Investigation of Radiocesium Migration from Land to Waterbody using Radiocesium Distribution and Soil to Sediment Ratio: A Case of the Steep Slope Catchment Area of Ogi Reservoir, Kawauchi Village, Fukushima

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In the present study, we reported the investigation of radiocesium migration from steep slope surrounding catchment to water body based on the radiocesium distribution and soil to sediment ratio. It was confirmed that the radiocesium was derived from the Fukushima accident by considering the 134Cs to 137Cs ratio corrected to the released day which is about 1. We found the higher radiocesium inventory in higher elevation area and steep slope forest catchment, revealing the atmospheric dry initial deposition-derived radiocesium and high retention of the forest zone. The radiocesium distribution data revealed that the radiocesium migrated more from the transition zone to the water body than from forest catchment, and accumulated in the deeper layer of sediment. The lower value of 137Cs soil to sediment ratio provided an evidence of radiocesium migration from the catchment and its accumulation in the sediment. The physicochemical property of surface soil was revealed as one of the possible factors of radiocesium high retention in forest catchment.

Key Words: radiocesium, migration, elevation, land-use, soil properties

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1. Introduction

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident that occurred in March, 2011, caused a substantial amount of radioactive materials released to the environment. Among them, radiocesium (134Cs and 137Cs) beside radioiodine (131I) is the most major released volatile radionuclide having a direct impact on land contamination.1) The longer 137Cs half-life (30.2 years) in comparison to 134Cs (2.06 years) and 131I (8 days) has made the 137Cs as the main radionuclide of interest to be studied.

The distribution and migration of FDNPP accident-derived radiocesium in the terrestrial environment over the period of time have been widely investigated. The understanding of radiocesium migration behavior is key to the assessment for the long-term radiation hazard risk and its countermeasures. Investigation of radiocesium migration from soil to the water body is one of the methods that can be used to analyze the rate of up-stream clean-up over time as well as to determine to what extent the radiocesium will contaminate the downstream. The soil-water body migration from forest catchment is the
main feature in Fukushima because the forest area covers more than 60% of the contaminated zone and has higher radiocesium inventory in comparison to the other land use.2,3) This area could be a long-term source of particulate radiocesium for the downstream area especially for the water body such as river, reservoir or lake, and sea.2,4)

Previous studies revealed the importance of elevation and slope aspects on radioactive materials migration in the catchment over several times following the accident.5–9) Other studies showed that, in the case of Fukushima, the lateral migration was promoted in the higher slope area and resulted in the radiocesium accumulation in the middle and bottom of the slope as well as in the lake sediment.8–10)

The methods to estimate the migration of 137Cs in the catchment are difficult and time consuming such as by field experiment that utilized the leachate or seepage water samples3,11) and by measuring the 137Cs in suspended solid of water sample12) Another method is performed by analyzing the vertical profile of the lake sediment. The idea is constructed by assuming that the catchment-derived 137Cs has a significant contribution to the total 137Cs inventories in the lake sediment cores13) and the vertical profile is time-dependent.14) The relatively easier method of estimation was performed by using 137Cs soil to lake sediment ratio to estimate the 137Cs initial accumulation and flushing in the sediment of the lake just after the accident, however, it needs more investigation regarding its robustness.15)

In the present study, we examined the 137Cs distributions and 137Cs soil to lake sediment ratio for investigating the radiocesium migration. Hereafter the sediment means the lake sediment. Furthermore, we discussed the high radiocesium retention of the steep forest zone catchment by considering the physicochemical property of the surface soil.

2. Materials and methods

2-1. Air dose measurement, soil and sediment sampling and sample preparation

The study area is in Ogi reservoir catchment area, Kawauchi Village, Fukushima, which is located about 18 km southwest from FDNPP. It is one of the areas that was passed by the radioactive plum following the accident, thus the area received a significant amount of radioactive material. There have been various researches about radioactive contamination and its consequences in this area.16–18)

Air dose measurement and soil and sediment core sampling were conducted during March 15~16, 2018 in a steep slope forest and transition zone of the catchment area. The slope catchment was divided into 3 air dose measurement area (high, middle and low elevation area), see Fig. 1. Air dose measurement points were selected to apart to each other by about 2 m. The soil sampling sites were selected purposively within each air dose measurement area as an imaginary transect 2 × 2 m². Sediment core samples were collected from the deeper part and most 137Cs contaminated area of the reservoir based on the previous study.19) Geographic position and elevation of the sites were recorded using GPS locator (Garmin, Oregon 650TCJ). The details of measurement and sampling sites are summarized in Table 1 and Appendix 1.

The air dose rate was measured by a portable gamma survey meter with a NaI detector (ALOKA MYRATE PDR-111) about 1 m above the ground. Regarding the core sampling, at least 3 samples for each soil sampling transect were collected in order to obtain the representative data.20) The data are shown as the average with accidental error. The hand core sampler (5 cm diameter, 25 cm long tube) and gravity core sampler (6.5 cm diameter, 60 cm long tube) were used to collect soil and sediment samples, respectively. The soil and sediment core samples were sent to the laboratory by keeping the natural vertical direction to preserve the vertical profile.21)

The soil and sediment core samples were cut each 2 cm and 1 cm increment, respectively. The samples were dried at room temperature and further dried in oven (105°C, 24 hours). The prior dry sieving (2 mm sieve) using electric horizontally rotating sieve (SKH-01, AS ONE) with mesh size 2 mm at about 280 rpm for 1 h was conducted to separate gravel and stone from the sample. The remaining samples were packed into the so-called U8 vessel (100 ml, 5 cm height, 5 cm diameter) for radioactivity measurement.

2-2. Radioactivity measurement and calculation

The activities of 134Cs and 137Cs were measured by gamma spectroscopy with HP Ge detector and multichannel analyzer (GEM 30-70, ORTEC), at energy peak of 604 keV and 662 keV for 134Cs and 137Cs, respectively. The measurement time was judged to be sufficient statistically when the error value of less than 5% was reached. Counting efficiency was calibrated using a set of standard sources (MX03U8PP) manufactured by the Japan Radioisotope Association. Particularly for 134Cs, as multiple gamma emitter, its activity was calculated by involving the correction due to the sum-effect. However, only
Fig. 1. The study area, Ogi reservoir catchment, Kawauchi Village, Fukushima, source: [http://mapps.gsi.go.jp/maplibSearch.do#1](http://mapps.gsi.go.jp/maplibSearch.do#1), accessed in September 2019.

| Area          | Samples | Location | Elevation | Core sample depth (cm) | Site details                                                                 |
|---------------|---------|----------|------------|------------------------|------------------------------------------------------------------------------|
| Catchment,    | 1       | 37.34363 | 140.90566  | 378 0~18               | The sampling area is in the middle to the top of the ridge, consisted of mixed forest type. |
| Area 1        | 2       | 37.34359 | 140.90571  | 376 0~18               |                                                                              |
|               | 3       | 37.34372 | 140.90567  | 379 0~16               |                                                                              |
|               | 4       | 37.34365 | 140.90559  | 382 0~18               |                                                                              |
| Catchment,    | 1       | 37.34364 | 140.90589  | 369 0~20               | The sampling area is in the middle of the ridge, consisted of mixed forest between coniferous and deciduous forest type. |
| Area 2        | 2       | 37.34363 | 140.90589  | 369 0~20               |                                                                              |
|               | 3       | 37.34368 | 140.90589  | 368 0~21               |                                                                              |
| Catchment,    | 1       | 37.34362 | 140.90615  | 365 0~20               | The sampling area is in between the catchment and reservoir or transition zone, there is no vegetation. |
| Area 3        | 2       | 37.34361 | 140.90613  | 365 0~20               |                                                                              |
|               | 3       | 37.34357 | 140.90623  | 365 0~7                |                                                                              |
| Reservoir     | 1       | 37.34426 | 140.90716  | 362 0~35               | The sampling area is in the deep area of the reservoir                        |
|               | 2       |          |            |                        |                                                                              |
long half-lived $^{137}\text{Cs}$ is reported in this paper as a dominant contributor of Cs isotopes (>90%) and a strong relationship was obtained between $^{137}\text{Cs}$ and $^{134}\text{Cs}$ ($R^2: 0.99$), see Fig. 3. The activity was decay corrected to the sampling day and decay corrected to the released day, on March 15, 2011, because the main plume reached on the day. However, in this paper, only the data of sampling day is presented to show the current status of radioactive contamination.

The total $^{137}\text{Cs}$ inventory (Bq • cm$^{-2}$) was calculated from the $^{137}\text{Cs}$ activity concentration (Bq • g$^{-1}$), the soil density (g • cm$^{-3}$) and physical depth (cm). In the calculation of $^{137}\text{Cs}$ vertical profile, the cumulative mass depth (kg • m$^{-2}$) was used instead of physical depth (cm or m), because the cumulative mass is more comparable among the samples than physical depth and was used by some previous researches.

The $^{137}\text{Cs}$ soil to sediment ratio was simply calculated by dividing the $^{137}\text{Cs}$ deposition density of soil by that of sediment. The ratio of the present study was compared with the previous

| Area | Measurement | Location | Elevation |
|------|-------------|----------|-----------|
|      |             | Latitude (N) | Longitude (E) |   (m) |
| Area 1, Mixed forest | 1 | 37.34359 | 140.90571 | 376 |
| Catchment | 2 | 37.34363 | 140.90566 | 378 |
|     | 3 | 37.34363 | 140.90565 | 378 |
|     | 4 | 37.34372 | 140.90567 | 379 |
|     | 5 | 37.34364 | 140.90542 | 389 |
|     | 6 | 37.34358 | 140.90554 | 383 |
|     | 7 | 37.34365 | 140.90559 | 382 |
|     | 8 | 37.34369 | 140.90559 | 382 |
|     | 9 | 37.34376 | 140.90550 | 389 |
|     | 10 | 37.34380 | 140.90549 | 391 |
|     | 11 | 37.34388 | 140.90543 | 394 |
| Area 2, Mixed forest | 1 | 37.34368 | 140.90589 | 368 |
| Catchment | 2 | 37.34356 | 140.90592 | 368 |
|     | 3 | 37.34367 | 140.90588 | 369 |
|     | 4 | 37.34364 | 140.90589 | 369 |
|     | 5 | 37.34363 | 140.90589 | 369 |
| Area 3, Transition zone | 1 | 37.34354 | 140.90634 | 365 |
|     | 2 | 37.34370 | 140.90617 | 365 |
|     | 3 | 37.34356 | 140.90628 | 365 |
|     | 4 | 37.34357 | 140.90623 | 365 |
|     | 5 | 37.34358 | 140.90620 | 365 |
|     | 6 | 37.34362 | 140.90615 | 365 |
|     | 7 | 37.34361 | 140.90613 | 365 |
2.3. Characterization of the physicochemical property of surface soil samples

Soil pH was measured by measuring the pH of soil sample-distilled water mixture (1/5 of w/v ratio), as previous work. 

The soil sample and distilled water were mixed by vortex for about 2 minutes and soil slurry was measured using pH meter.

Organic matter (OM) content fraction was determined by weight loss on ignition method according to the previous work. 

The samples were placed in muffle furnace (FO300, Yamato scientific) at 500°C for 12 hours after conducting the soil pre-treatment.

OM free samples were further wet sieved using 75 μm sieve to obtain silt and clay fraction (size fraction <75 μm) separated from the fine sand.

The exchangeable major cations and cation exchange capacity (CEC) of surface soil samples were determined by NH₄⁺ exchange (1 M Ammonium acetate, pH 7, 1/5 of w/v ratio), as previous research. 

The concentration of soil cations in solution was measured by ICP AES spectrometry (SPS3500, Hitachi). The soil precipitate of exchangeable cation measurement was used to determine the CEC, by exchanging the NH₄⁺ ion in soil sample matrix by using 1 M KCl. The CEC of the soil was determined as the concentration of ammonium ion released into the supernatant. The extracted NH₄⁺ concentration was measured by colorimetry (Methyl red, Thymol blue and titration of 0.01 M NaOH). The base saturation was further calculated based on the ratio of total exchangeable cation to CEC.

3. Results

3-1. Horizontal distribution of radioactive contamination along the gradient elevation

The data of air dose rate along the gradient elevation are presented in Fig. 2 (a). The air dose was ranged from 0.31 to 1.92 μSv・h⁻¹. Higher air dose rate was found in Area 1 (1.64 ± 0.07 μSv・h⁻¹) and Area 2 (1.4 ± 0.1 μSv・h⁻¹) which are the higher elevation area and mixed forest area in comparison to Area 3 (0.50 ± 0.07 μSv・h⁻¹) which is lower elevation and no forest or transition zone.

The ¹³⁷Cs inventory and air dose rate relationship is shown in Fig. 2 (b). This shows a linear relationship between ¹³⁷Cs inventory and the air dose rate, which could mean that the source of air dose rate was mainly from the gamma-ray, especially from ¹³⁷Cs, see below.

The ¹³⁴Cs and ¹³⁷Cs concentrations in the soil of the catchment area show a strong relationship, with its ratio of sampling day (March 15, 2018) about 0.11 and its ratio of decay corrected to the released day (March 15, 2011) about 0.93, see Fig. 3. The value of ¹³⁴Cs to ¹³⁷Cs ratio decay corrected to released day shows that the radiocesium was derived from FDNPP accident and more specifically was mostly from the reactor unit 1.

3-2. Vertical distribution of ¹³⁷Cs in soil and sediment

The vertical distributions of ¹³⁷Cs in soil and sediment are presented in Fig. 4. In the case of soil catchment, the ¹³⁷Cs was accumulated in the upper layer of soil as an overall tendency. However, in more detail, the ¹³⁷Cs penetrated more in the soil of Area 1 and Area 2 (forest soil) in comparison to the transition
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3-3. $^{137}$Cs soil to sediment ratio

Table 2 shows the data of $^{137}$Cs deposition density in soil and sediment and its ratio for some water bodies in the Fukushima area. The previous and the present study are compared and further presented in Fig. 5.9,15, 24)

Fig. 5 (a) shows the comparison of $^{137}$Cs deposition density in soil and sediment for some water bodies along its distance from FDNPP. It shows the location dependence of $^{137}$Cs deposition density in soil and sediment for some water bodies. The higher $^{137}$Cs deposition density was found in the water body area closer to FDNPP. However, there was no location dependence in the case of $^{137}$Cs soil to sediment ratio.

Fig. 5 (b) shows the comparison of $^{137}$Cs soil to sediment ratio for some water bodies along its sampling time. This shows that the ratio for the sample of longer sampling time from the accident is relatively smaller than that of the sampling time closer to the accident. The large variation among the data was most probably caused by the different characteristics of the catchment and water bodies, such as water resident time and particle size of the sediment.15) The characteristics probably governed the migration and accumulation of $^{137}$Cs. In the case of $^{137}$Cs deposition density in soil and sediment (Bq cm$^{-2}$), there was no time dependence.

3-4. Physicochemical property of the surface soil

Table 3 summarizes the physicochemical properties of the soil samples of the present study area. The measured soil samples are the surface layer of 0–2 cm depth. The value is a calculated mean value of samples and the error value is...
calculated based on the standard deviation (accidental error).

The bulk density, excluding the particle size >2 mm, ranged from 0.30 ± 0.10 to 0.46 ± 0.10 g cm⁻³. Transition zone (Area 3) surface soil has a higher bulk density (0.46 ± 0.10 g cm⁻³) than the forest zone surface soil, 0.30 ± 0.10 g cm⁻³ (Area 1) and 0.37 ± 0.20 g cm⁻³ (Area 2).

The soil pH of all area was acidic, ranged from 4.3 ± 0.1 to 4.7 ± 0.1. The pH of the transition zone is relatively higher (4.7 ± 0.1) than forest zone Area 1 (4.3 ± 0.1) and Area 2 (4.6 ± 0.3).

The CEC value of surface soil was relatively different between the transition zone 59 ± 8 cmol kg⁻¹ and forest zone Area 1 (99 ± 23 cmol kg⁻¹). However, the base saturation value of all samples was relatively low for all samples, ranged from 8.4 ± 3.7% to 12 ± 5% which means only about 8~12% of surface area was occupied by the major cation or there was still a large space of surface soil to hold additional cation.

Table 3 also shows a notable difference in organic matter content and soil fraction content <75 μm between the transition zone and forest zone. The soil of transition zone (Area 3) contains higher soil fraction <75 μm (about 66 ± 9%) and less organic matter (about 19 ± 4%) than forest soil that contains soil fraction <75 μm about 17 ± 7% (Area 1), 21 ± 3% (Area 2) and organic matter about 48 ± 18% (Area 1), 26 ± 11 (Area 2).

4. Discussion

4-1. Radiocesium horizontal distribution exhibiting the dry initial deposition origin and high retention by forest area

The results obtained in Fig. 2 showed that the air dose and 137Cs inventory distribution were to some extent heterogeneous. The air dose and 137Cs inventory in the higher elevation area (Area 1) were higher than the lower elevation area (Area 2 and Area 3). Moreover, in the lower elevation area (Area 3, transition area), there is a very large decrease in the air dose and 137Cs inventory compared to the higher areas (Area 1, Area 2).

The heterogeneous distribution with the tendency of higher inventory in higher elevation areas probably occurred by dry initial deposition process from the atmosphere. It has been discussed previously that the atmospheric wet initial deposition resulted in a more homogenous distribution of radiocesium than the dry initial deposition.\textsuperscript{31} Kawauchi area as the present...
study research area had received radioactive contamination through the dry initial deposition for about 60~70%. The higher radioactive contamination found in the higher elevation area was in line with some results of previous research that showed the elevation dependence of the radioactive contamination in dry deposition area, especially in the slope facing FDNPP. The obtained finding of horizontal distribution is not in line with some previous researches which showed the increase of radioactive contamination along with the decrease of elevation that probably corresponded to lateral indirect deposition or redistribution in slope area. Some previous researches show more radiocesium accumulation in the middle and bottom of the slope and accumulation in the water body in the steep.
4-2. Radiocesium migration based on $^{137}$Cs distribution and soil to sediment ratio

$^{137}$Cs horizontal and vertical distributions in soil catchment show the lesser $^{137}$Cs inventory in the soil layer of the transition zone than in forest soil, see Fig. 2 and Fig. 4. We used here the mass depth (kg · m$^{-1}$) instead of the depth, because the density of the soils is dependent on the area (which is shown in Table 3). This probably shows that the radiocesium in the transition zone was more depleted. And the radiocesium in the transition zone was probably more migrated to the water body than in the forest zone.

The $^{137}$Cs vertical profile of sediment in Fig. 4 shows some peaks in accumulation that might correspond to multiple times of radiocesium influx from the surrounding. Moreover, the vertical profile in sediment shows that the radiocesium was accumulated in the deeper layer of sediment and radiocesium inventory in the surface of sediment had a small contribution to the total $^{137}$Cs inventory. It might mean that, even though the catchment area, in particular forest catchment, still has relatively high radiocesium inventory, the amount of Cs influx onto sediment from surrounding catchment was decreasing sharply with time.

The low value of $^{137}$Cs soil to sediment ratio (0.53 ± 0.31), which means the higher radiocesium deposition in sediment than in soil catchment, showed the radiocesium accumulation in sediment, see Table 2. Furthermore, the $^{137}$Cs soil to sediment ratio of the present study was lower than some previous studies, suggesting that the soil to sediment ratio was time-dependent.

The $^{137}$Cs soil to sediment ratio of the present study was related to some previous studies, by neglecting the location and characteristics of the water body. The $^{137}$Cs soil to sediment ratio data of the present study and previous studies showed time dependency instead of location dependency, see Fig. 5(a), (b). Although there is a large variation of the ratio among the data which was probably caused by the different characteristics of the catchment and water bodies, such as water resident time and particle size of the sediment. It has been explained before that longer resident time of water body and fine particle in the sediment will promote the radiocesium retention and accumulation in the sediment. The higher radiocesium retention and accumulation in the sediment will lower the $^{137}$Cs soil to sediment ratio. The present ratio 0.53 ± 0.31 is relatively low, suggesting that radiocesium was retained for a long time in the sediment of Ogi reservoir.
the dissolved form, but later after the radiocesium interacted with the soil particles, the radiocesium was in the fixed form so that the radiocesium migration rate became slower. 30)

4-3. Physicochemical property of the forest surface soil and the inhibition of radiocesium migration from the catchment

The obtained vertical profile data show a deeper accumulation of $^{137}\text{Cs}$ in the sloped forest zone than in the transition zone (Fig. 4). This finding was in line with the previous research that showed deeper radiocesium accumulation in the forest than other undisturbed areas such as uncultivated cropland and grassland. 4) Fig. 4 also shows the very different depth profile between soil and sediment. The depth profile of soil probably showed the radiocesium inventory depletion that probably due to radiocesium migration and soil erosion. The sediment depth profile showed the radiocesium inventory accumulation along the time. This accumulation speed was faster than the speed of $^{137}\text{Cs}$ penetration in the soils.

Deeper radiocesium accumulation in the sloped forest zone than transition zone was probably one of the factors that could inhibit horizontal migration of particulate radiocesium into the water body, even in the very steep slope area. Besides the biophysical factor such as forest canopy protection, the deeper radiocesium accumulation in soil surface probably provided indirect protection from horizontal migration through surface water runoff mediating particulate cesium migration. The deeper the accumulation of radiocesium on the forest surface layer, the less reachable the radiocesium by surface water runoff.

Some physicochemical properties of surface soil such as bulk density, fine particle content, and organic matter content were probably the underlying factors that contributed to the radiocesium deeper accumulation in the forest zone. Table 3 shows the general tendency that the forest surface soil contained lower fine grain and bulk density and higher organic matter content than transition zone soil which probably promoted the radiocesium penetration depth. Previous research showed the positive correlation between $^{137}\text{Cs}$ retention in 0–1 cm layer and bulk density and negative correlation between $^{137}\text{Cs}$ retention in 0–1 cm layer and organic carbon content in clay. 4) The fast vertical migration of $^{137}\text{Cs}$ in the litter layer of the forest floor within one year after the accident was reported and the vertical migration becomes very slow once it reached the mineral layer. 11) However, previous research showed that $^{137}\text{Cs}$ retention in the litter layer of the forest floor can delay the radiocesium penetration onto the soil. 3)

Regarding the soil particle size dependence on radiocesium concentration, it has been widely known that finer particle size soil especially clay can adsorb and fix the radiocesium strongly. 9,31,32) However, the high content of organic matter could inhibit the sorption of radiocesium by the clay material. 11) The decomposition of litter in the surface soil of forest zone forms mobile soluble organic matter complex compound with cation including $^{137}\text{Cs}$. 4)

5. Conclusions

We showed the horizontal distribution of air dose and $^{137}\text{Cs}$ inventory in the steep slope catchment that was relatively heterogeneous and found a higher $^{137}\text{Cs}$ inventory in higher elevation and forest area. This probably corresponded to the atmospheric dry initial deposition-derived radiocesium and the high retention of radiocesium in the steep slope forest area. The migration of radiocesium from catchment to the reservoir was probably contributed more from the transition zone. The sediment vertical profile showed the accumulation of $^{137}\text{Cs}$ in the deeper layer of sediment. The low value of $^{137}\text{Cs}$ soil to sediment ratio provided the evidence of radiocesium accumulation in the sediment from the catchment. The physicochemical property of the forest surface soil could be a possible factor of high retention of the radioactive contaminants in the steep slope forest zone. Further research work should be done especially to analyze in more detail the robustness of $^{137}\text{Cs}$ soil to sediment ratio for investigation of the radiocesium migration from the catchment area.

Conflict of interest statement

There is no conflict of interest needed to be disclosed.

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