani

Host Researcher : Tomisato Miura

1. Report (about one page of A4 paper)

The Mamuju regency in West Sulawesi, Indonesia, is considered to have a high dose rate due to the high level of natural uranium content in the soil (Syaeful et al. 2014). A comprehensive dose evaluation including both of internal and external exposures by Hosoda et al. (2021) revealed that the annual effective dose in Botteng village, Mamuju could reached 27 mSv based on the median value of the measurements. Recent investigations revealed that Tande-Tande sub-village in Mamuju has the highest radon concentration that could reached 1644 Bq/m³. As a radioactive decay product of radium, radon, which has a short half-life (3.82 days), is a colorless, odorless gas that can easily move through the soil into the air before decaying by emission (Autsavapromporn et al. 2018; Ćujić et al. 2020). Therefore, the long-term exposure to radon and its progeny is the major contributor to exposure in people who live in areas with high natural background radiation, such as Tande-Tande sub-village inhabitants.

Radon can emitted alpha particles that capable to induce reactive oxygen species and DNA damage of respiratory cells and increase the lung cancer risk. Thus, it can be assumed that the antioxidant level in Tande-Tande sub-village inhabitants should be lower compare to control samples due to the utilization of antioxidants that could scavenge the higher free radical concentrations. In this study we will evaluate the activities of antioxidant enzymes to find out whether the level of antioxidant enzymes in Tande-Tande sub-village inhabitants are higher compare to control subjects.

Methods

The MnSOD and GPx activities from 17 healthy adult subjects from Tande-Tande Sub-village, Mamuju and 17 healthy adult subjects from normal background radiation areas (NBRAs) were measured using specific kit by visible spectrometer.

Results

The mean of MnSOD and GPx activities in the Tande-Tande sub-village inhabitants (8.47 ± 0.3611 , 0.374 ± 0.03508) were not statistically different than that found in the Topoyo village inhabitants (8.341 ± 0.4007 , 0.3511 ± 0.0323 ; $p=0.8133$, 0.633).

References

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2. Hosoda M, Nugraha ED, Akata N, Yamada R, Tamakuma Y, Sasaki M, et al. A unique high natural background radiation area – Dose assessment and perspectives. Sci Total Environ. 2021;750(142346):1–11.
3. Autsavapromporn N, Klunklin P, Threeratana C, Tuntiwechapikul W, Hosoda M, Tokonami S. Short telomere length as a biomarker risk of lung cancer development induced by high radon levels: A pilot study. Int J Environ Res Public Health. 2018;15(10).
4. Ćujić M, Janković Mandić L, Petrović J, Dragović R, Đorđević M, Đokić M, et al. Radon-222: environmental behavior and impact to (human and non-human) biota. Int J Biometeorol. 2020;

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【I-20-15】

Gross alpha and gross beta radioactivity concentration in environmental water

Principal Researcher : Fei Chen
Host Researcher : Hirofumi Tazoe

1. Report (about one page of A4 paper)

To improve the precision and accuracy of gross radioactivity detection in water, and to improve the radiological health and environmental monitoring level of each laboratory, it is necessary to carry out method verification and quality control. The thick source method for gross alpha and the thin source method for gross beta analysis were used to analyze the actual sample. After multiple test, the precision of the detection was calculated. Through the spike recovery test and the comparison sample analysis of the national radiological health in 2019, the accuracy analysis of the method was performed.

In the precision experiment, the relative standard deviations of the gross alpha and gross beta result were 23% and 13%, respectively. The three spike recoveries were 103.1%, 103.5%, and 105.8%, respectively. The relative errors of gross alpha and gross beta compared with the reference values were 0.92% and 9.4%, respectively. All the result reflected the high accuracy of the method. The relative standard deviations, recoveries, and relative errors in the method validation and quality control were below the limits of the criterion.

In the future, proposed method will be applied to river water sample collected in Ukedo river, Fukushima Prefecture in 2012 and 2013. These samples are registered as sample archive catalogue in ERAN. Since radioactive nuclides such as Cs-134 and Cs-137 and elemental concentration have been determined in IREM, Hirosaki University. Cs-137 concentrations range from 100 Bq/L to 2000 Bq/L, which exceed level of natural radionuclides such as K-40 and Ra-226 and become ideal sample to validate proposed method.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

陈飞，张震，王雪涛，邵宪章，李玉文（2020） Quality control of gross α and gross β analysis in water. 中华放射医学与防护杂志 Chin. J. Radiol. Med. Prot. 40(8) 65-67

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【I-20-16】

**Development of producing radioactive aerosol under a variety of
environmental conditions for dose assessment**

Principal Researcher : Rungroj Sakulnaeramit
Host Researcher : Chutima Kranrod • Yuki Tamakuma

1. Report (about one page of A4 paper)

OBJECTIVE

To develop a method for producing radioactive aerosols in 10 to 150 nm of radon progeny and estimate the lung effective dose.

RESEARCH SCOPE

- Using NaCl, Glucose and KMnO₄ solutions by different concentrations to produce radioactive aerosols.
- Study the effect of temperature at 25 and 35 ° C and suitable relative humidity in calibration chamber on radioactive aerosols.
- Estimate the equivalent dose from inhalation of radon progeny with the Integrated Modules for Bioassay Analysis (IMBA).

EXPERIMENTAL

At first, we use dryer in front of pump and generate pressure for Atomizer. The atomizer generates aerosols from water-soluble materials such as NaCl, Glucose and KMnO₄, the product is sent to calibration chamber with radioactive. After that, the attached decay products will be measured by PAMS.

In this time, The results of the experiment have the results obtained from the use NaCl, Glucose and KMnO₄. The next expectation is an experiment to control environmental conditions such as humidity and temperature to obtain complete results. And can summarize the results of the experiment completely.

RESULT

The procedure for producing radioactive aerosols of radon progeny in the range that can accumulate in the alveoli. It can be used to estimate the effective dose that humans will be exposed to due to the radioactive aerosols of radon progeny. And it can be applied to measure and assess the equivalent dose, radiation of radon progeny in various environments.

NOTE

Because of the COVID-19 epidemic situation Therefore it is a work limitation Causing the performance to not be as expected But the operation has made some progress. It is estimated from the results of the experiment that this research will be successful in meeting the goals.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

Kranrod C, Chanyotha S, Tokonami S, Ishikawa T. A simple technique for measuring the activity size distribution of radon and thoron progeny aerosols. J Environ Radioact. 2021 Apr;229-230:106506. doi: 10.1016/j.jenvrad.2020.106506. Epub 2021 Jan 17. PMID: 33472114.

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【I-20-18】

**A study of Japanese beaches for Fukushima Dai-ichi derived
radionuclides**

Principal Researcher : Ken O. Buesseler

Co-researcher : Nuria Casacuberta • Seiya Nagao

Host Researcher : Hirofumi Tazoe

1. Report (about one page of A4 paper)

The Fukushima Dai-ichi Nuclear Power Plants (FDNPPs) accident in 2011 led to an unprecedented release of radionuclides into the environment. Particularly important are Sr-90 and Cs-137 due to their known health detriments and long half-lives ($T \approx 30$ y) relative to ecological systems. These radionuclides can be combined with the longer-lived I-129 ($T = 15.7$ My) to trace hydrologic, atmospheric, oceanic, and geochemical processes. This study seeks to evaluate Cs-137, Sr-90, and I-129 concentrations in seawater off the coast of Japan, reconcile the sources of contaminated waters, and assess the application of Cs-137/Sr-90, I-129/Cs-137, and I-129/Sr-90 as oceanic tracers. We present new data from October 2015 and November 2016 off the coast of Japan, with observed concentrations reaching up to 198 ± 4 Bq/m³ for Cs-137, 9.1 ± 0.7 Bq/m³ for Sr-90, and $(114 \pm 2) \times 10^{-5}$ Bq-m³ for I-129. The utilization of activity ratios suggests a variety of sources, including sporadic and independent releases of radiocontaminants. Though overall concentrations are decreasing, concentrations are still elevated compared to pre-accident levels. In addition, Japan's Environment Minister has suggested that stored water from the FDNPPs may be released into the environment and thus continued efforts to understand the fate and distribution of these radionuclides is warranted. This result has been published in Kenyon et al. EST (2020).

Published Papers

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【I-20-19】

EVALUATION OF THE CONVERSION FACTOR OF RADON AND THORON PROGENY DETECTOR IN DWELLING USING TRACK ETCH DETECTORS

Principal Researcher : Chanis Pornnumpa

Co-researcher : Chanis Pornnumpa

Host Researcher : Chutima Kranrod • Yuki Tamakuma

1. Report (about one page of A4 paper)

In this study, radioactive aerosol generation system was developed to understanding the behavior of ambient aerosols for evaluating the conversion factor of Rn-Tn progeny by alpha-tract detector. The conversion factor values obtained could be used to calculate Rn-Tn progeny concentrations in the dwellings for internal dose assessment. In order to develop a high-performance of radioactive aerosol generation system, 1,000 ppm NaCl solution was used for generating particle in the chamber. For uniformity of the particles, a fan was installed upper side of a tank lid. The particles were neutralized by an electrostatic eliminator that was installed inside. The temperature was set around 20-21C under controlling of an air-conditioner. The system was divided into two parts; first part was an aerosol particles generator. The particle was generated and controls the concentration by flow rates before sending into the chamber. The amount of particle would be measured using a scanning mobility particle sizer (SMPS). Second part (Radioactive gases generator), radioactive gas from generator was produced and injects into the mixing chamber. Both of them were mixed with the optimum ratio for producing radioactive aerosols before entering into the exposure chamber for radiation instrument calibration.

All results revealed that the aerosol particles generating system can maintain the particle concentration between 1.7×10^3 particles per cm³ and 3.3×10^5 particles per cm³. The particles could be generated in the range between 60 and 80 nm for NaCl solution at concentration of 1,000 ppm. From this study, the condition with other flow rates should be carried out for observing the effect of flow rate to particle concentration for development of the calibration system and evaluation the conversion factor of Rn-Tn progeny by alpha-tract detector in the future.

Published Papers

Papers published by the research group between April 2020 and March 2021 (papers in which ERAN funding is stated in the acknowledgements, or the principal investigator and host researcher are co-authors. Accepted papers can be included).

Japanese journal of health physics

Journal of physics conference series

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【I-20-20】

Study on the migration law of radioactive aerosol

Principal Researcher : Zhou shumin

Co-researcher : Chen Rui · Zhang Xiongjie

Host Researcher : Chutima Kranrod · Yuki Tamakuma

1. Report (about one page of A4 paper)

In order to study on the migration law of radioactive aerosol under PM2.5 atmospheric pollution, the project established an experimental platform of PM2.5 combined radon progeny aerosol, mixed different types of PM2.5 aerosol with radon and it's progeny.

Studied the formation mechanism and decay law of PM2.5 combined radon progeny, the influence law of particle size, wind speed, humidity and other parameters on the equilibrium factor of PM2.5 and radon progeny was obtained. The radon-progeny equilibrium factor F is 0.56 ± 0.14 ($28.04\text{ C}/77.51\%$, $n = 120$) and 0.54 ± 0.15 ($15.50\text{ C}/50.00\%$, $n = 120$). The F value conforms with the indoor standard value recommended by UNSCEAR. The growth rate of F increases with the PM2.5 level. From moderate pollution to heavy pollution, the growth rate of F is significantly higher than the previous growth rate, but F remains within the reasonable range of 0.1–0.9. There is a positive correlation between the radon-progeny equilibrium factor F and the concentration of PM2.5 ($R^2 = 0.91805$), and these two parameters are closely related. The study of the radon-progeny equilibrium factor F is significant and can provide reference for environmental protection and radiation protection. The effects of different PM2.5 aerosol concentrations on the counting of ^{218}Po and ^{214}Po were studied based on the solid particles as aerosol vectors of radon. The results show that the total number of ^{218}Po and ^{214}Po increases with the increase of the concentration of aerosol in PM2.5, which reaches the maximum when the PM2.5 concentration is $309 \mu\text{g/m}^3$.

Keywords: radioactive aerosol, PM2.5, equilibrium factor, concentrations

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【I-20-21】

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

Principal Researcher : Wu men
Host Researcher : Jian Zheng

1. Report (about one page of A4 paper)

^{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, we plan to complete the remaining experiments in QST or in China.

Keywords: ^{137}Cs , ^{135}Cs , seawater, analytical method, ICP-MS

Published Papers

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【I-20-22】

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

Principal Researcher : Hai Wang

Co-researcher : Qianlin Li

Host Researcher : Jian Zheng

1. Report (about one page of A4 paper)

Groundwater in karst aquifers constitutes about 25% of drinking water sources globally. Soils on karst landscapes are typically thin, patchy and extremely fragile. The widespread existence of metal binding and transport by natural organic matter (NOM) in karst drip waters. The sediments, plants, water and other environmental sample were collection in karst region. The vertical distribution of ^{137}Cs activity in soils and lake sediment from Hunan karst environment were investigated. The ^{137}Cs activity was 15.11 ± 0.63 mBq/g in surface soil and 12.4 ± 0.53 mBq/g in lake sediment. The ^{137}Cs activity ratios in undisturbed soils decreased from 15.11 ± 0.63 mBq/g to 0.74 ± 0.14 mBq/g with increasing depth from 1cm to 10cm. The ^{137}Cs activity ratio in disturbed soils was a narrow range with increasing depth.

Published Papers

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【I-20-23】

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

Principal Researcher : Wang Zheng

Co-researcher : Yuanming Zhang

Host Researcher : Jian Zheng

1. Report (about one page of A4 paper)

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 the sediment cores near the Antarctic sea, where local anthropogenic Hg sources are absent. The total Hg concentration (THg) and Hg isotope ratios together reveal different Hg sources in different time periods. Period I (50-20 ka BP) is marked by low sedimentation rate, low THg, strongly negative $\delta 202\text{Hg}$ (as low as -2‰), and slightly positive $\Delta 199\text{Hg}$ and $\Delta 200\text{Hg}$. The low sedimentation rate and low THg is likely

due to extensive ice cover at the Antarctic sea during Last Glacial Maximum (LGM). The isotope signals in period I are consistent with modern seawaters that primarily receive deposition of background atmospheric Hg. Thus Hg during period I was most likely deposited from the atmosphere at ice-free area, and then transported by ocean circulation to the Antarctic sea. Period II (20-10 ka BP) shows a dramatic increase of the deposition rate and a slight increase of THg compared to period I, suggesting enhanced terrestrial weathering after the LGM due to the retreat of glacial. The Hg isotope signals are also consistent with enhanced input of terrestrial Hg. Period III (10 ka BP to present) shows no significant changes in the sedimentation rate, but a dramatic increase of THg, suggesting that the increased THg was caused by enhanced external input. $\delta 202\text{Hg}$ shows even more positive values compared to period II, and $\Delta 199\text{Hg}$ and $\Delta 200\text{Hg}$ both shift towards zero, which are consistent with volcanic or anthropogenic emissions. It is well known that there has been intensive volcanic activities surrounding the Antarctica since LGM 1. Thus we suggest that Hg in the last 10 ka was primarily contributed by volcanic or hydrothermal emissions near the Antarctica.

In the year of 2021 we will 1) continue to finish the analysis for Hg concentration and isotopes in the remaining sediment samples; 2) perform data analysis; 3) publish the results. We expect at least two publications based on these data.

1. Fraser, C. I., Terauds, A., Smellie, J., Convey, P. & Chown, S. L. Geothermal activity helps life survive glacial cycles. Proc. Natl. Acad. Sci. 111, 5634–5639 (2014).

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【I-20-24】

Method development for simultaneous analysis of ^{237}Np and Pu isotopes in the seawater by SF-ICP-MS

Principal Researcher : Shaoming Pan

Co-researcher : Shuai Zhang

Host Researcher : Jian Zheng

1. Report (about one page of A4 paper)

Plutonium in seawater is a well-known tracer in biogeochemical processes with its particle-reactive properties. Recent studies evidenced that another transuranic isotope, ^{237}Np ($t_{1/2}=2.14 \times 10^6$ y) has a similar behavior of ^{137}Cs , thus shows great potential to act as a tracer for water mass circulation for decadal and longer time scales. However, information on ^{237}Np presented in seawater, especially in open ocean is very scarce, the distribution and inventory of ^{237}Np in the ocean remains largely unknown due to its low concentration in seawater and great analytical challenge for its measurement. Therefore, a method for simultaneous analysis of ^{237}Np and Pu isotopes in the seawater by SF-ICP-MS was developed. The results show that almost 100% ^{237}Np and Pu isotopes were concentrated from small volume seawater (15L) using the $\text{Fe}(\text{OH})_2$ co-precipitation, the average chemical fractionation was 0.98 ± 0.03 ($n=9, 1\sigma$) for ^{237}Np and ^{242}Pu during the entire analytical method, which proved that ^{242}Pu can be used as a non-isotopic tracer for monitoring the recovery of ^{237}Np in this method, and the chemical recovery of ^{242}Pu yield tracer ranged 72%-76%. Moreover, a high U decontamination of $\sim 10^7$ was achieved, which made the U interference negligible for the determination of ultra-trace level ^{237}Np and Pu isotopes in seawater. To demonstrate the accuracy of our method, ^{237}Np and $^{239,240}\text{Pu}$ concentrations in IAEA-443 standard reference materials was determined. The results showed good consistency with the certificated values and the reported values.

Keywords: ^{237}Np , Pu isotopes, seawater, ICP-MS, IAEA-443

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【I-20-25】

**Source identification and environmental impact assessment of
actinides around Fukushima: Development of ultra-sensitive methods
in environmental samples**

Principal Researcher : Xiongxin Dai

Co-researcher : Shan Xing · Maoyi Luo

Host Researcher : Jian Zheng

1. Report (about one page of A4 paper)

Isotopic signatures of actinides have been used as a powerful tool to identify the source-term of anthropogenic radionuclides and to evaluate their radio-contamination contributions and environmental impact. Due to extremely low level of actinides present in complicated environmental sample matrices, ultra-sensitive radioanalytical techniques would be very much needed for analysis of actinides in environmental samples. Mass spectrometric measurements, particularly accelerator mass spectrometry (AMS) and inductively coupled mass spectrometry (ICP-MS) technologies, often provide superior analytical sensitivity for long- and intermediate-lived actinide nuclides in environmental samples. Based on ICP-MS and AMS, numerous methods have been developed and applied for measurements of ^{239}Pu and ^{240}Pu in soil and sediment samples. However, the measurement data on ^{237}Np and ^{241}Pu in soil and sediment samples are still very scarce largely due to the lack of sufficiently sensitive and reliable methods for these two radionuclides at the environmental levels. Therefore, a radiochemical separation method was developed for simultaneous measurement of plutonium isotopes (^{239}Pu , ^{240}Pu and ^{241}Pu) and ^{237}Np in soil and sediment samples by AMS and triple quadrupole ICP-MS (ICP-QQQ).

In this method, after the addition of the ^{242}Pu tracer, 5-10 g of soil/sediment sample is first fused with lithium metaborate and the fusion button is completely dissolved in concentrated acids. The digested solution is then processed through 3 precipitation steps, including NaOH precipitation, YF₃ and HTiO (hydrous titanium oxide) co-precipitation, to remove majority of the sample matrices. After dissolution of Pu and Np in the HNO₃, the sample is loaded

onto a solid-extraction (TisKem TK200 resin) column to simultaneously separate Pu and Np from Th and U. The Pu and Np are then eluted together off the resin, and finally measured with ICP-QQQ. The validation results of the spike samples showed no observable differentiation between Pu and Np through the entire procedure (with the average Np/Pu ratio of 0.972 ± 0.041). For 5 samples spiked with known amounts of ^{237}Np and Pu isotopes, good agreements between the expected and measured values by ICP-QQQ have been achieved with satisfactory chemical recoveries of $>70\%$, confirming the validity of the method. Due to the major delay of the installation of our new AMS (caused by the pandemic issue), the AMS validation tests are yet to be performed.

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Keywords: Pu isotopes, ^{237}Np , soil, sediment, triple quadrupole ICP-MS, accelerator mass spectrometry

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