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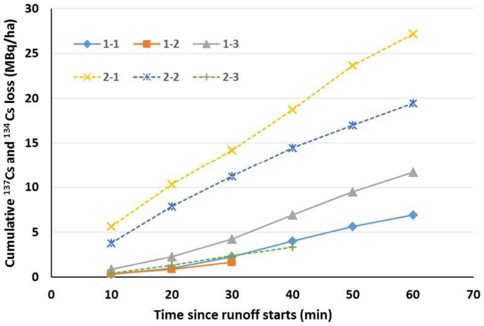

First detailed investigation of radioactive cesium export process from upland fields under simulated rainfall in Fukushima.

Environmental impact statement

Heavy rainfalls can cause severe runoff and soil erosion from agricultural fields in Fukushima. The eroded sediments carried high concentration of radioactive caesium that could pose a radioactivity risk to the organisms in the river system.

Export of radioactive cesium from agricultural fields under simulated rainfall in Fukushima.

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[1] The study was conducted during PT's visit to Tokyo University of Agriculture and Technology

ABSTRACT

In this study, we investigated the impact of rainfall on runoff, soil erosion and consequently on the discharge of radioactive cesium in agricultural fields in Fukushima prefecture using rainfall simulator. Simulated heavy rainfalls (50 mm/h) generated significant runoff and soil erosion. The average concentration of radioactive cesium (the sum of ^{134}Cs and ^{137}Cs) in the runoff sediments was ~3500 Bq/kg dry soil, more than double the concentrations measured in the field soils which should be considered in studies using ^{137}Cs loss to estimate long-term soil erosion. However, the estimated mass of cesium discharged through one runoff event was less than 2% of the cesium inventory in the field. This suggested that cesium discharge via soil erosion is not a significant factor in reducing the radioactivity of contaminated soils in Fukushima prefecture. However, the eroded sediment carrying radioactive cesium will deposit into the river systems and potentially pose a radioactivity risk for aquatic living organisms.

Keywords: radioactive cesium, rainfall simulation, suspended solid, erosion, sediment

1. INTRODUCTION

The nuclear accident at the Fukushima Daiichi nuclear power plant (FDNPP) following the massive earthquake and tsunami in March 2011 has released an enormous amount of radionuclides into the atmosphere corresponding to the highest level (Level 7) on the International Nuclear and Radiological Event Scale of the International Atomic Energy Agency (IAEA). Decay of some short-lived radionuclides such as ^{131}I (half-life = 8 days (d)), ^{129m}Te (34 d), and ^{136}Cs (13 d) and the remediation of contaminated soil has brought down the level of radiation in Fukushima prefecture. However, concern remains over the environmental radioactivity from relatively long-lived ^{134}Cs (2.07 years) and longer-lived ^{137}Cs (30 years), especially for agricultural products grown in the region. It is widely believed that people could be exposed to radioactivity through the consumption of ^{134}Cs and ^{137}Cs contaminated food[1] due to the transfer of radioactive cesium into the biomass of agricultural products.[2]

Many decontamination techniques have been employed in the contaminated zones in Fukushima to reduce the external exposure in different surface areas. In agricultural lands, current techniques include deep ploughing, topsoil removal and application of potassium fertilizer with the average cost of about 1,500 yen/m^2 in the Special Decontamination Area (area of high radioactive contamination where decontamination is implemented by the national government).[3] Most of the remediation efforts are focused on the Special Decontamination Area where the recommended threshold for topsoil removal is equal or higher than 5,000 Bq/kg (based on ^{137}Cs).[3] In other areas where the radioactive level is lower than the threshold, it would be too costly to apply soil removal techniques. It is important to implement/facilitate any measures to reduce and/or completely remove the radioactivity in these areas so that radioactivity could return to pre-accidental levels.

^{137}Cs is strongly adsorbed to soil particles, especially clay and organic matter[4] and thus can be carried out of the field with runoff sediment. Soil erosion carrying ^{137}Cs from agricultural lands could be considered a natural process to reduce the external dose of radioactivity in farmland, especially when at least half of the deposition of ^{134}Cs and ^{137}Cs from FDNPP occurred in the thin surface soil layer (0-4 cm)[5] and downward movements of ^{134}Cs and ^{137}Cs are restricted by strong adsorption behaviour.[6] Recent studies have indicated that ^{134}Cs and ^{137}Cs adsorbed to soil has been flushed by rainfall and snowmelt from the contaminated areas possibly to the surrounding decontaminated farmlands and to the river system in large quantities [7] under severe weather conditions such as typhoon.[8, 9] However, little detail on the relationship between the amount of radioactive cesium washed from contaminated farmlands and the rainfall amount is reported and there are very few plot measurements to quantify soil erosion and associated Cs export from cropland/paddies in this region.

In this study, we attempted to address this problem by i) investigating the level of soil erosion and radioactive cesium discharge from farmlands in Fukushima prefecture using simulated rainfalls and; ii) estimating the contribution of cesium discharge through runoff to the total reduction of radioactivity in Fukushima farmlands.

2. MATERIALS AND METHODS

2.1. Study site

This study was conducted in 2 upland fields in Nihonmatsu, Fukushima prefecture which are about 30 km from FDNPP (Fig. 1). The study sites are not within the evacuation zone so easy access was ensured during the study period. Based on the governmental air borne monitoring,

^{134}Cs and ^{137}Cs depositions around the study site were 60-100 and 100-300 kBq/m^2 respectively on 16 November, 2012.[10]

The fields were ploughed before the experiment to ensure homogenous soil conditions. There was no rainfall in the area between ploughing and the rainfall simulation. Rainfall data were retrieved from the Japan Meteorological Agency website for Nihonmatsu meteorological station. The mean annual precipitation of the area is 1199 mm with typhoon season from September to October.

At each site, three plots of 5 m^2 (1m x 5m) were prepared inside the well-plough field using plastic borders (Fig. S1). The borders were driven ≈10 cm into the soil and were supported with additional soil at the base. Runoff at the downhill side of the plots was collected using a stainless steel V-shape drain covering the full width of the plot. The slopes were measured and initial surface soil samples (0–5cm) were taken at each individual plot before the rainfall simulation occurred. The slope and soil properties of the sites are listed in Table 1.

Table 1. Soil properties and field characteristics of experimental sites.

	Site 1	**Site 2**
Slope (%)	8.4	8.2
Moisture content (vol/vol)	0.29	0.27
Organic matter content (%)	3.14	4.24
Bulk density (g/cm^3)	0.96	0.94
^{134}Cs radioactivity (Bq/kg dry soil)	484	450
^{137}Cs radioactivity (Bq/kg dry soil)	1109	1062
Soil type (USDA classification)	Sandy loam	Sandy clay loam
Sand (%)	72	66
Silt (%)	23	6
Clay (%)	5	29
pH	6.68	6.17
Total K (mg/g)	0.005	0.01

2.2. Rainfall simulation

A swing type rainfall simulator was used in this experiment. The silicone nozzles were mounted on a frame located approximately 1.6 m above the soil surface. To assess the rainfall intensity delivered by the simulator, a plastic sheet was used to cover the experimental plot to direct all simulated rainfall water into 10-L buckets and the time taken to fill the buckets was measured.

The performance of the rainfall simulator was assessed by rain drop measurements of the simulator after being calibrated at a rainfall intensity of 48.0 mm/hr. Drop sizes and rainfall kinetic energy were measured using a laser drop-sizing gauge (LDG) consisting of a laser transmitter and receiver (LX2-02; Keyence Corporation, Osaka, Japan), paired to an amplifier (LX2-V10; Keyence Corporation, Osaka, Japan) and an A/D Converter (AXP-AD02; Adtek System Science Co., Ltd., Yokohama, Japan) as described by Nanko et al. [11] Measurements were performed for five minutes at three points, respectively: 1.25, 2.5, and 3.75 m from the upstream edge on centre line of the plot.

The kinetic energy of natural rainfall in Japan was estimated from the equation with parametric values of Japan derived from van Dijk et al. [12] The kinetic energy estimation equation is expressed as:

$$KE = 23.7I\left(1 - 0.51e^{-0.019I}\right)$$

where KE is kinetic energy (J m^{-2} h^{-1}) and I is rainfall intensity (mm h^{-1}).

Field rainfall simulations were carried out in Site 1 in 15-16 November, 2013 and in Site 2 in 16-17 November 2013. Each simulation was designed to last for 60 min after the beginning of the runoff event and a rainfall intensity of 50 mm h^{-1} was achieved. The value of the rainfall intensity at 50mm h^{-1} was selected (as calibrated) – with a return period of approximately 15 years. Lower rainfall intensity in Plot 2-3 at Site 2 was applied partly due to the shortage of water supply. Lack of water also shortened the simulation time in Plot 1-2 at Site 1 (Table 2). During the simulation, all run-off water was collected and its volume was recorded. Two litres of run-off samples were taken every 10 min after run-off began. The samples were stored in plastic bottles and immediately transferred to the laboratory for analysis.

Table 2. Parameters of the rainfall simulations in different plots.

	Plot	Rainfall intensity (mm/h)	Rainfall amount (mm)	Runoff amount (mm)	Runoff coefficient (%)	Cumulative sediment loss (t/ha)
Site 1	1-1	50	55	26	47	1.41
	1-2*	50	23	5	22	0.42
	1-3	50	55	34	62	4.51
Site 2	2-1	50	55	42	76	7.18
	2-2	50	57	45	79	6.62
	2-3*	30	40	15	38	0.7

*Simulations during which water shortage occurred

2.3. Sample analysis

In the laboratory, run-off samples were filtered through a 1.1 µm glass microfiber filter paper (Whatman, GF/C) to separate the sediment phase from the liquid phase. Both soil and sediment samples were dried at 105 °C for two days. The sediment samples were regularly checked and stirred to ensure homogenous drying. Dry soil and sediment samples were grounded using a mortar and pestle, and preserved in tightly closed glass bottles prior to analysis.

About 5 g of dry sample was transferred to a plastic cup for measurement. The radioactivity of ^{134}Cs and ^{137}Cs in the samples was determined using gamma-ray spectroscopy. Gamma-ray emissions at energies of 604.7 keV (^{134}Cs) and 661.6 keV (^{137}Cs) were measured using a high-purity germanium coaxial detector system (Ortec, GEM20-70) coupled to a multi-channel analyzer (Ortec, DSPEC jr 2.0). The energy and efficiency calibrations for this detector were performed using standard and background samples. All activities were decay-corrected to the sampling date prior to statistical analyses. Particle size distribution of soil and sediment samples were analysed using a laser diffraction particle size analyser (SALD-2300, Shimadzu Co., Ltd., Kyoto, Japan).

Organic matter content of the samples was determined using loss-on-ignition (LOI) method.[13] Briefly, around 3 g of air-dried sample were placed in a ceramic crucible of the electric muffle furnace (Advantec. Ful230 FA) and heated to 105 ^{o}C for 12h. The sample was then weighed to measure mass of dry sample. After that the dry sample was heated to 400 ^{o}C overnight and weighed again. The value of organic matter content was calculated as the difference between the initial and final weights of the dry sample divided by the initial dry sample weight times 100%.

3. RESULTS AND DISCUSSION

3.1. Rainfall characteristics.

During the performance assessment of the rainfall simulator, a total of 12,364 rain drops were measured. The mean (± standard deviation) of the maximum drop diameter and mean median drop size (D_{50}) at three points were 2.93 (± 0.03) mm and 1.63 (± 0.34) mm respectively. The mean (± standard deviation) of rainfall intensity obtained from the LDG was 43.8 (± 11.1) mmh^{-1}. The mean (± standard deviation) of rainfall kinetic energy was 702 (± 171) Jm^{-2}h^{-1}.

The kinetic energy of the rainfall simulator was compared to that of natural rainfall in Japan. From the estimation, the kinetic energy produced by the rainfall simulator was equal to the natural rainfall with intensity from 31 to 47 mm h^{-1} which was considered acceptable for the experiment.

3.2. Runoff volume and soil erosion.

3.2.1. Runoff volume

Rainfall simulations were carried out at the beginning of the dry season so there was no significant rainfall before the experiment and soil moisture was considered lower than infiltration capacity.

Runoff started within less than 10 minutes after the rainfall was initiated (average of 7 minutes at Site 1 and 8 minutes at Site 2), equivalent to less than 10 mm of rainfall. Such a low threshold to generate runoff significantly increases the risk of runoff in this area. In comparison, a similar rainfall simulation experiment on Andisol soil (light clay) in Tokyo region required 30-50 minutes of rainfall with similar intensity (50 mm/h) to generate runoff (unpublished data).

Fig. 2 showed that the runoff rates were high at both sites but they were stronger at Site 2 than at Site 1. The runoff rate increased quickly and plateaued when it approached the rainfall rate (50 mm/h or 30 mm/h for plot 2-3). The runoff coefficients ranged up to a maximum of 62% in Site 1 (plot 1-3) while it reached 79% in Site 2 (plot 2-2) (Table 2).

3.2.2. Soil erosion

As shown in Fig. 3a, the concentrations of sediment in runoff water were relatively stable throughout the rainfall event except at the time when runoff started (first flush effect). On average, the sediment concentration measured in runoff water of Site 1 (sandy loam) was significantly lower than that of Site 2 (sandy clay loam) ($p<0.05$) but the effect should be taken into account together with the sediment flux (sediment loss) from those plots. The difference mostly caused by the lower sediment concentrations from plot 1-1 while sediment concentrations from plot 1-3 were similar to those from 2-1 and 2-2 (data from 1-2 and 2-3 were excluded as the simulation condition were different). It is suggested that the variation in field condition between the two Sites that caused such difference.

The rate of sediment loss in the experimental plots was shown in Fig. 3b. There was significant difference between the erosion rates in two sites which was attributed mainly to the difference in runoff rates between them (Fig. 2). The first flux effect also contributed considerably to the difference in sediment loss as occurred in plot 2-1 and 2-2 compared to other plots (Fig. 3b).

The maximum cumulative sediment loss at Site 2 was 7.18 t/ha with 42 mm of runoff amount while the corresponding value at Site 1 was 4.51 t/ha with 34 mm of runoff amount (Table 2). These values are comparable with the erosion rates reported for similar rainfall events [14] and fall within the range of the estimated annual soil erosion rates of 3.54 – 34 t/ha/year in Japan.[15] These estimations are reasonable since it came from a single rainfall event. Several or longer heavy rains will probably increase the erosion rate.

Rainfall events of shorter duration (plot 1-2) or of lower intensity (plot 2-3) expectedly produced a lower loss because they produced less runoff.

It should be noted that the runoff rate measured in this study was representative of the worst case scenario since the fields at both sites were ploughed only a few days before the experiment and had no vegetation cover available in all the plots. The bare and well ploughed sandy loam/sandy clay loam soils were thus highly prone to runoff during high intensity rainfall events.

3.3. The discharge of cesium through runoff/erosion.

Cesium (^{134}Cs and ^{137}Cs) was discharged from the field to streams mainly through sediment carried by runoff water because they are strongly adsorbed by organic matter or fine soil particles. [16] According to a previous study on the distribution of cesium in surrounding water streams after the accident at FDNPP, the concentrations of cesium in the dissolved phase were considered to have little impact on the total mass of cesium discharge and thus were not measured in this study. [17]

The total concentrations of ^{134}Cs and ^{137}Cs in runoff sediments were presented in Fig. 4. The concentrations of ^{134}Cs and ^{137}Cs in runoff sediment were higher (up to three folds) than the average values measured in the plots before the experiment (Table 1). It could be attributed to the smaller particle size of the runoff sediment as well as the higher concentration of organic matter in the runoff sediments than those of the field soils (at both sites) as reported earlier. [16, 18] In fact we observed a smaller average particle size of sediment samples than that of the initial field soil samples (Fig. S2). We also found that there was a correlation between the concentrations of cesium and the organic matter content of the sediments (Fig. 5) which was in agreement with the results of previous studies. [16, 19]

Cumulative losses of cesium (MBq/ha) during runoff were calculated and presented in Fig. 6. As most cesium was carried by runoff sediment, the profile of cesium losses was very similar to those of sediment as presented in Fig. 3b. The extent of cumulative losses of cesium from the experiment plots was also similar to the extent of sediment losses in those plots (Fig. 3b and Fig. 6).

Runoff volume and sediment concentrations are the two main factors that control the discharge of cesium from the experimental plots. The findings of this study indicated that compared to a large rainfall simulated in this study (~60 mm at 50 mm/h), shorter or smaller rainfalls would result in a disproportionally smaller amount of cesium discharged to the environment (see Fig. 6). Major rainfall events such as storms will certainly cause a surge in the amount of cesium discharged to the environment as reported previously. [20]

3.4. Implication for the field inventory of radioactive cesium and the environmental fate and transport of cesium

3.4.1. Effect of runoff on the inventory of radioactive cesium

The ratio of ^{134}Cs and ^{137}Cs measured in this study (Table 1) was expectedly in agreement with the estimated ratio using natural decay rate (with the initial deposition ratio of approximately 1:1). [21] This ratio remained the same in the runoff sediment samples, i.e. that the distribution of cesium in samples was relatively homogeneous. According to the measurements in this study, the radioactivity of cesium has decreased by about 33% since the nuclear incident with about 30% of the decrease due to the decay of ^{134}Cs.

During rainfall events, it was found that the average radioactivity of cesium in runoff sediment (Bq/kg) was more than double that of the experimental field soil. The higher radioactivity in runoff compared with field background means that the discharge of cesium radioactivity through runoff may act as a way of land rehabilitation from cesium contamination. However, compared to the total radioactivity of the field the maximum amount of radioactive cesium discharged through runoff in this study was about 2% assuming the depth of the contaminated soil layer of 10 cm due to the cultivation practice.[9] Smaller and shorter rainfalls would produce considerably lower discharge amounts. Therefore, runoff is not considered an effective way to reduce the inventory of radioactive cesium in the field (field decontamination). This finding is in agreement with data from previous study where no evidence of change in the total inventory of ^{137}Cs could be found between the pre- and post-rainy season of 2011 in different land uses in Fukushima prefecture.[22] The same study also suggested that rainfall events including intense rainfalls did not significantly modify the profile of cesium with depth in the soil.[22] Our estimation indicated that natural decay of radioactive cesium, especially ^{134}Cs, will have larger impact on the field radioactivity than rainfall runoff events (33% reduction by natural decay in 2.5 years against ~2% by one large runoff event of 15-year return period).

It should be noted that the phenomenon that eroded soils contain higher concentrations of ^{137}Cs than the bulk field soil would be considered in future studies where ^{137}Cs is used as tracer to assess soil erosion. [23]

3.4.2. Environmental fate and transport of cesium discharged from fields.

Soil erosion and subsequent sediment transport can play a major role in the dispersion of the radioactive cesium in the natural environment because this element is strongly adsorbed to the fine particle and the organic matter in the field. [7] This study confirmed that the majority of cesium mass transported out of the field was through sediment in runoff water. The sediment eroded from fields loaded with radioactive cesium will be transported to the river stream and deposited behind the dams before being released into the ocean.[15, 17, 24, 25] Peak levels of cesium radioactivity were observed in river water during the heavy rainfall events with almost 100% contribution from particulate adsorbed cesium.[20] Ueda et al. reported that the estimated discharge of radioactive cesium from river catchments in Fukushima prefecture in 2011 was about 0.3-0.5% of the total cesium deposited on the catchments.[25] This reported value was in agreement with the estimation from this study (~2%) considering that our data was based on erosion mass from bare fields not from multiple land-use catchments (consisting of e.g. forestry and meadow) in the previous study. [25]

Although the discharge of radioactive cesium by soil erosion has no effect on the field inventory of cesium, it may have significant consequences on water quality with high concentrations of cesium (especially ^{137}Cs due to its long half-life) in runoff during extreme rainfall events [15]. Our experiments also confirmed that soil erosion was a way of concentrating radioactive cesium in sediment, making the radioactivity of sediments found in rivers higher than those measured in nearby soil. [24] The flux of highly contaminated sediment along rivers, especially during heavy rainfall events, will shift the load of radioactive soil from upstream regions to coastal plains and eventually to the ocean.[8, 20] The peak load of highly radioactive sediment in the rivers and coastal areas due to heavy rainfalls should be considered carefully by local authorities to manage fishing and recreational activities in such areas since contamination of sediment can also propagate to living organisms via changes in food and habitat. [8, 9]

4. CONCLUSIONS

This study evaluated the impact of heavy rainfall to the runoff of radioactive cesium using rainfall simulators. In the studied fields, a simulated heavy rainfall of 60 mm can cause significant runoff with high runoff coefficients of up to 0.71. Results of sediment and radioactive cesium analysis for runoff samples in this study can lead to the following conclusions:

- Runoff caused by heavy rainfall can discharge large amount of radioactive cesium from the field to the river system. In such a runoff event, almost all radioactive cesium was transported out of the field via sediment.
- Concentrations of cesium in runoff sediment were several times higher than in the field soil due to their higher organic matter content and smaller particle sizes.

- Soil erosion does not significantly change the field inventory of cesium in the field but it may contribute significantly to the contamination of radioactive cesium in the river system.

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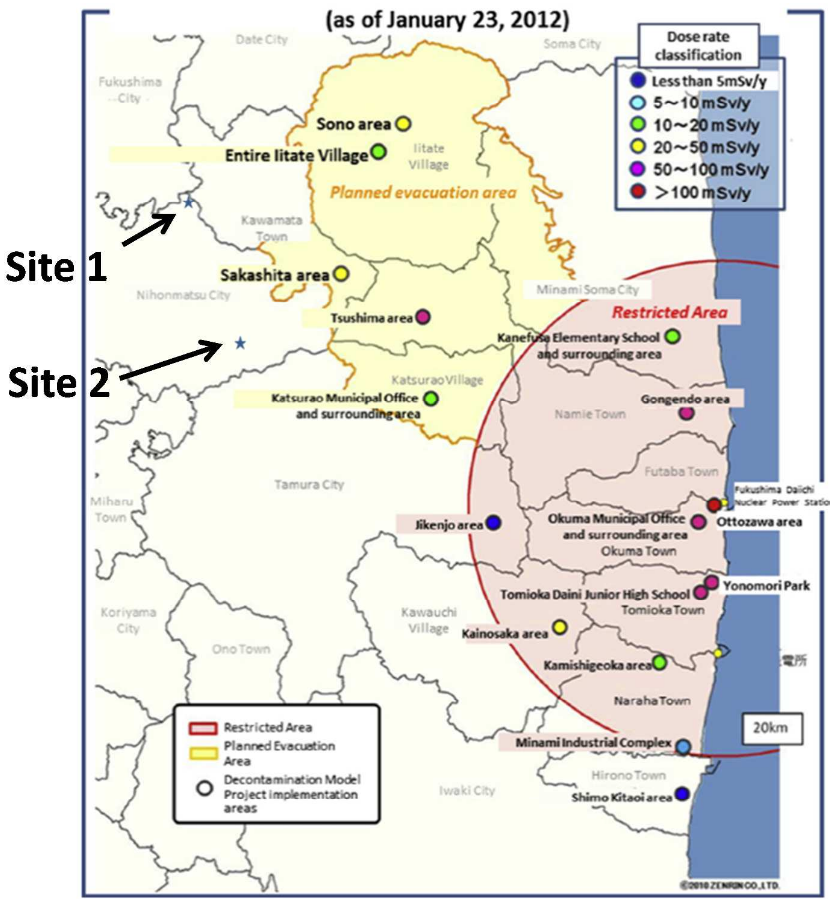


Figure 1. Location of the experimental sites in the Fukushima Prefecture (adapted from Hardie and McKinley, 2014).

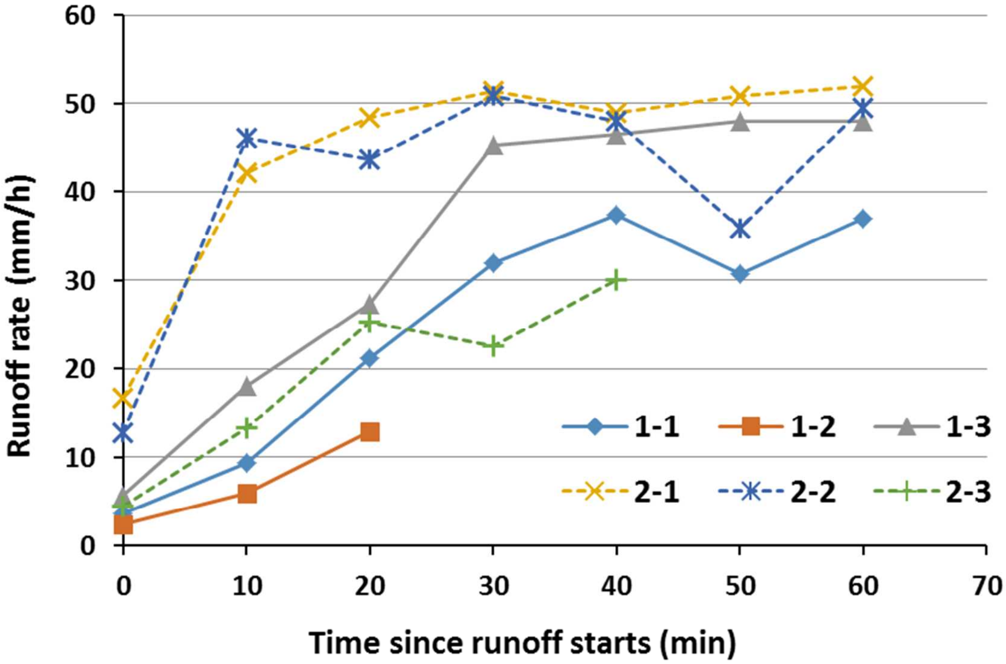
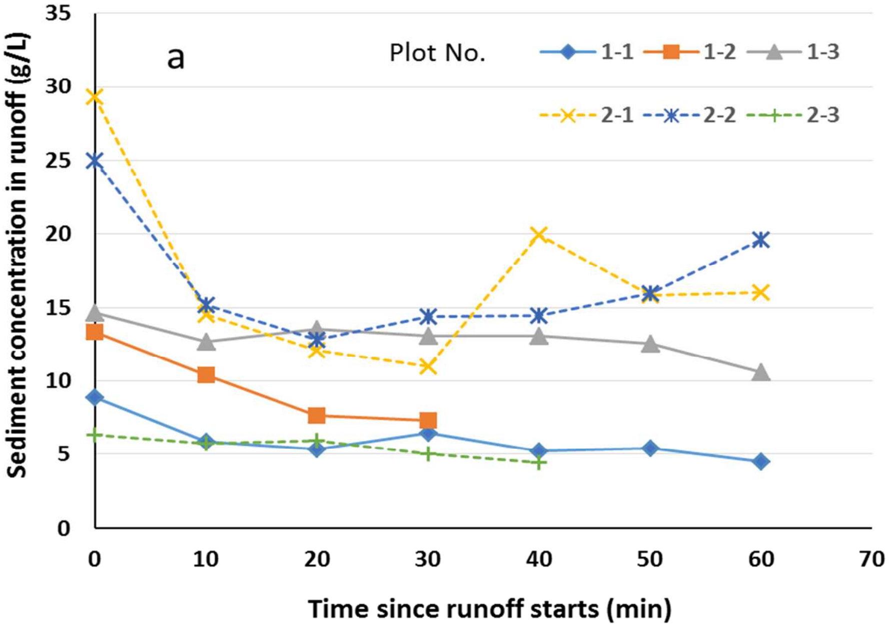


Figure 2. Runoff profile during the rainfall simulation in Site 1 (solid lines) and Site 2 (dashed lines).



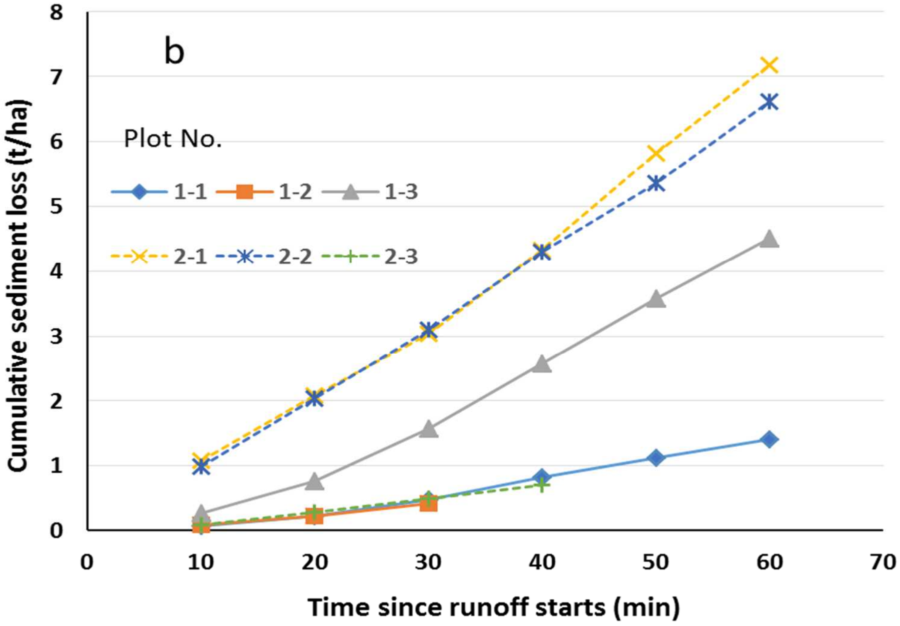
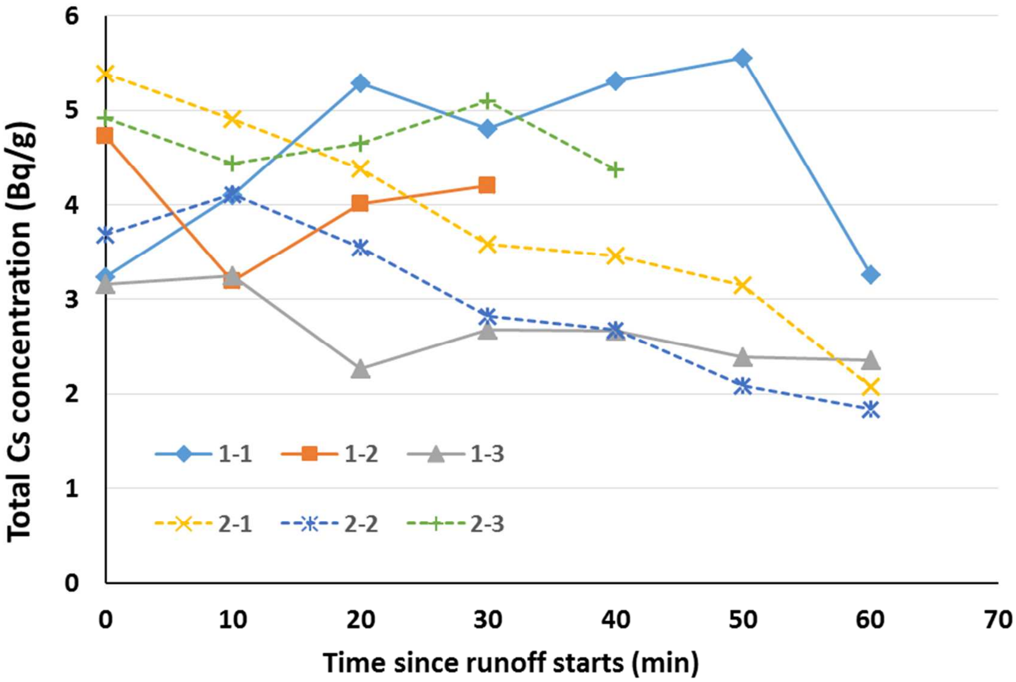


Figure 3. Concentration (a) and cumulative loss (b) of sediment through runoff water during the experiments.



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390 **Figure 4**. Total concentration of ^{134}Cs and ^{137}Cs in sediment samples during the runoff events.

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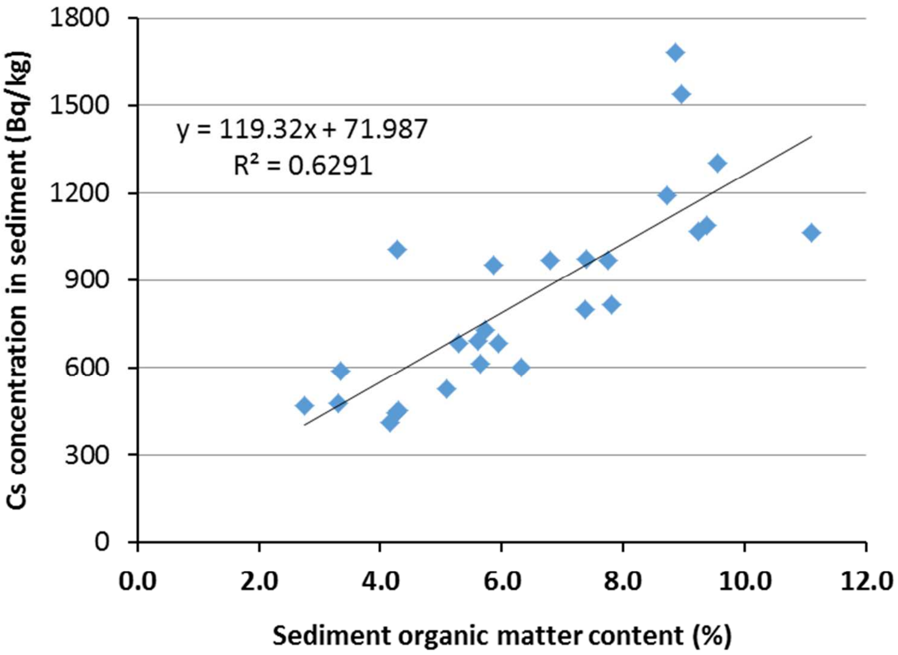

Figure 5. Correlation between Cs concentration and organic matter content of sediment samples in runoff water.

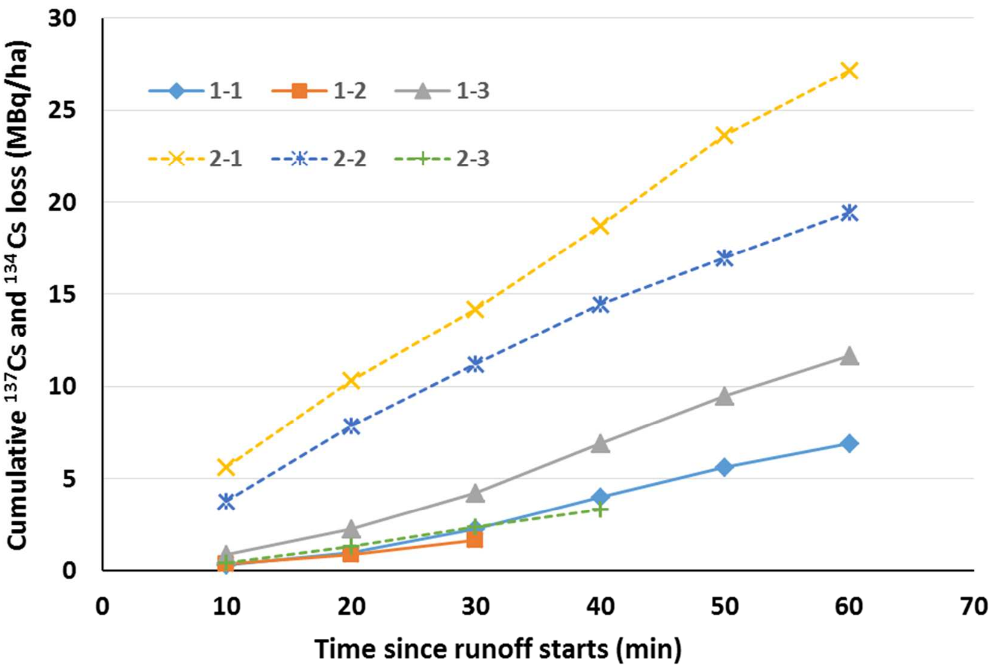

Figure 6. Cumulative loss of radioactive caesium (^{134}Cs and ^{137}Cs) during the runoff events.