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### Fate and Transport of Fukushima-Derived Radiocesium in the Environment: Key Findings and Challenges for the Future

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#### Abstract

Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in March 2011 led to extensive environmental contamination by radiocesium isotopes <sup>134</sup>Cs (half-life  $T_{1/2}=2.06$  years) and <sup>137</sup>Cs ( $T_{1/2}=30.17$  years). Numerous research studies of environmental behavior of radiocesium in the context of geoclimatic conditions of Japan were undertaken. A lot of thought was given to what makes Fukushima environmental impacts different from previous nuclear disasters. This review paper summarizes key findings of post-Fukushima studies of radiocesium fate and transport in abiotic soil-water environment and discusses some challenges for future research. After the Fukushima accident scientific evidence was obtained to confirm that radiocesium behavior in the environment is governed by its speciation in fallout and site-specific environmental characteristics. Given strong binding of Fukushima-derived <sup>137</sup>Cs to soil and sediment particles, its potential bioavailability appeared to be reduced. Incorporation of the deposited <sup>137</sup>Cs in glassy hot microparticles insoluble in water and slowly decomposing in the environment was another salient feature. The Fukushima contaminated areas are noted by relatively high annual precipitation and steep slopes, resulting in significant erosion and intensive radiocesium wash-off, especially during devastating typhoons. Extreme floods during typhoons Etou in 2015 and Hagibis in 2019 caused major redistribution of <sup>137</sup>Cs on river watersheds and floodplains, and, in some cases, natural self-decontamination. Significantly, for all special features and characteristics of Fukushima areas, the recently obtained knowledge is in line with the basic perceptions of modern environmental chemistry regarding radionuclide behavior in the environment. Challenges for future research include, to name a few, tackling radiocesium leaching from glassy hot particles and studying dynamics of radiocesium in the environment over long term, better understanding of dissolved radiocesium seasonality in water bodies and addressing radiocesium remobilization from rivertransported sediments at the freshwater-seawater interface.

**Keywords:** Fukushima; Radiocesium; Speciation; Fate; Transport; Selfpurification; Challenges

#### Introduction

The Great East Japan Earthquake and the following tsunami on 11 March 2011 caused the accident at FDNPP which led to extensive local soil contamination by <sup>134</sup>Cs (half-life  $T_{1/2}$ =2.06 years) and <sup>137</sup>Cs ( $T_{1/2}$ =30.17 years). Radiocesium (r-Cs) deposition north-west of the NPP resulted in a trace of contamination 50-70 km long and 20 km wide [1-4]. The initial ratio of <sup>134</sup>Cs/<sup>137</sup>Cs isotopes in the Fukushima fallout was about one [2,5]. The



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contribution of <sup>134</sup>Cs to the radioactive contamination and local air dose rate, as compared to <sup>137</sup>Cs, decreased over time due to its more rapid decay [6,7].

The behavior of accidentally released r-Cs in the environment is determined by speciation in the fallout and site-specific environmental characteristics which are key for its mobility and bioavailability [8-10]. The areas of Japan contaminated as a result of FDNPP accident are characterized by wet monsoon climate with highly variable total annual precipitation (1200-1800 mm/yr). Less than 5-7% of precipitation falls as snow. Extreme rains with precipitation amount > 200 mm event occur once every 5-7 years according to meteorological observations. Several rains >50 mm fall each year, most frequently during the monsoon season between June and October. Some heavy rains can however occur in April-May, when soil surface on cultivated fields is less protected by vegetation [11]. Occurrence of high flow events in rivers especially during typhoon's season facilitates r-Cs wash-off from contaminated catchments and its lateral migration [7,12-16].

The geology of the contaminated territory is highly heterogeneous, and the mountains serve as a partition between the Abukuma river valley, the largest in the region, and the ocean. The proportion of clays is 20-30%. These mountains are 1200-1300 m high and folded metamorphic and sedimentary rocks with numerous magmatic intrusions. Parent rock materials in the Fukushima contaminated areas are primarily granites and volcanic ashes that are subject to physico-chemical weathering in the humid monsoon climate conditions. In the region there are several active and dormant volcanoes; hot springs of different geochemical composition are abundant. Soil diversity is great due to the combination of mountain rocks of different lithological composition, intense weathering and denudation from high seismicity, and the steep inclination of mountain slopes. The interfluve areas include brown soils (under beech forest), ashy-volcanic, rich in humus, acidic allophonic (andosol) and leached brown soils. The valleys floors are used mainly for growing rice and are represented by alluvial soils strongly modified as a result of many years land use. Undisturbed alluvial soils occur on the leveed parts of river valleys and along the canalized parts of stream channels typical of intermountain depressions. The arable lands, mainly paddy fields, occupy about 12% of the total territory in the region, and occur primarily on extensive depressions and piedmont plain [17,18]. After the Fukushima accident, cultivation on arable land with high radiocesium deposition levels has been banned and decontamination activities have been implemented to remove contaminated soil layers [19,20].

The contaminated territory of Fukushima Prefecture is characterized by an expansive and differentiated hydrographic network, dominated by the largest river of the area – Abukuma. Other rivers include Ukedo, Niida, Uda, Mano, Ota, Odaka, Maeda, Kuma, Tomioka, Ide, Kido, Natsui and Same with their tributaries [21,22]. All these rivers ultimately end in the Pacific Ocean. Thus, the river catchments contaminated from the FDNPP accident became a long-term source of secondary contamination of water bodies by surface runoff and radiocesium flux to the Ocean [23,24]. Moreover, surface runoff and river transport results in the transfer of radiocesium from contaminated evacuated areas to cleaner populated regions, and the settling of radiocesium in bottom sediments of river dam reservoirs and on floodplains (mostly during rainy seasons) [7,25].

The objective of this review paper is to synthesize key findings regarding behavior of Fukushima-derived r-Cs in soil-water environment including catchment soils, sediments, rivers, ponds and dam reservoirs as well as to formulate relevant challenges for post-Fukushima environmental radioactivity studies in the future.

# **Radiocesium Speciation in Deposition and its Transformation with Time**

#### Release of hot microparticles following FDNP accident

Immediately after the accident it was hypothesized that accidentally released r-Cs was transported in the atmosphere and deposited as part of sulfate aerosol particles of 0.5-0.6 µm diameter. R-Cs in these particles was assumed watersoluble and washable by precipitation [26]. Yet, waterinsoluble spherical glassy hot particles of a few micrometers in diameter were discovered later as far as 170 km from the FDNPP, containing, apart from r-Cs, uranium, and other elements representative of reactor materials [27,28]. Later, Igarashi et al. [29] reported about determination of Pu isotopes in hot particles derived from Unit 1 of the FDNP. Similar particles have been identified by Niimura et al. [30] using autoradiography of soils, plants, and mushrooms. Near the FDNPP, even coarser particles (up to hundreds of µm) were identified with higher r-Cs activity (sometimes more than 1000 Bq/particle) and irregular shape [29]. These radiocesium-bearing microparticles (CsMPs) are primarily composed of silicates [31], and most likely were the product of molten core-concrete interaction during the accident, which was confirmed by laboratory experiments [32].

In terms of r-Cs fate and transport, it is important that CsMPs are insoluble in water and persistent in the environment [33,34]. For adequate modeling and prediction of Fukushima-derived r-Cs behavior in the environment, it is necessary to know the fraction of CsMPs activity in the total radiocesium release and deposition at different locations and the rate of radiocesium leaching from CsMPs due to decomposition [10].

Ikehara et al. [35,36] succeeded to characterize quantitatively the content of CsMPs in soils at different

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directions from the FDNPP. They found that CsMPs account for a significant fraction of FDNPP-deposited r-Cs. The number of CsMPs in surface soils at various directions, distances and deposition levels varied from few to more than 300 particles/g, and radioactivity fraction of CsMPs from the total deposition was in range 15-80%. Occurrence of CsMPs in soils and sediments can substantially impact the r-Cs solid-liquid distribution in the soil-water environment [37]. However, the variability of CsMPs fraction in deposition in a relatively wide range complicates accounting for CsMPs in assessment of mobility and bioavailability of r-Cs in the soilwater environment and its dynamics.

Miura et al. [33] discovered CsMPs in suspended sediments collected from Kuchibuto River (right bank tributary of the Abukuma River flowing from the most contaminated section of the Abukuma River catchment) during 2011-2016. The fraction of radiocesium incorporated in CsMPs to the total radiocesium in the sediments was found to be up to 67%.

### Transformation of r-Cs chemical forms in the environment

After the deposition, the initial chemical forms of radionuclides are subject to transformation [10,38-40]. With CsMPs disintegrated in the environment due to weathering, r-Cs incorporated in these particles transfers to the solution [41,42]. In the solution, radiocesium is sorbed by the soil and sediment particles by ion exchange on selective frayed edge sites (FES) and regular exchange sites (RES) [43-45]. Exchangeably sorbed r-Cs is fixed by micaceous

clay minerals, i.e., transferred into nonexchangeable r-Cs, due to the replacement of the interlayer K-cations by Cscations [46-50]. The mechanism of r-Cs fixation consists of the replacement of interlattice  $K^+$  by Cs<sup>+</sup> ions due to the collapse of the expanded edges of the mineral's crystal interlayers and/or the slow long-term solid-state diffusion of Cs<sup>+</sup> ions through the interlayer inside the particle [38,51,52]. Data about long-term dynamics of r-Cs chemical forms in the soil and sediments are indicative of the existence of the remobilization process, which is the reverse of fixation [8,10,53-56].

Radiocesium can be uptaken from solution by biota and reversely be released due to excretion or decomposition of dead organic matter [57-60]. Rates of these processes are variable and highly uncertain. However, in many cases they do not play critical role in r-Cs fate and transport [61]. Conceptual model of all major transformation processes for Fukushima-derived r-Cs chemical forms in the environment is presented in Figure 1.

### Solid-liquid distribution of r-Cs in the soil-water environment

Solid-liquid distribution of r-Cs is a governing factor for its fate and transport in the soil-water environment. It is characterized by the apparent distribution coefficient  $K_d$  (L/kg) which is the ratio of the particulate radionuclide activity concentration  $[R]_p$  (Bq/kg) to its dissolved activity concentration  $[R]_d$ (Bq/L) at equilibrium [62]:

$$K_d = \frac{|R|_p}{[R]_d} \tag{1}$$



Figure 1: Conceptual model of transformation processes for Fukushima-derived radiocesium speciation in soil-water environment (updated after Konoplev et al. [42]).

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 $[R]_p$  includes the r-Cs embedded in CsMPs  $(R_h)$ , the exchangeably sorbed r-Cs  $(R_{ex})$ , radiocesium fixed by clay minerals  $(R_f)$ , and organically bound r-Cs  $(R_{org})$  (see Figure 1). The exchangeable r-Cs occurs at instantaneous ion-exchange equilibrium with the dissolved r-Cs, whereas the non-exchangeable form does not take part in the r-Cs exchange with the solution in immediate term. Therefore,  $K_d^{ex}$  worth using the exchangeable distribution coefficient , which is the ratio of the exchangeable radionuclide activity concentration in soil or sediments  $[R]_{ex}$  to its activity concentration in solution at equilibrium  $[R]_d$  [8,54]:

$$K_d^{ex} = \frac{[R]_{ex}}{[R]_d} = \alpha_{ex} K_d \tag{2}$$

where  $\alpha_{ex}$  is the exchangeable fraction of r-Cs in soil or sediments, and  $K_d$  is the apparent (or total) distribution coefficient.

Soon after the FDNPP accident it was discovered that Fukushima-derived r-Cs is strongly bound to soil and sediment particles, and its apparent distribution coefficient  $K_{d}$  in Fukushima rivers is at least order of magnitude higher than in pre-Fukushima observations [22,37,63-65]. This fact was then confirmed by longer term studies [61,66-69]. Table 1 presents summary of typical available data on <sup>137</sup>Cs distribution in suspended sediment-water system of rivers in Fukushima contaminated areas. The strong binding of r-Cs with soils and sediments seems to be associated with two factors: 1) relatively high fraction of micaceous clay minerals in Fukushima soils and sediments capable to selectively sorb and fix radiocesium [48,49,70-79] and 2) occurrence of high proportion of insoluble in water and persistent in the environment glassy hot particles CsMPs in FDNPP release [10,33]. It should be mentioned here that nuclear weapons testing derived r-Cs was not characterized by elevated values of  $K_d$  in rivers of Japan [80,81], which also confirms the role of FDNP-derived CsMPs for r-Cs solid-liquid distribution in Fukushima rivers.

Due to the fact that river water differs from marine water by hydrochemistry an interesting feature is observed such as  $K_{d}$  jump on the "freshwater-marine water" interface. The point is that freshwater differs markedly from seawater in terms of cation's concentrations in solution and, in the first place, concentration of potassium, which is the key competitor of r-Cs for sorption sites FES. On the interface between fresh and coastal waters, the apparent  $K_d$  value of <sup>137</sup>Cs jumps down by two orders of magnitude due to a sharp increase of potassium concentration from several mg/L in rivers to about 350-400 mg/L in coastal waters [82-84]. An obvious increase in potassium concentration also occurs from upstream to the mouth in a number of rivers in Fukushima contaminated areas. In Abukuma River, as an example, [K<sup>+</sup>] changed from 3.5 mg/L upstream to 44 mg/L at the mouth at tide, or in Maeda River [K<sup>+</sup>] changed from 0.7 mg/L upstream to 11.5 mg/L at the mouth in 2018 [84]. This leads to enhanced ion exchange desorption and remobilization of r-Cs from sediments to solution in the coastal areas of the Ocean. As a result, more than 95% of radiocesium in the ocean occurs in solution [85,86], while in rivers and other freshwater bodies a major part of radiocesium is bound to sediments [24,37,61,67,87]. A massive delivery of contaminated suspended material from accidental zone catchments by rivers during extreme floods in the course of typhoons causes an intense desorption and remobilization of <sup>137</sup>Cs and its concentration increase in estuary [86].

#### Leaching of r-Cs due to CsMPs weathering

The processes of decomposition of CsMPs are difficult to model due to diversity of their size, shapes, and chemical characteristics [10,41]. Therefore, an integral parameter is often used such as the first order rate constant  $k_i$  (yr<sup>-1</sup>) accounting for the rate of radionuclide leaching from hot particles, i.e. radionuclide transfer from hot particle to solution [10,88]. It can be expected that r-Cs leaching from CsMPs is very slow process. At the same time, when released

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River-site	Observation period	Mean value	Reference
Abukuma River-Kuroiwa	2012-2020	(6.5±3.0) × 10 <sup>5</sup>	lgarashi et al. [63]
Kuchibuto River upstream	2011-2016	(4.9±2.1) × 10 <sup>5</sup>	Taniguchi et al. [68]
Ukedo River downstream	2015-2018	(2.2±0.3) × 10 <sup>5</sup>	Nakanishi and Sakuma [67]
Ukedo River at Ogaki dam inflow	2014-2019	(6.3±2.0) × 10 <sup>5</sup>	Funaki et al. [69]
Kodeya River at Ogaki dam inflow	2014-2019	(8.6±2.1) × 10 <sup>5</sup>	Funaki et al. [69]
Ukedo River at Ogaki dam outflow	2014-2019	(4.5±1.8) × 10 <sup>5</sup>	Funaki et al. [69]
Takase River (Ukedo River system)	2011-2016	(6.6±1.2) × 10 <sup>5</sup>	Taniguchi et al. [68]
Ohta River downstream	2015-2018	(2.4±0.6) × 10 <sup>5</sup>	Nakanishi and Sakuma [67]
Niida River Haramachi	2011-2016	(6.3±2.5) × 10 <sup>5</sup>	Taniguchi et al. [68]
Hiso River (Niida River system)	2011-2020	(4.6±3.0) × 10 <sup>5</sup>	Ueda et al. [70]
Wariki River (Niida River System)	2011-2020	(7.7±6.3) × 10 <sup>5</sup>	Ueda et al. [70]

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[10].

from CsMPs, r-Cs in the soil-water environment is fairly quickly fixed by clay minerals of soils or sediments [47]. Based on laboratory experiments on r-Cs leaching from individual CsMPs by Okumura et al. [41] average values of rate constants of r-Cs leaching from CsMPs were estimated as  $k_i \ge 0.043$  yr<sup>-1</sup> in freshwater, and  $k_i \approx 0.44$  yr<sup>-1</sup> in seawater

The rate constant can also be estimated from long-term monitoring data indirectly. Monitoring of <sup>137</sup>Cs in heavily contaminated ponds in the vicinity of FDNPP revealed a gradual decline of its apparent solid-liquid distribution coefficient  $K_d$  from 2015 to 2019 [42], which is not typical of r-Cs dynamics in water bodies in mid- and long-term [23]. The initial and intermediate phases after the Chernobyl accident, and after nuclear weapon tests (NWT), were characterized by a slight increase of r-Cs apparent  $K_{d}$  in soils and sediments as a result of its fixation or aging [8,53,55]. Assuming that the  $K_d$  decline trend is associated with gradual remobilization of <sup>137</sup>Cs due to leaching from CsMPs in the ponds  $k_i$  was estimated to be in the range 0.12-0.18 yr<sup>-1</sup> [42]. A similar trend for the radiocesium apparent  $K_d$  in 2012-2020 with the rate constant  $k_i=0.05$  yr<sup>-1</sup> has been recently reported for suspended sediment-water system in Abukuma River at Fukushima city (Kuroiwa) [61]. Thus, the estimates based on long-term monitoring data are consistent with those derived from laboratory experiments of Okumura et al. [41] with individual CsMPs. It can be expected that  $k_i$  obtained for natural freshwaters is higher than in laboratory experiments with pure water and lower than with marine water. This applies to the estimates presented above.

#### Fixation of r-Cs by clay minerals and remobilization

As it appears from laboratory experiments and field observations with a variety of soils and sediments [47,54,55,89-91] the timescale of radiocesium fixation equals to weeks or months ( $k_f$ =4-20 yr<sup>-1</sup>), depending on environmental conditions, while the timescale of remobilization can be up to a few years ( $k_r$  = 0.4-2 yr<sup>-1</sup>). Based on data on speciation of Fukushima-derived r-Cs [78,79,92,93], it is expected that these processes are characterized by similar rates in Fukushima soils and sediments [10].

## Natural attenuation of radioactive contamination after FDNPP accident

### Vertical and lateral redistribution of radiocesium in catchment soils

Processes of natural attenuation besides radioactive decay do not reduce the total amount of radionuclides in the environment, rather they affect radionuclide distribution over space and time [94]. Physical processes involved in natural attenuation (advection, diffusion, dispersion) dilute radionuclides in the environment or partially remove/relocate and spread them (wash-off, erosion, sedimentation and river

transport) [7]. Processes of r-Cs vertical and lateral migration lead to gradual reduction in contamination of catchment soil, particularly its top layer [95-98]. This, in turn, results in a gradual decrease of r-Cs concentrations in both particulate and dissolved forms of surface runoff and river water [23,61,87].

Climate and geomorphological conditions essentially influence the rate of natural attenuation processes [7]. Fukushima's watersheds are mostly hilly with steep slopes. Annual precipitation is relatively high with annual averages of about 1500 mm/year for Fukushima according to the Japan Meteorological Agency [11,65].

After deposition of radionuclides on the ground surface, over time the contamination migrates down through the soil profile [6,10,95,97,98]. The dynamic pattern of vertical distribution of radionuclides in soil is critical from the standpoint of external dose rate, availability of radionuclides for transfer to surface runoff and wind resuspension in the boundary atmospheric layer, availability of radionuclides for root uptake by plants and percolation to groundwater. Radionuclides migrate vertically in solution and as colloids with infiltration water flow or attached to fine soil particles. Transport of r-Cs in solution by infiltration is slower than the water flow because of its sorption-desorption and fixation on soil particles [99]. Fine soil particles containing radiocesium can move by penetrating through pores, cracks and cavities, as well as with infiltration flow (lessivage), and as a result of vital activity of plants and biota (bioturbation) [58,96]. Nevertheless, the vertical migration of radionuclides in undisturbed soils can be described by the advection-dispersion equation [57, 99,100]. Simultaneous solution of respective equations written separately for specific radiocesium chemical forms in soil  $R_i$  ( $R_h$ ;  $R_{org}$ ;  $R_f$ ;  $R_{ex}$  and  $R_d$  - see Figure 1) with allowance for their transformation [54,89,101] is the most accurate way of representing <sup>137</sup>Cs migration by the advection-dispersion model:

$$\frac{\partial R_i}{\partial t} = \frac{\partial}{\partial x} \left( D_i \frac{\partial R_i}{\partial x} \right) - v_i \frac{\partial R_i}{\partial x} + \sum k_{ji} R_j - \sum k_{ij} R_i - \lambda R_i$$
(3)

with the initial conditions

 $R_i = R_i^0 \delta(x - 0)$  and boundary conditions  $R_i |_{x=\infty} = 0$ 

where  $D_i$  and  $v_i$  are the effective dispersion coefficient and effective advective velocity for each chemical form *i*, respectively;  $k_{ij}$  and  $k_{ji}$  are the rate constants of respective transformation process; and  $\lambda$  is the rate constant of radioactive decay equal 0.023 yr<sup>-1</sup> for <sup>137</sup>Cs.

Importantly, most vertical distributions of Fukushimaderived r-Cs demonstrated maximum of its activity concentrations in the topsoil layer for undisturbed land [6,95,97,98]. This means that <sup>137</sup>Cs vertical distribution can be roughly approximated by simple diffusion equation with effective dispersion coefficient varying mostly in the range 1-9 cm<sup>2</sup>/yr [65,95,96].

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Vertical distribution of r-Cs in floodplain soils differs significantly from that in undisturbed catchment soils because of erosion of topsoil and/or accumulation of sediments during floods [7,17,65]. Significant floods occur in Fukushima Prefecture during typhoon seasons, usually from mid-August to October. A very rare extreme floods occurred in September 2015 (Typhoon Etau) and in October 2019 (Typhoon Hagibis). The probability of such extreme floods in Fukushima contaminated area can be estimated at 4-5% per year [7]. During the floods, water flows washed out unsupported sections of stream banks. Also, some landslides occurred, mainly on very steep slopes. Sediments originated from the bank erosion, and landslides have very low concentration of <sup>137</sup>Cs because of high proportion of material from deep soil layers. Therefore, extreme flood events in Fukushima contaminated areas very often cause substantial natural decontamination of river floodplains because of erosion of contaminated particles from the top layer and additional burying contaminated surface particles by deposited clean sediments [15,17]. In many cases this is followed by significant drop of air dose rate. Estimated rate constants of dose rate reduction for the sites without contaminated sediments accumulation in 2016 were in range of 0.2-0.4 year<sup>-1</sup> [7]. Therefore, typhoons passing on Fukushima contaminated area result in relatively fast and efficient natural attenuation.

At the same time, in case of extreme floods on relatively large rivers, sediments with relatively high <sup>137</sup>Cs concentrations can deposit on floodplain downstream influx of tributaries flowing through areas with elevated <sup>137</sup>Cs levels [17]. This was the case after Typhoon "Hagibis" in October 2019 on the sections of Abukuma River floodplain downstream of Kuchibuto river catchment characterized by elevated depositions of <sup>137</sup>Cs. Figure 2 shows vertical profiles of <sup>137</sup>Cs for floodplain sediments of Abukuma River

at two monitoring sites Fukushima (Kuroiwa) and Fushiguro located downstream of the mouth of the Kuchibuto River, before Typhoon "Hagibis" in 2018 and right after it in November 2019. The top layer of the floodplain sections was contaminated by sediments deposited after typhoon's flood, while previous top layer sediments were removed by powerful water flow during the typhoon. It can be suggested that during the typhoon "Hagibis" the sediments from contaminated Kuchibuto River catchment were transported and deposited onto Abukuma River floodplain downstream [84].

Accounting for soil erosion and sediment accumulation within river catchment and in particular, river floodplain, is key for predicting redistribution of radioactive contamination after the FDNPP accident on the contaminated territories, as well as for decision making about their remediation and cleanup. In many cases monitored natural attenuation along with appropriate restrictions can be the most optimal option for water remediation in Fukushima contaminated areas [7,94].

# **Radiocesium accumulation in bottom sediments of dam reservoirs**

Knowing temporal changes of <sup>137</sup>Cs particulate concentrations in rivers is critically important for better understanding and prediction of radionuclide behavior in the soil-water environment. In the absence of systematic monitoring data, this gap can be filled by reconstructing particulate <sup>137</sup>Cs concentrations based on <sup>137</sup>Cs depth distribution in the bottom sediment column. Bottom sediments in dam reservoirs represent the final chain in the geological cycle of the matter within a river basin [102,103]. In the Fukushima contaminated areas, at least 10 major dams are in operation to support agricultural and surface water management [104,105]. Apparently, these reservoirs have trapped substantial fraction of transported radiocesium-contaminated sediments [106,107].



**Figure 2:** Depth distribution of <sup>137</sup>Cs in accumulation zones of floodplain sediments of Abukuma River at sites located downstream for the mouth of its right side tributary Kuchibuto River flowing through the FDNPP contaminated zone – Fukushima (at Kuroiwa) and Fushiguro, based on the data from Golosov et al. [17].

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Depth distributions of radionuclides in bottom sediments of dam reservoirs can be used to reconstruct concentrations of strongly bound to sediment radionuclides occurring in rivers [108,109]. Sediments depositing in deep-water zones of reservoirs are delivered from the river and therefore they can serve to retrieve time changes of radionuclide concentrations in the river [25]. For reconstruction, however, it is required that mixing of sediments after deposition of radionuclides on the bottom be negligible, so that the radionuclide concentration in each individual sediment layer correspond to concentration in river suspended sediments during the respective time interval. Another requirement is that radionuclide should be strongly bound by sediments ( $K_d >> 10^4$  L/kg) [25,109] which is valid for Fukushima-derived r-Cs [37].

This approach was used for bottom sediments core from accumulation zone of Ogaki Dam at Ukedo River [25]. Figure 3 presents time dependencies of particulate and dissolved <sup>137</sup>Cs in Ukedo River after the Fukushima accident reconstructed on the basis of bottom sediments core collected in 2019 and their comparison with monitoring data [66,68] and with empirical two-exponential model [25]. Time changes of particulate and dissolved <sup>137</sup>Cs activity concentration in Ukedo River, reconstructed from the depth distribution in bottom sediments of deep-water accumulation zone in Ogaki Dam reservoir, are in good agreement with monitoring data [66,68] and are adequately approximated by the simple empirical two-exponential model. This means that the described method can be used in the future to reconstruct r-Cs dynamics in rivers of Fukushima contaminated areas.

# Time Changes of <sup>137</sup>Cs in Freshwaters and their Modelling

Initial radioactive contamination of water bodies after the nuclear FDNPP accident was relatively high, owing to direct fallout onto the rivers and lakes surfaces [18,22,63,64,110]. The contamination of water bodies was then decreasing sharply due to fast processes of sorption and fixation of radionuclides to sediments, as well as sedimentation of particles to the bottom [10,24,61,66,67,111-113]. Yet, extensive territories contaminated due to the FDNPP accident continue to serve as a long-term source of r-Cs to natural waters and aquatic ecosystems. Wash-off driven by surface runoff is the primary pathway for contamination of water bodies in mid- and long-term [11,18,21,24,87,114,115].

Prior the Fukushima accident, temporal changes in <sup>137</sup>Cs activity concentrations in rivers were often described by the empirical fitting model using a series of exponential functions [116-118]:



**Figure 3.** Reconstruction of time changes in <sup>137</sup>Cs activity concentrations in the Ukedo River based on its depth distribution in Ogaki dam reservoir's bottom sediments. A – location of Ukedo River and Ogaki dam in FDNPP contaminated area; B - <sup>137</sup>Cs depth distribution profile for bottom sediment core from the Ogaki dam reservoir taken for reconstruction; C - time dependence of reconstructed particulate <sup>137</sup>Cs activity concentration in the Ukedo river versus monitoring data [66,68] and approximation by the empirical two-exponential model; D - time dependence of reconstructed dissolved <sup>137</sup>Cs activity concentration in Ukedo River versus monitoring data [66,68] and approximation by the empirical two-exponential model.

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$$c(t) = \sum_{i} c_i^0 e^{-(\lambda + k_i)t} \tag{4}$$

where c(t) is the current <sup>137</sup>Cs activity concentration in river (for particulate <sup>137</sup>Cs in Bq/kg, and for dissolved in Bq/ m<sup>3</sup>);  $\lambda$  is the <sup>137</sup>Cs decay rate const<sub>c</sub> ot equal 0.023 yr<sup>1</sup>;  $k_i$  are empirically fitted rate constants; are fitting parameters, and t is time. This approach was followed by a number of researchers in post-Fukushima studies of r-Cs (both dissolved and particulate) dynamics in rivers [24,61,66-69].

Alternative way to model mid- and long-term dynamics of radiocesium in rivers is semiempirical diffusional approach [23,135]. The key assumption of the model is that the main source of suspended particles for surface runoff is the top layer of catchment soil, and r-Cs vertical distribution in the topsoil layer is described by the diffusional equation [96]. In this case time changes of <sup>137</sup>Cs activity concentration in topsoil layer and thus in suspended sediments can be approximated by the following equation:

$$C_p(t) = \frac{\sigma_0}{\rho \sqrt{\pi D_{eff}t}} e^{-\lambda t} = C_p^0 \frac{e^{-\lambda t}}{\sqrt{t}}$$
(5)

where  $\sigma_{\theta}$  is the initial average deposition of <sup>137</sup>Cs on the catchment;  $D_{eff}$  is the effective dispersion coefficient, averaged over the catchment area; is the radioactive decay rate constant;  $\rho$  is the average bulk density of the topsoil over the catchment; and *t* is time.

With allowance for equations (1) and (5), the time dependence of dissolved <sup>137</sup>Cs concentration in a river can be approximated by the equation [23]:

$$c_{\rm d}(t) = \frac{\sigma_0}{\rho K_{\rm d} \sqrt{\pi D_{\rm eff} t}} e^{\lambda t}$$
(6)

The advantage of this approach is that the same equation can be used for middle- and long-term phases after a nuclear accident with the same values of physically based parameters which can be estimated or determined by field or laboratory studies. More simply, decay corrected particulate r-Cs activity concentrations in surface runoff and rivers are described by the inverse square root of time function.

Figure 4 presents time changes of annual mean activity concentrations of dissolved 137Cs in Abukuma River at Fukushima city (Kuroiwa) in 2012-2019 [61] and their simulation by semi-empirical diffusional model. It should be noted that the <sup>137</sup>Cs activity concentrations in rivers and lakes in Fukushima contaminated areas quite often decline a bit faster than predicted by the semi-empirical diffusional model [10]. There are two major potential reasons for the observed discrepancy and difference with modelling results. First, extensive remediation program implemented by Japanese government on the contaminated catchments after the accident has effectively reduced average <sup>137</sup>Cs deposition on the catchment soils and especially decreased <sup>137</sup>Cs content in topsoil layer [19,20,24]. Second, the basic processes of surface runoff development in Fukushima are quite specific due to relatively high intensity of precipitation, especially during typhoons, and high energy of surface runoff flows [11,105]. As a result, deeper soil layers become to be involved in exchange with surface runoff which causes faster decline of particulate and dissolved radionuclide concentration in surface runoff [7].

To characterize catchment-to-river transfer of radionuclides by surface runoff the particulate and dissolved wash-off ratios  $n_p$  and  $n_d$  are widely used [18,22,23,61,87,115]:

$$n_p = \frac{\overline{c_p}}{\sigma}; \ n_d = \frac{\overline{c_d}}{\sigma} \tag{7}$$

where  $\overline{c_p}$  and  $\overline{c_d}$  are the particulate and dissolved radionuclide annual mean activity concentrations in river or surface runoff, respectively;  $\sigma$  is the current average radionuclide deposition on the catchment.

The wash-off ratios enable predicting the radionuclide wash-off from contaminated catchment and its concentration in rivers and other water bodies. To estimate the fraction of the radionuclide washed off in solution, the dissolved washoff ratio is multiplied by the expected runoff depth for a given runoff event or period of interest. The fraction of radionuclide



**Figure 4:** Time changes of annual mean particulate and dissolved <sup>137</sup>Cs activity concentrations in Abukuma River at Fukushima city (Kuroiwa) in 2012–2019 based on the data of [61] against diffusional model prediction (dotted line).

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washed off with sediments is estimated by multiplying the particulate wash-off ratio by the predicted sediment yield during the runoff event or period of interest [119,120].

Time dependence of <sup>137</sup>Cs particulate and dissolved washoff ratios for mid- and long-term phases after the FDNP accident according to the semi-empirical diffusional model can be described as follows [121]:

$$n_p(t) = \frac{1}{\rho_{\sqrt{\pi D_{eff}t}}} = \frac{n_p^0}{\sqrt{t}}; n_p^0 = (\rho_{\sqrt{\pi D_{eff}}})^{-1}$$
(8)

$$n_d(t) = \frac{1}{\rho K_d \sqrt{\pi D_{eff} t}} = \frac{n_d^0}{\sqrt{t}}; n_d^0 = (\rho K_d \sqrt{\pi D_{eff}})^{-1}$$
(9)

Decline of particulate wash-off ratio with time for Fukushima rivers can be described by diffusional model where  $D_{eff}$ =3.5-5.0 cm<sup>2</sup>/yr, whereas dissolved wash-off ratios follow the model at  $K_d$ =2.5×10<sup>5</sup> L/kg for Ukedo and Ohta rivers and at  $K_d$ =6×10<sup>5</sup> L/kg for Abukuma River at Fukushima, which values correspond to experimental data for these rivers [61,65,66].

# Seasonal Variations of <sup>137</sup>Cs in Contaminated Water Bodies

Rivers of the Fukushima contaminated areas demonstrated clear seasonal variations of dissolved <sup>137</sup>Cs, showing pronounced minimum in winter and maximum in summer [61,66,87,122]. Similar seasonal variations were observed in Fukushima ponds, lakes, and dam reservoirs [42,123-125]. The amplitude of such variations, i.e. ratio of maximal summer and minimal winter dissolved <sup>137</sup>Cs concentrations within the same year, is usually about 2-3 times. Figure 5 presents seasonal variations of dissolved <sup>137</sup>Cs concentration in water of the pond Suzuuchi in Okuma town in 2016 and 2017 [42].

Conceivably, the seasonal changes in water temperature could have an impact on ion-exchange desorption of <sup>137</sup>Cs which proceeds according to the Gibbs–Helmholtz equations for <sup>137</sup>Cs exchangeable distribution coefficient  $K_d^{ex}$  [61,126]:



**Figure 5:** Seasonal variations of dissolved <sup>137</sup>Cs activity concentration in water of Suzuuchi irrigation pond in Okuma town at FDNPP exclusion zone in 2016 and 2017 [42].

$$\frac{\partial lnK_d^{ex}}{\partial(\frac{1}{T})} = -\frac{\Delta H}{R}$$
(10)

where  $\Delta H$  is the enthalpy change for <sup>137</sup>Cs desorption from sediments to solution at temperature *T*, and *R* is the gas constant.  $\Delta H = -E_A$ , where  $E_A$  is Arrhenius activation energy [127-129]. The obtained estimates of  $E_A$  were around 20 kJ/mol for Fukushima rivers [61,87] and ponds [42], which is consistent with results of laboratory experiments on sediments from Hanford site [126].

Seasonal variations of dissolved Chernobyl-derived <sup>137</sup>Cs in lakes and ponds [113,130,131] were attributed to the variations in ammonium concentration in the water. Generally shallow and fast-flowing Fukushima rivers are characterized by negligible ammonium concentrations. At the same time, in the Fukushima stagnated water bodies such as lakes, ponds, and dam reservoirs the decomposition of organic matter in the reducing conditions of bottom-sediments could also have led to ammonium generation in pore water, which could have contributed to increased dissolved <sup>137</sup>Cs in the water column in summer [42,123,124]. According to the r-Cs ion exchange theory [38,43,132,133], exchangeable distribution coefficient of <sup>137</sup>Cs at constant water temperature depends on ammonium concentration as follows:

$$K_d^{ex} = \frac{RIP^{ex}(K)}{[K^+] + K_c(NH_4/K)[NH_4^+]}$$
(11)

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where  $RIP^{ex}(K)$  is exchangeable r-Cs interception potential, intrinsic property of soils and sediments accounting their ability to selectively and reversibly sorb r-Cs on frayed edge sites (FES) of micaceous clay minerals [113,132,133];  $K_c(NH_4/K)$  is selectivity coefficient of ammonium sorption on FES in relation to potassium. According to Wauters et al. [132,133], for more than hundred studied soils and sediments  $K_c(NH_4/K)$ =5±2. Equation (11) clearly shows that looking for correlations of dissolved <sup>137</sup>Cs activity concentration with single concentrations of potassium or ammonium in water is hopeless, when both these r-Cs competitors for sorption sites FES have comparable contribution in r-Cs desorption from sediments.

Combining equations (10) and (11) seasonal variations of  $K_d^{ex}({}^{137}Cs)$  and dissolved  ${}^{137}Cs$  activity concentration  $[{}^{137}Cs]_d$  in water body for mid- and long-term phases after the accident can be approximated by the following equations:

$$K_{d}^{ex}(^{137}CS) = \frac{K_{0}}{([K^{+}] + 5[NH_{4}^{+}]) \times e^{-\frac{E_{A}}{RT}}}$$
(12)

$$[^{137}Cs]_d = C_0 e^{\frac{E_A}{RT}} \times ([K^+] + 5[NH_4^+])$$
(13)

where  $K_0$  and  $C_0$  can be taken as a constant within the single year

Most likely, seasonality of dissolved <sup>137</sup>Cs in Fukushima rivers is determined exclusively by changes of water

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temperature, since ammonium concentration in river water is normally negligible, and potassium concentration does not exhibit any regular seasonal variations. For ponds, lakes, and dam reservoirs the situation is more complicated when contribution of ammonium to <sup>137</sup>Cs desorption is higher or comparable with contribution of potassium. It is expected that using of equations (12) and (13) should reduce the scattering of experimental points at the  $K_d^{ex} - ([K^+] + 5[NH_4^+])$  and  $lnK_d^{ex} - \frac{1}{T}$  plots for field observations especially in ponds and dam reservoirs [42,68,123,124]. However, applicability of equations (12)  $\mu$  (13) should be further investigated and tested in field observations.

#### Conclusions

The FDNPP accident in March 2011 led to extensive environmental contamination by radiocesium. Numerous studies of the radionuclide fate and transport in the environment in the context of geoclimatic conditions of Japan were carried out. The scientific evidence from conducted research confirmed that behavior of accidentally released radiocesium in the environment is governed by its speciation in fallout and site-specific environmental characteristics, even though the Fukushima-derived radiocesium has certainly distinct features and characteristics.

The Fukushima-derived <sup>137</sup>Cs was found to be strongly bound to soil and sediment particles, which reduces its potential bioavailability. Also, in surface waters faster scavenging of radiocesium is observed and its transport primarily occurs on suspended material, which is important to bear in mind in modeling and prediction.

Up to 80% of the deposited <sup>137</sup>Cs on the soil of the contaminated area were reported to be incorporated in glassy hot microparticles insoluble in water. These particles decompose in the environment very slowly, and long-term radiocesium leaching from these particles remains to be understood in future.

The frequently used in modeling parameter of apparent distribution coefficient  $K_d$ <sup>(137</sup>Cs) in the suspended sediments—water system of Fukushima rivers and ponds was shown to decrease with time after the accident. From the decrease in  $K_d$  estimates of the timescale of <sup>137</sup>Cs leaching from glassy microparticles were obtained, ranging from 6 to 20 years, which is consistent with laboratory findings.

The high annual precipitation and steep slopes in Fukushima contaminated areas are conducive to significant erosion and intensive radiocesium wash-off, especially during devastating tropical storms. Typhoons Etou in 2015 and Hagibis in 2019 were reported to bring about major redistribution of <sup>137</sup>Cs on river watersheds and floodplains, and in many cases natural self-decontamination was noticed.

With Fukushima dam reservoirs acting as traps,

preventing radiocesium transport downstream, the <sup>137</sup>Cs vertical distributions in bottom sediments of dam reservoir deep zones can be regarded as representing time changes in river water contamination.

The observed regular seasonal variations of dissolved <sup>137</sup>Cs in water bodies (increase in summer and decrease in winter) can be attributed to two major factors: 1) temperature dependence of radiocesium desorption rate from sediments, and 2) generation of ammonium as a result of organic matter decomposition in anoxic conditions. Hence, a key mechanism of radiocesium seasonality in aquatic ecosystems is an important issue to be tackled in future.

Another challenge for contemporary environmental chemistry understands mechanisms and prediction of radiocesium remobilization from river-transported sediments at the freshwater-seawater interface.

Environmental radioactivity research following the Fukushima accident has made a major contribution to gaining greater knowledge about the processes of interest [134-136]. Nevertheless, unforeseen results can occur and require further investigation in future. Thus, new challenges are emerging, and environmental radiochemistry has yet to respond to them.

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