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研究成果の概要(和文)：気候、地形、降下物における放射性セシウム(r-Cs)の存在形態の特性が環境中r-Csの移行と自然減衰の速度の違いにつながることを示された。福島はチェルノブイリに比べ年間降水量が多く、台風シーズンに最大の暴風雨が発生する。2019年台風ハギビスは、河川流域や氾濫原にr-Csの再分布を引き起こし、場合によっては自然除染をもたらした。さらに集水域が急傾斜のため浸食性が高く、粒子状r-Csの流出が多い。河川・湖沼における放射性核種長期動態の半経験的拡散モデルを開発し、福島とチェルノブイリのデータセットを用いて検証を行った。河川・湖沼における放射性核種長期動態の再構築手法を提案し底質コアに適用した。

研究成果の学術的意義や社会的意義

The project provided assessment of the current state and prediction of r-Cs long-term dynamics in aquatic ecosystems of the Fukushima contaminated areas. Data obtained can be used by decision makers. Results were disseminated through mass media and meetings with people in Fukushima Prefecture.

研究成果の概要(英文)：The peculiarities in climate, geomorphology and radiocesium (r-Cs) speciation in the fallout were demonstrated to lead to differences in migration rates of r-Cs in the environment and rates of its natural attenuation. The climate conditions for the Fukushima Prefecture of Japan are characterized by higher annual precipitation as compared with Chernobyl with maximum rainstorm events during typhoon season. Typhoons Hagibis in 2019 demonstrated the substantial redistribution of r-Cs on river watersheds and floodplains and in some cases natural self-decontamination occurred. Steep slopes of Fukushima catchments are conducive to higher erosion and higher particulate r-Cs wash-off. A semi-empirical diffusional model for radionuclide long-term dynamics in rivers and lakes was further developed and validated using the Fukushima and Chernobyl datasets. A methodology for reconstruction of radionuclide long-term dynamics in rivers and lakes was proposed and applied for bottom sediment cores.

研究分野：Dynamics of environmental radioactivity

キーワード：Fukushima Chernobyl environment contamination radiocesium fate and transport rivers lakes

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1 . 研究開始当初の背景 / Background at the Beginning of the Study

Research of long-term dynamics of radionuclides in the environment continues to be of high relevance, both scientifically and practically, today when over 36 years have passed after the Chernobyl accident and 11 years after the Fukushima Dai-ichi nuclear power plant (FDNPP) accident. Climate and geographical conditions for Fukushima Prefecture and Chernobyl zone differ essentially and hence migration rates and solid-liquid distribution of r-Cs are different in Fukushima and Chernobyl areas (Konoplev et al., 2022). What is more, r-Cs deposited on the catchments and water bodies in Fukushima Prefecture is different from Chernobyl depositions in terms of speciation. Because of the complexity of processes involved, there are still many open questions as to how radionuclides in Fukushima aquatic ecosystems will behave in future. This project provided prediction of radionuclides dynamics in Fukushima water bodies based on obtained data and results. Also, given that fish is an important component of Japanese diet, we studied r-Cs transfer to various kinds of fish as a function of its age and size, and clarified prediction of r-Cs dynamics in fish.

2 . 研究の目的 / Purpose of the Study

The proposed project was aimed to study and compare long-term dynamics of radiocesium in rivers and other water bodies (reservoirs, lakes and ponds) of two areas contaminated following the major nuclear accidents (Chernobyl and Fukushima). Experimental studies were conducted to identify similarities and differences in the behavior of Chernobyl- and Fukushima-derived r-Cs in these water bodies, with a view to understand how environmental conditions such as climate, geology and geomorphology influence the fate and transport of accidentally released r-Cs in soil-water environment and its transfer to fish. Integrating research of different types (environmental media and biota) in the framework of one project was carried out in framework of the project.

Specific tasks of the project included:

- Characterization and prediction of r-Cs vertical distribution in catchment soils of contaminated areas both in Chernobyl and Fukushima.
- Assessment and prediction of chemical speciation of r-Cs in soils and sediments of aquatic ecosystems.
- Quantification and predict the long-term dynamics of dissolved and particulate r-Cs in rivers, reservoirs and lakes/ponds of the contaminated areas and its solid/water distribution as a function of time and solid and water phase chemical composition.
- Quantification of r-Cs transfer from water and sediments to fish as a function of fish species, fish size and water chemistry.
- Identification of similarities and differences in behaviour of Fukushima- and Chernobyl-derived r-Cs in aquatic ecosystems to improve the predictive power of semi-empirical and numerical models for long-term.

3 . 研究の方法 / Research Methods

Samples of water from rivers, lakes, reservoirs and ponds, suspended and bottom sediments, fish and soils on the catchments were periodically collected. Soils were collected on catchments and floodplains using 30-cm core sampler. Bottom sediments were collected by conventional method using standard sediment corers and grab samplers. Sediments/soil cores were sliced at 1 to 2 cm intervals and the dry solid were measured for ^{137}Cs and ^{134}Cs to obtain an activity-depth profile. R-Cs speciation were characterized in terms of proportion of exchangeable, bound by organic matter and nonexchangeable forms. Dissolved K^+ , NH_4^+ , Na^+ , Ca^{2+} , Mg^{2+} in water were measured by ion chromatography, and stable $^{133}\text{Cs}^+$ was measured by ICP-MS. Soils and sediments were characterized in terms of organic matter content and mineralogy. R-Cs deposition was assessed on the base of inventories in soils of catchments. Water samples of suitable volume (to make sure r-Cs detection both in solution and on suspended matter) were be filtered *in-situ* using "Midia" system and/or in laboratory using 0.45 μm membrane filter. Fish was be collected by conventional techniques using gill nets. ^{137}Cs and ^{134}Cs activity concentrations in all collected samples was measured by gamma-spectrometry using a high-purity germanium detector (HPGe) CANBERRA GC3018 (Konoplev et al., 2021a; Konoplev et al., 2021b).

Data on r-Cs dynamics in Chernobyl rivers (Pripyat, Sakhan, Braginka etc.) and lakes (Glubokoe etc.) were taken from the literature and data bases available to project participants. Dynamics of

r-Cs both in Fukushima and Chernobyl were be modelled and predicted using analytical, empirical and semi-empirical models (Konoplev et al., 2021b; Konoplev et al., 2022a; Konoplev et al., 2022b) as well as numerical models (MOIRA, COASTOX and THREEETOX) integrated in the Hydrological Dispersion module of EC RODOS system (Zheleznyak et al., 2022).

4 . 研究成果 / Research Results

It was shown that wash-off is the principle long-term process responsible for r-Cs secondary contamination of surface waters on accidentally contaminated areas. For its characterization the particulate and dissolved wash-off ratios were proposed. Both of them were found to decrease in the mid- and long-term as a result of r-Cs depletion in the topsoil layer due to its vertical migration in catchment soils.

The processes of wash-off, river transport and radionuclide vertical migration in catchment soils were considered in an integrated way by the proposed semi-empirical diffusional model. This is a novel approach enabling description of changes in the particulate and dissolved ^{137}Cs wash-off ratios using only two physically meaningful parameters D_{eff} - the ^{137}Cs effective dispersion coefficient in the topsoil layer and K_d - the ^{137}Cs apparent distribution coefficient.

The mid- to long-term monitoring data for both Chernobyl and Fukushima rivers did not reveal any time trend in the apparent ^{137}Cs distribution coefficient K_d despite distinct interannual variations. Comparison of collected data set for Chernobyl rivers Pripyat and Dnieper with obtained and available data for Fukushima rivers Ukedo and Ohta confirmed that the apparent $K_d(^{137}\text{Cs})$ value for Fukushima rivers is about an order of magnitude higher than for Chernobyl rivers.

The radiocesium dynamics was analyzed based on results of sampling campaigns in high-flow events. The episodic sampling at the river mouth of the Abukuma river during typhoon Hagibis revealed that substantial amount of ^{137}Cs is desorbed from suspended solids into seawater, elevating ^{137}Cs concentration in seawater. From the results obtained in three events in Niida, Ukedo and Takase rivers during 2019-2020, variations in both the dissolved and particulate ^{137}Cs concentrations appeared to reflect the spatial pattern of the ^{137}Cs inventory in the catchments, rather than variations in physico-chemical properties of suspended sediment and water.

Sampling campaigns during high-flow events were conducted on three major rivers flowing in the southern part of coastal area: the Natsui, Fujiwara, and Same rivers. A small river flowing in vicinity of Fukushima Dai-ichi Nuclear Power Plant, the Ottozawa river, has been monitored with hydrological monitoring instruments.

Data on changes in radiocesium inventories on different floodplain levels were collected and generalized for the Niida River basin and along the valley bottom of the Abukuma River. The data was derived using a variety of techniques, including analysis of ^{137}Cs depth distributions, evaluation of sediment and sediment-associated ^{137}Cs at artificial plastic lawn-grass mats, assessment of river bottom transformation based on satellite image interpretations and field survey with measurement of air dose rates. All these findings helped us to better understand the transformation of floodplain contamination levels after ordinary and extreme floods.

Particulate ^{137}Cs wash-off ratios from the catchments of the Fukushima area display only minor differences compared to those in the Chernobyl area, being at the lower limit of the Chernobyl values. Somewhat lower values of $N_p(^{137}\text{Cs})$ in the Fukushima area were explained by higher values of the effective dispersion coefficient $D_{\text{eff}}(^{137}\text{Cs})$ in typical Fukushima soils.

Dissolved ^{137}Cs wash-off ratios for Fukushima catchments were found to be at least an order of magnitude lower than those for Chernobyl, mainly due to an order of magnitude difference in the ^{137}Cs distribution coefficients for the Fukushima and Chernobyl rivers.

The temporal trends for the ^{137}Cs wash-off ratios, both in Chernobyl and Fukushima areas were satisfactorily described by the proposed model. This proposed model can be used as a tool to predict ^{137}Cs wash-off after a nuclear accident.

Long-term regular monitoring at the ponds in Okuma Town was carried out. Important parameters of radiocesium behavior such as rate constant of ^{137}Cs leaching from glassy hot particles (k_l) in soil-water system and exchangeable radiocesium interception potential (RIP^{ex}) have been determined. Scientific rationale was provided for the observed seasonal variation of dissolved ^{137}Cs in ponds: it appeared to be associated with temperature dependence of ^{137}Cs desorption from frayed edge sites (FES) of micaceous clay minerals. Based on data of the conducted field works, the activation energy (E_a) of this process was also estimated. The value of E_a was confirmed by Fukushima rivers observations jointly with members of Radioecology Project.

It was shown that bottom sediments of lakes and dam reservoirs can provide an insight into understanding the dynamics of ^{137}Cs strongly bound to sediment particles. A number of cores of bottom sediments were collected in deep parts of lakes Glubokoe, Azbuchin, and Cooling Pond in close vicinity of the Chernobyl NPP in Ukraine, in Schekino reservoir (Upa River) in the Tula

region of Russia (2018) and in Ogaki reservoir (Ukedo River) in Fukushima contaminated area (2019). Each layer of bottom sediments could be attributed to a certain time of suspended particles sedimentation. With ^{137}Cs activity concentration in a given layer of bottom sediments corresponding to ^{137}Cs concentration on suspended matter at that point in time, we were able to reconstruct the post-accidental dynamics of particulate ^{137}Cs activity concentrations. Using experimental values of the distribution coefficient K_d , changes in the dissolved ^{137}Cs activity concentrations were estimated. The estimates of particulate and dissolved ^{137}Cs concentrations in Chernobyl cases were in reasonable agreement with monitoring data and predictions using the semi-empirical diffusional model. However, both the particulate and dissolved ^{137}Cs activity concentrations and wash-off ratios in the Ukedo River declined faster during the first eight years after the FDNPP accident than predicted by the diffusional model, most likely, due to greater natural attenuation and, to some extent, remediation measures implemented on the catchments in Fukushima.

5. 主な発表論文等

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1. 著者名 S. H. ...	4. 巻 59
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〔産業財産権〕

〔その他〕

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6. 研究組織

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7. 科研費を使用して開催した国際研究集会

〔国際研究集会〕 計0件

8. 本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関
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